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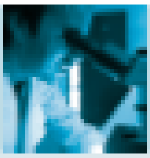

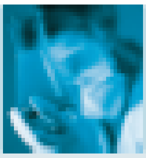

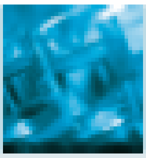

Volume 7

Membrane Technologies for Wastewater Treatment and Reuse

Proceedings of 2nd IWA National Young Water Professionals Conference, 4–6 June 2007, Berlin, Germany

Boris Lesjean (ed.)

Under patronage of: IWA International Water Association

Conference sponsors: VEOLIA WATER Berliner Wasserbetriebe KOCH A3

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Volume 7

CONFERENCE PROCEEDINGS

2nd IWA National Young Water Professionals Conference, Germany

“Membrane Technologies for Wastewater Treatment and Reuse”
4-5 June 2007, Berlin (Germany)

Under patronage of:

International Water Association

Conference sponsors:

VEOLIA WATER

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2^{nd} IWA National Young Water Professionals Conference, Germany
"Membrane Technologies for Wastewater Treatment and Reuse"
4-5 June, Berlin (Germany)

Berlin 2007

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About NYWPC

The Berlin Centre of Competence for Water organised, together with the International Water Association, the 2nd National Young Water Professionals Conference in Germany.

This event was held on 4-5 June 2007 in Berlin and was following up on the first NYWP conference organised in Aachen in October 2005. It provided a forum for young researchers and professionals working in the membrane sector of the wastewater industry to present their work and meet their peers. The conference consisted on formal presentation of papers and posters, and an exchange with water industry professionals providing information on water career opportunities in Germany.

This event was an initiative of "MBR-Network", the European cluster on the membrane bioreactor technology, gathering about 50 European and international companies and institutions within the four FP6 projects Amedeus, Eurombra, MBR-Train and Puratreat (more info at www.mbr-network.eu).

The technical program of the conference consisted mainly of contributions from German and international young water professionals including students, recent graduates and other professionals under the age of 35.

This book contains most of the papers and posters which were presented at the Conference. Wishing you a pleasant lecture,

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Conference Program

MONDAY 4 JUNE 2007 – IWA NYWP CONFERENCE DAY 1

Session I – Welcome address and key notes

Chairman: L. Pawlowski
(Berlin Centre of Competence for Water, Germany)

Welcome addresses
L. Pawlowski
(Director of Berlin Centre of Competence for Water, Germany)

I. Strube
(Ruhrverband, representative of IWA Germany)

Prof. R. Ben-Aim
(INSA Toulouse, Chairman of IWA Membrane Technology Specialist Group)

The young professionals programme of the International Water Association
Dr. T. Wintgens
(*RWTH Aachen University, representative of IWA Young Water Professionals in Germany*)

Overview of membrane business for wastewater and reuse applications in Europe
Dr. J.C. Schrotter
(*Anjou Recherche – Veolia Water, France*)

Future of membrane technologies in worldwide sanitation
Dr. P. Côté
(IWA Specialist Group on Membrane Technologies)

Session II – MBR process Optimisation and Wastewater Reuse

Chairman: Prof. M. Kraume
(TU Berlin, Germany)

TUESDAY 5 JUNE 2007 – IWA NYWP CONFERENCE DAY 1

Session VI – Career Development and Awards

Chairman: B. Lesjean
(Berlin Centre of Competence for Water, Germany)

WEDNESDAY 6 JUNE 2007 – TECHNICAL WORKSHOP
‘PERFORMANCES AND ECONOMICS OF MEMBRANE-BASED CONCEPTS FOR DECENTRALISED WASTEWATER TREATMENT’

(The corresponding articles for the presentations of this technical workshop are not included in this book, but they are available on www.kompetenz-wasser.de (Project ENREM))

ENREM demonstration project: decentral wastewater scheme with low pressure sewer and advanced biological nutrients removal in MBR

Chairman: L. Pawlowski
(Berlin Centre of Competence for Water, Germany)

Welcome Address
L. Pawlowski
(director of Berlin Centre of Competence for Water, Germany)

Context of the ENREM project
R. Gnirss
(Berliner Wasserbetriebe, Germany)

Design parameters and construction of the ENREM demonstration plant
B. Lesjean
(*Berlin Centre of Competence for Water, Germany*)

Operational issues and process optimisation of containerised MBR plant
C. Lüdicke
(Berliner Wasserbetriebe, Germany)

Results of process for advanced biological nutrients removal in MBR
M. Vocks
(*Berlin Centre of Competence for Water, Germany*)

Evaluation of concept and outlook
R. Gnirss
(Berliner Wasserbetriebe, Germany)

Four German projects of membrane-based processes for decentral wastewater treatment

Chairman: B. Lesjean
(Berlin Centre of Competence for Water, Germany)

Semi-decentralized wastewater management with membrane technology and large-scale operation experiences
T. Zech, W. Sternad, W. Trösch
(Fraunhofer Institute for Interfacial Engineering and Biotechnology, Stuttgart, Germany)

LAGOON-MEMB – a trend-setting concept for economical wastewater handling in rural area
H. D. Janke, A. Ante, R. Hasselbach
(*Gesellschaft für umweltkompatible Prozeßtechnik mbH i.L., Saarbrücken, Germany*)

Innovative decentralised wastewater treatment concepts

Dr. M. Wade, Dr. G. Schories

(*TTZ-Bremerhaven, Germany*)

Overview of domestic MBR plants for decentralised wastewater treatment in Germany

Dr. E. Dorgeloh, A. Kaiser

(PIA / RWTH-Aachen, Germany)

Poster Session

Membrane bioreactor technology for agricultural reuse of municipal wastewater: a comparative study

G. Guglielmi*, D. Chiarani*, G. Andreottola* and G. Ziglio*

* Dipartimento di Ingegneria Civile e Ambientale, Università degli Studi di Trento, Via Mesiano, 77, 38050 Trento, Italy
(E-mail: giuseppe.guglielmi@ing.unitn.it, daniele.chiarani@ing.unitn.it, gianni.andreottola@ing.unitn.it, giuliano.ziglio@ing.unitn.it)

Abstract A comparative experimentation was carried out within the research project AGRESTE, aimed to assess the reliability of membrane bioreactor technology for agricultural reuse of treated wastewater. The MBR optimisation was carried out under steady-state biological process conditions, with a special focus on membrane scouring aeration supply. The tests showed a steady permeability during the whole duration of the research; moreover, short-term critical flux tests permitted to evaluate the best hydrodynamic conditions in terms of energy saving, which was equal to 0.5 $Nm^3\ m^{-2}\ h^{-1}$ under 50% intermittent aeration. Effluents characteristics were routinely (weekly or bi-weekly) monitored; both processes guaranteed the required quality standards proposed by the Italian Law D.Lgs. 185/03 for wastewater reuse. Although, microbiological parameter were constantly lower than the Law limits for Escherichia Coli and Salmonella for both treatment schemes, the MBR technology resulted more reliable than the conventional tertiary process concerning parasites and protozoa removal.

Keywords municipal wastewater; agricultural reuse; MBR; air-sparging; sand filtration; UV disinfection.

INTRODUCTION

Membrane processes are currently considered among the most promising and complete solutions for wastewater reuse. As recently summarised by Wintgens *et al.* (2005), applications involving and integrating membrane technologies include aquifer recharge for tackling seawater intrusion (Flemish coast, Belgium), indirect potable reuse (NEWater Project in Singapore), direct potable reuse (Windhoek, Namibia) and a large number of dual water systems in small communities residential areas and industrial processes. Membrane bioreactors, coupling conventional suspended growth biological processes with either micro- or ultra-filtration, nowadays represent an optimal solution for retrofitting and upgrading existing wastewater treatment plants, also thanks to the excellent effluent quality which is often suitable for wastewater reclamation. MBRs have been proven to achieve good removal efficiency for microbiological parameters (Ueda and Noran, 2000), organic and inorganic micro-pollutants (*inter alia* Joss *et al.*, 2004; Innocenti *et al.*, 2002). Moreover the high compactness and low footprint requirement make such technology playing a key-role in the so-called “soft path” to water management, which is based on small and de-centralised systems rather than large centralised treatment facilities (Gleick, 2003).

The water deficit of a given country is usually expressed by the water stress index (WSI), calculated on a yearly basis as ratio between the total water withdrawal and the total water renewable resources. A WSI value lower than 10% indicates a non-stressed condition while WSI higher than 20% implies the need for improved water management in order to avoid social conflicts. In the range between 10% and 20%, water availability is commonly assumed to set some constraints for social and economical development. As most Southern Europe and Mediterranean countries, Italy suffers a gradual increase of the water stress index that is typically thought to involve the coastal semi-arid regions of South; this has been widely discussed in recent research papers dealing with wastewater reuse projects carried out in South of Italy (Pollice *et al.*, 2004; Lopez *et al.*, 2006).

However, the drought event of 2003 showed that water scarceness can affect even regions which are typically assumed to be rich in availability of water resources; this leaded to the emergency decree D.Lgs. 185/03 for wastewater treatment and reclamation, issued in June 2003.

The paper shows results obtained during the last year of a three years research project named AGRESTE (AGricultural REuse of Secondary post-Treated Effluents), which was started-up in 2003. The project was financially supported by the Autonomous Province of Trento and was aimed to investigate the reliability of two different technologies in wastewater reclamation for apple orchards irrigation. Different partners were involved, including the Department of Civil and Environmental Engineering of the University of Trento (DICA-UNITN), the Agricultural Research Institute of San Michele all'Adige (IASMA), the Public Health Laboratory (APSS-LIESP) and the Provincial Office for Wastewater Treatment (PAT-SOIS). The performances of a large pilot scale membrane bioreactor and of a conventional tertiary treatment systems were compared over the whole irrigation period (spring-autumn 2005) in term of removal efficiencies for macro- and micro-pollutants and for the microbiological parameters mentioned by D.Lgs. 185/03. Furthermore, aeration for membrane fouling mitigation was optimised, in order to identify the most suitable membrane operation for full scale installations working under similar conditions. Particularly, the critical flux concept proposed by Field *et al.* (1995) was used to investigate the impact of different aeration rates on the membrane performances.

MATERIALS AND METHODS

Experimental set-up

A 1000 m^2 area was divided into six lisymeters, two for each kind of water source considered (MBR, tertiary effluent and well water). Drip irrigation and sprinkle irrigation were tested in order to investigate the impact of water distribution systems and water source on the plants growth and the soil characteristics (data not shown in this paper). Both the membrane bioreactor and the conventional tertiary process were run continuously during the whole experimental period irrigation periods; no storage tank were used and the produced effluents were directly pumped in the pipeline to the specific lisymeters.

The large pilot scale MBR consisted of a biological process tank, with an anoxic compartment (2.8 m^3) and a oxidation chamber (5.1 m^3), in which a three-modules cassette of the Zenon hollow fibre membrane is immersed (see flow-scheme in Figure 1.). The nominal pore size of the PVDF membrane (ZW 500c) is 0.04 µm, and the overall membrane surface are is 69.6 m^2.

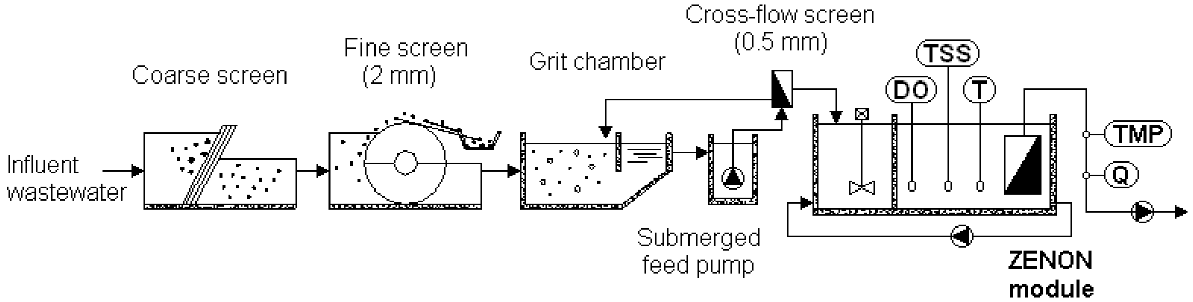


Figure 1 Scheme of the pilot scale MBR and the up-stream preliminary treatment units

Feed-water was pumped after the grit chamber and passed through an additional cross-flow fine-screen (500 µm) before entering the biological process unit; its composition is shown in Table 1. The system was entirely controlled and operated by a PLC, with on-line acquisition of both hydraulic (permeate flow-rate, TMP, level) and biological (dissolved oxygen DO, MLSS concentration) parameters. Two distinct air supply sources were installed, one for the biological process requirement and another for the membrane scouring. The DO concentration in the nitrification tank was regulated at the set-point value of 2 g m^{-3}, by a frequency converter coupled with the blower for the biological process. During the whole experimentation, sludge wasting was carried out in order to keep a constant suspended solids concentration of ~ 12 kgTSS m^{-3}; this resulted in a sludge age ranging between 12 and 15 days. Recycle ratio r ($Q_{recycle}/Q_{permeate}$) from the aerated to the anoxic compartment was tuned between at 3 and 5.

Table 1 Characteristics of influent wastewater for the pilot scale MBR

Parameter	**Mean value ± σ (g m^{-3})**	**Max (g m^{-3})**	**Min (g m^{-3})**
COD	777 ± 329	1654	358
Soluble COD	75 ± 22	120	35
TKN	69.9 ± 23.5	126	39.8
N-NH_4	28.7 ± 6.0	37.5	13.8
Total P	16.9 ± 7.6	35.2	7.3
TSS	598 ± 303	1416	211
VSS	427 ± 195	854	199

COD fractionation carried out by respirometric tests showed the following composition:

- soluble biodegradable COD: 9.1%
- soluble un-biodegradable COD: 2.0%
- particulate biodegradable COD: 61.8%
- particulate un-biodegradable COD: 27.0%

Membrane aeration was supplied intermittently (10 s ON/10 s OFF) in a range of gross specific aeration demand SAD_m between 0.5 and 1.0 Nm^3 m^{-2} h^{-1}. Permeate suction was applied according to a relaxation cycle with 9 minutes of extraction and 1 minute of pause; no cyclic backwash with permeate was operated but once every month a weak maintenance cleaning was performed with a 100 g m^{-3} sodium hypochlorite solution.

The conventional activated sludge wastewater treatment plant (CAS-WWTP) of Pietramurata has a 5000 PE capability and its biological process compartment is configured according to the modified Edzack-Luttinger scheme for nitrogen removal, with an anoxic tank of 400 m^3 and an oxidation/nitrification tank of 600 m^3; during the experimental activity, the plant was operated at a steady sludge age of about 23-25 days. Under normal operation, effluent from the secondary settling tank is filtered on surface filtration units and, after chlorine disinfection, discharged to the surface water body. During the experimental period, a centrifugal pump was immersed in between the sedimentation and the filtration compartment, in order to feed the pilot unit for wastewater reclamation; the composition of the secondary effluents during the experimental period is summarised in Table 2.

Table 2 Characterisitcs of the secondary effluent fed to the sand/anthracite filtration unit

Parameter	**Mean value ± σ (g m^{-3})**	**Max (g m^{-3})**	**Min (g m^{-3})**
COD	23 ± 6	33	15
Soluble COD	10 ± 4	15	4
BOD$_5$	295 ± 141	745	170
TKN	1.8 ± 1.3	6.3	0.4
N-NH$_4$	0.3 ± 0.2	1.6	0.1
Total N	16.8 ± 3.9	29.7	11.1
Total P	0.7 ± 0.4	2.7	0.3
TSS	10 ± 7	28	1
VSS	7.1 ± 4.9	19.8	0.7

The traditional tertiary treatment pilot plant consisted a pressurised two-stages sand/anthracite filter coupled with UV disinfection. The flow diagram of the filtration unit (Culligan OFSY-WGR 20) is shown in Figure 2.; two columns worked in series, in both cases with a down-stream flow. Low density granular anthracite (ø ~ 2 mm) and sand (ø ~ 0.8 mm) were used to fill the first column whereas smaller granulometry sand (ø ~ 0.6 ÷ 0.8 mm) was used to fill the second column. Before entering the first stage, a on-line addition of commercial aluminium poly-chloride Culligan PF60 (2% v/v) was operated in order to promote coagulation/flocculation of colloidal substances. Removal of smallest diameter particles was then enhanced by dosing commercial poly-acrilammide Culligan PF81 (1% v/v) up-stream the second filtration stage. The influent flow-rate was varied between 1 and 2.5 m^3 h^{-1}, corresponding to an hydraulic cross-sectional rate of about 5-15 m h^{-1} in the filtration columns; once every day, a 30 minutes backwash was automatically carried out with feed-water and with on-line addition of 12% w/w solution of NaOCl, in order to get an active chlorine concentration of 100 mg L^{-1} entering the columns.

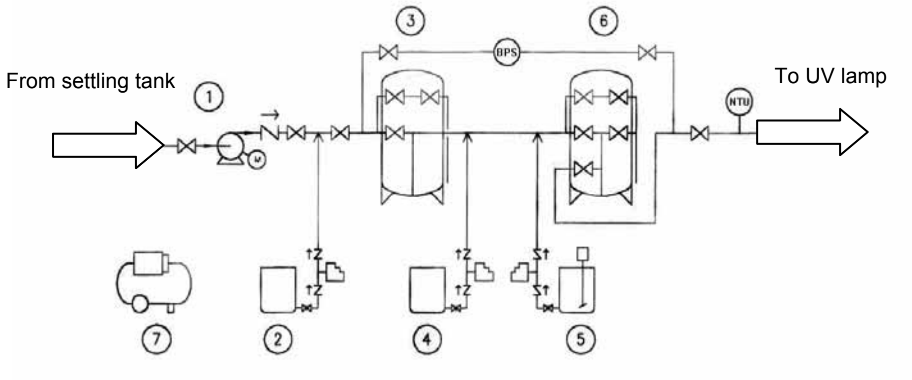


Figure 2 Schematic of the two-stages sand filter Culligan OFSY-WR 20 (1: feed pump; 2: storage tank for aluminium poly-chloride, PF60; 3: 1st filtration stage; 4: storage tank for NaOCl; 5: stirred storage tank for poly-acrilammide PF81; 6: 2nd filtration stage; 7: air compressor for pneumatic valves)

Filtered effluent was disinfected with UV-C rays through a low pressure-low intensity mercury lamp (Prominent Dulcodes, schematic in Figure 3.) generating a monochromatic radiation ($\lambda = 254$ nm). The lamp is placed in a stainless steel chamber (ø = 0.09 m), parallel to the effluent flow. Radiation intensity depends on the water transmittance which, on its own, is a function of water characteristics; in our case, the intensity was 4 mW cm^{-2} on average and, given the exposure time of 7-18 s; therefore, UV dose was ranging between 28 and 72 mJ cm^{-2}.

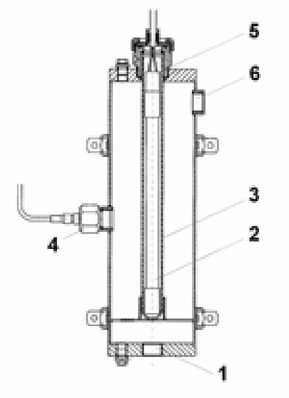


Figure 3 Longitudinal section of the UV disinfection chamber 1: inlet; 2: UV lamp; 3: quartz chamber; 4: UV-C probe; 5: electrical connection; 6: outlet

Analytical methods

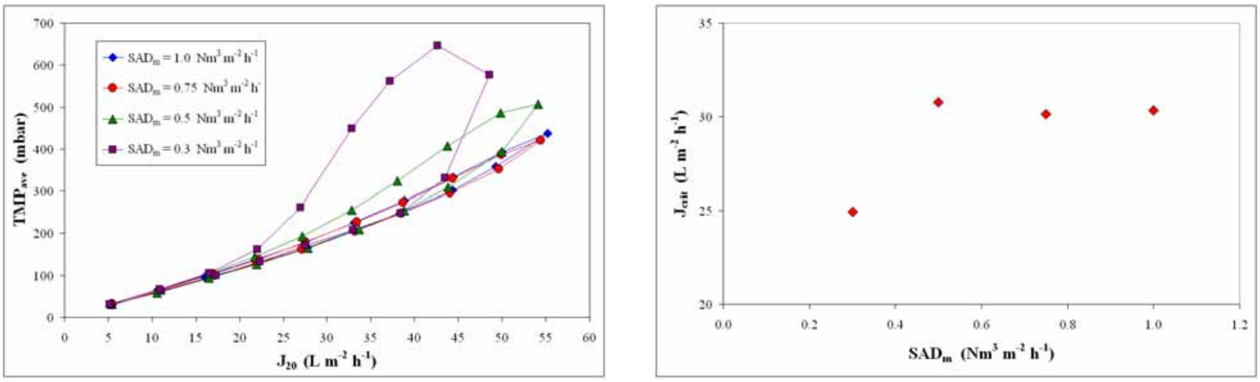
APHA Standard Methods were used for the analytical determination of most important macro-pollutants: COD, nitrogen forms, suspended solid and phosphorus were routinely analysed twice a week at DICA-UNITN and weekly at PAT-SOIS. Microbiological analyses were carried out by PAT-SOIS laboratory to detect *Total Coli*, *Escherichia Coli* and *Faecal Streptococci*. *Salmonella* bacterium, *Campylobacter bacterium*, protozoa as *Giardia* and *Cryptosporidium* and parasites as *Heminths* (eggs and larvae) were weekly searched in the effluents at the APSS-LIESP laboratory; sample volume for these analysis were 5 L and 100 mL for *Salmonella* and *Campylobacter*, 3 L for *Helminths* and 50 L for protozoa species. Therefore, the microbiological analyses carried out were more intense than required in the D.Lgs. 185/03, which imposes only monitoring of *Escherichia Coli* and *Salmonella*. In particular, the limit value for *E. Coli* is 10 CFU/100 mL in the 80% of samples and 100 CFU/100 mL as maximum value which, when exceeded, obliges to stop the irrigation practice; concerning *Salmonella*, the national regulation states that it must be absent in all samples.

RESULTS AND DISCUSSION

MBR performances

During the whole experimentation, the MBR was operated under sub-critical flux conditions, tuning permeate flux between 10 and 20 L m^{-2} h^{-1} (values normalised at 20°C). Under such conditions, the TMP value ranged between 70 mbar and 150 mbar, the normalised permeability being consequently ~ 140 L m^{-2} h^{-1} bar^{-1}. The residual fouling, calculated according to Howell *et al.* (2004), varied as a function of flux between 0.002 and 0.011 mbar min^{-1}, regardless with the SAD_m applied.

Critical flux tests were carried out under different aeration rates in order to investigate possible strategies for fouling mitigation. Flux was gradually step-increased up to a maximum value of about 55 L m^{-2} h^{-1}; step duration was 15 minutes while step height was steadily kept at 5 L m^{-2} h^{-1}. The criticality threshold was evaluated by TMP-derived parameters, including the fouling rate dTMP/dt during each step and the average permeability; more in detail, the critical flux J_{crit} was calculated as the average between the highest flux at which membrane permeability was higher than 90% of first step permeability and the next one. Data elaboration indicates that, under the applied biological process conditions, J_{crit} ranged between 25 and 32 L m^{-2} h^{-1}. Interestingly, Figure 4.(graph B) shows that it was possible to find out an optimal gross aeration rate (0.5 Nm^3 m^{-2} h^{-1}) above which no further increase of the critical flux was achieved; such result agrees with other study carried out on bench scale MBRs (Bouhabila *et al.*, 1998). On the other hand, graph A suggests also that the aeration can significantly affect the amplitude of the hysteresis loop in the super-critical region, which indicates the membrane capability to restore its pre-critical performances in a short time; from this point of view, the optimal aeration rate resulted to be 0.75 $Nm^3\ m^{-2}\ h^{-1}$, whose effect was almost identical to the 1 $Nm^3\ m^{-2}\ h^{-1}$ test. Being membrane aeration the most significant cost item in MBR operation, such results can be usefully adopted as operating values for full scale MBRs working under similar biological process conditions.



A B

Figure 4 Hysteresis loop (A) and impact of specific aeration demand on the critical flux (B)

The effluent quality and removal efficiencies for the analysed macro-pollutants are summarised in Table 3.

Table 3 Permeate characteristics in terms of macro-pollutants

Parameter	**Mean value ± σ (g m^{-3})**	**Max (g m^{-3})**	**Min (g m^{-3})**	**Removal efficiency (%)**	**National Law D.Lgs. 185/03**
COD	12.9 ± 3.0	21	6	98.1	100
Soluble COD	10.2 ± 3.7	16	5	85.6	-
BOD_5	2.0 ± 0.5	4	1		20
TKN	1.9 ± 1.5	5.9	0.6	97.3	-
$N\text{-}NH_4$	0.6 ± 0.5	1.9	0.1	97.7	2
N-NO2-	0.1 ± 0.1	0.5	0.01	-	-
N-NO3-	6.0 ± 4.0	14.7	0.9	-	-
Total N	8.1 ± 4.5	21.2	2.6	88.2	15
Total P	1.2 ± 0.8	4.7	0.2	91.4	2
TSS	< 1	-	-	>99.9	10

Compared with the quality standards required in the national Law D.Lgs. 185/03, permeate characteristics resulted constantly suitable for agricultural reuse, even if once the total nitrogen value was higher than the permitted value. However, as the regulation imposes the respect of all macro-pollutants as mean values calculated on a yearly basis (or on the duration of the irrigated season), the observed exceeding can be neglected in the evaluation of nitrogen removal performances. Moreover, since an increase of total nitrogen concentration up to 35 $g\ m^{-3}$ is allowed, recycle ratio was varied in the range between 3 and 5, always achieving an excellent nitrification efficiency; in full scale systems, this would result in relevant savings of energy costs and, from the agronomic standpoint, reduction of fertilizer consumption.

Although no specific biological phosphorus removal was implemented, total P concentration in the effluent was generally compatible with the required quality; most of phosphorus removal was in fact achieved by sludge wasting operation, with an average content of P in the sludge 0.035 gP $gSSV^{-1}$, higher than conventional activated sludge. Average sulphate concentration (27.1 g SO_4^- m^{-3}) and average chloride concentration (46.6 gCl^- m^{-3}) resulted significantly lower than the imposed values (500 g SO_4^- m^{-3} and 250 gCl^- m^{-3}, respectively).
Microbiological analyses always showed a complete rejection by the membrane for *Escherichia Coli* and *Faecal Streptococci*; for both parameters the highest value was 1 CFU/100 mL which was observed just once over the whole experimental activity. Analyses carried out at APSS-LIESP also showed the always complete removal of *Salmonella* and searched parasites as *Helminths* (both eggs and larvae) and protozoa *Giardia* and *Cryptosporidum*.

Conventional tertiary treatment performances
As shown in Table 2., the secondary effluent from the WWTP in Pietramurata was already suitable for effluent reuse in terms of macro-pollutants. In any case, an improvement was observed concerning suspended solids and all particulate matter related components: COD removal was approximately 50% (average effluent COD: 14 ± 5 g m^{-3}) while TSS abatement was 80% (average effluent TSS: 2.3 ± 0.5 g m^{-3}). However, in two cases the effluent TSS concentration was higher than the required standard (14 g m^{-3} and 20 g m^{-3}) because of the increase in suspended solids content of the secondary effluents during two intense rainy events. Moreover, filter was steadily able to ensure a good turbidity removal (Figure 5.), compatible with optimal working conditions of the UV-C lamp.

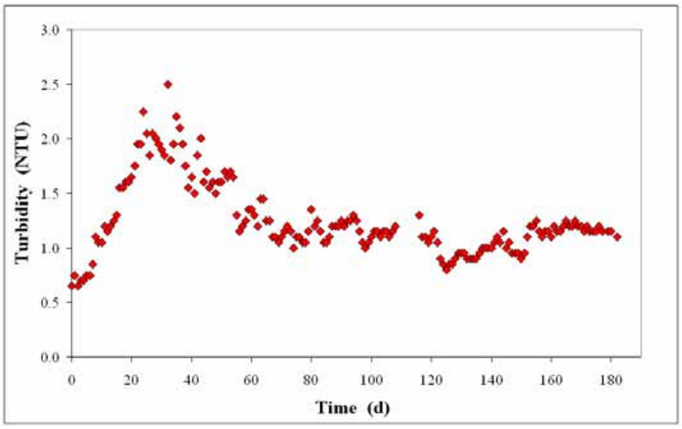


Figure 5 Turbidity trend over the experimental period

Over the entire experimentation, the average *Escherichia Coli* content in the secondary effluent was 12891 CFU/100 mL and *Salmonella* was often observed; conversely, neither *E. Coli* nor *Salmonella* were ever detected in the effluent, thus confirming the suitability of treated wastewater for reuse, according to the national regulation. Lower removal efficiency was observed for protozoa (98.7% for *Giardia* and 96% for *Cryptosporidium*) and parasites (77% for *Helminths* eggs and 96% for *Helminths* larvae). These results can be explained by the less effective rejecting action of filtering mechanisms (straining, adhesion, interception, sedimentation and flocculation) compared with membrane filtration. When not removed by the filtration unit, such complex microrganisms pass through the disinfection chamber, where the UV-C dose is not able to achieve the DNA-inactivation.

CONCLUSIONS

The experimental investigation was focused on process optimisation of the MBR technology aimed to production of treated wastewater suitable for apple orchards irrigation. Effluent wastewater quality was also compared with a traditional tertiary treatment scheme with coagulation/flocculation/filtration and UV-C disinfection. Major inferences are summarised below:

- the membrane critical flux, determined according to the flux step method, ranged between 25 and 32 L m^{-2} h^{-1}. Its value was not affected by the imposed specific aeration demand above a given value (0.5 Nm^3 m^{-2} h^{-1}), that resulted the most effective one in terms of energy saving for the process;
- under sub-critical flux regime, the membrane permeability was constantly around 140 L m^{-2} h^{-1} bar^{-1} and no sudden increase of TMP were never observed between two subsequent chemical backwash, therefore indicating that the imposed fluxes were constantly below the sustainable threshold for the considered operational conditions (MLSS, SRT, etc.);
- permeate quality was always compatible with the strict limitations of D.Lgs. 185/03 for all the analysed macro-pollutants and for all microbiological parameters investigated; this confirms the strong reliability of membrane ultrafiltration, when adequate pre-treatment units avoid debris matter to damage the membrane surface integrity;
- the characteristics of the conventionally treated wastewater (sand/anthracite plus UV-C disinfection) were always below the limits of the considered regulation; however, sand pressurised filtration resulted less effective than MBR concerning the protozoa and parasites removal.

On the basis of the obtained results, feasibility studies of possible full scale wastewater reclamation projects in apple orchards cultivation are currently under development. Besides, further scenarios are going to be evaluated for MBR permeate reuse in Trentino Province, including toilet flushing in decentralised household like alpine refuges and water sources for snow production during skiing season.

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REFERENCES

Bouhabila, E.H., Ben Aim, R., Buisson, H. (1998), Microfiltration of activated sludge using submerged membrane bioreactor with air bubbling (application to wastewater treatment), Desalination 118, 315-322

Field, R.W., Wu, D., Howell, J.A., Gupta, B.B. (1995), Critical flux concept for microfiltration fouling, J. Membr. Sci., 100, 259-272

Gleick, P.H. (2003), Global freshwater resources: soft-path solutions for the 21st century, Science, 302, 1524-1528

Howell, J., Chua, H.C., Arnot, T.C. (2004), In situ manipulation of the critical flux in a submerged membrane bioreactor using variable aeration rates, and effects of membrane history, J. Membr. Sci., 242, 13-19

Innocenti, L., Bolzonella, D., Pavan, P., Cecchi, F. (2002), Effect of sludge age on the performance of a membrane bioreactor: influence on nutrient and metals removal, Desalination, 146, 467-474

Joss, A., Andresen, H., Ternes, T., Richle, P.R., Siegrist, H. (2004), Removal of estrogens in municipal wastewater treatment under aerobic and anaerobic conditions: consequences for plant optimisation, Environ. Sci. Technol., 38, 3047-3055

Lopez, A, Pollice, A. Lonigro, A, Masi, S., Palese, A.M., Girelli, G.L., Tosano, A., Passino, R. (2006), Agricultural wastewater reuse in Southern Italy, Desalination, 187, 323-334

Police, A., Lopez, A., Laera, G., Rubino, P., Lonigro, A. (2004), Tertiary filtered municipal wastewater as alternative water source in agriculture: a field investigation in Southern Italy, Sci. Tot. Environ., 324, 201-210

Ueda, T., Noran, N.J. (2000), Fate of indigenous bacteriaphage in a membrane bioreactor, Wat. Res., 34, 2151-2159

Wintgens, T., Melin, T., Schäfer, A., Khan, S., Muston, M., Bixio, D., Thoye, C. (2005), The role of membrane processes in municipal wastewater reclamation and reuse, Desalination, 178, 1-11

Impact of different recirculation schemes on nutrients removal in a laboratory scale MBR

U. Bracklow*, L. Manigas**, A. Drews***, M. Vocks****, M. Barjenbruch* and M. Kraume***

* Dept. of Urban Water Management TIB 1B-16, TU Berlin, Gustav-Meyer-Allee 25, 13355 Berlin, Germany (E-mail: ute.bracklow@tu-berlin.de, matthias.barjenbruch@tu-berlin.de)
** Dept. of Geoengineering and Envir. Technol., University of Cagliari, Piazza d'Armi 1, 09123, Italy (E-mail: lmanigas@unica.it)
*** Dept. of Chemical Engineering MA5-7, TU Berlin, Str. des 17. Juni 135, 10624 Berlin, Germany (E-mail: anja.drews@tu-berlin.de;matthias.kraume@tu-berlin.de)
*** Berlin Centre of Competence for Water, Cicerostr. 24, 10709 Berlin, Germany (E-mail: martin.vocks@kwb.de)

Abstract For membrane bioreactors (MBR) with enhanced nutrient removal, commonly rather complex recirculation schemes based on the biological requirements are recommended. The aim of this work was to evaluate other recirculation options. For a laboratory scale MBR, four different recirculation schemes were tested. The MBR was operated with COD degradation, nitrification, post-denitrification without carbon dosing and biological phosphorus removal. For all configurations, efficient COD, nitrogen and phosphorus removal could be achieved. However, there were no big differences in elimination efficiency between the configurations (COD elimination: 96.6 - 97.9 %, nitrogen removal: 89.7 - 92.1 % and phosphorus removal: 97.4 - 99.4 %).. Changes in the degradation, release and uptake rates were levelled out by the changes in contact time and biomass distribution. With relatively constant outflow concentrations, different configurations are still interesting regarding oxygen consumption, simplicity of plant operation or the special support of certain degradation ways like biological phosphorus removal of nitrification.

Keywords biological phosphorus removal; denitrification; membrane bioreactor; nitrification; recirculation scheme; synthetic wastewater

INTRODUCTION

Like traditional wastewater treatment plants, membrane bioreactors (MBR) require sludge and mixed liquor recirculation for a better/even biomass distribution and to enable denitrification. For biological phosphorus removal and denitrification it is generally recommended to protect the anaerobic and anoxic zones from oxygen entrainment due to recirculation flows out of the strongly aerated membrane chamber. The biomass concentration in the membrane chamber is higher than in the other degradation zones. Nevertheless, when compared to the rest of the plant, the biomass concentration in a secondary settler of a conventionally wastewater treatment will be at a higher concentration than in a membrane chamber. Therefore, for a good biomass distribution, the recycled volumetric flow rate in a MBR must be higher than in a conventional plant which contributes to the rather complex recirculation configurations in MBR.

A laboratory scale MBR (Fig. 1) was operated at the Dept. of Chemical Engineering at the Berlin University of Technology. This MBR featured a post-denitrification process without carbon dosing and biological phosphorus removal. The aim of this work was to find other options for recirculation loops besides the one usually recommended for MBR with carbon, nitrogen and phosphorus removal (MUNLV, 2002; Vocks et al., 2005). Four different recirculation configurations (Fig. 2) were tested, particularly varying the location of the recirculation and for the last configuration (Fig. 2 d) also the volumetric flow rate. The influence on degradation efficiency, degradation rates and biomass distribution were investigated.

MATERIALS AND METHODS

Laboratory scale membrane bioreactor

The cascaded plant (volume 50 L) was designed for carbon and enhanced nutrients removal with post-denitrification without any additional carbon dosing and has been working for more than one year. The plant consisted of three anaerobic (AN0, AN1, AN2), two aerobic (AE1, AE2), three anoxic (AX1, AX2, AX3) and one membrane chamber (MF). The feed (concentrate + tap water) consisted of a complex synthetic wastewater resembling municipal wastewater (Table 1). The total COD of the feed amounted to 730 mg/L. Mineral and trace metal contents were adapted according to Brand (2003): H_3BO_4 300 µg/L, $CuCl_2$ 40 µg/L, KI 60 µg/L, $MnSO_4{}^{\bullet}H_2O$ 320 µg/L, $NaMoO_4{}^{\bullet}2H_2O$ 120 µg/L, $ZnCl_2{}^{\bullet}2H_2O$ 140 µg/L and $CoCl_2{}^{\bullet}6H_2O$ 300 µg/L.

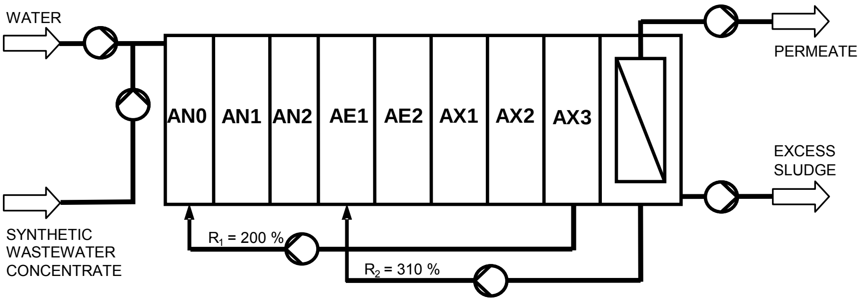


Figure 1 Basic flow sheet of the membrane bioreactor

Table 1 Composition of synthetic wastewater

Ingredient	[mg/L]	Ingredient	[mg/L]
Peptone	25	KH_2PO_4	26
Yeast extract	80	$MgHPO_4{}^{\bullet}3H_2O$	3.1
Milk powder	180	$MgSO_4*7H_2O$	4.2
Starch	200	K_2HPO_4	29
Sunflower oil	30	Urea	40
Ammonium acetate	140	$(NH_4)Cl$	38

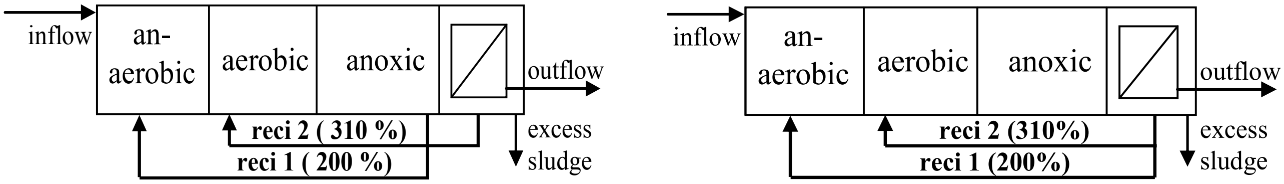
Sludge retention time (SRT) was set to 25 d. The flow rate was between 3.5 and 4.2 L/h which led to a hydraulic retention time of 12.0 to 14.4 h. Contact times in the different zones changed with different recirculation schemes (see Table 2). The most significant changes happened between configuration 2 and 3 (double contact time in the aerobic zone) and between configuration 3 and 4 (decreased contact time in anaerobic zone and increased time in anoxic zone and membrane chamber).

Table 2 Contact times and hydraulic retention time (HRT)

	Anaerobic zone AN [min]	Aerobic zone AE [min]	Anoxic zone AX [min]	Membrane chamber MF [min]	HRT [h]
Config. 1	64	18	30	33	13.2
Config. 2	69	21	35	26	12.6
Config. 3	74	48	40	29	13.3
Config. 4	54	35	59	43	14.2

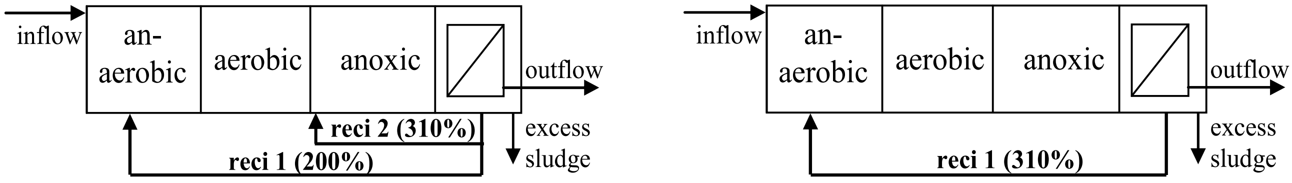
Investigated recirculation schemes

Figure 2 a-d shows the investigated recirculation schemes. Configuration 1 (Fig. 2 a) is the one recommended in Vocks et al. (2005), with a recirculation (reci 1) out of the last anoxic chamber into the first anaerobic chamber (appoximately 200 % of the volumetric flow rate of the inflow) and the main recirculation (reci 2) from the membrane chamber to the first aerobic chamber (approximately 310 % of the volumetric flow rate of the inflow). This configuration guaranteed optimum protection of the anaerobic zone from oxygen input and relatively good biomass distribution but the recirculation is rather complex for a post-denitrification process. In configuration 2 (Fig. 2 b) the recirculation 2 stayed as it was in configuration 1 but recirculation 1 was changed to MF to AN1, aiming for smoother biomass distribution and also testing the influence of aerated sludge recirculation on biological phosphorus removal. In configuration 3, recirculation 2 went into a small degassing chamber in front of the anoxic zone and no longer into the first aerobic chamber. Due to lower biomass concentration in the aerobic chambers, it demanded a lower amount of oxygen. Since there is no need for a second recirculation in a plant with post-denitrification, in configuration 4 only one recirculation from the membrane chamber to the first anaerobic chamber was implemented, a logical and technically simple arrangement for this plant.



a) Configuration 1

b) Configuration 2



c) Configuration 3

d) Configuration 4

Figure 2 a – d: Investigated recirculation arrangements

Analyses

Profile measurements of COD, nitrogen and phosphorus compounds were conducted every 3-4 days with a certain adaptation time for each configuration. Anions were measured using a Dionex DX 100 ion chromatograph with an IonPac AS 4a column for NO_3-N, NO_2-N, oPO_4-P (ortho-phosphat) and an IonPac CS12a column for ammonia-nitrogen. For the determination of COD, total phosphorus (TP) and total nitrogen (TN) Dr. Lange cuvette test kits LCK 114, 314, 414, 350, 349, 338, 238 were used. All cuvette tests comply in calibration, detection and quantitation limits with ISO 8466-1, DIN 38402 A51 and DIN 32645. For suspended solids, 100 mL sludge samples were taken and dried at 105 °C until constant weight was reached. The dried sample was heated at 600 °C for 3 h, and the amount of volatile suspended solids was calculated from the weight of the residue (compare DIN 38409 part 1).

RESULTS AND DISCUSSION

Distribution of biomass

Fig. 3 shows the averaged volatile suspended solids (VSS) of the different degradation zones as a portion of the VSS concentration in the membrane chamber.
The distributions developed according to the expectations due to the change of the recirculations. The slightly higher amounts of VSS in the aerobic zone compared to anoxic zones in configurations 1, 2 and 4 are most likely the result of the small degassing zone between AE2 and AX1. This zone became partially blocked and the VSS concentration in the second aerobic chamber was much higher than the VSS concentration in the first (and following) anoxic chambers.

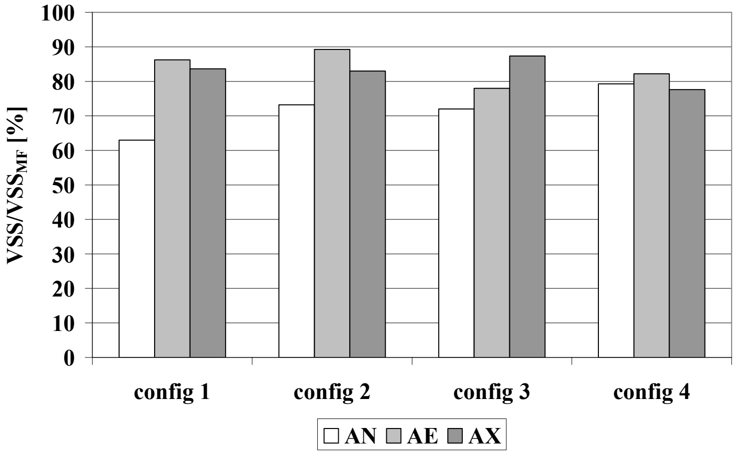


Figure 3 Portion of VSS [%] in comparison to the VSS in the membrane chamber

As in configurations 2, 3 and 4 the sludge was recirculated out of the most concentrated membrane chamber, a much higher biomass concentration in the anaerobic chamber could be reached. This was also the reason for the increase in the aerobic zone (when configuration 1 was switched to 2) and in the anoxic zone in configuration 3. The higher flow rate for recirculation 1 (MF to AN0) led to a further increase of the VSS concentration in the anaerobic zone in the last configuration. As expected, the smoothest distribution was reached for the last configuration with only one recirculation from the membrane to the first anaerobic chamber.

Table 3 gives the average values for suspended solids (SS) and volatile suspended solids (VSS) concentration for all configurations. Due to a computer failure the SS concentration during configuration 1 was still a bit low and did not reach the target value of about 10 g/L

Table 3 SS and VSS concentration

	SS [g/L]	VSS [g/L]	VSS [%]
Config. 1	7.7	5.9	77
Config. 2	10.6	8.6	81
Config. 3	9.6	7.5	79
Config. 4	9.7	7.8	80

COD degradation

As expected, the COD outflow concentration for all configurations mostly remained lower than 30 mg/L which led to a stable elimination above 95 %. These are routinely reachable values for MBR (Kraume et al., 2005). The averaged COD inflow concentrations were 908, 764, 718 and 786 mg/L for configurations 1, 2, 3 and 4, respectively. Standard deviation was calculated to 170 mg/L. The averaged outflow concentrations were 28, 23, 15 and 22 mg/L for configuration 1, 2, 3 and 4, respectively, with a standard deviation of 10 mg/L. On average the outflow concentrations were lower with lower inflow concentration and higher biomass concentrations (compare Tab. 3) and not depending on different recirculation schemes.

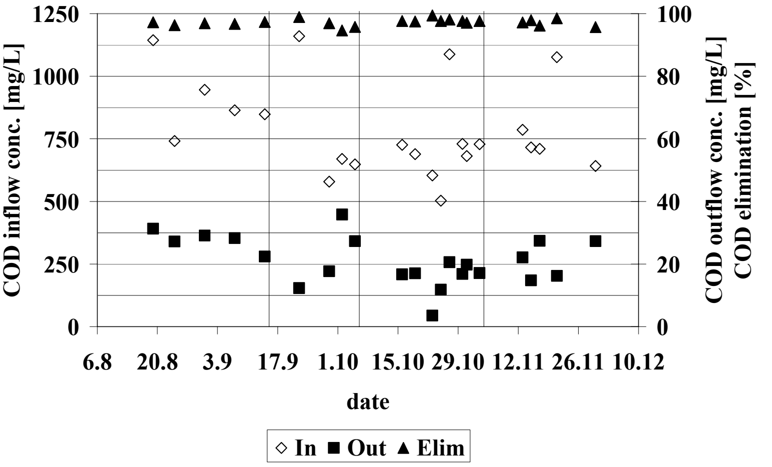


Figure 4 COD concentration and elimination

Nitrogen elimination

Total nitrogen elimination. For total nitrogen removal, a larger change was expected with the different recirculation configurations than the one observed for COD. As shown in Fig. 5, the inflow concentration averaged to 71 mgN$_t$/L. Outflow concentrations were mostly lower than 10 mgNO$_3$-N/gVSS/h and they averaged very close to each other to 6.5, 6.6, 6.9 and 6.5 mgNO$_3$-N/gVSS/h for configuration 1, 2, 3 and 4 whereas the elimination was around 91, 90, 90 and 92 %. These values are lower than those given in Gnirss et al. (2003), who found a nitrogen elimination of around 96 % and an outflow concentration of around 2.5 mgN$_t$/L for a post-denitrification process without carbon dosing. However, it was found in this study that the elimination was in the same range as for post-denitrification with a slightly different wastewater (Bracklow et al., 2007) and therefore as well higher than what was previously measured for this laboratory plant working with pre-denitrification. For the combination biological phosphorus removal and pre-denitrification an elimination of 76% was reached. This was the expected value for a recirculation of about 300 % into the anoxic zone (Bracklow et al., 2007)

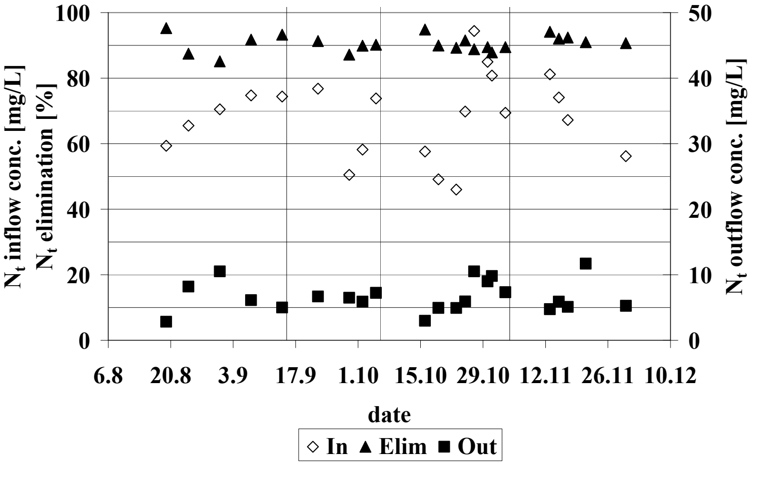


Figure 5 Nitrogen removal

Nitrification. The nitrogen in the inflow consisted of a higher portion of organically bound nitrogen (milk powder) than of ammonia-nitrogen (approx. 50 % NH_4-N). Therefore, an ammonification was necessary before nitrification could be obtained. Usually ammonification was not completed until the aerobic zone was reached, but due to the aerated membrane chamber the outflow concentration of NH_4-N was always lower than 0.2 mg/L. The nitrified ammonium in the membrane chamber amounted to 3 – 5 mgNH_4-N/L.

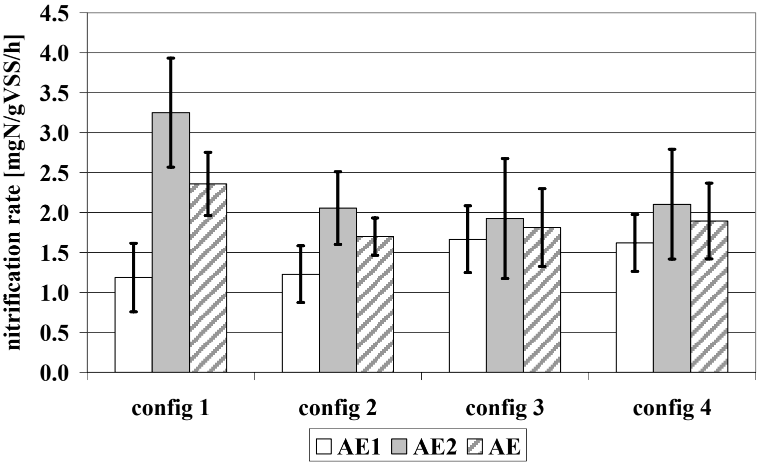


Figure 6 Nitrification rates

The nitrification rates for the different aerated chambers were mostly within the range given in literature (Kraume et al. (2005) 0.8 – 2 mgNH4-N/gVSS/h). One reason for the different nitrification rates observed in the different configurations could be the variation of the oxygen supply. Wiesmann (1986) stated that for degradation of organic substrates, about 1 mgO_2/L is needed whereas for nitrification 2 – 4 mgO_2/L are necessary to avoid limitations. Usually the oxygen concentration in the first aerobic chamber was about 2 – 3 mgO_2/L, whereas the oxygen concentration in the second aerobic chamber was between 4 - 5 mgO_2/L. The best nitrification rates (NR) were reached in the second aerobic chamber. It is assumed that a lag/adaptation phase which the microorganisms had to undergo when subjected from anaerobic to aerobic conditions in the first aerobic chamber resulted in the lower nitrification rates. This hypothesis is also supported by the observation that the shorter the contact time in aerobic zone (Table 4: Config 1, Config. 2, Config. 4, Config.3), the bigger the difference in nitrification rate between AE1 and AE2 (Fig. 6 and Table 4). Since the aerobic zone was split into two chambers a lag/adaptation time would influence only the first chamber and particularly when contact times are short. However, the nitrification rates did not change significantly with the change of the recirculation configuration.

Table 4 Total amount of nitrified and denitrified nitrogen

	AE VSS [g/L]	NR [mg/g/h]	t_C AE [min]	Δ N in AE [mg/L]	AX VSS [g/L]	DNR [mg/g/h]	t_C AX [min]	Δ N in AX [mg/L]
Config. 1	6.4	2.4	18	4.4	6.2	0.9	30	3.1
Config. 2	9.2	1.7	21	5.3	8.6	0.8	35	4.0
Config. 3	6.4	1.8	48	9.1	8.0	0.6	40	5.1
Config. 4	7.6	1.9	35	8.3	7.2	0.9	59	6.5

Degradation rates, biomass concentrations and contact times together are responsible for the degradation results. Table 4 shows that due to the different contact times, very good conversions in terms of total amounts were reached, i.e. even with low conversion rates.

Denitrification. As with the nitrification rates, the denitrification rates (DNR, see Fig 7 and Table 4) did not vary significantly between the different configurations. Almost all rates are in a similar range, confirming the rates given for post-denitrification without carbon dosing (Adam, 2004; Vocks et al. 2005). Notably is that, except for the last configuration, the first anoxic chamber always showed the lowest denitrification rates. A possible reason is the small degassing zone between the last aerobic and first anoxic chamber. The partial blocking of this zone could have produced short cuts between these two chambers which rather introduced a flow with high dissolved oxygen concentrations than protected the first anoxic chamber from oxygen. Also the blocking sludge got rapidly anaerobic which might have had negative effects on the denitrification. The complete deterioration of the denitrification in the first anoxic chamber in configuration 3 is interesting. As the denitrification rates of configurations 1 and 2 are all within the range of denitrification based on hydrolysis products (Sieker, 1999), this configuration should have obtained more hydrolysable substrate in the denitrification zone, with the sludge being recirculated not into the aerated but in the anoxic zone. With starch as the main slowly biodegradable substrate, an increase of denitrification rate was expected, especially since San Pedro et al. (1994) found no significant difference in hydrolysis rates for starch under anaerobic, aerobic or anoxic conditions (San Pedro, 1994). However, in the former configurations, hydrolysis products might not have been completely consumed in the aerobic zone as expected. The relatively high denitrification rates (1.9 $mgNO_3$-N/gVSS/h) in AX3 would underline this assumption. San Pedro et al. (1994) also found a build up of storage compounds under aerobic conditions for starch.

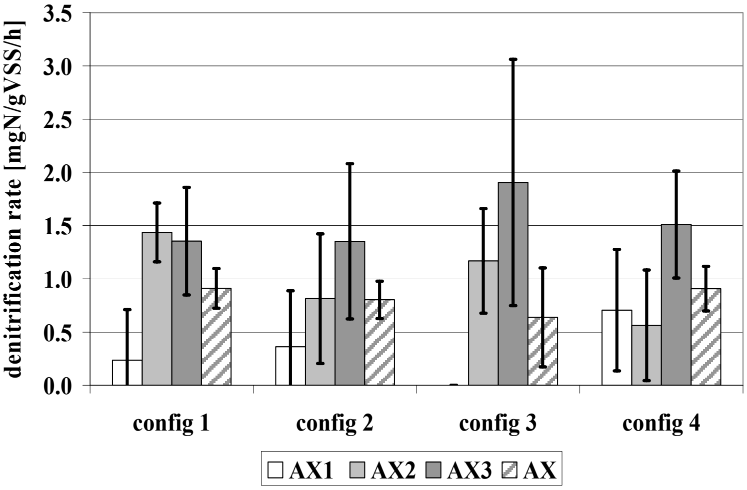


Figure 7 Denitrification rates in each anoxic chamber

Phosphorus removal

Enhanced biological phosphorus removal worked stably and efficiently throughout this study. During the experimentation two problems occurred. First, a change in the wastewater composition due to the introduction of saccharide, led to a complete deterioration of the biological phosphorus removal (not shown). Following the substitution of the sugar with acetate and milk powder, biological phosphorus removal recovered rapidly, but slight problems could still be seen in the first configuration. The second problem was a worm bloom, which led to lower eliminations for all parameters including phosphorus (end of measurement in last configuration). During the study the averaged outflow concentration was less than 0.4 mgP_t/L and the averaged eliminations were always higher than 97 %. It was expected that the introduction of recirculation sludge with high oxygen concentration would negatively affect the phosphorus release. This was not the case, on the contrary, the change of the main recirculation of the sludge between anaerobic and anoxic conditions (config. 1: reci 1 from AX3 to AN0), to anaerobic and aerobic conditions (config. 2 – 4:

reci 1 from MF (aerobic) to AN0), led to higher phosphorus release rates in the first anaerobic chamber. The design of the membrane chamber offers a good protection against oxygen entrainment since the recirculation outlet is on the side where no direct aeration is placed. Measurements of oxygen concentration at this point showed a variation between 0 and 4.5 mgO_2/L.

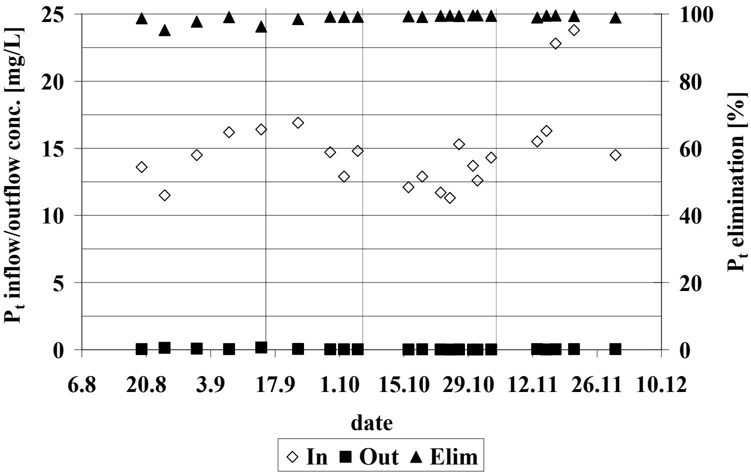


Figure 8 Biological phosphorus removal

Phosphorus release rates (PRR) were 8.7, 16.3, 26.9 and 33.1 mgP/gVSS/h for configurations 1, 2, 3 and 4, respectively (Manigas et al. 2007, in preparation). There are different reasons for the increase of the PRR in configuration 2 – 4. The alternation between anaerobic and aerobic conditions yields better metabolic conditions than the alternation between anaerobic and anoxic conditions (Kuba et al., 1996) and also increases the contact times in the aerobic zones in configuration 3 and 4 which could lead to a higher build up of storage compounds involved in the biological phosphorus removal process. This resulted in a higher rate of phosphorus release in the anaerobic zone, where the compounds stored in the aerobic zone are used (Manigas et al., 2007, in preparation).

SUMMARY AND CONCLUSIONS

In a laboratory scale membrane bioreactor for enhanced nutrient removal, four different mixed liquor recirculation schemes were tested.

The operation of the four different configurations had no strong impact on COD, nitrogen or phosphorus elimination in general. Decreases of degradation rates were levelled out by higher contact times or different biomass concentrations which resulted from the differences in recirculation location or flow rate.

Configurations 2 – 4 showed a better phosphorus release in the anaerobic zone due to the greater amount of energy involved in the phosphorylation during the metabolic processes, triggered by the alternation from anoxic-anaerobic to aerobic-anaerobic conditions. This could be important for plants with weak biological phosphorus removal.

Due to the cascaded design of the plant, the influence of lag/adaptation phases in combination with the contact times of the single chambers could be seen.

The overall performance of the plant remained relatively constant with no big differences of the eliminations between each configuration. The values for COD elimination were 96.9 %, 96.6 %, 97.9 % and 97.1 % for nitrogen removal 90.6 %, 89.7 %, 90.2 % and 92.1 % and for phosphorus removal 97.4 %, 99.0 %, 99.4 % and 99.3 % for configuration 1, 2, 3 and 4, respectively. However, each configuration was found to have its specific advantages depending on the specific requirement:

- Higher simplicity in recirculation scheme
- Lower oxygen consumption due to lower biomass concentration in aerated zone
- Longer contact times coupled with smoother biomass concentration
- Support of certain degradation ways.

REFERENCES

Adam, C. (2004). Weitgehende Stickstoff- und Phosphorelimination in Membranbelebungen mit nachgeschalteter Denitrifikationsstufe (Enhanced nitrogen and phosphorus removal in membrane bioreactors with post-denitrification). PhD thesis, Umwelttechnik 15, VDI Verlag, Düsseldorf.

Bracklow, U., Drews, A., Vocks, M. and Kraume, M. (2007, in press). Comparison of nutrients degradation in smallscale MBR fed with synthetic/domestic wastewater. Hazardous Materials.

Brand, D. (2003). Möglichkeiten und Grenzen von Biofilmverfahren zur Kombination von biologischer Stickstoff- und Phosphorelimination (Options and limits of biofilm reactors with combination of denitrification and enhanced biological phosphorus). PhD thesis, Berichte zur Siedlungswasserwirtschaft 22, TU Berlin, Berlin.

Gnirss, R., Lesjean, B. and Buisson, H. (2003). Biologische Phosphorentfernung mit einer nachgeschalteten Denitrifikation im Membranbelebungsverfahren (Biological phosphorus removal combined with post-denitrification process in a membrane bioreactor). In: 5. Aachener Tagung für Siedlungswasserwirtschaft und Verfahrenstechnik, Aachen.

Kraume, M., Bracklow, U., Vocks, M. and Drews, A. (2005). Nutrients removal in MBRs for municipal wastewater treatment. Water Sci Technol, 51(6-7), 391-402.

Kuba, T., Murnleitner, E., Loosdrecht, M. C. M. v. and Heijnen, J. J. (1996). A Metabolic Model for Biological Phosphorus Removal by Denitrifying Organisms. Biotechnology and Bioengineering, 52 (6), 685-695.

Manigas, L., Bracklow, U., Drews, A., Vocks, M. and Kraume, M. (2007, in preparation). Design Optimization of a Membrane Bioreactor for Enhanced Nutrients removal.

MUNLV (2002). Abwasserreinigung mit Membrantechnik. NRW, Aachen.

San Pedro, D. C., Mino, T. and Matsuo, T. (1994). Evaluation of the rate of hydrolysis of slowly biodegradable COD (SBCOD) using starch as substrate under anaerobic, anoxic and aerobic conditions. Water Science Technology, 30(11), 191-199.

Sieker, C. (1999). Kombination der Denitrifkation und der vermehrten biologischen Phosphorelimination in einer alternierend betriebenen Biofiltrationsanlage (Combination of denitrification and enhanced biological phosphorus removal in an alternating working biofiltration plant). PhD thesis, Berichte zur Siedlungswasserwirtschaft 12, TU Berlin, Berlin.

Vocks, M., Adam, C., Lesjean, B., Gnirss, R. and Kraume, M. (2005). Enhanced post-denitrification without addition of an external carbon source in membrane bioreactors. Water Research, 39 (14), 3360-3368.

Wiesmann, U. (1986). Kinetik der aeroben Abwasserreinigung durch Abbau von organischen Verbindungen und durch Nitrifikation (Kinetic of degradation of organic compounds and nitrification in aerobic wastewater treatment). Chemie Ingenieur Technik, 58(6), 464-474

A comparative study between multi-zone and alternating anoxic/aerobic MBRs for municipal wastewater treatment and reuse

Giacomo Carletti*,****, Francesco Fatone*, Emanuela Cola**, Enrico Maria Battistoni***

*Department of Science and Technology, University of Verona. Strada Le Grazie,15 - 37134 Verona, Italy. (E-mail: giacomo.carletti@virgilio.it; fatone@sci.univr.it;)
**Institute of Hydraulics and Transportation Infrastructures, Marche Polytechnic University. Via Brecce Bianche-60131 Ancona, Italy. (E-mail: e.cola@univpm.it;)
***University of Venice “Cà Foscari”, Department of Environmental Sciences, Calle Larga santa Marta, Dorsoduro, Venice - Italy
**** Interuniversity Consortium “Chemistry for the Environment” (INCA), via delle Industrie 21/8 Marghera, Venice, Italy.

Abstract A comparison between multi-zone and alternating anoxic/oxic membrane bioreactors has been carried out at pilot scale, treating real municipal wastewater. The two systems have been tested under the same loading conditions and similar operating parameters, so to isolate and investigate the effect of the different biological process. The alternating membrane bioreactor showed better nitrogen removal performances respect to the multi-zone thanks to the better utilization of the carbon source, which was oxidized almost anoxically. Also, the alternating MBR demonstrated the potential to enhance the biological phosphorus removal, while the multi-zone scheme was able to remove phosphorus only by the normal biomass assimilation. As for the metals, the different redox regimes of the two membrane bioreactors, the first stable tank by and the second fluctuating in the same tank, did not show significant impact on the removal performances. As for the organic hazardous compounds, while the Polynuclear aromatic hydrocarbons were removed at same level, the organic solvents were volatilized more in the multi-zone scheme according to the longer aerobic time.

Keywords membrane bioreactors; multizone and alternating processes, nutrients, hazardous compounds

INTRODUCTION

The drivers for the widespread implementation of membrane bioreactors (MBRs) for municipal wastewater treatment are becoming more and more remarkable (Judd, 2006). However, high power requirements for the plant operation represent still a restrain, which not only increases the operating costs, but also places the MBR technology in disagreement with the current energy saving policies. Since major power is required for the aeration of the system, a good design and operation practice should be based on the optimization of the air utilization. This can be done intervening both on the membrane section (air for membrane scouring) and on the biological process (air for biological oxidization). As for the biological process, the alternating aerobic/anoxic systems demonstrated to be able to reduce the energy consumptions thanks to best utilization of the nitrogen-bound oxygen that enable to degrade the organic matter anoxically (Charpentier et al., 1987; Olsson et al., 2005; Battistoni et al., 2002, 2003). To date, this solution of intermittent aeration has been successfully applied from pilot, to demonstration up to full scale MBRs, pointing out relevant and reliable savings of power requirements (Battistoni et al, 2006; Fatone et al., 2005; 2007). However, actually there is still a lack of knowledge about the real convenience, in terms of process performances, of alternating systems with respect to the more conventional multi-zone processes. Therefore, this paper deals with a comparative study where two parallel pilot MBRs have been used: the first operating the alternating anoxic-oxic process and the second with the more classical multi-zone predenitrification-nitrification scheme. The experimental results following presented deal with the behaviour of both nutrients and some xenobiotics (Heavy Metals, Polynuclear Aromatic Hydrocarbons, Volatile Organic Compounds). Furthermore, the applied approach of the research will finally allow to outline, with good degree of reliability for full scale applications, the advantages of one plant configuration with respect to the other. From the practical point of view, the obtained results may give useful addresses regarding the technology which is most suitable to upgrade the existing obsolete treatment facilities, aiming to water reclamation and reuse.

MATERIALS AND METHODS

The pilot plants

The Treviso membrane demonstration plant (Figure 1) was composed of two parallel lines completely equal for features such as: (a) total reaction volume (11 m^3 per line, including the membrane tank); (b) membrane modules (hollow fiber membranes -ZeeWeed®500c- with a total area of 69,90m^2 per line); (c) feeding wastewater. The main difference between the two parallel MBRs was the biological process operated before the ultrafiltration section: multi-zone predenitrification (DN) system, for the first, and Alternate Cycles (AC) process (Battistoni et al., 1999), for the second. As for the AC-MBR line, the plant, the process and the control automation have been already exhaustively described in previous papers (Fatone et al., 2005, 2006, 2007). On the other hand, the DN-MBR is divided into the first anoxic tank of 4,8 m^3 followed by the aerobic tank of 4,8 m^3 and the final ultrafiltration section, which is also an effective aerobic bioreactor (1,4 m^3 inclusive of the membrane overall dimensions). Therefore, in practice the DN-MBR is anoxic and aerobic for 43% and 57%, respectively.

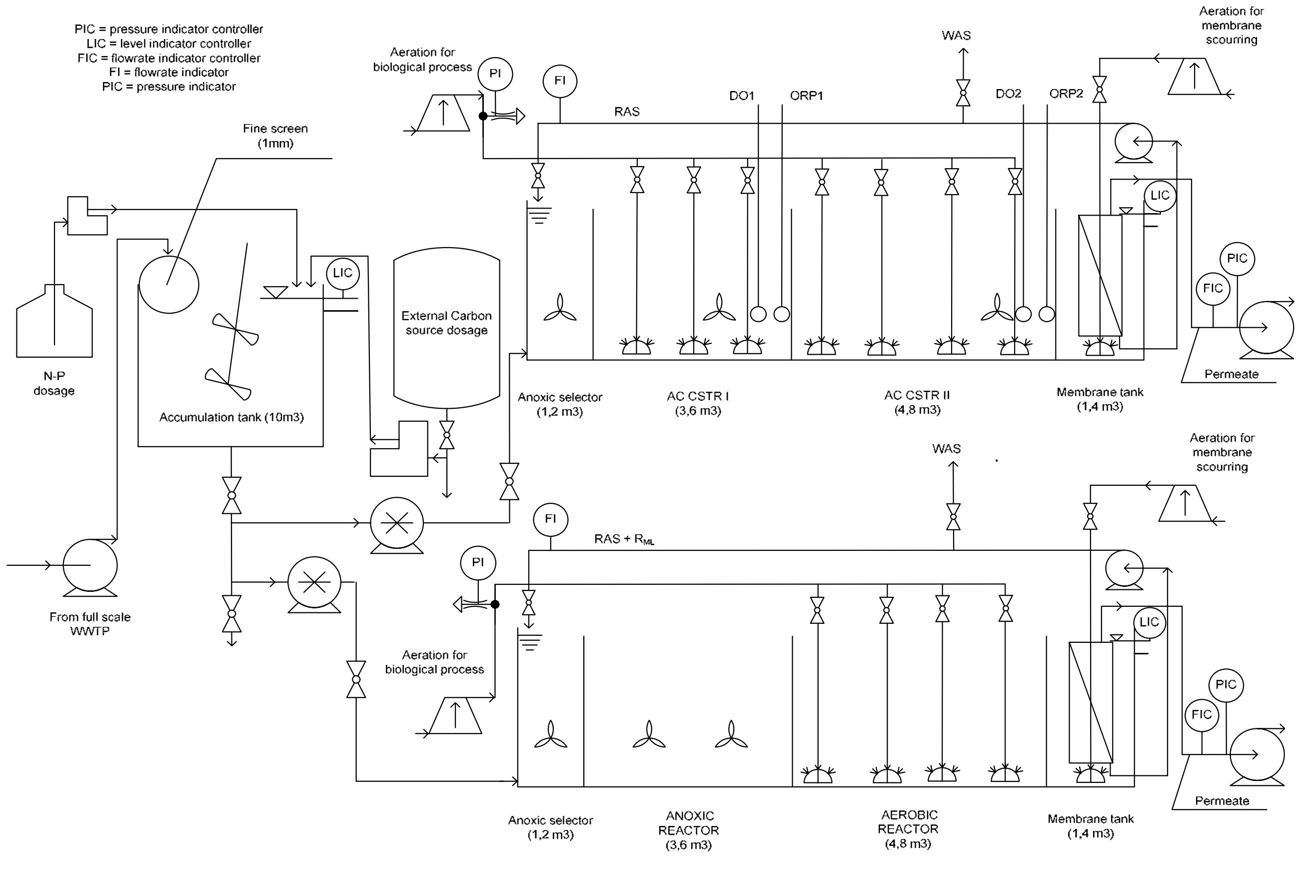


Figure 1. Block flow diagram of the AC and DN lines demonstrative plant.

MBRs monitoring and sampling methods

The two MBRs were monitored focusing the attention not only on the more “classic” pollutants like carbon, nutrients and suspended solids, but also on a number of hazardous and persistent substances. In particular, the presence of some heavy metals, Polynuclear Aromatic Hydrocarbons (PAHs), Volatile Organic Compounds (VOCs) was investigated in the main streams of the two parallel MBRs (influent, permeate and waste activated sludge). The conventional chemical-physical parameters like TSS, COD, soluble COD, TKN, NH_4-N, Total P, were determined according to the Standard Methods. Anion contents (NO_3-N, NO_2.N, PO_4-P) were determined with high performance liquid chromatograph (HPLC–Dionex ICS90). Metals, PAHs and VOCs were determined according to the EPA methods.

All the samples were collected as average over 24 hours. Moreover, a particular procedure was adopted to investigate the partition of influent hazardous compounds between the fractions associated to the suspended particulate dissolved in the liquid phase. In particular, a membrane-equipped automatic sampler was used to filter 350÷450 $L_{influent}$ per day and, by this way, to concentrate enough suspended particulate to obtain a significant and reliable samples, averaged over 24 hours. These were analysed to quantify the fraction of the before mentioned hazardous substances which is associated to the suspended particulate.

Raw municipal wastewater and external dosages

The MBRs was fed with real wastewater (see Table 1 for the main characteristics) coming from the Treviso (northern Italy) municipal WWTP.

Table 1. Characteristics of the raw influent wastewater

pH	Alkalinity mg$CaCO_3$/L	TSS mg/L	COD mg/L	rbCOD mg/L	NH_4-N mg/L	TKN mg/L	NO_3-N mg/L	PO_4-P mg/L	TP mg/L
7,4÷7,8	230÷350	80÷100	90÷160	17÷21	11÷15	20÷23	0,4÷1	0,7÷1,2	1,8÷2,2

Further, over the experimentation the influent loadings were adjusted by the dosage of external carbon, nitrogen and phosphorus. In particular, a solution of NH_4Cl and $(NH_4)_2HPO_4$ was used for nutrients dosage, while two different external carbon source were used: the acetic acid and the liquid part of the Organic Fraction of the Municipal Solid Waste ($OFMSW_L$ – see table 2 for the characteristics), which was taken directly from the screw press of the full scale co-digestion system of Treviso.

Table 2. Characteristics of the $OFMSW_L$ dosed as external carbon source

TSS mg/L	COD mg/L	rbCOD mg/L	NH_4-N mg/L	TKN mg/L	PO_4-P mg/L	TP mg/L
200÷1000	7000÷15000	2500÷3000	80÷120	400÷500	4÷8	5÷10

RESULTS AND DISCUSSION

The experimental approach: influent loadings and operating parameters

The experimental approach was addressed from the preliminary considerations over the loading conditions of the existing municipal treatment facilities. In fact, municipal WWTPs are usually loaded with Nitrogen Loading Rates (NLRs) in the range 0,06÷0,08 kgN $m^{-3}d^{-1}$, and can reach values of 0,03÷0,05 kgN $m^{-3}d^{-1}$ during wet weather periods. In this study, eight steady state experimental runs were carried out increasing the nitrogen loading rates (NLRs) from 0,05 to 0,25 kgN $m^{-3}d^{-1}$ (Table 3). From a practical viewpoint, this allowed to evaluate the potential of the existing biological reaction volumes to be exploited for the co-treatment of liquid wastes rich in ammonia nitrogen and municipal wastewater.

As for the process operating parameters (Table 3), the MBRs was managed taking into account that too high biomass concentration can drastically influence the oxygen transfer (Germain et al., 2007) and, consequently, the power requirements. Therefore, the MLSS were in the range 5÷10 g L^{-1} and the SRT in the range 13÷50 days. Table 3 summarises the main loading conditions and operating parameters for the two MBRs, reported as average over the whole experimental run.

Table 3. Operational conditions and process parameters

	Operating parameters[a]			Volumetric Loadings Rates for C, N, P			Influent characteristic ratios	
	T °C	MLSS DN g/L	MLSS AC g/L	OLR kgCOD m^{-3}d^{-1}	NLR kgN m^{-3}d^{-1}	PLR kgP m^{-3}d^{-1}	COD/ TN	rbCOD/ COD
Run 1	20	-	4,8	0,24	0,05	0,004	5,1	0,19
Run 2	13	-	8,5	0,29	0,07	0,006	3,9	0,19
Run 3*	11	9,2	9,0	0,67	0,07	0,009	9,2	0,48
Run 4**	19	5,3	5,5	0,59	0,07	0,010	8,4	0,19
Run 5	23	7,0	6,6	0,35	0,09	0,022	4,5	0,17
Run 6**	22	7,7	8,0	0,61	0,11	0,019	5,1	0,14
Run 7**	19	8,2	8,2	0,76	0,12	0,020	6,7	0,15
Run 8**	15	9,6	10,0	1,35	0,25	0,048	5,2	0,07

External Carbon Source: *Acetic Acid; **OFMSW$_L$

[a] The DN-MBR was not operating over for the first two runs

The experimentation was carried out over some 20 months and involved temperature of the mixed liquor in range (11÷23 °C) typically observed over the whole year. Furthermore, the availability of carbon source was always adjusted in a way to be critical for the removal of the total nitrogen, so to study the boundary performances of the two systems. The process operating parameters may seem not consistent with the liquor temperature or the loading conditions. Actually, when the OFMSW$_L$ was dosed as external carbon source, the MLSS content was influenced by the influent solids, escaped from the screw press of the full scale co-digestion plant. As a consequence, the sludge age was adjusted changing the waste activated sludge appropriately. However, this apparent drawback did not show impacts nor on the removal performances, as further discussed below, nor on the membrane fouling, as will be discussed in a forthcoming paper.

Nutrients removal

Phosphorous removal. Fatone et al. (2006) showed that the AC-MBR can enhance the phosphorus biological removal by performing (a) the release in the deep anoxic phases (when the reactor environment is on the boundary line between the anoxic and anaerobic conditions) and (b) the luxury uptake in the aerobic phase and in the final membrane tank. Of course, the authors pointed out that this phenomenon is possible only with a carbon source suitable for the PAOs and DPAOs growth and metabolism. In this study the phosphorus biological removal in the two systems was performed mainly by biomass assimilation (table 4). However, detailed studies on the two MBRs pointed out that phenomena of luxury uptake were occasionally observed only in the AC-MBR system. Actually, when the plant could always rely on sufficient amount of volatile fatty acids for PAOs and DPAOs activity (run 3 – table 4), these phenomena turned from occasional to stable. As a consequence, in this run the P removal in the two reactors was significantly different: the DN-MBR went on removing phosphorus by classical biomass assimilation, while the AC-MBR was an effective system for biological nutrients removal (BNR). As for the carbon source effect, unexpectedly the effect of the $OFMSW_L$ was not very significant for P biological removal, probably because the organic waste was not enriched in VFA because it was not fermented before the entry in the MBRs.

Table 4 Influent/effluent characterization and removal of COD and total P

	COD					P				
	AC-D/N In Kg d^{-1}	AC Out Kg d^{-1}	Rem. %	D/N Out Kg d^{-1}	Rem. %	AC-D/N In Kg d^{-1}	AC Out Kg d^{-1}	Rem. %	D/N Out Kg d^{-1}	Rem. %
Run 1	2,69	0,26	90	-	-	0,04	0,02	47	-	-
Run 2	3,16	0,17	95	-	-	0,06	0,04	32	-	-
Run 3	7,38	0,14	98	0,14	98	0,10	0,04	63	0,6	40
Run 4	6,54	0,18	97	0,46	93	0,11	0,05	55	0,05	55
Run 5	3,90	0,45	89	0,33	91	0,24	0,17	27	0,19	21
Run 6	6,70	0,79	88	0,77	88	0,21	0,09	56	0,09	57
Run 7	8,32	0,81	90	0,58	93	0,22	0,11	50	0,09	59
Run 8	14,88	0,26	98	0,26	96	0,53	0,20	62	0,17	68

Carbon and TSS removal. The comments on COD and TSS removal are quite obvious. In fact the low amount of carbon source led to almost complete removal of the biodegradable COD and only the soluble non biodegradable COD was found in the permeate (TSS effluent < 1 mg/L). However, it is interesting to note that nor the dosage of $OFMSW_L$ and the increasing OLRs influenced significantly the COD removal, even at OLR of 1,35 kgCOD m^{-3} d^{-1} (run 8). This may demonstrate that the particulate COD is hydrolyzed and assimilated in the MBR, which is able to retain the macromolecules allowing the further and complete biodegradation.

Nitrogen removal. The nitrogen mass balance was calculated according to Battistoni et al. (2002) and the nitrification and denitrification performances were studied according to four parameters: the nitrifying efficiency referring to the total incoming nitrogen (En) and to the amount of the only form of nitrogen that can be nitrified (Enn); the nitrogen removal efficiency referring either to the total incoming nitrogen (Ed) or to the nitrified nitrogen, NOx-N (Edd). Edd% and Enn% have been calculated and reported in Figure 2.
Figures 2 a) and b) show the way in which the processes has been loaded, with particular concern to COD/TKN ratios and NLRs, and as a consequence the behaviour of the two lines. It can be noted how much nitrogen has been removed: up to NLR=0,11÷0,12kg$m^{-3}d^{-1}$ the removal of total nitrogen was very satisfactory for the AC-MBR process, for higher values (unusually for municipal WWTP) DN-MBR process had better performances.
Considering that the influent COD is the same both for AC line and for DN line, the alternating process is able to guarantee higher total nitrogen removal because of the better utilization of the carbon source which is mostly degraded anoxically, so to improve the nitrogen removal performances and the reduce the power requirements. Of course, this peculiatity falls for C:N < 4 when the carbon source is really too low for the biological nitrogen removal (figure 3c)
The AC nitrogen removal efficiency referring to the total incoming nitrogen compared with the same calculated for DN line (Figure 2-d)) is a reliable proof of this fact. An important consequence of these aspects is the optimization of the air supplying and the low recycle ratio required for the best performances of biological process; under the economical point of view the energy saving is remarkable: 20%÷40% (Fatone et al., 2005; 2007).

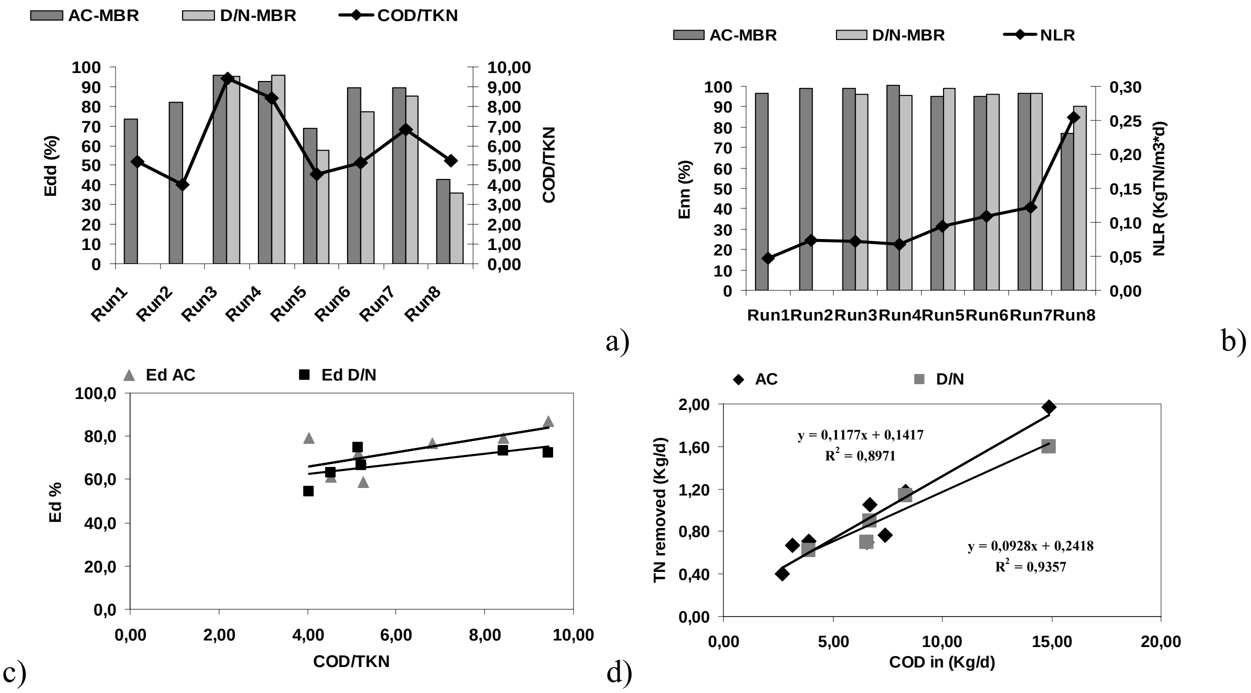


Figure 2. Nitrogen removal in AC and DN processes under different conditions

Micropollutants removal

A number of papers have been written about micropollutants removal in conventional biological process and, generally, one can observe that an activated sludge process is able to guarantee the biosorption or bioaccumulation of a large part of these compounds. However, the chemical-physical form of the hazardous compound is fundamental to understand the potential of one molecule to be removed biologically. In particular, the Redox Potential (ORP) may play an important role with regard to the influence on the chemical form of many xenobiotics substances. With concern to the biological reactors for wastewater treatment, ORP is usually in the range± 250 mV. However, while the multi-zone processes keep stable ORP values tank by tank, the alternating processes let the ORP fluctuate continuously in the same reactor. Since relationship between xenobiotics removal and ORP has been found out by several authors (Patrick and Verloo, 1998; Carbonell-Barrachina et al., 1999; Meng et al., 2001), aim of this study was also to investigate and compare the removal of hazardous compounds in alternating and multi-zone MBRs.

To better understand the role of the particular biological process, also the solid/liquid partition of the compounds have been determined by the coupled sampling method before described. Table 5 shows the main results obtained and the solid and liquid fraction. PAHs and VOCs are usually detected at very low levels and this is the reason why the table below reports the total amounts. However, the major frequency of occurrence among polycyclic aromatic hydrocarbons have been detected for Naphthalene, Acenaphtene and Anthracene; while organic solvents such as benzene, toluene and xylens (BETX) have been detected for organic volatile compounds.

Table 5. Influent/effluent waste activated sludge (WAS) characterization; solid liquid portioning and removal efficiencies of organic and non organic compounds during runs 3-4.

	INFLUENT				OUT AC--D/N	REMOVAL		
	Average (μg L^{-1})	Min÷Max (μg L^{-1})	Sol. (%)	Liq. (%)	Average (μg L^{-1})	AC (%)	D/N (%)	CASP (%)
As	38,8	27,2÷50,5	62	38	<2,5-2,7*	23	23	-
Hg	7,8	6,4÷9,2	31	69	<2,5	37	35	57÷92
Cu	17,0	9,4÷23,6	71	29	<2,5	>93	>93	54÷82
Pb	10,6	3,2÷18	27	73	1,2*-0,6	>85	>85	68÷100
Cd	3,8*	-	3	97	2,4*-1,5	>30	>30	25÷74
Ni	2,3	1,3÷3,5	31	69	1,2-1,8	89	94	43÷95
Cr	0,2*	-	nd	nd	<0,25	>70	>70	68÷85
Zn	350	229÷571	35	65	<50	86	86	87÷88
Fe	1967	1362÷2380	74	26	<50-62*	97	97	67÷90
Al	2391	1973÷2824	93	7	<50-70*	98	97	70-80
PAH	1,73	0,9÷2,5	-	-	0,2-0,1	90	95	63÷90
VOC	1,5	0,05÷15,4	-	>99	0,5-0,6	63	79	15÷99

* only one sample higher than the limit of quantification

As for the metals, table 5 shows as the alternating and multi-zone scheme have comparable removal, notwithstanding the almost different redox regime. Even removals of compounds like the As, which chemical form and potential to be biosorbed demonstrated to be influenced from the pH and ORP values (Carbonell-Barrachina et al., 1999; Meng et al., 2001), were not significantly influenced by the stable or alternating environment. This, probably, can depend on the low metals concentration present in municipal wastewaters which do not allow for significant differences according to biological process.

About the final fate of the PAHs and VOCs, hypothesis regarding the removal mechanisms are almost impossible, because these compounds were always under our limit of quantification in the waste activated sludge. On the other hand, it is possible to note that while PAHs are removed similarly in the two MBRs, the VOCs are less present in the permeate of the DN-MBR. This fact is reasonably explained with the lower volatilization in the alternating system, which have longer anoxic phases. Of course, this is just a particular scenario of the wastewater with low C:N ratio, while an opposite situation could be observed for high C:N which would involve longer aerobic phases, and major VOCs volatilization, in the AC-MBR.

CONCLUSIONS

A comparative study was carried out between two twin MBRs for municipal wastewater treatment: on adopting a conventional multi-zone scheme, another the alternate cycles process.

The main conclusions are following itemized:

- generally, alternating system resulted more appropriated than multi-zone anoxic/oxic for the treatment and reuse of municipal wastewater. In fact, although both were able meet the Italian standard for water reuse, the alternating processes optimize both removal performances and power requirements;
- nitrogen removal performances were higher in the alternating system thanks to the better utilization of the carbon source. In fact, this was adequately oxidized anoxically, allowing for higher nitrates denitrification;
- the alternating MBR demonstrated the potential of being also a BNR system in the influent contains the suitable biodegradable carbon. On the other hand, the multi-zone MBR was never able to enhance the biological phosphorus removal;

- using real municipal wastewater coming mainly from domestic dwellings, the metals contents were low and the removals were similar in the two systems, in spite of the different redox regimes: stable in the multi-zone and fluctuating in the alternating MBR;
- the removal of Polynuclear Aromatic Hydrocarbons was also similar in the two systems, while the organic solvents were better volatilized in the multi-zone system. However, this fact must be considered a particular peculiarity proper of low C:N wastewater, which involve short aerobic phases.

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REFERENCES

Battistoni P. and Chemitec. (1999). Metodo e dispositivo di controllo di un processo di trattamento biologico, a cicli alternati, di acque reflue., Italian Patent N°RM99A0000182. 6.99.

Battistoni P., Boccadoro R., Innocenti L., Bolzonella D. (2002). Addition of an external carbon source to enhance nitrogen biological removal in the treatment of liquid industrial wastes. Ind. Eng. Che. Res., 41(11), 2805-2811.

Battistoni P., De Angelis A., Boccadoro R., Bolzonella D. (2003). An automatically controlled alternate anoxic-oxic process for small municipal wastewater treatment plants. Ind. Eng. Chem. Res., 42 509-515.

Battistoni P., Fatone F., Bolzonella D., Pavan P. (2006). Full scale application of the coupled alternate cycles-membrane bioreactor (AC-MBR) process for wastewater reclamation and reuse. Wat. Prac. and Tech. 1 (4).

Carbonell-Barrachina A., Jugsujinda A.,. DeLaune R.D, and Patrick, Jr W.H. (1999). The influence of Redox chemistry and pH on chemically active forms of arsenic in sewage sludge-amended soil. Environmental International, 25 (5), 613-618.

Charpentier J., Florenz M., David G., Oxidation-reduction potential (ORP) regulation: a way to optimize pollution removal and energy savings in low load activated sludge process, Wat. Sci. Tech., 19 (1987) 645-655.

Fatone F., Bolzonella D., Battistoni P., Cecchi F. (2005). Removal of nutrients and micropollutants treating low loaded wastewaters in a membrane bioreactor operating the automatic alternate-cycles process Desalination 183, 395-405.

Fatone F., Battistoni P., Pavan P., Cecchi F. (2006). Application of a membrane bioreactor for the treatment of low loaded domestic wastewater for water re-use. Water Science and Technology 53 (9), 111-121.

Fatone F., Bolzonella D., Battistoni P., Pavan P., Cecchi F. (2007). Long-term experience with an automatic process control for nitrogen removal in membrane bioreactors. Desalination, submitted.

Germain E., Nelles F., Drews A., Pearce P., Kraume M., Reid E., Judd S., Stephenson T. (2007). Biomass effects on oxygen transfer in membrane bioreactors. Water Research, 41 (5), 1038-1044.

Judd. S., The MBR Book: Principle and Applications of Membrane Bioreactors in Water and Wastewater Treatment, Elsevier, Oxford, 2006.

Karvelas M., Katsoyiannis A., Samara C. (2003). Occurrence and fate of heavy metals in the wastewater treatment process. Chemosphere 53 1201-1210.

Meng X., Korfiatis G.P., Jing C., Christodoulatos C. (2001). Redox transformations of arsenic and iron in water treatment sludge during aging and TCLP extraction. Environmental Science and Technology, 35 (17), 3476-3481

Olsson G., Nielsen M.K., Yuan Z., Lynggaard-Jensen A., Steyer J.P. (2005). Instrumentation, Control and Automation in Wastewater Systems. Scientific and Technical Report No. 15. IWA publishing

Rogers H. R. (1996). Sources, behaviour and fate of organic contaminants during sewage treatment and in sewage sludges The Science of the total Environment, 185, 3–26

Analysis of long term performance of a full-scale MBR plant treating domestic and industrial wastewater

V. Ferre*, S.J. Churchouse**, P. Jeffrey***, S. Judd***, S. Warren****

* Kubota Membrane Europe Ltd, 8 Hanover Street, London W1S 1YE, UK
(E-mail: victor@kubotalon.co.uk)
** Sci Eng Consultancy Services, Bristol, UK
*** Building 39, Cranfield University, Cranfield, Bedfordshire MK43 0AL, UK
**** Wessex Water Plc, Claverton Down, Bath BA2 7WW, UK

Abstract This paper provides long-term fouling correlations for a full-scale membrane bioreactor (MBR). It is based on the experience of 4 years' operation with Kubota membranes in Westbury, UK.
A statistical analysis was undertaken based on instantaneous telemetry data, which was appropriately selected and averaged. Potential fouling factors such as trans-membrane pressure (TMP), mixed liquor suspended solids (MLSS), hydraulic retention time (HRT) and food-to-microorganisms (F:M) ratio were compared against membrane permeability. The results showed positive correlation for TMP, weak positive correlation for HRT and F:M ratio, and approached zero correlation for MLSS. A potential degree of long term irreversible fouling was also detected, although it was not possible to determine how much of it was recoverable with modified chemical cleaning procedures and improved plant operation.
Proposed further work included research on inorganic fouling. The ultimate aim of this paper is to contribute to strengthen the confidence of end users and decision-makers towards MBR.

Keywords Membrane bioreactor; fouling control; full-scale; long-term

NOMENCLATURE

A	Area, m^2
BOD	Biochemical oxygen demand, kg O_2 m^{-3}
F:M	Food-to-microorganisms ratio, s kg_{MLSS} kg_{BOD}^{-1}
HRT	Hydraulic retention time, s
J	Flux, m s^{-1}
MLSS	Mixed liquor suspended solids, kg m^{-3}
Q	Permeate flow, m^3 s^{-1}
SRT	Solids retention time, s
SS	Suspended solids, kg m^{-3}
TMP	Trans-membrane pressure, Pa

INTRODUCTION

Westbury MBR in the UK was designed to assist the existing trickling filters in treating increased domestic flows and the industrial effluent produced by a new dairy plant. The combined effluent from the conventional works and the MBR would be compliant with increasingly stringent discharge consents without the need to upgrade or replace the conventional plant. The MBR consisted of 4 reactors in parallel, each of them provided with fine bubble pre-aeration and Kubota flat-sheet membrane technology. Approximate design flow and load were 5,000 m^3d^{-1} and 2,500 kg biochemical oxygen demand (BOD) per day respectively. The carbonaceous organic load entering the MBR was calculated from periodic BOD analysis and daily permeate flows, and ranged from 450 to 3,800 kg BOD per day, 1,300 kg BOD being the daily average. Hydrodynamic design set points were 54 Ls^{-1} maximum permeate flow, 0.65 $m^3m^{-2}d^{-1}$ maximum flux (0.46 $m^3m^{-2}d^{-1}$ average) and 0.1 bar maximum TMP. Analysis was made possible by the availability of abundant telemetry data for the last 3 years, making this site ideal for analysing trends on a commercial MBR plant.

MATERIALS AND METHODS

Compilation of historical data

Available trending data at Westbury MBR was downloaded from the operating company's telemetry system. Seven parameters were considered for each tank (Figure 1):

- *Dissolved Oxygen (DO) (1)*, measured via a sensor located beneath surface level in each membrane tank,
- *Permeate flow (2)*, measured by means of a magnetic flow meter downstream the permeate collection manifold and the permeate control valve,
- *Permeate valve position (3)*, registered as % opening by the valve actuator,
- *Permeate pressure (4)*, measured using a pressure transducer located upstream the permeate automated valve,
- *Trans-membrane pressure (TMP)*, calculated through the programmable logic controller (PLC) as the difference between the inlet pressure transducer (5) and each permeate pressure transducer,
- *Recycled Activated Sludge (RAS) flow (6)*, measured via a magnetic flow meter after each RAS pump,
- *RAS valve position (7)*, registered as % opening by the valve actuator.

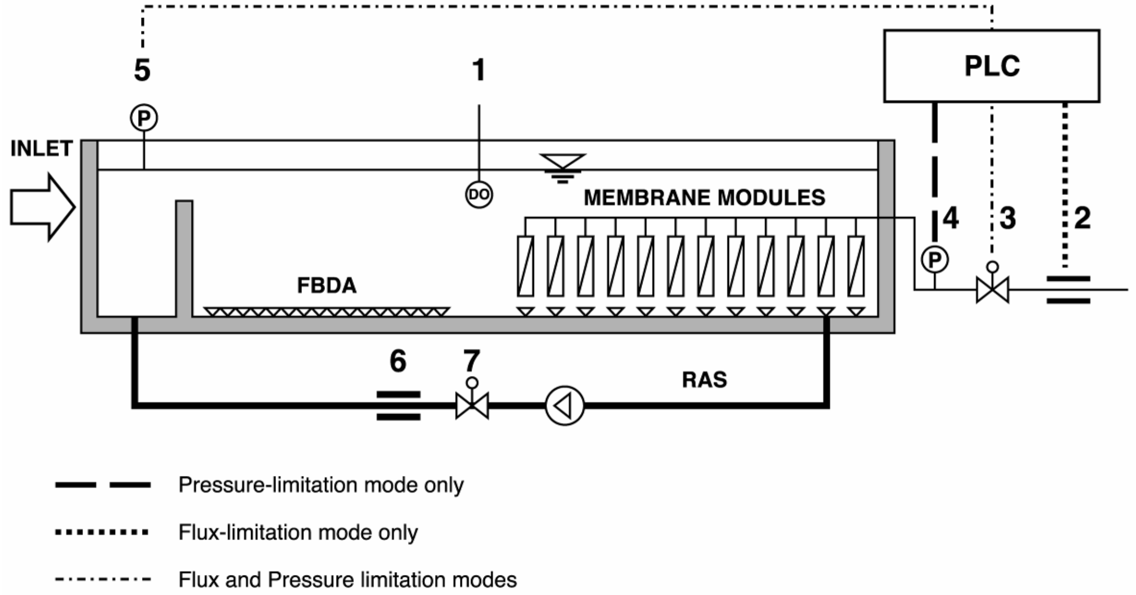

Figure 1 Diagram showing an MBR tank with control equipment.

Each set of data consisted of a series of instantaneous readings registered in real time by the telemetry system every 15 minutes. Most parameters had available records from March 2003 to May 2006, making it a 3-years'-worth database.

Additionally, analytical results were available at four different points of the process: dairy influent, combined influent, liquors and permeate. Basic process parameters such as BOD, suspended solids (SS) or pH were measured daily at the dairy influent, whereas MLSS in the liquors and routine discharge consents were checked once per fortnight and once per week respectively. Operators' log books from the works provided chemical cleaning dates, tasks schedules and de-sludging rates for the last 2 years.

Two modes of operation were available, as shown on Figure 1:

- Flux-limitation mode matched the permeate flow to the liquid level by regulating the valve aperture,
- Pressure-limitation mode maintained TMP below a setpoint (typically 0.1 bar) by adjusting the valve opening.

Data processing protocol

Due to the vast amount of labour work required to process the raw data, the analysis was firstly limited to a single tank, under the assumption that all four tanks had the same pattern of functioning. Raw data entries were organised chronologically and arranged into tables so as to allow comparisons between parameters. Excessive scatter was avoided by filtering-out transient periods during which parameters get adjusted (e.g. valve variations or physical clean protocols) and by ranging the data in accordance with the set points specified on the PLC. To illustrate this, any erroneous readings such as negative inlet pressure would be discarded with this protocol.

When pressure data were not available, these were estimated using the preset look-up table to which the PLC executes. This table unequivocally correlates the tanks' liquid level and the permeate flow under flux-limiting operation mode.

Permeability

Instantaneous values for permeability were obtained from the filtered data using equation [1],

$$\text{Permeability (m (Pa s)}^{-1}\text{or l (m}^{2}\text{ h bar)}^{-1}\text{)} = \frac{\text{Flux (m}^{3}\text{ m}^{-2}\text{ s}^{-1}\text{)}}{\text{TMP (Pa)}} \qquad [1]$$

Flux and TMP were previously obtained out of equations [2] and [3] respectively,

$$\text{Flux (m}^{3}\text{ m}^{-2}\text{ s}^{-1}\text{)} = \frac{\text{Flow (m}^{3}\text{ s}^{-1}\text{)}}{\text{total membrane area (m}^{2}\text{)}} \qquad [2]$$

$$\text{TMP (Pa)} = \text{Inlet pressure (Pa) - Permeate pressure (Pa)} \qquad [3]$$

where total membrane area in one single tank is calculated as 12 modules of 200 panels each, each panel being 0.8 m^2 surface area. The design threshold value for Westbury MBR was **260 LMH bar^{-1}**. This was obtained as the quotient of the design flow rate for the whole plant (54 Ls^{-1}), the total membrane area (7,680 m^2) and the maximum recommended operational TMP for Kubota flat sheet membranes (0.1 bar). Figure 2 shows an increase in permeability after a chemical clean. Each point is a 12-hour averaged value calculated from instantaneous readings taken every 15 minutes. Short term variations were generally due to differing daily flow and loading conditions and other operational variables (DO, temperature, etc) whereas the longer term trend reflected the underlying membrane fouling behaviour.

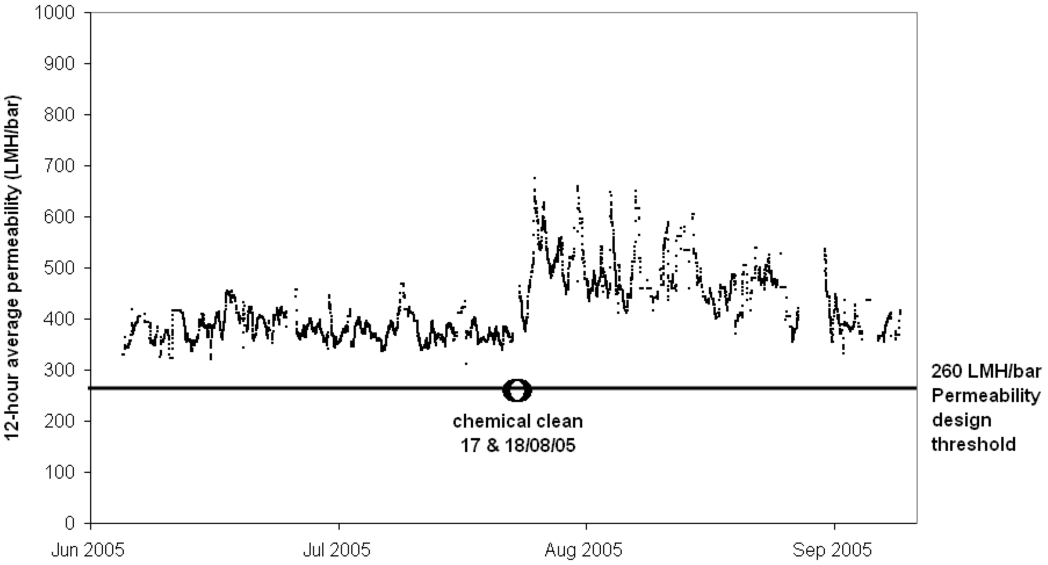

Figure 2 Long-term average permeability for Tank 2.

Fouling analysis

The abundance of data provided a unique opportunity to look for long-term trends in MBR operation. Parameters with potential influence over fouling were averaged and plotted against permeability using 3-years' worth data. The fouling factors considered were as follows:

- TMP
- MLSS concentration
- HRT
- F:M ratio

Other parameters with proven influence on fouling could not be fully assessed because of incompleteness or lack of relevant data. These were aeration rate, solids retention time (SRT), temperature and viscosity.

RESULTS

TMP

Average TMP readings were plotted against 3-year timescales for two extreme scenarios:

- Recently recovered membranes, which showed high permeability (>800 LMHbar^{-1} on average) and high flux (>0.41 m^{3}m^{-2}d^{-1} on average).
- Severely fouled membranes, which showed low permeability (<260 LMHbar^{-1} on average) and high flux (>0.41 m^{3}m^{-2}d^{-1} on average).

As per the equations shown in Figure 3, TMP increased 3 times more rapidly in a severely fouled membrane than in a recovered one. Although significance was high for a linear correlation (correlation coefficients or R^2 were 0.997 and 0.827) , more data points would be required to confirm this hypothesis.

The data indicated a significantly greater rate of long term irreversible fouling at higher TMP, emphasising the importance of operation at lower pressures to optimise performance. The lower curve indicated the potential irrecoverable fouling of the membranes over the period. The figures showed a rise of up to 0.0036 bar/year (~36 mm of water head per year) under the prevailing flow conditions.

Figure 3 showed that some long term irreversible fouling had occurred at Westbury MBR over a 4-year period. As defined by Judd (2006), irrecoverable or absolute fouling is the long-term and insidious fouling that cannot be removed by any cleaning regime. However, the extent that the fouling observed was truly irrecoverable could only be determined following more intensive/alternative cleaning tests. Hypothetic irrecoverable foulants could be inorganic compounds such as scaling caused by dairy influent (Churchouse and Wildgoose, 1999) and/or ferric sulphate dosed for phosphorus (P) removal.

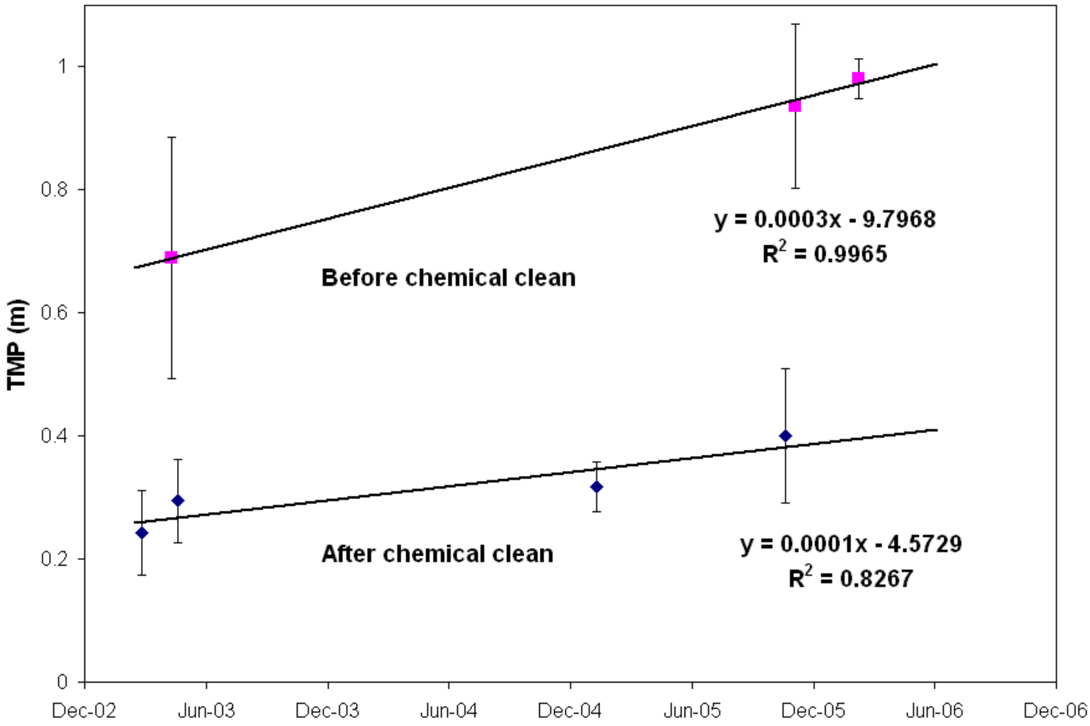

Figure 3 Observed long term change in TMP over time before and after chemical cleaning

MLSS concentration

An analysis of permeability against MLSS at design flux showed no influence between parameters. The fouling state seemed not to be influenced by the sludge concentration within the operating range of the plant over the period (figure 4).

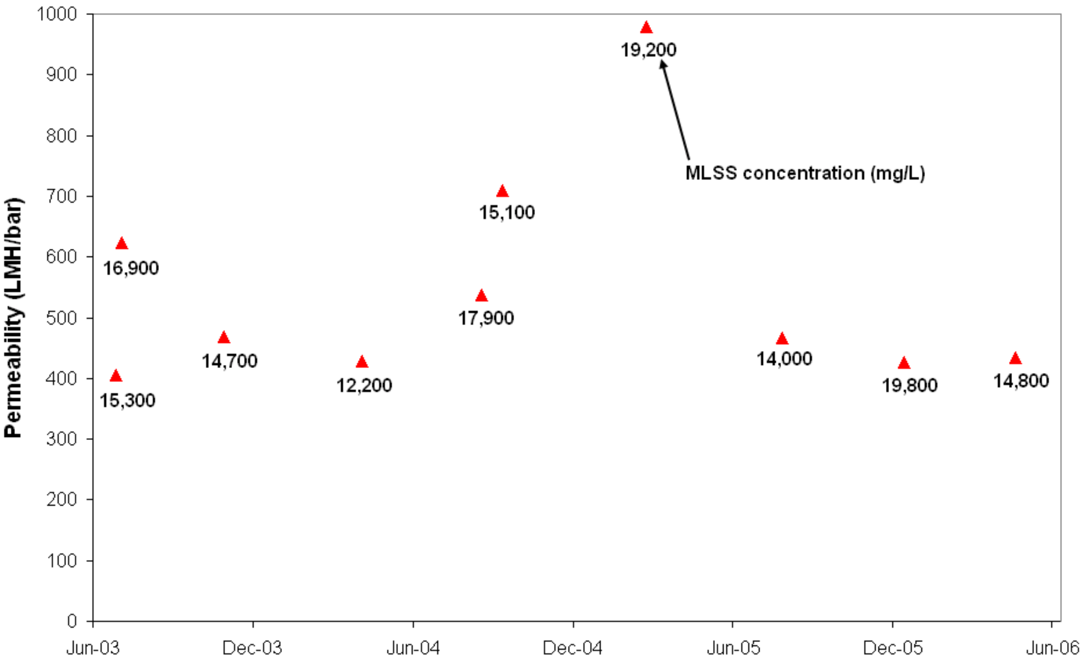

Figure 4 Permeability and MLSS approaching zero correlation.

HRT

Long-term trending data distribution showed that most records were in the HRT region between 13 and 20 hours. It was found that the permeability trend was to increase with HRT between 15 and 20 hours, however no definite linearity was found. From 25 hours onwards permeability increased linearly ($R^2 = 0.956$) and had a tendency to stabilize around 700 LMH bar^{-1} at 30 hours HRT.

F:M ratio

It was found that typical F:M values for Westbury MBR were between 0.02 and 0.04 kg BOD per kg MLSS, although values up to 0.12 had been observed. Permeability was noted to increase from 350 to 500 LMH bar^{-1} when F:M increased from 0.04 to 0.10 kg BOD (kg MLSS $day)^{-1}$. However, linear regression significance was rather weak ($R^2 = 0.417$).

DISCUSSIONS

Permeability vs. TMP.

The increased fouling rate detected at high TMP suggests that low TMP is a more sustainable operation mode. TMP control is possible with more frequent chemical cleans or different chemical cleaning solutions. This can be combined with limiting the maximum TMP possible by both software and/or mechanical means (raising pipework, or lowering hydraulic level). One drawback of altering the frequency of chemical cleans is the risk of reducing the membranes' life (Parameshwaran *et al.*, 2001). The use of different chemical solutions may have a positive influence on recovery; unlike sodium hypochlorite, oxalic acid can remove inorganic fouling (personal communication, Mr. H. Sakai, August 18, 2006). Limiting the maximum liquid level might have further effects on solids production, HRT, SRT and F:M ratio.

Permeability vs. MLSS.

The lack of influence of MLSS on fouling in this paper agrees with the general scientific knowledge compiled by Judd (2006). However, experimental error in MLSS measurements or the effect of non-monitored parameters such as temperature (Chiemchaisri and Yamamoto, 1994) might hinder a positive correlation. MLSS at Westbury MBR had been varying between 14 and 18gL^{-1} and no trends in permeability were observed. This partially contradicted the conclusions of Howell *et al.* (2004) and Melin *et al.* (2006), who suggested that optimal MLSS operating windows should not exceed 15gL^{-1} because of risk of membrane fouling. However, sludge filterability varies considerably between different MBR pants depending on operating conditions and effluent type so one rule for all is unlikely (personal communication, Dr S. J. Churchouse, September 1, 2006).

Permeability vs. HRT.

As pointed out by Chae *et al.* (2006) and Leiknes *et al.* (2006), HRT may be closely linked to the propensity to release extra-cellular polymeric substances (EPS). Although the increasing interest in HRT as a main factor in fouling limitation, the design of Westbury MBR did not allow modulation of this parameter. The hydraulic retention time was completely subordinated to the control system's flux-regulating strategy. When tanks were full, HRT was around 12 hours. For lower levels, the flow was reduced and HRT could increase up to 50 hours. The trends observed in this study were largely linked to the flux (or flow) rate rather than biology. An experiment to separate the effect of HRT from flux and F:M ratios could be carried out, but being a full scale operational plant, such a test at Westbury would need to be carefully controlled if permission were given.

Permeability vs. F:M ratio.

F:M ratio is a process parameter that usually complements SRT (Metcalf & Eddy, 2003). Normal observed conditions for Westbury MBR sludge were F:M ratio 0.02-0.04kg BOD (kg MLSS)$^{-1}$ and 47 days SRT. This review showed that higher F:M ratios up to 0.10kg BOD (kg MLSS)$^{-1}$ improved permeability. According to the combined work of various authors (Lee *et al.*, 2003; Han *et al.*, 2005; Grelier *et al.*, 2006) the optimal SRT range for fouling minimization should be around 15-20d. Further research combined with SRT readings might be required.

CONCLUSIONS

Westbury MBR correlation analysis, based on long-term recorded data, revealed a number of process trends that influenced the membranes' permeability;

- High TMP significantly accelerated fouling over time. Albeit a good correlation coefficient backing up this result, more data points were a requisite to confirm the hypothesis.
- Irrecoverable fouling was identified. Although its nature was not revealed, it would possibly hold some relation with inorganic compounds. Hypotheses could be scaling caused by dairy influent and/or ferric sulphate dosed for P removal.
- No influence of MLSS was observed upon filtration performance within the plant's normal operation range. This may confirm that sludge concentration alone is a poor indicator of fouling propensity.
- Certain positive linearity was observed between permeability and HRT. This could be largely related to MBR design, as HRT was closely linked to the reciprocal of the flow rate.
- Permeability was detected to increase from 350 to 500 LMH bar^{-1} when F:M increased from 0.04 to 0.10 kg BOD (kg MLSS day)$^{-1}$. The correlation was rather weak, but does suggest a possible relationship.
- Effluent quality remained constantly high during the 4 years of operation.

REFERENCES

Chae, S.R., Ahn, Y.T., Kang, S.T. and Shin, H.S. (2006). Mitigated membrane fouling in a vertical submerged membrane bioreactor (VSMBR). Journal of Membrane Science, Article in press.

Chiemchaisri, C. and Yamamoto, K. (1994). Performance of membrane separation bioreactor at various temperatures for domestic wastewater treatment. Journal of Membrane Science, 87 119-129.

Churchouse, S. and Wildgoose, D. (1999). Membrane bioreactors progress from the laboratory to full-scale use. Membrane Technology, 111 4-8.

Grelier, P., Rosenberger, S. and Tazi-Pain, A. (2006). Influence of sludge retention time on membrane bioreactor hydraulic performance. Desalination, 192 10–17.

Han, S.S., Bae, T.H., Jang, G.G. and Tak, T.M. (2005). Influence of sludge retention time on membrane fouling and bioactivities in membrane bioreactor system. Process Biochemistry, 40 2393–2400.

Howell, J.A., Chua, H.C. and Arnot, T.C. (2004). In situ manipulation of critical flux in a submerged membrane bioreactor using variable aeration rates, and effects of membrane history. Journal of Membrane Science, 242 13–19.

Judd, S. (2006). The MBR book. Elsevier, Oxford.

Le-Clech, P., Fane, A. and Leslie, G. (2005). MBR focus: the operators’ perspective, Filt., Sep. 42, 20-23.

Lee, W., Kang, S. and Shin, H. (2003). Sludge characteristics and their contribution to microfiltration in submerged membrane bioreactors. Journal of Membrane Science, 216 217–227.

Leiknes, T., Ivanovic, I. and Ødegaard, H. (2006). Investigating the effect of colloids on the performance of a biofilm membrane reactor (BF-MBR) for treatment of municipal wastewater. In: The Water Institute of Southern Africa (WISA) Biennial Conference & Exhibition, Durban, South Africa, 21st - 25th May 2006, 26 - 36.

Melin, T., Jefferson, B., Bixio, D., Thoeye, C., De Wilde, W., De Koning, J., van der Graaf, J. and Wintgens, T. (2006). Membrane bioreactor technology for wastewater treatment and reuse. Desalination, 187 271–282.

Metcalf & Eddy. (2003). Wastewater Engineering Treatment and Reuse. 4th international ed. McGraw-Hill, New York.

Parameshwaran, K., Fane, A. G., Cho, B. D. and Kim, K. J. (2001). Analysis of microfiltration performance with constant flux processing of secondary effluent. Water Research, 35(18), 4349–4358.

Membrane bioreactor biological nutrient removal activated sludge systems: Performance and kinetics

Valentina Parco[*,**], Geoff du Toit[**], Mark Wentzel[**], and George Ekama[**]

[*] Hydraulic and Environmental Engineering Department, University of Palermo, 90100, Italy
(Email: parco@idra.unipa.it)
[**]Water Research Group, Civil Engineering Department, University of Cape Town, Rondebosch, 7701, South Africa (Emails: *geoff.dutoit@shands.co.za*; *MARKW@EBEFAC.uct.ac.za*; *George.Ekama@uct.ac.za*)

Abstract
A membrane bioreactor (MBR) and a conventional (CAS) UCT lab-scale biological nutrient removal (BNR) activated sludge system were operated in parallel and set up to have identical design parameters such as reactor mass fractions, recycles sludge age, and influent feed strength and composition. The performances of the two systems were extensively monitored and compared to identify and quantify the influence of the membranes on system response. From batch tests on mixed liquor drawn from the two BNR systems, the kinetics rates were delineated. In terms of COD, TKN, FSA (free and saline ammonia), total nitrogen, total soluble phosphorus, TSS and faecal coliforms, the MBR system produced an effluent that is equivalent or superior to the conventional system but had a greater sludge production. It was found that the specific nitrification rates differed between MBR and CAS BNR activated sludge systems, and that the rate in the MBR system was influenced by the mixed liquor solids concentration, probably due to ammonia diffusion limitations. In contrast to the nitrification process, for the denitrification and P removal processes, the kinetics and stoichiometry incorporated in the design procedures and simulation models for CAS BNR systems can be applied directly to MBR BNR systems..

Keywords
Membrane Bioreactor, Biological Nutrient Removal Systems, Nutrient removal kinetics

INTRODUCTION

With increasingly stringent water quality standards internationally for discharge and reuse (Howell, 2004) there is a need for wastewater treatment plants to provide effluents of a reliable and excellent standard. In recent years, new membranes specially developed for the use in waste water treatment have made Membrane BioReactors (MBRs) become a promising alternative to the well-known Biological Nutrient Removal Conventional Activated Sludge (BNR-CAS) systems.

Compared to CAS systems, MBRs offer several advantages: possibility of elevated solids concentrations resulting in reduced reactor volume requirements; high quality treated effluent for possible re-use; insensitivity to sludge flocculation, settleability and filamentous bulking; lower footprint due the omission of secondary settling tanks; favourable conditions for growth of organisms with low growth rates; and compactness of the whole system. These advantages, coupled with the robustness, simplicity to operate and increasing affordability of membranes, are making membranes increasingly attractive as the solid/liquid separation process of choice in the BNRAS systems.

For CAS BNR systems, considerable knowledge has been accumulated on their performance, design and operation. However, the correct application of these existing design procedures and models to MBR systems requires additional information about the population composition and kinetics in BNR MBRs. Much of the research has been focused on the membrane performance, to understand the mechanisms of fouling and minimize them thus improving the lifespan of membranes and their cost, or on the membrane performance in removing organic compounds or nitrogen compounds. However only a limited number of studies have been conducted on BNR system performance with membranes, despite speculation that the inclusion of membranes may affect the nature of the activated sludge (AS) biomass, such as (i) floc structure (Zhang et *al*., 1997; Huang et *al,*. 2001; Yamamoto et *al*., 2002; Gao et *al*., 2004; Manser et *al,*. 2005a), (ii) bacterial communities (Ghyoot et *al*., 1999; Luxmy et *al.,* 2000; Liebig et *al,*. 2001; Smith et *al.,* 2002;

Manser et *al,*. 2005b), (iii) metabolic activities (Witzig et *al,*. 2002; Rosengerber et *al*., 2002; Lee et *al*., 2003; Han et *al*., 2005; Sperandio et *al.,* 2005; Li et *al*., 2005) and (iv) sludge production (Cicek et *al*., 1999; Smith et *al.,* 2002; Holbrook et *al.,* 2005; Monti et *al.,* 2005). The impact of the membranes on the design approach and concepts for BNRAS systems also has been established (Ramphao et *al*., 2005). However, possible impacts on the operation and performance (N and P removal) of the BNRAS system remain to be quantitatively determined.

Accordingly, the aims of this study were to investigate the long-term treatment performance of parallel conventional and membrane BNRAS systems and to investigate the impact of including membranes for solid liquid separation on the kinetics of BNR-AS systems.

MATERIAL AND METHODS

Two parallel lab-scale membrane (MBR) and conventional (CAS) activated sludge systems (Figs. 1 and 2) were operated, in the Water Research Laboratory at UCT (South Africa), for 449 days under laboratory conditions allowing their behaviour to be monitored and their performance compared.

Both systems were in UCT configurations (Fig 1, Table 1) so that denitrification and BEPR (Biological Excess Phosphorus Removal) could function independently, provided the recycles do not overload the anoxic reactor with nitrate. System design and operational parameters such as zone mass fractions, inter-reactor recycles and sludge ages were kept the same in both systems (Table 1).

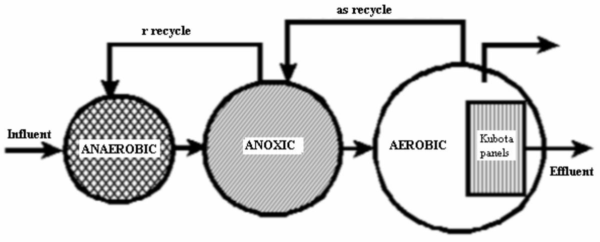

Figure 1 Schematic layout of MBR UCT system

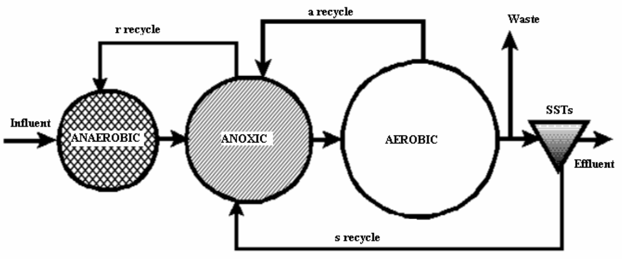

Figure 2 Schematic layout of CAS UCT system

MBR-UCT system was an adaption of the UCT configuration with membranes (Kubota® panel) in the aerobic reactor replacing the function of an SST.

Table 1 MBR and conventional UCT systems' design and operating parameters..

System parameters	MBR UCT	CAS UCT
Sludge age (d)	20	20
Anaerobic (R1) volume (ℓ)	19	5.6
Anoxic (R2) volume (ℓ)	21	6.2
Aerobic (R3) volume (ℓ)	35	13.2
Anaerobic (R1) mass fraction (%)	12.6[1]	12.6[1]
Anoxic (R2) mass fraction (%)	27.9[1]	27.9[1]
Aerobic (R3) mass fraction (%)	59.5[1]	59.5[1]
a-recycle (R3 to R2)	3:1	2:1
r-recycle (R2 to R1)	1:1	1:1
s-sludge Return Recycle (SST to R2)	-	1:1
Hydraulic retention time (d)	0.53	1.67
MLVSS concentration (mg/ℓ)	12 500	3 600
MLTSS concentration (mg/ℓ)	18 000	5 000
Influent flow (ℓ/d)	140	15
Feed COD concentration (mg/ℓ)	1000	1000
Membrane flux ($m^3/m^2/d$)	0.239	

[1] For the given a- and r-recycle ratios

The systems were fed screened (1mm mesh) raw unsettled municipal wastewater (800 mgCOD/ℓ) from the Mitchell's Plain Wastewater Treatment Plant (Cape Town, South Africa), augmented with sodium acetate (200 mgCOD/ℓ, to accentuate BEPR), ammonia (20 mgN/ℓ, to increase TKN/COD), phosphorus (sufficient to ensure systems not P limited) and sodium bicarbonate (to provide alkalinity for pH buffering). The sewage was collected in batches, stored in stainless steel tanks at 4°C and served as feed for both systems for 10 to 14d.

The two systems were monitored daily via the parameters in Table 2. Additionally, recycle flow rates and transmembrane pressure (TMP, constant flux) were monitored daily. Once monthly mixed liquor samples were analysed by a microbiologist for filament identification and floc morphology. Moreover the samples were sent fortnightly to the Durban Institute of Technology for FISH analysis (Maharaj et *al.*, 2006). Sewage readily biodegradable COD (RBCOD) was measured daily according to Ekama et *al.* (1986).

Table 2 Sampling position and parameter measurement.

Test	**COD**	**TKN**	**FSA**	**NO_3**	**NO_2**	**T-P**	**TSS**	**VSS**	**OUR**	**DSVI**	**pH**
Influent	F; UF	UF	F			UF					
Anaerobic				F	F	F	UF	UF			
Anoxic				F	F	F	UF	UF			
Aerobic	UF	UF		F	F	F	UF	UF	D	D	D
Final effluent	F; UF	F; UF	F	F	F	F; UF					

F = 0.45 µm filtered; UF = Unfiltered samples; D = Direct measurement taken. COD; TKN; FSA (Free and Saline Ammonia); T-P (Total Phosphorus); TSS; VSS (Standard Methods, 1985). DSVI = Dilute Sludge Volume Index; (Ekama and Marais, 1984); OUR = Oxygen Utilization Rate (Randall et *al*., 1991).

For each sewage batch (steady-state period) the daily results were averaged (after analysis for outliers). These steady-state averages were used to assess the performance of the systems and the following process characteristics were calculated: system COD and N mass balances; influent unbiodegradable soluble and particulate COD fractions ($f_{S,us}$ and $f_{S,up}$ respectively); RBCOD fraction, mixed liquor VSS/TSS, COD/VSS and TKN/VSS ratios; nitrate and P mass changes across each reactor and sludge production (Parco V., 2006; Du Toit G. et *al*., 2006).

To determine the kinetics rates, specific batch tests on the mixed liquor harvested from the two BNR systems, were run (Parco, 2006; Parco et *al*., 2006, 2007). Particularly on the MBR BNR system the influence of the biomass concentration and of the limitation of substrate concentrations on biological activity were examined. Moreover, to provide additional information on the anoxic behaviour of phosphate accumulating organisms (PAO), the ability of AS in MBR BNR systems to denitrify under anoxic conditions with simultaneous phosphate uptake was investigated and quantified.

RESULTS AND DISCUSSION

System responses

The average removals and effluent quality of both the MBR and conventional UCT systems are summarised in Table 3.

Table 3 Summary of the influent and effluent qualities, and the resultant removals, of both UCT systems.

Parameter		Influent	MBR UCT		CAS UCT	
			Effluent	Efficiency	Effluent	Efficiency
COD	mgCOD/l	950,.9	41 (115^1, 69^2)	95.70% (88%^{1}, 92.7%^{2})	73.4^1 (50.7^2)	92.3%^{1} (94.7%^{2})
TKN	mgN/l	106.5	1.52	98.60%	3.34^1 (2.24^2)	96.9%^{1} (97.97%^{2})
FSA	mgN/l	81.5	0.74	99.10%	0.99	98.8%
NO3	mgN-NO_3/l	0	16.53	-	16.8	-
TN	mgN/l	106.5	18.05	83.10%	20.1	81.10%
TP	mgP/l	30.7	8.4	22.5 mgP/l_{inf}	12.06	17.4 mgP/l_{inf}
TSS	mgTSS/l	N/A	0	-	19.3	-
E. coli	CFU/100ml	N/A	0	-	2250	-

[1] unfiltered sample; [2] 0.45 filtered sample; N/A = value not available

The MBR UCT system exhibited removals that were equivalent or superior to that produced by the conventional UCT system (Table 3).

The COD removal efficiency of the MBR system (96%) was superior to that of the CAS system (92% unfiltered, 95% 0.45 µm filtered). However the MBR UCT system produced an effluent 0,45 µm filtered (measured from the supernatant of the MBR aerobic sludge) COD concentration that was consistently higher than the effluent filtered (0.45 µm) COD concentration in the conventional UCT system, Table 3. Similarly the MBR unfiltered "effluent" COD values (measured from the supernatant of the DSVI test on MBR aerobic sludge) were consistently higher than those from the CAS system.

From these observations, the following aspects can be highlighted:

- the membranes, because of smaller pore size, retain organics that would be considered "soluble" in a conventional system with SSTs;
- despite the total COD removal of the MBR showed better performance due to the filtration effect of the membranes, in terms of biological removal the CAS system seem to have better performance. The FISH analysis results confirmed this observation since the samples of MBR system on average has indicated that only 50% of total DAPI (total DNA) was eubacteria (EUB probe) compared to 80% in the CAS system;
- although the nominal pore size of the Kubota® membranes used in this study were 0.4µm, the considerably lower MBR effluent COD (41 mgCOD/l) than the 0.45 µm filtered COD (69 mgCOD/l) indicates that a dynamic gel layer forms on the membrane which reduces their effective pore size.

The differences in the MBR UCT effluent COD and the conventional UCT effluent are accommodated in the steady-state and dynamic simulation models as differences in the soluble unbiodegradable COD fractions ($f_{S,us}$) which were 0.045 and 0.066 respectively.

The MBR UCT system produced a solids free effluent, whereas in the conventional UCT system there was a continual loss of solids to the effluent (19.3 mgTSS/ℓ), quantified as the difference between filtered and unfiltered effluent COD concentrations (Table 3) and via the VSS/COD and VSS/TSS ratios. This confirms that the MBR UCT system effluent quality is independent of the flocculation characteristics of the mixed liquor. The microbiological quality (faecal coliforms) of the MBR UCT system effluent was superior, the results indicated pathogen counts were not detectable in the MBR UCT system whereas in the conventional UCT system pathogen counts an average of 2250 CFU/100ml (Table 3). Clearly, from the removals described above, due to complete retention of solids, and pathogens, the membrane effluent has a higher quality for reuse purposes. The TKN removal efficiency of the MBR system was marginally better than that of the CAS system (Table 3). This is again attributed to the retention of solids by the membranes that were lost in the effluent of the CAS system. In both systems, nitrification was virtually complete, as indicated by the low residual free and saline ammonia concentrations and similar total N removals was exhibited, indicated by the similar effluent nitrate concentrations (Table 3).

In both systems TP was dosed in excess of the amount the system could remove in order to observe the maximum BEPR possible. Thus P removal performance is represented by P removals. System average P removals of 22.5 mgP/l_{inf} and 17.4 mgP/l_{inf} were achieved in the MBR and CAS systems indicating that total P removal would have been possible in both systems with influent P concentrations of up to ~23 mgP/l_{inf} and ~18 mgP/$l_{inf.}$ Clearly however, the P removal performance of the conventional UCT system was inferior to that of the MBR system. Reasons for this difference was because anoxic P uptake was more prevalent in the CAS system with 22.1% of P total uptake in the anoxic reactor compared with the MBR system with 8.5%. With anoxic P uptake by denitrifying PAOs, significantly reduced BEPR has been reported (Ekama et *al*., 1999; Hu et *al*., 2002) probably due to less efficient utilization of stored COD by PAO (Hu et *al*., 2002). This was also evident in this investigation in the 2 systems and in P removal batch tests results (Parco, 2006; Parco et *al.*, 2007). The nitrate load on the anoxic reactor of the MBR system was less than this reactor's denitrification potential identified by zero nitrate concentrations in this reactor. In contrast, denitrification was not complete in the anoxic reactor of the CAS system, identified by non-zero nitrate concentrations in this reactor. This was the main factor stimulating anoxic P uptake in the CAS system. Moreover, the loading of nitrate on the anaerobic reactor from the anoxic reactor in the CAS reduced the RBCOD available for PAOs and consequently additionally reduced P removal. The observations above suggest that the nitrate load in the CAS should have been reduced, via reducing the a-recycle ratio to maximize BEPR. While reducing the a-recycle ratio does not change the anoxic mass fraction and denitrification potential, the N removal performance will decrease, i.e. more nitrate out with the effluent rather than recycled to the anaearobic reactor. In contrast, in the MBR UCT system, from the design procedures of Ramphao et *al*., (2005) reducing the a-recycle ratio causes a corresponding reduction in the anoxic mass fraction and nitrate load and hence makes greater decrease on N removal performance. This highlights the conflicting requirements between N and P removal in BNRAS systems at elevated influent TKN/COD ratios, which is by no means new but operates differently in the MBR BNR configuration. The balance between optimizing N and P removal will be influenced by a number of factors, such as influent TKN/COD ratio, N and P removal requirements, influent RBCOD.

At present anoxic P uptake BEPR is not explicitly incorporated in the steady-state design procedures for BEPR systems (Wentzel et *al*., 1990), as quantitative relationships linking the extent of anoxic P uptake to the system design or operational parameters have not been established.

For all the duration of the investigation, as regards COD, N and P removal, the MBR system showed a greater stability than the CAS system (Parco, 2006).

The MBR UCT system had a greater sludge production per COD load (0.31 kgVSS wasted/kgCOD load) than the conventional UCT system (0.20 kgVSS wasted /kgCOD load). This increased sludge production can be explained in part by the retention of solids that would normally be lost to the effluent in a conventional system with SSTs, and that the membranes retain unbiodegradable organics that would be considered "soluble" in a system with SSTs (above) and hence also would be lost to the effluent. Additionally other factors could contribute to the higher sludge production in the MBR system:

- the higher P removal in the MBR UCT system suggests a greater PAO population which would produce more sludge per unit influent COD than OHOs due to their lower endogenous respiration rates (Wentzel *et al*., 1990);
- particulate organics that are biodegradable in the conventional UCT system are no longer biodegradable in the MBR system due to factors such as high MLSS concentrations, or different floc morphology.

More comprehensive data at different sludge ages are required to determine if this increase is consistent, and to identify the underlying cause.

In the steady-state design procedures, the increased sludge production in MBR UCT systems is accommodated by increasing the influent unbiodegradable particulate COD fraction ($f_{S,up}$), determined as 0.241 mgCOD/mgCOD versus 0.084 mgCOD/mgCOD for the conventional UCT system. Similarly the unbiodegradable soluble COD fraction ($f_{S,us}$) must be decreased to account for the additional retention of "soluble" COD which is attributed to the finer membrane pore size.

In the literature previous studies comparing conventional and MBR BNR systems run under the same operating conditions (sludge age, influent wastewater) have indicated similar results (Cicek et *al*., 1999; Smith et *al*., 2002; Holbrook et *al.,* 2005).

Nutrient removal kinetics

Nitrification rates were determined from aerobic batch tests conducted on mixed liquor drawn from the two systems. In the MBR BNR system: 1) as the sludge concentration was increased, the specific nitrification rate (and $\mu_{max,n}$) decreased, apparently due to ammonia transfer limitations and 2) as the ammonia concentration was increased for similar sludge concentration, the specific nitrification rates (and $\mu_{max,n}$) increased, in agreement with the observations above of possible ammonia transfer limitations. Additionally at the same mixed liquor concentration the conventional BNR system exhibited higher specific nitrification rates than the parallel MBR BNR system, apparently due to different selection pressures imposed by membranes versus SSTs (Parco, 2006, Parco et *al.*, 2006).

To determine the specific denitrification constants K (mgNO_3-N/d per mg active organism (OHO or PAO) VSS), nitrate utilization rates (NUR) were measured in anoxic batch tests The results obtained with different initial concentrations of added nitrate, show the K_{2OHO} denitrification rate to be zero order with respect to nitrate concentration, in agreement with past work (van Handel et *al.*, 1981, Clayton et *al.*, 1991; Ekama et *al.*, 1999). For the MBR system, the K_{2OHO} rates observed at different mixed liquor concentrations indicate no effect of ML on the rate, in contrast to the observations in the nitrification tests. From all the batch tests on MBR system, an average K_{2OHO} = 0.264 mgNO_3-N/(mgOHOVSS d) was obtained. This is very close to the range of values in the literature for conventional BNR systems, e.g. 0.255 (Clayton et *al.*, 1991, see Ekama et *al.*, 1999). Moreover the data relating to the denitrification rates in the MBR and Conventional systems indicate that the systems exhibited similar specific denitrification rates (Parco, 2006; Parco et *al.*, 2007).

To determine the biological excess phosphorus removal (BEPR) kinetics, batch test were conducted on the mixed liquor drawn from the UCT systems. The mixed liquor was exposed to definite sequential phases: in the first phase the anaerobic condition was established, after that the mixed liquor was divided in two parts, one exposed to aerobic condition and the other exposed to anoxic condition. From the batch tests results seems that the BEPR was mediated by 2 different PAO populations: denitryfing PAO and aerobic PAO. The specific denitrification rates of OHOs were significantly higher than those of PAOs, to confirm the greater affinity of OHOs than PAOs for nitrate (Parco, 2006; Parco et *al.*, 2007). The anaerobic P release and HAc consumption rates, the $P_{relase}/HAc_{consumption}$ ratio, as well the aerobic and anoxic P uptake rates obtained for different volatile suspended solids (VSS) concentrations indicate no effect of the sludge concentration on these rates. The results obtained with different concentrations of HAc added show HAc consumption rates to be zero order respect to HAc concentration in agreement with the literature studies, and in conformity with the observations in a single batch test where a constant HAc consumption rate was observed. Further the anaerobic P release and as well the aerobic and anoxic P uptake rates obtained with different concentrations of HAc added indicate no influence of the substrate concentrations on these rates. (Parco, 2006; Parco et *al.*, 2007). The following average values for each rate were obtained: 1) Anaerobic P release: 17.78 mP/gVSS h (69.41 mgP/gPAOVSS h); 2) Anaerobic HAc consumption: 29.27 mHAc/gVSS h (115.12 mgHAc/gPAOVSS h), 3) Aerobic P uptake: 9.77 mP/gVSS h (38.13 mgP/gPAOVSS h), 4) Anoxic P uptake: 2.89 mP/gVSS h (11.42 mgP/gPAOVSS h) and 5) $P_{release}/HA_{cconsumption}$: 0.544 mmolP/mmolC. These values (on the basis of VSS) are very close to the range of values in the literature for conventional BNR systems with mixed cultures and very low compared with investigations on enhanced cultures (Kuba et *al.*, 1994; Kuba et *al.*, 1997; Wachtmeister et *al.*, 1997, Hu et *al.*, 2003). This confirms the importance of specifying kinetic rates of bioprocesses with respect to the actual biomass which performs that particular biodegradation process. Rates with respect to a lumped parameter like VSS are not comparable. The concentration of OHOs and PAOs in the VSS was determined with the aid of the Activated Sludge Model N° 2 (Henze et *al.,* 1995) calibrated to conform to the UCT MBR BNR system performance results.

CONCLUSIONS

This study clearly demonstrates that BNR in membrane activated sludge systems is entirely feasible, and offers considerable advantages over conventional BNR activated sludge systems with SSTs, such as improved effluent quality, operation independent of sludge flocculation or settling characteristics. Thus the membrane effluent is safer and more viable for reuse purposes.

Higher sludge productions were observed in the MBR system. The increased sludge production is explained in part by the retention, by the membranes, of additional COD and non-settleable solids which would otherwise flow through SSTs, however this cannot explain the magnitude of difference in sludge production between the two systems. More comprehensive data possibly at different sludge ages are required.

The introduction of membranes into BNR systems may cause associated changes in the nitrification kinetics of BNR: 1) in the MBR BNR system: i) as the sludge concentration is increased to that present in the system (18 gTSS/l), the specific nitrification rate decreases, apparently due to ammonia transfer limitations and ii) as the ammonia concentration is increased for similar ML concentration, the specific nitrification rates increases, in agreement with the observations above of possible ammonia transfer limitations; 2) at the same mixed liquor concentration the conventional BNR system exhibits higher specific nitrification rates than the parallel MBR BNR system, apparently due to different selection pressures imposed by membranes versus SSTs.

In contrast to the nitrification process, for the denitrification and P removal processes, the kinetics and stoichiometry incorporated in the design procedures and simulation models for CAS BNR systems can be applied directly to MBR BNR systems. The denitrification and P removal batch test results obtained for different ML concentrations indicate no effect of ML on the specific denitrification rates and on the P removal rates. Specific denitrification rates are zero order with respect to nitrate concentration and HAc consumption rates are zero order respect to HAc concentration in agreement with previous observations on conventional BNR systems. Anoxic P uptake has been consistently observed and the existence of 2 group of PAO bacteria has been demonstrated. Anoxic P uptake is detrimental to the BEPR performance in a BNR system. However, quantitative links between design and operational parameters and the extent of anoxic P uptake have not been established. This has hindered incorporation of anoxic P uptake in the design and simulation models for BNR systems, with or without membranes, and requires resolution.

REFERENCES

Cicek N., Franco J.P., Suidan M.T., Urbain V. and Manem J. (1999). Characterization and comparison of membrane bioreactor and a conventional activated sludge system in the treatment of wastewater containing high molecular weight compounds. Wat. Envir. Res. 71(1): 64-70.

Clayton J.A., Ekama GA, Wentzel MC, and Marais GvR, (1991). Denitrification kinetics in biological N and P removal activated sludge systems treating municipal wastewaters. Wat. Sci. Tech. 23: 1025-1035

Du Toit GJG.; Wentzel M.C. and Ekama G.A. (2006). Design and performance of BNR activated sludge systems with flat sheet membranes for solid-liquid separation. MSc (Eng) Thesis, Department of Civil Engineering, University of Cape Town, Rondebosch 7701, RSA.

Ekama G.A., Dold P.l .and Marais Gv.R. (1986). Procedures for determining influent COD fractions and the maximum specific growth of the heterotrophs in activated sludge systems. Wat. Sci. Tech. 18(6), 91-114.

Ekama G.A .and Wentzel M.C. (1999). Denitrification kinetics in biological N and P removal activated sludge systems treating municipal wastewaters. Wat. Sci. Tech. 39(6): 69-77.

Gao M., Yang M., Li H., Yang H. and Zhang Y. (2004). Comparison between a submerged bioreactor and a conventional activated sludge on treating ammonia-bearing inorganic wastewater. Jour. Biotech. 108: 265-268

Ghyoot W., Vandale S., and Verstraete W. (1999). Nitrogen removal from sludge reject water with a membrane assisted bioreactor. Wat. Res.. 33(1): 23-32.

Han S.S., Bae t.H., Jang G.G. and Tak T.M. (2005). Influence of sludge retention time on membrane fouling and bioactivities in membrane bioreactor system. Proc. Biochem. 40: 2393-2400.

Henze M., Gujer W., Mino T., Matsuo T., Wentzel M.C. and Marais Gv.R. (1995). Activated sludge model N°2. IAWQ Scientific and Technical Report N°3, IAWQ, London 32 pp.

Holbrook R.D., Massie K.A. and Novak J.T. (2005). A comparison of membrane bioreactor and a conventional activated sludge mixed liquor and biosolids characteristics. Wat. Envir. Res. 77(4): 323-360.

Howell J.A. (2004). Future of membranes and membrane reactors in green technologies and for water reuse. Desalination 162(1), 1-11.

Hu Z-R, Wentzel M.C. and Ekama G.A. (2002) The significance of denitrifying polyphosphate accumulating organisms in biological nutrient removal activated sludge systems. Wat. Sci. Tech. 46(1/2), 129-138.

Hu J-H., Ong S.L., Lu N. F. and Fan X.J. (2003). A new method of characterizing denitrifying phosphorus removing bacteria by using three different types of electron acceptors. Wat. Res. 37: 3463-3471.

Huang X., Gui P and Qiuan Y. (2001). Influence of sludge retention time on microbial behaviour in a submerged membrane bioreactor. Proc. Biochem. 36(10):1001-1006.

Kuba T., Wachmeister A.., van Loosdrecht M.C.M. and Heijen J.J. (1994). Effect of nitrate on phosphorus release in a biologial phosphorus removal systems. Wat. Sci. Tech. 30: 263-269

Kuba T., van Loosdrecht M.C.M., Brandse F.A .and Heijen J.J. (1997). Occurrence of denitrifying phosphorus removing bacteria in modified UCT-type wastewater treatments plants. Wat. Sci. Tech. 31(4): 777-786.

Lee W., Kang S. and Shin H. (2003). Sludge characteristics and their contribution to microfiltration in submerged membrane bioreactors. J. Membr. Sci. 216: 217-227.

Li H., Gao M., Yang H., Zhang Y, and Kamagata Y. (2005). Comparison of nitrification performance and microbial community between a submerged bioreactor and a conventional activated sludge. Wat. Sci. Tech. 51(6-7):193-200.

Liebig T., Wagner M., Bjerrum L., and Denecke M. (2001). Nitrification performance and nitrifier community composition of a cheostat and a membrane-assisted bioreactor for the nitrification of sludge reject water. Bio. Bioststem Eng. 24: 203-210.

Luxmy B.S., Nakajima F. and Yamamoto K. (2000). Analysis of bacterial community in a membrane separation bioreactors by fluorescent in situ hybridization and DGGE techniques. Wat. Sci. Tech. 41(10-11): 259-268.

Maharaj S., Naidoo A. and Bux F. (2006). Comparison of the nitrifying population dynamics from an MBR and conventional activated sludge system. MSc Thesis, Department of Microbiology, Durban Institute of Technology, Durban.

Manser J., Gujer W. And Siegrist H. (2005a). Consequence of mass transfer effects on the kinetics of nitriefiers. Wat. Res. 39: 4633-4642.

Manser J., Gujer W. And Siegrist H. (2005b). Membrane bioreactor versus conventional activated sludge system: population dynamics of nitriefiers. Wat. Sci. Tech. 52(10-11): 417-425.

Monti A., Hall E., Dawsin R.N., Husain H. And Kelly H.G. (2005). Comparative study of biological nutrient removal (BNR) processes with sedimentation and membrane based separation. Manuscript 05-608, Biotech & Bioeng. - John Wiles & Sons.

Parco V.; Du Toit GJG.; Wentzel MC.; and Ekama GA. (2006). Evaluation of kinetics in a membrane bioreactor biological nutrient removal activated sludge system. Proc. WISA Biennial Conference 2006, Durban, South Africa.

Parco V. (2006). Bioreattori a membrana per la rimozione biologica dei nutrienti: cinetiche di processo ed efficienze. PhD thesis, Department of Hydraulic and Environmental Engeeniring, University of Palermo, Italy.

Parco V., Du Toit G.J.G., Wentzel M.C. and Ekama G.A. (2007). Biological nutrient removal in membrane bioreactors: Denitrification and Phosphorus removal kinetics. Proc. IWA 4th International Membranes Conference, 15-17th May 2007 – Harrogate (UK).

Ramphao M., Wentzel MC., Merritt R., Ekama GA., Young T., and Buckley CA.. (2005). Impact of membrane solid-liquid separation on design of biological nutrient removal activated sludge systems. Biotechnol. and Bioeng., 89(6): 630-646.

Randall E.W., Wilkinson A.., and Ekama G.A. (1991). An instrument for the direct determination of oxygen utilization rate. Water SA 17(1): 11-18.

Rosengerger S., Kuger U., Witzig R., Manz U, Szewzyk M., and Kraume M. (2002). Performance of a bioreactor with submerged membranes for aerobic treatment of municipal wastewater. Wat. Res. 36: 413-420.

Smith S., Jefferson B. and Judd S.J. (2002). Membrane bioreactors: hybrid activated sludge or a new process?. CHISA August 2002, Prague

Sperandio M., Mass M., Espinoza Bouchot C. and Cadassud. (2005). Characterization of sludge structure and activity on submerged membrane bioreactor. Wat. Sci. Tech. 52(10-11):410-408

Van Haandel; Ekama G.A., and Marais Gv.R. (1981). The activated sludge process part 3 – single sludge denitrifcation. Wat.Res. 15, 1135-1152.

Wachmeister A., Kuba T., van Loosdrecht M.C.M. and Heijen J.J. (1997). A sludge characterization assay for aerobic and denitrifying phosphorus removing sludge. Wat. Res. 31(3): 471-478.

Wentzel M.C., Ekama G.A., Dold P.l. and Marais Gv.R. (1990). Biological excess phosphorous removal-steady state process design. Water SA 16(1): 29-48.

Witzig R., Manz W. , Rosenberger S. , Krüger U. , Kraume M. and Szewzyk U. (2002). Microbiological aspects of a bioreactor with submerged membranes for aerobic treatment of municipal wastewater. Wat. Res. 36: 394-402

Yamamoto K. (2002). Membrane Bioreactor: an advanced wastewater treatment/reclamation technology and its function in excess sludge minimization. In Advances in Water and Wastewater Treatment Technology. Amsterdam,229-237

Adsorption of Selected Organic Micropollutants from WWTP Effluent on Powdered Activated Carbon and Retention by Nanofiltration

Kai Lehnberg[1], Lubomira Kovalova[1], Christian Kazner[2], Thomas Wintgens[2], Thomas Melin[2], Juliane Hollender[3], Wolfgang Dott[1]

[1] Institute of Hygiene and Environmental Health, RWTH Aachen, Pauwelsstr. 30, D-52074 Aachen, Germany. E-mail:Kai.Lehnberg@rwth-aachen.de
[2] Institute of Chemical Engineering, RWTH Aachen, Turmstr. 46, D-52056 Aachen, Germany
[3] Swiss Federal Institute of Aquatic Science and Technology (Eawag), Überlandstr. 133, CH 8600 Dübendorf, Switzerland

Nowadays, it is possible to detect organic substances in aquatic systems in the lower ng/l range. Thus the number of anthropogenic micropollutants detected in surface, ground and even drinking water increased considerably. Most of these organic compounds are found in concentrations below acute toxic concentrations or, for pharmaceuticals, below medical dosage. However, micropollutans like endocrine disrupting compounds (EDCs) and cytostatics are cause of environmental concern due to their endocrine disruptive or (geno)toxic potential even if they occur in the environment at low concentrations.

In this project, we evaluate the possibilities of removal of micropollutants from municipal wastewater treatment plant (WWTP) effluent using a pilot plant combining sorption to powdered activated carbon and retention by nanofiltration (Figure 1). The effluent of the municipal WWTP at Aachen Soers, Germany, reaches the pilot plant after the sand filtration step. The pilot plant will be tested for removal of micropollutants, more specifically two EDCs, Bisphenol A and 17α-Ethinylestradiol, and two cytostatics, 5-Fluorouracil and Cytarabine. These substances have been chosen because of their quite different sorption abilities (K_{OC} 2 to 6800) and their molecular weights (130 to 296 g mol^{-1}) near to the membrane cuttoff of 200 Da.

Preliminary adsorption experiments in laboratory scale were performed to specify the amount of micropollutants that can be adsorbed to powdered activated carbon at operating conditions of the pilot plant. Figure 2 shows adsorption isotherms to powdered activated carbon in the effluent. The ongoing operation of the pilot plant demonstrates that the combination of powdered activated carbon and nanofiltration reduces the amount of dissolved nonpurgeable organic carbon up to 93 %. Further experiments will evaluate adsorption kinetics, performance of different types and particle size of carbon, influence of temperature, concentration of the micropollutants and matrix effects due to the effluent. The operation of the pilot plant is continued to optimize operation conditions and, after finishing the laboratory scale experiments, spiking experiments in the pilot plant will take place.

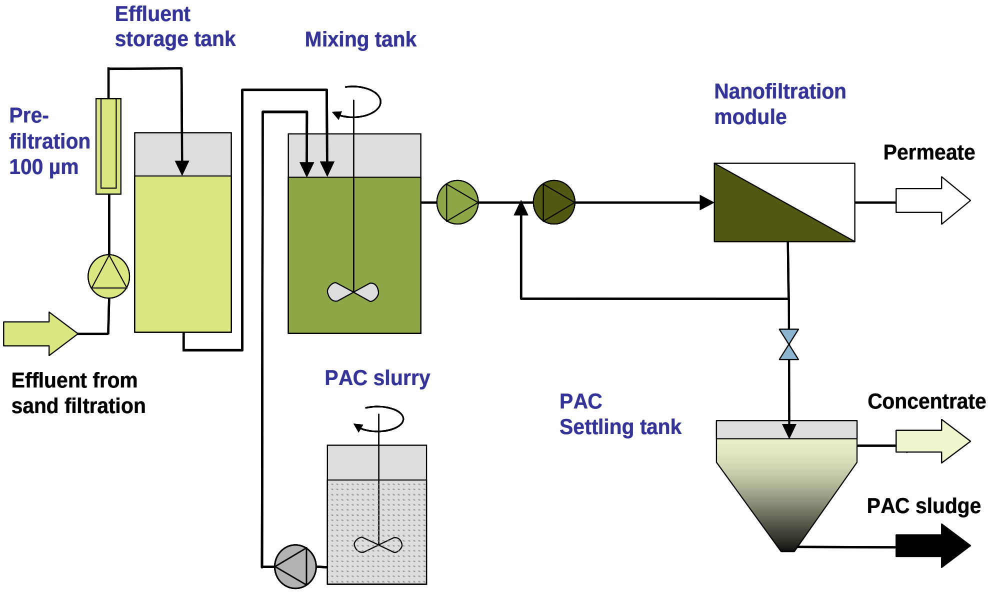

Figure 1: Schematic view of the pilot plant at Aachen Soers.

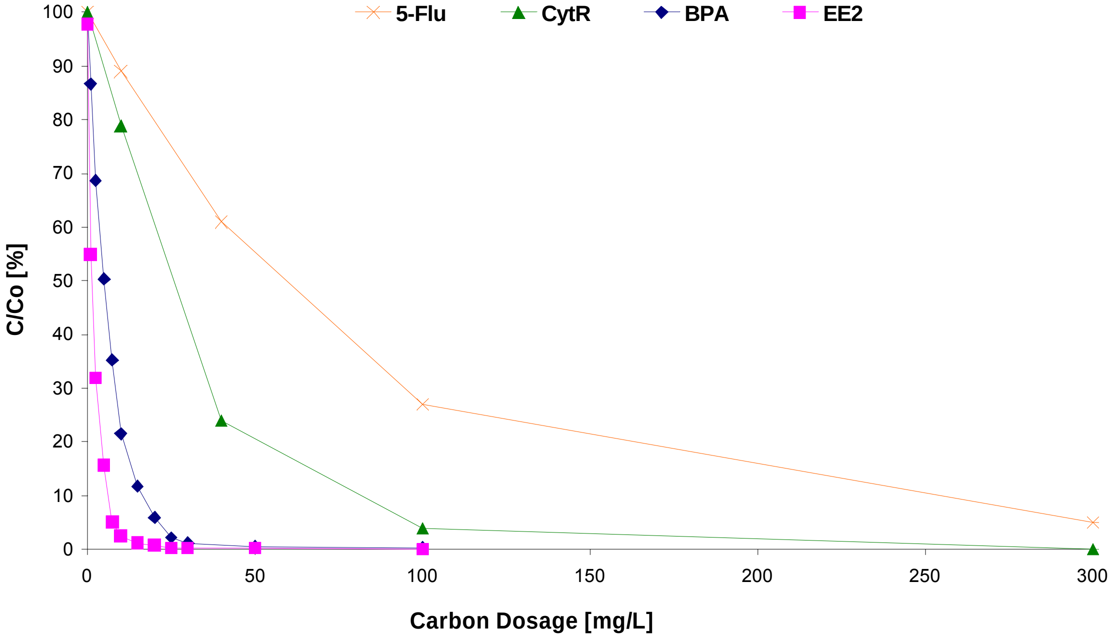

Figure 2: Adsorption isotherms of 5-Fluorouracil (5-Flu), Cytarabine (CytR) Bisphenol A (BPA) and 17α-Ethinylestradiol (EE2) to powdered activated carbon (Norit SAE Super) in Soers WWTP effluent. Effluent was spiked with 200µg/l of either compound and incubated at 20°C.

Fluorescence Spectroscopy as a Tool for Performance Monitoring of Membrane Bioreactors in Wastewater Treatment Systems

C. F. Galinha*, C. Portugal*, **, G. Carvalho*, **, J.G. Crespo* and M. A. M. Reis*

* REQUIMTE/CQFB, Chemistry Dept., FCT, Universidade Nova de Lisboa, 2829-516 Caparica, Portugal.
(E-mail: claudia.galinha@dq.fct.unl.pt, cmp@dq.fct.unl.pt, gildacarvalho@dq.fct.unl.pt, jgc@dq.fct.unl.pt, amr@dq.fct.unl.pt)
** IBET/ITQB, UNL, 2870-156 Oeiras, Portugal

Abstract In this work 2D-fluorescence spectroscopy was evaluated as a tool for monitoring EPS production in MBR for domestic wastewater treatment. Impact of humic- and fulvic-like substances in protein detection by fluorescence was assessed, since these fluorescent compounds can be present in wastewater in high concentrations, with a widely variable composition. Also differentiation and characterisation of different water types using 2D-fluorescence spectroscopy was studied. 2D-fluorescence spectroscopy proved to be sensitive to differences in water composition when samples from different sources were analysed. The main fluorescence peak of a model protein showed decreased fluorescence intensity in the presence of spring water, surface water and wastewater, due to quenching effects. A MBR system for wastewater treatment will be monitored using 2D-fluorescence maps as fingerprints of the system status. Fluorescence maps can be correlated with the operational parameters using non-mechanistic methods, such as artificial neural networks and principal component analysis, for performance monitoring and process optimisation.

Keywords 2D-fluorescence; EPS; Excitation-emission-matrix; MBR; quenching effect; domestic wastewater treatment

INTRODUCTION

Membrane bioreactors (MBRs) are increasingly employed as an alternative to conventional activated sludge processes in wastewater treatment, particularly due to the lower space requirements and the low solids content of the effluent. However, membrane fouling, and its consequent costs, are still limiting a larger application of MBR processes. Extracellular polymeric substances (EPS) are generally assumed to play a major role in membrane fouling and thus EPS monitoring and control in MBR operation is essential. EPS contain large amounts of proteins and aromatic organic substances that possess natural fluorescence properties, making fluorescence a promising technique to monitor the production of these compounds in MBRs. Additionally, conventional bioreactor monitoring, such as chemical analyses of the bulk liquid or transmembrane pressure, does not give information about the processes happening at the membrane surface (biofilm, pore clogging or membrane fouling). Fluorescence scanning can also be used *in situ*, at the membrane surface to provide direct information about the factors that lead to fouling.

Fluorescence spectroscopy is a highly sensitive, selective and non-invasive technique that can provide rapid information about the composition of complex media such as biological systems. The excitation-emission matrices (EEMs) obtained by scanning the spectra wavelengths can cover a wide diversity of natural fluorophores. The maps obtained from the EEMs can thus correspond to a fingerprint of the physiological activity of a biological system.

Fluorescence spectroscopy was previously applied for on-line monitoring of an extractive MBR for degradation of volatile organic compounds in aqueous media (Wolf et al., 2001). The biofilm formed in the membrane surface was monitored *in situ* using an optical fibre cable for delivering excitation light and capturing emission light. The 2D-fluorescence maps obtained were analysed together with operational parameters using artificial neural networks (ANN) for system monitoring and modelling. 2D-scanning fluorometry together with ANN, as a nonlinear and non-mechanistic technique for deconvoluting fluorescence information, proved to be suitable for monitoring this complex biosystem.

In the present study a tool is being developed to monitor MBR systems with submerged hollow-fibre membranes for domestic wastewater treatment. These systems contain suspended activated sludge as well as biofilms attached to the membranes, immersed in a complex matrix. Among other compounds, this matrix contained EPS and natural fluorophores present in the wastewater, such as humic- and fulvic-like compounds. Humic and fulvic substances, which are present in surface water and wastewater in high concentrations, with a widely variable composition, are heterogeneous mixtures of organic compounds with high molecular weight, both aromatic and aliphatic, rich in oxygen-containing functional groups. Due to these characteristics, humic and fulvic compounds are very reactive with light and are able to absorb in a wide range of wavelengths, which means that they can interfere in fluorescence measurements of proteins (both have absorbance at the wavelength of 280 nm) and thus interfere in 2D-fluorescence analysis of the system.

The aim of this study is to assess the feasibility of using on-line 2D-fluorescence to monitor EPS production in MBR systems. The characterisation of EPS in biological wastewater treatment systems by fluorescence sludge has been previously proposed in literature (Sheng and Yu, 2006, Esparza-Soto and Westerhoff, 2001). However, due to the possibility of interferences, it is of high importance to assess the impact of humic- and fulvic-like substances in protein detection by fluorescence. Additionally, characterisation and differentiation of water of different sources using 2D-fluorescence spectroscopy was carried out in this study.

METHODS

Solutions of commercial humic acids (Sigma, USA) in distilled water were used to confirm the excitation/emission regions typical from humic acids. 2D-fluorescence maps of a model protein, bovine serum albumin (BSA, Fluka, Switzerland), were obtained with and without commercial humic acids (10 mg/L) to test for interference of the presence of humic acids in protein spectra. BSA was employed at concentrations of 10 and 50 mg/L. Subsequently, 2D-fluorescence maps were obtained for different water samples with and without addition of BSA at the same concentrations to assess the interference of natural humic/fluvic acids in protein fluorescence.

Water samples

All the water samples were collected in Portugal. Surface water was collected from river Tagus on 13 November 2006, spring water was collected in Alenquer on 24 October 2006, and domestic wastewater was collected at the entrance of a wastewater treatment plant (WWTP) in the region of Lisbon (Almada) on 4 December 2006. The wastewater sample was previously centrifuged at 9000 rpm, 10 min at 10 ºC. All water samples were filtered using a filter with 0.20 μm pore diameter before the fluorescence scanning to remove bacteria.

2D-fluorescence

2D-fluorescence spectra maps where obtained with a fluorometer computer-interface Perkin-Elmer LS 50. The excitation source was a pulsed Xenon UV lamp, and the detector was a gated photomultiplier. Reflection grating monochromators were used on both excitation and emission sides. The scanning speed was 1500 nm/min; excitation and emission slits were 10 nm. Fluorometric spectra were generated in a range of 200 and 600 nm (excitation) and 225 and 625 nm (emission), in synchronous scanning mode with a constant wavelength difference (starting wavelength difference of 25 nm between excitation light and read emission).

RESULTS AND DISCUSSION

Interferences in protein detection were firstly studied in this work by using commercial humic acids and BSA. Fluorescence spectra of proteins have a characteristic profile with two peaks, the main one at excitation wavelength of 280 nm and emission wavelength at around 345 nm, and a scatter peak at the same emission wavelength of 345 nm. Figure 1 shows the scanning spectra of the model protein, BSA, displaying the two characteristic protein peaks.

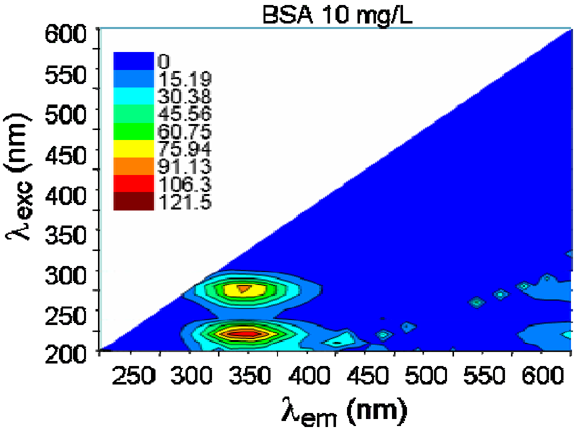

Figure 1. 2D-fluorescence spectrum of a BSA solution with a concentration of 10 mg/L.

BSA and commercial humic acid were scanned individually and combined at the same concentrations. The obtained fluorescence spectra (Figure 1 and 2) showed that the commercial humic acids had a strong effect on the fluorescence spectrum of BSA, reducing the protein peak in the 2D fluorescence map. This loss of fluorescence intensity is called quenching and results from the interaction of the excited molecule of protein with other molecules, in this case humic acids, whatever the nature of the competing intermolecular process. Usually quenching mechanisms involve a fast process of transfer of electrons, protons or energy between quenchers and quenched solutes.

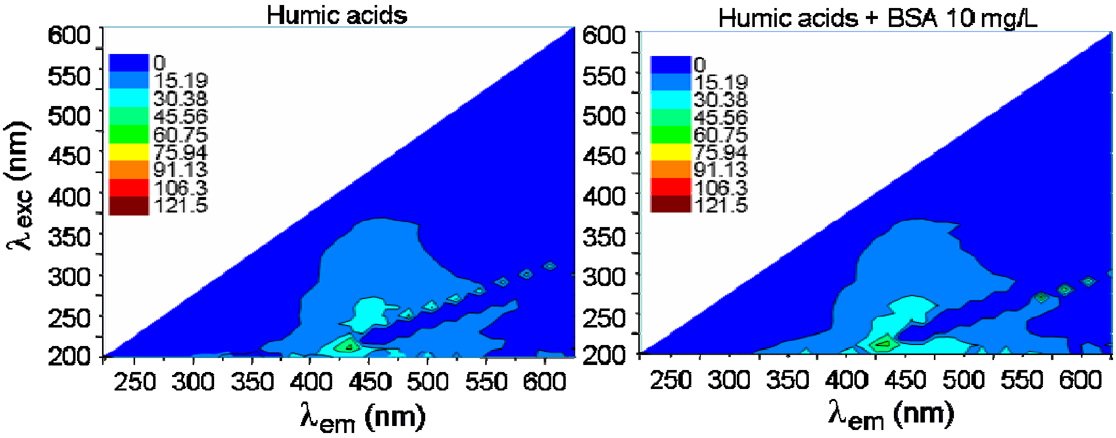

Figure 2 2D-fluorescence spectra of commercial humic acid solution (10 mg/L) and a mixed solution of humic acids and BSA, both at 10 mg/L.

In order to assess the interference of humic acids present in real waters, samples of domestic wastewater, surface and spring water were analysed together with standard BSA solutions. The quenching effect was also observed in these samples (Figure 3). Intensity at Excitation /Emission of 280/345 nm corresponding to the main peak of proteins were measured in water samples with added BSA and compared to a BSA solution in distilled water. To understand if quenching effect is constant for each type of water, independently of protein concentration, two different concentrations of BSA (10 and 50 mg/L) were tested. The decrease of intensity at 280/345 nm excitation/emission is shown in table 1. We can conclude that the degree of interference in real waters was lower than with commercial humic acids, most likely due to different composition and vast nature of the fulvic- and humic-like compounds present in real waters. Most probably, some fractions of this wide range of compounds have lower quenching efficiency in protein detection by fluorescence.

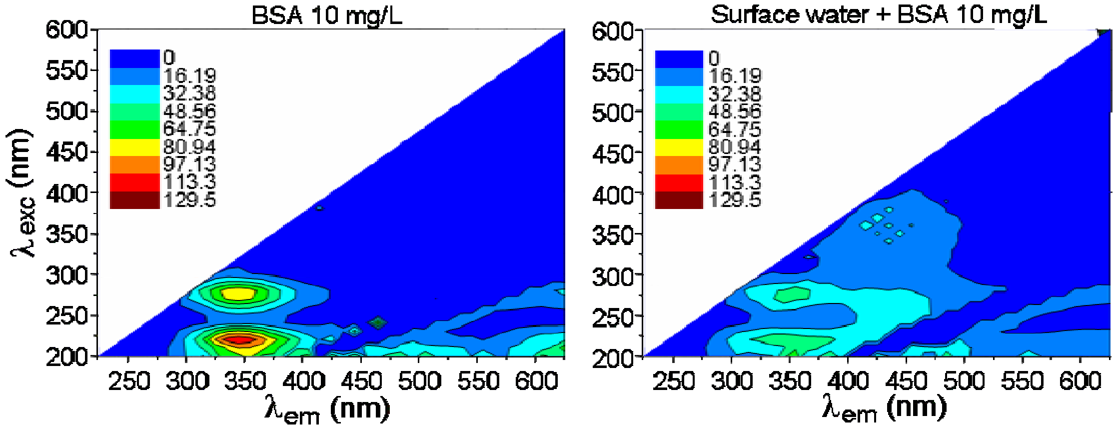

Figure 3 Example of quenching effect in surface water: 2D-fluorescence spectra of BSA 10 mg/L solution and BSA 10 mg/L solution with surface water.

The extent of the quenching effect depends on the quenching efficiency of each quencher, their concentration and, in some cases, on the concentration of fluorophore. Results obtained with wastewater show that, in this case, the quenching effect depends on the protein concentration and not only on concentration of quenchers (Table 1). This phenomenon is likely due to the fact that quenching happens in the short period of molecular excitation (nearly 10^{-12}s) and is therefore strongly dependent on the probability of molecular collision during the excited state. With less molecules of fluorophore, the distance between the fluorophore and quencher is, in average, higher than for higher concentrations of fluorophore. Thus, the presence of a higher fluorophore concentration can favours the efficiency of quenching. Regarding the complexity of the medium and the complex quenching effects observed in wastewater media, quantitative determination of proteins and development of direct correlations using deterministic methods does not seem to be a feasible approach.

Table 1. Decrease of the fluorescence intensity at λ_{exc} = 280 nm and λ_{em} = 345 nm (maximum emission of BSA), upon addition of water from different sources.

	BSA 10 mg/L	BSA 50 mg/L
Humic Acids	95 %	96 %
Surface water	52 %	57 %
Spring water	33 %	31 %
Wastewater	37 %	58 %

2D-fluorescence spectra of spring water, surface water, and wastewater were compared (Figure 4). The significant differences between the spectra obtained show the sensitivity of this technique to distinguish waters with different composition. Thus the maps obtained by fluorescence scanning may be used as fingerprints of the system.

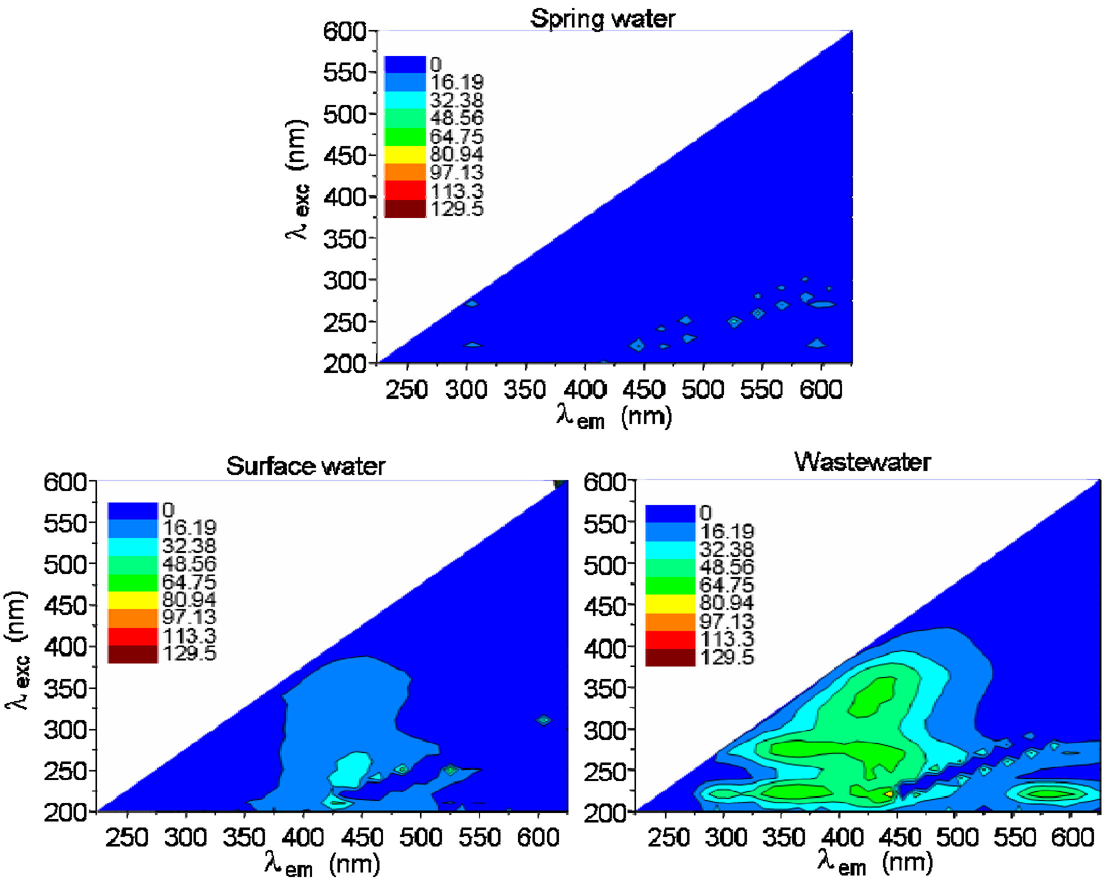

Figure 4 2D-fluorescence spectra of spring water collected in Alenquer, Portugal; surface water collected from the river Tagus; and domestic wastewater collected at the entrance of a WWTP in the region of Lisbon, Portugal.

Since fluorescence maps can act as fingerprints of the system and deterministic methods seem not to be suitable to analyse and interpret them, non-mechanistic methods should be used. Principal Components Analysis (PCA) can be employed to interpret the maps, and based on the obtained information, correlations may be established with the operational parameters and process performance parameters using artificial neural networks (ANN). Ultimately, a mathematical tool can be developed to be used in monitoring the MBR operation and increase membrane performance.

CONCLUSIONS

This work assesses the feasibility of using 2D-fluorescence to monitor EPS production in MBR for domestic wastewater treatment. Based on previous studies, this technique seems to be suitable for feed, sludge, membrane biofouling and permeate characterisation. The results obtained in this study showed that humic and fulvic substances present in the three water types analysed have a strong quenching effect on proteins assessment by 2D-fluorescence, reducing fluorescence intensity of protein peak. 2D-fluorescence spectroscopy also proved to be sensitive to differences in water composition. Due to quenching effects in such complex media, development of direct correlations between EPS content and the fluorescence data using deterministic methods seems to be an unfeasible approach. The use of non-mechanistic methods may involve complex data processing, but are able to reveal the system status and possible interactions between species, even if not fluorescent, due to quenching. Quenching can, therefore, be useful to obtain more information on compounds present in media and consequently on system status. 2-D fluorescence technique, combined with non-mechanistic methods, seems to be a powerful tool to obtained information on compounds present in MBR systems, such as EPS, to monitor and control MBRs performance decreasing membrane fouling. Non-mechanistic analyses of the 2D maps have been used for similar situations described in literature. This approach will be employed to monitor an MBR system for wastewater treatment using 2D-fluorescence maps as fingerprints of the system status. A correlation between the operation conditions and the system performance will be established by non-mechanistic methods, such as principal components analysis and artificial neural networks, with the ultimate objective of using 2D-fluorescence for process optimisation.

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REFERENCES

Esparza-Soto M. and Westerhoff P. K. (2001). Fluorescence spectroscopy and molecular weight distribution of extracellular polymers from full-scale activated sludge biomass. Water Science and Technology, 43(6), 87-95.

Marose S., Lindemann C. and Scheper T. (1998). Two-dimensional fluorescence spectroscopy: A new tool for on-line bioprocess monitoring. Biotechnology Progress 14(1), 63-74.

Sheng G. P. and Yu H. Q. (2006). Characterization of extracellular polymeric substances of aerobic and anaerobic sludge using three-dimensional excitation and emission matrix fluorescence spectroscopy. Water Research 40(6), 1233-1239.

Wolf G., Almeida J. S., Pinheiro C., Correia V., Rodrigues C., Reis M. A. M. and Crespo J. G. (2001). Two-dimensional fluorometry coupled with artificial neural networks: A novel method for on-line monitoring of complex biological processes. Biotechnology and Bioengineering 72(3), 297-306.

Composition of membrane fouling layers for different filtration modes in MBR

U. Metzger* , P. Le-Clech** , R.M. Stuetz*** , F.H. Frimmel * and V. Chen**

* Engler-Bunte-Institut, Chair of Water Chemistry, University of Karlsruhe, Engler-Bunte-Ring1, 76131 Karlsruhe, Germany (E-mail: *ulrich.metzger@ebi-wasser.uni-karlsruhe.de*, fritz.frimmel@ebi-wasser.uni-karlsruhe.de)
* * UNESCO Centre for Membrane Science and Technology, School of Chemical Sciences and Engineering, University of New South Wales, Sydney 2052, Australia (E-mail: *p.le-clech@unsw.edu.au*, v.chen@unsw.edu.au)
*** Centre for Water and Waste Technology, School of Civil and Environmental Engineering, University of New South Wales, Sydney 2052, Australia (E-mail: r.stuetz@unsw.edu.au)

Abstract The fouling behaviour of membranes in membrane bioreactors (MBR) and the composition of membrane fouling layers have been observed for three different modes of operation, i.e. relaxation, backwash, and continuous. The same net productivity flux of 20 l/m^{2}h was used for each mode. It was found that the fouling behaviour was strongly dependent on the applied instantaneous flux rather than the filtration modes themselves. The transmembrane pressure (TMP) obtained after 24 hours of filtration was dominated by the fouling rates calculated within the first hour of the experiment. After the fouling experiments, the fouling layers were separated in three different fractions based on their affinity for the membrane. The lower fractions, representing the irreversible fouling, were predominantly composed of soluble molecular biopolymers (SMB) and featured the highest specific biopolymeric resistance. They contributed approximately 50 % to the total hydraulic resistance for all modes of operation. Intermediate fractions were found to be composed of equal parts of SMB and biomass flocs, with an strong enrichment of carbohydrates. The upper fractions, the loosely bound upper cake layer, were predominantly composed of attached biomass flocs and featured a specific biopolymeric resistance closely related to the applied instantaneous flux.

Keywords filtration modes, fouling layer, MBR, soluble molecular biopolymers

INTRODUCTION

Membrane bioreactors (MBRs) are receiving increasing interest for the treatment of municipal and industrial wastewaters. Compared to current treatment technologies, however, the operational costs for membrane technologies still remain considerably higher. Membrane fouling remains a major operational issue and to sustain an efficient process membranes may require frequent cleaning either by physical or chemical means (Judd et al., 2006). In-situ physical cleaning techniques for MBRs typically involves aeration, membrane relaxation and membrane backwashing (Le-Clech et al., 2006). To compensate for the filtration downtime due to relaxation and backwashing, higher instantaneous fluxes are required to maintain a given flux productivity.

Recent studies have indicated that next to the suspended biomass, soluble molecular biopolymers (SMB) are the main foulants in MBR. SMB originate from the biomass, diffuse into the liquid phase and can adsorb to the membrane surface. Once on the membrane surface, they block membrane pores and/or form a gel-like structure, resulting in a hydraulic resistance to permeate flow (Rosenberger et al., 2005). It has been observed that half of the total fouling resistance could be caused by SMB, even though their concentration is rather small compared to the biomass concentration (Huang et al., 2000). The composition and the distribution of different fouling layers and their fouling compounds in MBR systems, such as pore foulants and cake foulants, and their influence on the hydraulic performance, are not well understood and are the subject of this paper.

In this study, the fouling behaviors of MBRs operated with different filtration modes (i.e. relaxation, backwash, and continuous) but all generating the same productivity flux were evaluated and a detailed analysis of the obtained fouling layers has been conducted.

METHODS

Membrane bioreactor

The fouling experiments were conducted on a lab-scale aerobic MBR with a working volume of 30 L. The MBR was composed of an aeration bioreactor and a membrane tank equipped with six submerged hollow-fiber membrane modules, each with an approximate surface of 0.05 m². Hydrophilic polyvinylidene fluoride (PVDF) membranes (0.2 µm) (*Memcor Ltd, Windsor, Australia*) were used. Four of the membrane modules were used for long term operation of the MBR, and two others were operated in 24-hours experiments. Permeate produced during the 24-hours experiments was recycled into the bioreactor to maintain the hydraulic retention time (HRT) set up by the long term modules. The reactor was originally seeded with sludge collected in a local sewage treatment pant and fed with synthetic waste water. Prior to fouling experiments, the system was acclimatized for 3 months until steady state conditions were reached (Table 1).

Table 1: Steady state conditions of the MBR.

HRT (h)	COD removal (%)	MLSS (g/L)	MVSS (g/L)	SMP (mg/L)
15	97	5.5 ± 0.2	5.3 ± 0.2	4.5 ± 0.6

Fouling experiments

Three modes of filtration (i.e. relaxation, backwash, and continuous) were compared producing the same net flux of 20 L/m^2 h (Table 2).

Table 2: Details of the three filtration modes (net flux is 20 L/m^{2}h in all configurations).

Filtration mode	Continuous	Relaxation	Backwash
Details of operation (Flux in L/m^2 h)	20	On for 220s (21.8) Off for 20s	On for 220s (25) Backwash 20s (34)

The effects of these filtration modes on the MBR performances have be assessed by the continuous measurement of the transmembrane pressure (TMP) during operation and a detailed analysis of the hydraulic resistances based on Darcy's law.

Characterization of the fouling layer

After 24 hours of operation, a detailed analysis of the membrane fouling layers and their effect on the total hydraulic resistance was conducted. For this, fouling layers were separated into three fractions based on their affinity for the membrane. An upper fraction was separated by thoroughly rinsing the membrane with 50 mL Milli-Q water. An intermediate fraction was separated by backwashing the membrane with 50 mL Milli-Q water with an equivalent flux of 37 L/m^{2}h. The lower fraction, irreversibly attached to the membrane surface, was removed by soaking the membranes in NaOH solution (pH ~11) for 24 h. The cleaning solutions were collected and analysed in terms of carbohydrate concentration applying the Dubois method and protein concentration applying the Lowry method. Parts of the cleaning solutions were filtered through a 0.2 µm filter and analyzed in the same way. Proteins and carbohydrates that passed through the filter were considered SMB. After each cleaning step, a clean water resistance test was conducted to calculate the actual hydraulic resistance caused by each fraction of the fouling layer, using the following equations:

Hydraulic resistance total fouling layer: $R_{TF} = R_{total}-R_{membrane}$ (Equation 1)
Hydraulic resistance upper fraction: $R_{UF} = R_{total}-R_{rinsed}$ (Equation 2)
Hydraulic resistance intermediate fraction: $R_{IF} = R_{rinsed}-R_{backwash}$ (Equation 3)
Hydraulic resistance lower fraction: $R_{LF} = R_{backwash}-R_{membrane}$ (Equation 4)

The hydraulic resistances of the different fouling layer fractions were related to the concentration of total biopolymers in the fouling layer fraction and were expressed as the specific biopolymer resistance (α) (Nagaoka et al., 1998):

$$\alpha = R/Dp$$

where α is the specific resistance of the biopolymers ($m.kg^{-1}$), R is the resistance caused by each fraction (m^{-1}) and Dp is the biopolymer density in each layer on the membrane (kg biopolymers.m^{-2}). From this parameter information about the structure and density of the different fractions could be derived.

RESULTS AND DISCUSSION

The continuous filtration mode featured the lowest TMP of around 125 mbar after 24 hours of operation, whereas the backwash mode featured the highest TMP of around 190 mbar. The relaxation mode featured a TMP of around 150 mbar.

The differences in TMP were mainly due to the different fouling behaviour for each of the modes within the first hours of operation. Fouling rates in the first hours differed strongly between the different filtration modes (backwashing 57 mbar/h, relaxation 40 mbar/h, continuous 29 mbar/h) and appear to be caused by different applied instantaneous fluxes. Similar observations have been made by Hong et al. (2002) who reported that the initial permeation rate is especially important in controlling membrane fouling. It also has been revealed in other studies that there is an exponential relationship between membrane fouling rates and permeate flux (Germain et al., 2005). In this study, a difference in flux of 25 % led to an almost doubling of the fouling rate in the first hour of the experiment. The fouling rates of all filtration modes decreased significantly with time and after only one hour they decreased to around 30 % of the initial fouling rate. After 20 hours, the fouling rates were negligible (around 1 mbar/h), with the continuous operation mode showing the largest fouling propensity.

Characterisation of the fouling layer

Total biopolymer and SMB content, as well as the protein to carbohydrate ratio (P/C) of the fouling fractions of the different operations modes are displayed in Figure 1 A,B.

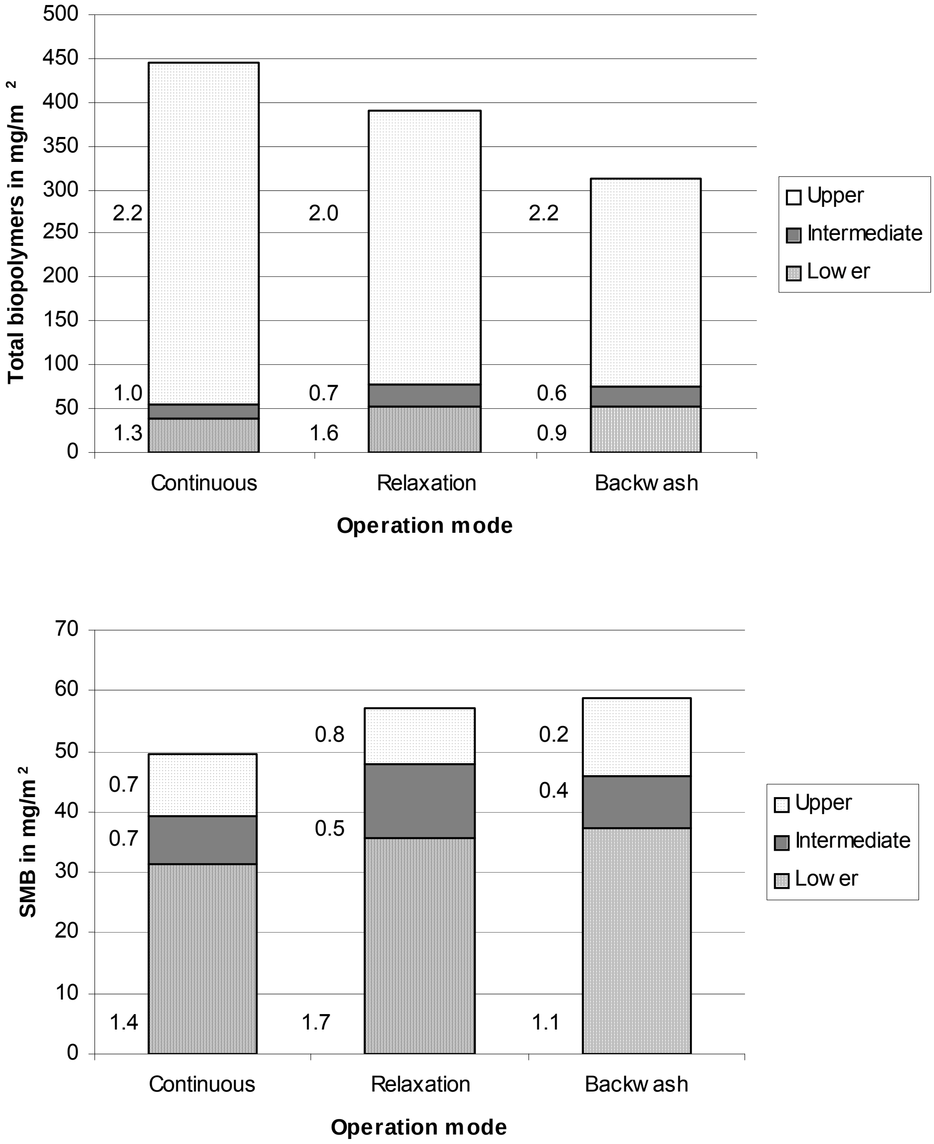

Figure 1 A) Total biopolymer concentration (protein + carbohydrate, P/C) B) SMB concentrations in the three cleaning solutions for the different filtration modes experimented. Numbers indicate the P/C of the different fractions.

The upper fraction, originating from the most easily removed layer, contained the largest fraction of total biopolymers for all filtration modes and only contained small amounts of SMB. The P/C of this fraction was around 2:1 for all filtration modes, which is similar to the P/C of the biomass in the MBR. Hence, it can be concluded that the upper fraction consists mostly of attached biomass flocs, which also has been previously observed in the study of Chu and Li (2005). A larger content of biopolymers observed in the upper fraction of the continuous mode was expected, as neither relaxation nor backwash took place to remove the loosely bound cake layer.

The concentration of biopolymers in the intermediate fraction was the lowest for all tested operation modes in the range from 14 to 26 mg/m^2. The intermediate fraction was composed of equal parts of the biomass aggregates and SMB for the relaxation and continuous modes, whereas the backwash mode featured a lower content of SMB (37 %). For all operation modes, soluble carbohydrates were strongly enriched in this fraction. Hence, carbohydrates appear to loosely adsorb to the membrane and to play a significant role in the formation of consecutive cake layer.

The irreversible fractions featured a concentration of biopolymers within the range of 40 mg/m^2 (continuous) and 50 mg/m^2 (backwash and relax). These fractions were composed predominantly of SMB (68-79 %) and were strongly attached to the membrane. In comparison to the intermediate fraction, a higher concentration of soluble proteins was observed in the lower fractions, especially in filtration modes featuring no backwashing. Proteins appear to have higher potential for deposition/adsorption directly on the membrane surface within this layer and appear to be more strongly attached to the membrane than carbohydrates during filtration. Although the proportions of biopolymers in each fraction are dependent on the method used to obtain each fraction, these results allow for a better understanding of the relative distribution of biopolymers within the fouling layers.

Relation between hydraulic resistances and total biopolymeric concentration of the fouling layer fractions

The backwash mode produces the highest total fouling resistances R_{TF} (2.4 x 10^{12} m^{-1}) and the continuous the lowest (1.7 x 10^{12} m^{-1}) after 24 hours operation. The lower fractions for all operation modes had the highest contribution to the total fouling resistances, ranging from 48 % for the continuous mode to 56 % for the relaxation mode. The intermediate fractions contribute a relatively small resistance of about 12 % of the total fouling resistance for all operation modes. Even the upper fraction possesses the largest biopolymer content, it contributes only 32 to 38 % (continuous, backwash) of the total fouling resistance.

Hence, the biopolymer compounds of the upper fractions (reversible resistance) have a lower impact on the fouling resistance than those in the lower fractions (responsible for the irreversible resistance). For example, approximately 45 mg/m² biopolymers in the lower fraction corresponds to a larger resistance than 300 mg/m² biopolymers of the upper fraction. The relation between the hydraulic resistances of the different fouling layer fractions and the concentration of total biopolymers in those fractions can be expressed as specific biopolymer resistance (α) and is displayed in Figure 2.

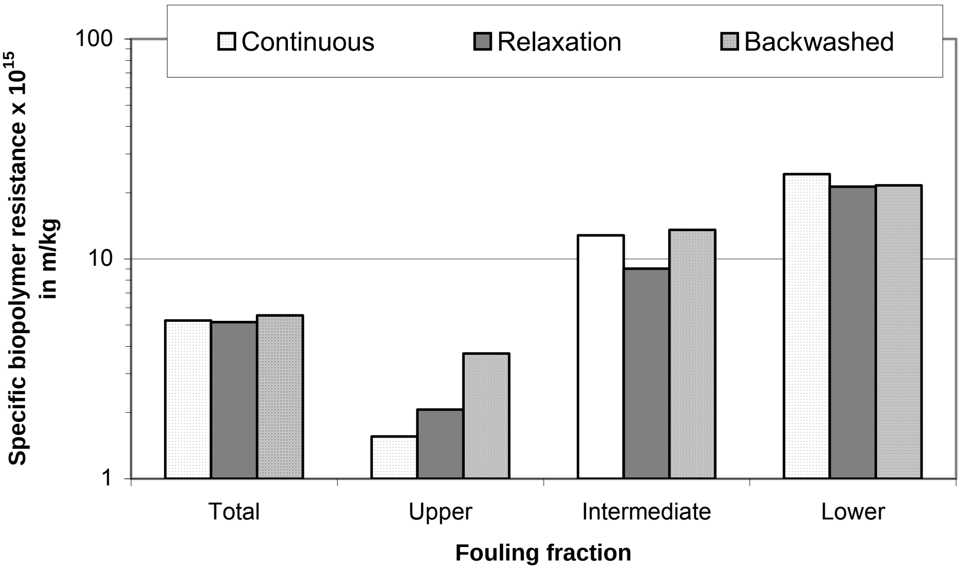

Figure 2 Specific biopolymer resistances for the different fouling layers obtained for different modes of filtration (Metzger et al., 2006).

The α value calculated for the total fouling layer ranged between 10^{15} and 10^{16} m.kg^{-1}. It can be noticed that α increases for fouling fractions closer to the membrane. The biopolymer compounds of the lower fractions had the highest *α* for the different operation modes at 2.2 x 10^{16} m.kg^{-1}, whereas α of the upper fraction featured the lowest *α* of around 0.3 x 10^{16} m kg^{-1}. The *α* value of the intermediate fraction ranged from 0.8 to 1.4 x 10^{16} m.kg^{-1}. The *α* values of the different fouling fractions and the analysis of their compositions provides a basis to draw conclusions about their structure. The upper fraction, which features the loosely bound cake layer, is assumed to have a porous structure containing voids and allowing water to pass through easily, as also has been proposed in a recent publication (Wang et al., 2007). The specific biopolymer resistance caused by the upper fraction seems closely related to the instantaneous flux applied during the filtration. As the flux increases, a more compressed cake biomass flocs is formed, resulting in a dense and compact upper cake layer. Hence, *α* of the upper fraction was largest for the backwash mode and smallest for the continuous mode.

The intermediate fraction represents the interface between the loosely and the firmly bound fractions. In this fraction, biomass and SMB clusters fill up membrane pores and are attached to the firmly bound layer. Soluble carbohydrates accumulate in this fraction. The fraction has a denser matrix and is expected to behave like a gel-like layer between the lower irreversible membrane fouling fraction and the upper cake fraction. Its composition is little influenced by the different operation modes.

The lower fraction is compact and tight and features a dense structure with a very low permeability. It is likely that this layer completely covers the membrane surface and its pores and is mainly responsible for irreversible pore fouling. This fraction consists predominantly of soluble polymers, which are directly attached to the membrane and are partly located in the pores, and hinders water permeation, leading to high α values for all filtration modes. Compared to the two other fractions, it contains a higher concentration of soluble proteins strongly bound to the membrane. The total biopolymer concentration is similar for all filtration modes.

CONCLUSION:

The results from this study indicate that different applied instantaneous fluxes for different filtration modes have a larger impact on the fouling behavior than the application of different physical cleanings when filtration is operated under the same net flux in short term experiments. With continuous filtration showing the highest hydraulic performance, the application of physical cleaning is not recommended for these relatively short-termed experiments. The variation in fouling behaviour were due to the different pore blocking and cake formation mechanisms occurring within the first few hours for each mode. After total coverage of the membranes with biopolymers and the formation of a cake layer, equilibrium is reached and fouling rates for all modes are similar. At this stage, similar pore fouling layers have been established for all different filtration modes and the main differences in fouling layers can now be found in the upper fouling fractions. The upper fouling fraction for the continuous mode features the highest concentration of biopolymers. The backwash mode features the most compact and dense cake layer, which was caused by exposure to higher instantaneous fluxes (Metzger et al., 2006).

REFERENCES

Chu H.P. and Li X. (2005). Membrane Fouling in a Membrane Bioreactor (MBR): Sludge Cake Formation and Fouling Characteristics, Biotechnology and Bioengineering, 90, 323-331.

Germain E., Stephenson T. and Pearce P. (2005). Biomass characteristics and membrane aeration: Toward a better understanding of membrane fouling in submerged membrane bioreactors (MBRs), Biotechnology and Bioengineering, 90, 316-322.

Hong S.P., Bae T.H., Tak T.M., Hong S. and Randall A. (2002). Fouling control in activated sludge submerged hollow fiber membrane bioreactors, Desalination, 143, 219-228.

Huang X., Liu R. and Qian Y. (2000). Behavior of soluble microbial products in a membrane bioreactor, Process Biochemistry, 36, 401-406.

Judd S. (2006) The MBR book: Principles and applications of membrane bioreactors in water and wastewater treatment, Elsevier, Oxford.

Le-Clech P., Chen V. and Fane T. (2006). Fouling in membrane bioreactors used in wastewater treatment - A review, Journal of Membrane Science, 284, 17-53.

Metzger U., Le-Clech P., Stuetz R., Frimmel F.-H., and Chen V. (2006), Characterisation of polymeric fouling in membrane bioreactors and the effect of different filtration modes, Journal of Membrane Science, submitted 2006

Nagaoka H., Yamanishi S. and Miya A. (1998). Modeling of biofouling by extracellular polymers in a membrane separation activated sludge system, Water Science and Technology, 38, 497-504.

Rosenberger S., Evenblij H., te Poele S., Wintgens T. and Laabs C. (2005). The importance of liquid phase analyses to understand fouling in membrane assisted activated sludge processes-six case studies of different European research groups, Journal of Membrane Science, 263, 113-126.

Wang X.-M., Li X.-Y. and Huang X. (2007). Membrane fouling in a submerged membrane bioreactor (SMBR): Characterisation of the sludge cake and its high filtration resistance, Separation and Purifification Technology, 52, 439-445.

Standard assays and metaproteomes as new approaches for functional characterization of membrane bioreactor biomass

R. Kuhn*, A. Pollice*, G. Laera*, L.L. Palese**, R. Lippolis***, S. Papa*, ***

*IRSA CNR, Viale F. De Blasio 5, 70123 Bari, Italy (e-mail: alfieri.pollice@ba.irsa.cnr.it)
**Dep. of Medical Biochemistry, Biology and Physics, University of Bari, Policlinico, P.zza G. Cesare, 70124 Bari, Italy
***Institute of Bioenergetics and Biomembranes, CNR, Bari, Italy

Abstract
EPS and SMP were often reported as a possible cause of membrane biofouling, but unequivocal relationships in this respect have hardly been demonstrated. A closer examination of EPS and SMP as possible tools to investigate the linkage between biofouling and enzymatic activity may provide interesting information. In particular, the high abundance of high molecular weight polysaccharides and proteins in the matrix of EPS and SMP has made DNA and RNA examination impractical for functional gene expression studies. Therefore a new approach called "metaproteomics" has been recently proposed for investigating environmental samples and activated sludge. This study provides background information for an on-going experimental work on MBR biomass characterization with innovative tools.

Keywords Membrane bioreactors, biomass characterization, EPS/SMP, metaproteomes.

INTRODUCTION: MBR - MINI REVIEW

Membrane bioreactors (MBR) are the combination of a bioreactor and a set of membrane modules separating the treated water (permeate) from the activated sludge and the concentrated wastewater feed (retentate).

Since the introduction of side-stream and submerged configuration in the early 1990s MBR technology was rapidly improved and advanced. Henceforth MBR are applied to the treatment of industrial wastewater with high organic loading rates, domestic wastewater, nitrate removal from drinking water (denitrification), pesticide removal, and expansion including food-processing wastewater, slaughterhouse wastewater and landfill leachates, and specific biodegradation processes (Yang *et al*., 2006).

The MBR technology provides many advantages, but its major feature is the support of both non-flocculating and flocculating biomass. Thus MBR can be operated on higher biomass concentrations since biomass loss through wash-out effects no longer occurs and this enables operation at higher loading rates. Furthermore, no primary sedimentation and secondary clarifier are required before and after the biological wastewater treatment which results in smaller footprints. The latter has high importance for countries with land limitations. Rapid start-up of MBR and fast stabilization of performances leading to relatively easy management practices were also reported (Pollice *et al*., 2004).

MBR can provide very high effluent quality by retaining all components with particle size larger than the membrane cutoff, i.e. micro-organisms, cell debris, extracellular polymeric substance (EPS), soluble microbial particles (SMP), colloids and organic matter. Problems related to high biomass production can be limited operating the system with longer sludge retention times (SRT) up to complete retention, independent of the hydraulic retention time (HRT). Rosenberger *et al.* (2002) reported that MBR performance on very long SRT significantly decreased biomass production, supported lower operation costs, and resulted in fewer problems in disposal and solids management. The disposal has important commercial impacts since wastewater sludge normally contains heavy metals and cannot be discharged on agricultural land (Gander *et al.,* 2000). Laera *et al.* (2005) have clearly demonstrated that the operation on complete retention results in high sludge concentration with a very limited biomass production, described as zero net growth. While the biomass activity is low under these conditions, the bioconsortium performs complete nitrification and COD removal, and it is capable to withstand loading variation. In contrast, How *et al.* (2005) investigated the operation at very short SRT and different HRT. He and his colleagues reported complete nitrification and excellent organic removal efficiency regardless of the short SRT. Generally speaking both the SRT and the HRT contribute to the removal efficiency of organic compounds from feed water.

However, despite these and many other remarkable advantages MBR technology still has a couple of relevant drawbacks which prevent further process optimization. The tendency of membrane biofouling is still one of the major limitations. In particular, the complete retention of bioflocs and soluble particles mentioned above has a series of very complex interactions to the membrane as consequences. The production of EPS and SMP by micro-organisms leads to blockage onto and into the membrane. This causes the hydraulic resistance to quickly increase whereas the permeate flux decreases. The resistance can be subdivided into reversible (removable by back-flushing) and irreversible (non-removable) resistance. The removable fraction is caused by absorption of suspended solids *onto* the membrane surface, cake layer formation *onto* the membrane layer and concentration polarization directly *in front* of the membrane. Non-removable resistance is caused by clogging *into* the membrane pores. Judd et al. (2006) described the fouling mechanism as a combination of standard blocking, cake filtration, intermediate blocking and complete blocking, respectively. Interactions and/or overlapping of all these phenomena must be considered.

Physical cleaning through aeration, backflushing and/or backpulsing, and also the use of strong chemicals like bases, acids and oxidants must be periodically performed to support the recovery of the membrane-permselectivity. Unfortunately, start-up permeability and flux normally can not be achieved due to internal membrane blockage and clogging.

Until recently, much research has been carried out to minimize such resistance effects with regard to different aspects. The main influences reported in the literature are: The particle size distribution and concentration (Cicek *et al.* 1999, Chang *et al.,* 2005); Influences of the dissolved oxygen concentration and aeration (Yu-Lan *et al.,* 2006); The operation below critical and sub-critical flux (Field *et al.,* 1995; Pollice *et al.,* 2005); Influences of permeate flux and mixed liquor suspended solid (MLSS) concentration (Rosenberger *et al.,* 2006); The viscosity of mixed liquor (Ueda *et al.,* 1996); The use of different membrane polymer materials (Yamato *et al.* 2006); The organic load and the food-to-micro-organism ratio *(F/M)* (Trussell *et al.,* 2006); Impacts of cations such as Ca^{2+} and Mg^{2+} (Kim *et al.,* 2006), and many other important operation parameters. Despite all these comprehensive investigations, the occurrence of biofouling due to the production of EPS/SMP is still poorly understood. There exists a lack of sufficient knowledge between the linkage between microbial dynamics, enzymatic activities and the interactions to their environment. In the end interdisciplinary research is required not only to characterize, but also to control and reduce biofouling.

UNDERSTANDING IS ALL ABOUT: THE FUNCTION OF EPS/SMP

The key in understanding microbial dynamics and activities finally leads to the functional expression of genes of all present micro-organisms. Extracellular material expressed can be subdivided into proteins and polysaccharides (carbohydrates) followed by lower quantities of lipids, RNA, and DNA. The latter has been often related to lysis of cells, but newer studies have already shown that micro-organisms also tend to use nucleic acids as EPS nano-structure elements (Böckelmann *et al.,* 2006).

Microrganisms in MBRs adapt their metabolic activity to current operation conditions, and there the expression of EPS contributes to the bioflocculation mechanism. In comparison, in environmental ecosystems microrganisms express EPS mainly due to the major advantage to attach onto surfaces. This is followed by the formation of a complex three-dimensional biofilm structure capable to protect micro-organisms against environmental changes such as salinity, pH, pressure, fluctuations in nutrient available and attacks by toxic substances (Wimpenny *et al.,* 2000). EPS are widely distributed in marine environments and enable micro-organisms to use every available niche. Therefore they can occur in very different structures such as in dissolved forms, colloids, and discreet particles like transport exopolymeric particles (TEP) and/or associated with particular matters. Accordingly the functions of EPS are diverse and generally contribute to the survival strategies of microbial life (Fux *et al.,* 2005).

Microbial survival strategies also take place in MBR. The formation and growth of biotic communities in heterogeneous bioflocs is comparable to surface attached biofilm structures. Flocs provide the same properties and represent niches to main microrganisms of the MBR bioconsortium. Similar to microrganisms observed in the environment, also those present in MBR compete for all biodegradable substrates and terminal electron acceptors such as oxygen. Thus, microrganisms adapt their metabolisms and express specific catalytic enzymes or release for example functional EPS-proteins into the surroundings (Ogunseitan, 1997). It is well known that EPS-proteins play an important role in the process of bioflocculation by influencing the hydrophobicity. They can directly respond to operational changes such as fluctuation in oxygen and/or nutrients available. Moreover, due to the fact that most material present in the EPS/SMP matrix consists of proteins and carbohydrates, the understanding of their nature and function has high priority.

DNA, RNA & PROTEINS – STANDARD ASSAYS

Over the last two decades molecular assays such as analysis, cloning, and sequencing of DNA fragments of abundant bacterial groups and/or single species have been frequently applied in order to understand microbial community diversity and function. Especially polymerase chain reaction (PCR) has become one of the most popular applications for identification, quantification and monitoring microbial communities not only in the field of MBR.

The direct amplification of mostly 16S rRNA genes from environmental samples provides a first identification of present microrganisms. Unfortunately, PCR is based on the amplification of known gene sequences. Extracted microbial DNA and/or RNA of unknown origins is consequently identified as unknown culture and the function of these microrganisms to the bioconsortium is uncertain (Rodríguez-Valera, 2004). Sabehi *et al.* (2003) showed that PCR was successfully applied to detect new and different types of a gene encoding a retinal protein, but at the same time others were completely missed. Amplifying DNA, however, has become a standardized experimental procedure. The technique is simple to perform, unexpensive and the results can show possible mutations in genes/genome (Bro and Nielsen, 2004).

Another significant assay was introduced in 1988 by Stahl and his colleagues: Fluorescence *in situ* hybridization (FISH). Henceforward, this culture-independent method has found widespread application in science for identification and quantification of microbial groups or single strains of environmental samples. Group-specific fluorescently labelled oligonucleotide probes are applied to target the ribosomal RNA (rRNA) present in all organisms. This semi-quantitative assay has been successfully utilized to determine microbial diversity and was an important achievement almost 20 years ago. But still, same limitation as for PCR occurs when the target sequence is unknown. Finally for microbial communities the presence of a single strain or species has less relevance compared to interrelations which takes permanently place in biocenose.

These standard assays provide some kind of pre-identification and they are therefore indirect approaches towards the functional diversity of microrganisms within a community, their metabolic activity and their responses (Ogunseitan, 1996). In particular, the DNA is the carrier of the complete genetic information for all ways of life, and in case of operational changes, relevant genes of the DNA will be expressed by transcription and translation mechanisms resulting in gene products such as functional proteins. Thus, DNA and/or RNA sequences provide indirect information on possible environmental changes/stress, whereas protein expressions are directly related to the biological state of the consortium.

With respect to MBR and the major interest in EPS and SMP (that mainly consist of proteins and carbohydrates), studies on functional gene expression of DNA and RNA have become impractical. A new approach called "metaproteomics" has been recently proposed for investigating environmental samples and activated sludge (Wilmes and Bond, 2004). This approach does not only lead to the identification of uncultured micro-organisms and their produced proteins, but also provides important information about the microbial enzymatic activity of the biomass. Thus its application can be helpful in the investigation of the interrelations between enzymatic activities, microbial dynamics and biofouling.

A NEW APPROACH TO FUNCTIONAL BIOMASS CHARACTERIZATION: METAPROTEOMS!

Basically proteins are synthesized through the expression of genes and therefore the term "proteome" describes the "*prote*in complement of the gen*ome*". This definition was first introduced by Wilkins and Williams in 1994. The qualitative/quantitative analysis of proteins provides direct information of organisms. Different to gene analysis (genomics) where only a single molecule of a high molecular weight (the DNA) is targeted, proteomic analysis is focussed on a various number of more than 1000 proteins for prokaryotes. The number can easily increase to more than 10,000 of proteins for eukaryotes.

According to their heterogeneous physical and chemical state, protein examinations are performed with difficulties. Hagenstein *et at.* (2006) indicated that proteomes are dynamic systems strongly influenced by small environmental changes such as microbial stress. Therefore different microbial protein profiles can be expected for different conditions in MBR for example. On the other hand, among biological wastewater treatment technologies MBR are better suited for maintaining steady process conditions, thus they provide better opportunities for monitoring microbial communities and their relationship with easily controllable environmental conditions.

For the separation, identification and quantification of protein samples different techniques are applicable. Two dimension polyacrylamide gel electrophoresis (2-D) first introduced by O'Farrell in 1975 is performed to separate very complex protein mixtures. The technique delivers information about intact proteins, isoforms or post-translational modifications. For 2D electrophoresis proteins are first extracted and purified, and later separated on two dimensions. The first dimension is based on the isoelectric focussing (IEF) in an immobilized pH gradient (IPG) strip. The protein sample, an amount of 200 µg approximately, is loaded on the IPG-strip and will be separated depending to their intrinsic electrical charge (p*I*) and molecular mass. Further separation is conducted at the second dimension using SDS-polyacrylamide gel electrophoresis (SDS-PAGE). The gradient gel contains sodium dodecyl sulfate (SDS) masking the p*I* of the proteins (Görg *et al.,* 2004). The polypeptides are then only separated due to their molecular weight. The resolution of 2D electrophoresis was continuously improved, but Hagenstein et al. (2006) pointed out that important protein classes like membrane-associated proteins can be only hardly detected due to resolution limitations of the method. Therefore the combine of SDS-PAGE with reversed-phase liquid chromatography-tandem mass spectrometry (LC-MS/MS) analysis is frequently carried out.

Another method applied for protein separation is fluorescent 2D differential gel electrophoresis (DIGE) where two different protein samples are compared due to the labelling of their different proteomes using fluorescent cyanine minimal dyes. Methods such as multidimensional protein identification technology (MudPIT) and isotope-coded affinity tagging [(c)ICAT] are "gel-free" methods and based on the separation on liquid chromatography (LC), but can only be applied to cystein-containing proteins and peptides.

For the identification of the selected protein spots obtained for example by SDS-PAGE are following digested and analysed. Therefore frequently matrix assisted laser desorption ionization time-of-flight mass spectrometry (MALDI-ToF MS) is favoured. The resulting mass spectra of the proteins will be then matched using sequence databases. Another method can be performed by using liquid chromatography mass spectrometry (LC-MS/MS).

This short introduction in the field of scientific proteomic laboratory work shows only a few parts of the diversity to protein analysis. However, pioneers like Wilmes and Bond (2004, 2006) applied the proteomic approach to a mixed community of prokaryotic micro-organisms. They proposed the term “metaproteomics” for the characterization of environmental microbiota at a given point in time according to the term “metagenome” by Rodríguez-Valera (2004). They performed metaproteomics first in 2004 and later in 2006 to a biomass of a laboratory-scale sequencing batch reactor (SBR). They successfully demonstrated the use of 2D-PAGE for protein separation and analysed strongest protein spots obtained with MALDI-ToF MS. They were capable to isolate and identify several proteins from the microbial community. Later in 2006 they applied metaproteomes to two different sludges and matched 630 individual protein spots. They showed significant differences between the two sludges due to the different metaproteomic maps for both sludges.

Metaproteomic analysis of Chesapeake Bay microbial community was reported by Kan *et al.* in 2005. He and his colleagues investigated the expressed protein profiles of natural microbial communities. The authors clearly pointed out the difficulties for protein identification by use of MALDI-TOF MS. They obtained distinct mass spectra form 34 protein spots, but did not find significant sequences by searching in the databases due to the fact that the applied method requires 97% of amino sequence identity to find a significant match. Thus, post-translational modification of proteins will be hardly identified due to MALDI-TOF MS. Therefore LC-MS/MS was conducted to improve metaproteomes identify. The protein identification for the Chesapeake Bay microbial communities, however, was limited due to the fact that at least two peptides per protein must be matched in order to avoid false-positive identifications. Finally the authors concluded the metaproteomic approach was applicable to microbial communities, but the interpretation of results obtained especially for the protein identification requires not only a good and comprehensive knowledge about applied mass spectrometry, but also a realistically point of view for their limitations and possible pitfalls.

Banfield *et al.* (2005) used genomic and mass spectrometry-based methods including proteomic methods to a natural microbial biofilm of an acid mine drainage (AMD). He and his colleagues compared different microbial growth stages in different times of the AMD. They were capable to identify most abundant microbes, but pointed out that for better a understanding of the role and function of proteomes to the community further improvements with regard to more precise measurements are strongly required.

Gu *et al.* applied the metaproteomic approach in 2006 in order to study the protein profiles of EPS and activated sludge of an MBR. They operated the reactor with two different feed solutions (glucose or acetate) and compared protein profiles obtained. They were able to demonstrate distinct protein profiles regarding to the two different types of carbon source in the feed solution. They also concluded that physical and chemical properties such as the total organic carbon (TOC), relative hydrophobicity (RH) and EPS analysis with respect to the portion of carbohydrates, proteins and humic substances did not provide sufficient information about the difference due to the two different fed conditions.

However, summarizing all available information reported throughout in the last few years in the young field of metaproteomic it can be concluded that metaproteomes as new approach has been successfully applied to link taxonomic diversity, functional diversity and biological processes to different aquatic ecosystem. But still, there exist some relevant limitations not only for the analysis and identification of proteins, but also for the procedure of sample preparation.

In comparison, all presented reports above in this review have shown significant differences in manner of samples pre-treatment. The sample preparation is the base for the work with proteomes and following mistakes cannot be compensated later by applying further analysis methods. Some authors, for example, utilize protease inhibitors and treat their samples with DNase and RNase, others relinquish. Both DNase and RNase are enzymes and will therefore occur on the SDS-PAGE.

The use of protease inhibitors has high importance to the identification of the proteins and it is therefore strongly suggested. The analysis of protein samples without protease inhibitors is impractical, because a lysis of the protein through the present proteases in the samples cannot be excluded. In this case the origin of the protein cannot be determined due to the incomplete occurrence of polypeptides.

Some authors propose very short protein extraction protocols; others conduct very long and intensive procedures. Sometimes filtration is performed as first sample pre-treatment which can dramatically change the composition of the sample and the interpretation of results is highly hypothetic. Further differences were found to initiate the cell lysis which is carried out through sonication, French press, lysis buffer or sample boiling for short periods. Concluding, all reports based on metaproteomic approaches distinguish slightly in the extraction protocols. For further metaproteomic developments and technical improvements standard methods for the sample pre-treatment is strongly recommended. Finally limited protein identification as indicated by Kan and also by Banfield should be always carefully considered when applying metaproteomes to a functional biomass characterization.

ON-GOING EXPERIMENTAL ACTIVITY (WITHIN MBR TRAIN PROJECT)

This review is part of a larger project started in October 2006 within the 6th FP Project MBR-TRAIN and focusing on the functional characterization of MBR biomass, with the aim to achieve a better understanding of microbial dynamics and biofouling mechanisms, especially EPS/SMP-protein expression.

Accordingly, in the first experimental period a procedure for the protein extraction from activated sludge is being developed and optimized in order to define a protocol. The biomass is currently obtained from a bench scale plant fed with a synthetic wastewater. The protein samples are separated by performing 2D electrophoresis and this is currently being optimized. Achieved protein profiles will be analysed and quantified, and selected protein spots will be identified by MALDI-TOF MS.

Subsequent, a pilot plant will be operated as a submerged MBR equipped with a hollow fibre module for performing COD and nitrogen removal. Initially, in order to avoid having a biomass with a very complex nature the reactor will be also fed with synthetic wastewater. The plant operation will be optimized with respect to operating parameters such as the flux and the sludge retention time, and biomass yield and treatment efficiencies will be monitored by measuring the following parameters: TSS, VSS, COD, N-NH_4, N-NO_2, N-NO_3, and TMP. Microbial dynamics and composition during the start-up and at the steady state will be studied by performing metaproteomic approach and standard assays to compare and complement the study. Final aim of the research activity would be to monitor environmental variations and/or stress on the biomass through the changes of protein profiles.

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REFERENCES

Banfield .JF., Ram R.J., VerBerkmoes N.C., Thelen M.P., Tyson G.W., Baker B.J. Blake R.C. II, Shan M., Hettich R.L. (2005). Community proteomics of a natural microbial biofilm. Science, 308, 1915-1920.

Böckelmann U., Janke A., Kuhn R., Neu T. R., Wecke J., Lawence J.R., Szwezyk U. (2006). Bacterial extracellular DNA forming a defined network-like structure. FEMS Microbiol. Lett., 262, 31-38.

Bro C. and Nielsen J. (2004). Impact of 'ome' analyses on inverse metabolic engineering. Metabolic Engineering, 6, 204-211.

Cicek N., Franco J.P., Suidan M.T., Urbain V., Manem J. (1999). Characterization and comparison of a membrane bioreactor and a conventional activated-sludge system in the treatment of wastewater containing high-molecular-weight compounds. Water Environ. Res., 71(1), 64-70.

Chang I.-S., Kim S.-N. (2005). Wastewater treatment using membrane filtration – effect of biosolids concentration on cake resistance. Process Biochemistry, 40, 1307-1314.

Field R.W., Wu D., Howell J.A., Gupta B.B. (1995). Critical flux concept for microfiltration fouling. J. Membrane Sci., 100, 259-272.

Fux C.A., Costerton J.W., Stewart P.S. and Stoodley P. (2005). Review, Survival strategies of infectious biofilms. TRENDS in Microbiology, 13, (1), 34-40.

Gander M., Jefferson B., Judd S. (2000). Aerobic MBRs for domestic wastewater treatment: A review with cost consideration. Sep. and Purif. Technol., 18, 119-130.

Görg A., Weiss W., Dunn MJ. (2004). Current two-dimensional electrophoresis technology for proteomics. Proteomics, 4, 3665-3685.

Gu J.D., Li X.Y., Xie B. (2006). Protein profile of extracellular polymeric substances and activated sludge in a membrane biological reactor by 2-dimensional gel electrophoresis. Water Science and Technol.: Water Supply, 6(6), 27-33.

Hagenstein M.C., Sewald N. (2006). Chemical tools for activity-based proteomics. J. of Biotechnol., 124, 56-73.

How Y. Ng, Hermanowicz S. (2005). Membrane bioreactor operation at short solids retention times: performance and biomass characteristics. Water Res., 39, 981-992.

Judd S. (2006). The MBR Book: Principles and applications of membrane bioreactors in water and wastewater treatment. Elsevier, London

Kan J., Hanson E.T., Ginter J.M., Wang K., Cheng F. (2005). Metaproteomic analysis of Chesapeak Bay microbial communities. Saline Systems, 1(7), doi: 10.1186/1746-1448-1-7.

Kim In S., Jang Namjung (2006). The effect of calcium on the membrane fouling in the membrane bioreactor (MBR). Water Res. 40, 2756-2764.

Laera G., Pollice A., Saturno D., Giordano C., Lopez A. (2005). Zero net growth in a membrane bioreactor with complete sludge retention. Water Res., 39, 5241-5249.

Ogunseitan O.A. (1996). Protein profile variation in cultivated and native freshwater microorganisms exposed to chemical environmental pollutants. Microbial Ecol., 31, 291-304.

Ogunseitan O.A. (1997). Direct extraction of catalytic proteins from natural microbial communities. J. of Microbiol. Methods, 28, 55-63.

Pollice A., Laera G., Blonda M. (2004). Biomass growth and activity in a membrane bioreactor with complete sludge retention. Water Res., 38, 1799-1808.

Pollice A., Brooks A., Jefferson B., Judd S. (2005). Sub-critical flux fouling in membrane bioreactors – a review of recent literature. Desalination, 174, 221-230.

Rodríguez-Valera F. (2004). Environmental genomics, the big picture? FEMS Microbiol. Lett., 231, 153-158.

Rosenberger S., Krüger U., Manz W., Szewzyk U., Kraume M. (2002). Performance of a bioreactor with submerged membranes for aerobic treatment of municipal waste water. Water Res., 36, 413-420.

Rosenberger S., Laabs C., Lesjean B., Gnirss R., Amy G., Jekel M., Schrotter J.-C. (2006). Impact of colloidal and soluble organic material on membrane performance in membrane bioreactors for municipal wastewater treatment. Water Res., 40, 710-720.

Sabehi G., Massana R., Bielawski J P., Rosenberger M., Delong E.F., and Béjà O. (2003). Novel proteorhodopsin variants from Mediterranean and Red Seas. Environ. Microbiol., 5, 842-849.

Stahl D.A., Flesher B., Mansfield HR., Montgomery L. (1988). Use of phylogenetically based hybridization probes for studies of ruminal microbial ecology. Appl. Envrion. Microbiol., 54, 1079-1084.

Trussell S.R., Merlo R., Hermanowicz S.W., Jenkins D. (2006). The effect of organic loading on process performance and membrane fouling in a submerged membrane bioreactor treating municipal wastewater. Water Res., 40, 2675-2683.

Ueda T., Hada K., Kikuoka Y. (1996). Treatment of domestic sewage from rural settlements by a membrane bioreactor. Water Sci. Technol., 34(9), 189-196.

Wilmes P. and Bond P.L., (2004). The application of two-dimensional polyacrylamide gel electrophoresis and downstream analyses to a mixed community of prokaryotic microorganisms. Environ. Microbiol., 6(9), 911-920.

Wilmes P. and Bond P.L. (2006). Towards exposure of elusive metabolic mixed-culture processes: the application of metaproteomic analyses to activated sludge. Water Science and Technol., 54(1), 217-226.

Wimpenny J., Manz W., Szewzyk U. (2000). Heterogeneity in biofilms. FEMS Microbiol. Reviews, 24, 661-671.

Yamato N., Kimura K., Miyoshi T., Watanabe Y. (2006). Difference in membrane fouling in membrane bioreactors (MBRs) caused by membrane polymer materials. J. Membrane Sci., 280, 911-919.

Yang W., Cicek N., Ilg J. (2006). State-of-the-art of membrane bioreactors: Worldwide research and commercial applications in North America. J. Membrane Sci, 270, 201-211.

Yu-Lan J., Woo-Nyoung L., Chung-Hak L., In-Soung C., Xia H. (2006). Effect of DO concentration on biofilm structure and membrane bioreactor. Water Res., 40, 2829-2836.

Use of water soluble polymers to reduce capital and operating costs of Membrane Bioreactors (MBR)

Jeroen Koppes*, Seong-Hoon Yoon **, John Collins** and Bhasker Davé **

*European Water R&D Department, Nalco Europe B.V., Ir. G. Tjalmaweg 1, 2342 BV Oegstgeest, the Netherlands (E-mail: *jkoppes@nalco.com*)
**Nalco Company, 1601 Diehl Rd., Naperville, IL 60563-1198, USA (E-mail: *syoon@nalco.com*)

Abstract Recently developed Membrane Performance Enhancer (MPE) product PermaCare® MPE30 was tested on a full-scale membrane bioreactor (MBR) treating landfill leachate wastewater. The full-scale MBR was suffering from reduced flux due to severe membrane fouling, possibly caused by the high salt concentration in the influent. This MBR was facing membrane expansion requirements as it could only treat ½ -2/3 of the wastewater flow. The purpose of this study was to investigate the effect of MPE on the flux and consequently on the capital expansion savings. With the addition of 400 mg/l MPE30 to the MBR it was possible to increase the flow by 50% and to reduce the trans membrane pressure (TMP) by a factor of two. PermaCare® MPE30 could prevent the otherwise required capital expansion.

Keywords: Flux increase, Membrane Bioreactor (MBR), Membrane fouling reduction, Membrane Performance Enhancer (MPE), Peak flow

Introduction

Membrane Bioreactors (MBRs) are increasingly replacing conventional activated sludge processes for municipal and industrial wastewater treatment. In spite of several benefits of MBRs over conventional activated sludge, including better effluent quality and smaller footprint, is membrane fouling still a major operational problem in the MBR process. Because of membrane fouling, which is mainly caused by colloidal material and extra-cellular polymeric substances (EPS), plants have to be operated at lower fluxes, which in turn results in both higher capital (more membrane area required) as well as operating cost. Currently, fouling is minimized by the combination of excess membrane area, scouring by course bubble aeration, back-pulsing with filtrate water, periodic membrane relaxation (stopping of filtration), and chemical cleanings. These techniques, although effective to varying extents, increases the investment and operating costs of MBRs.

Traditionally, water-soluble polymers, especially cationic polymers, have not been used directly in membrane systems because it is generally understood that such cationic polymers foul the negatively charged surfaces of membranes, causing the drastic reduction in fluxes. Specially modified cationic polymers were developed to increase the flux with 30-100 %, resulting in significant reduction of the operational and investment costs. The polymers used in this study are designated as Membrane Performance Enhancer (MPE). These products can, for example, reduce the amount of excess membrane area when applied during high fouling conditions, peak flow events or cold wastewater temperature. MPE polymers incorporate the smallest colloidal fraction into the sludge flocs, including free EPS. The reduced fouling rate can also be attributed to a increased membrane cake layer porosity (B.-H. Hwang *et al.* 2006)

In the example a full-scale leachate MBR was suffering from reduced flux due to severe membrane fouling. The purpose of this study was to investigate the effect of MPE on the flux and consequently on the reduction of the MBR capital and operational costs.

Methods

Laboratory scale filtration tests were used to evaluate the filterability of the sludge and the effect of MPE on the filtration characteristics. The blank was prepared by taking 50 ml sludge from the MBR. This sludge was vigorously mixed on a magnetic stirrer for 10 minutes at 500 rpm. After mixing, the sludge was filtered through a wetted Whatman 42 filter paper. The filtered water volume was recorded after 5 minutes. The same procedure was followed with addition of a variety of MPE concentrations. A graph was used to plot the filtrate volume as a function of the MPE dosage.

During full-scale experiments, PermaCare MPE30 was added directly to the aeration basin or to the mixed liquor recycle flow going into the aeration basin, depending on the turbulence at the injection point. The extent of membrane fouling was monitored by measuring the wastewater extraction flow rate and the trans membrane pressure (TMP). If necessary, additional MPE was added daily or intermittently to improve performance and to compensate for the loss of MPE due to mixed liquor removal or degradation.

Theory

A higher membrane flux can prevent both capital spending for membrane modules and operational costs.

For a given MBR plant having a flow rate of Q (l/h) and a design flux of J (l/m^2/h), the total membrane area necessary to treat the flow can be calculated as Eq. (1), where A_0 is the original membrane area in m^2.

$$A_0 = \frac{Q}{J} \qquad (1)$$

The membrane area savings (ΔA) with flux increase can be calculated as Eq. (2), where ε is the flux increase. For instance, if flux increases by 50% (or $\varepsilon = 0.5$), savings in membrane area will be 0.33 A_0 (Yoon *et al.*, 2007).

$$\Delta A = \frac{\varepsilon\, A_0}{1 + \varepsilon / 100} \qquad (2)$$

If required membrane area to treat certain flow decreases, all associated costs with the membrane such as module frames, piping, blowers, flow meters, labour for maintenance, *etc*. should decrease. For simplification, these associated costs were included in membrane costs using a multiplication factor, α, where $\alpha = 3$ means that the total costs for certain membrane areas triple the pure membrane costs. Finally the savings in membrane system can be calculated as Eq. (3), where ΔS is a membrane cost saving (€) and M is a unit membrane costs (€/m^2) (Yoon *et al.*, 2007).

$$\Delta S = \frac{Q}{J}\left(\frac{\alpha\varepsilon}{1 + \varepsilon / 100}\right)M \qquad (3)$$

Though costs for membrane tank also can decrease as a result of the less membrane frames, this portion was not considered for this calculation for the sake of simplicity.

In terms of operating cost savings, there are four major savings, i.e. 1) aeration costs for membrane scouring, 2) membrane replacement costs, 3) membrane cleaning costs and 4) labour costs for maintenance. In this discussion, only the aeration costs and membrane replacement costs will be considered because the other two costs are relatively minor.

If membranes are submerged in an aeration tank directly, reduction of scouring air may or may not result in more fine bubble aeration to maintain a required dissolved oxygen (DO) level such as 1-2 mg/L. However, the current trend is to separate the aeration tanks and membrane tanks to make the maintenance easier. In these systems, the DO level in the aeration tank is hardly affected by the reduction of scouring air in membrane tank (Yoon *et al*., 2007).

Since the volume of scouring air is proportional to the membrane area, savings of scouring air should be proportional to the membrane savings. The equation to calculate the savings in aeration (ΔQ_{air}, m^3/min) can be written as Eq. (4), multiplying unit aeration rate, q_{air} (m^3/min/m^2) by membrane savings shown in Eq. (2).

$$\Delta Q_{air} = \frac{\varepsilon\, A q_{air}}{1 + \varepsilon / 100} \qquad (4)$$

As a result of the aeration savings, power consumption for blowers should decrease. Considering the head loss in the aeration, H (m H_2O), power savings, ΔP (kW), can be calculated as Eq. (5) (modified from an equation for screw compressors, Wagner and Pöpel, 1998).

$$\Delta P = 0.306\, \Delta Q_{air}\, H^{0.8} \qquad (5)$$

Finally, annual savings can be calculated by multiplying unit power costs (€/kWh), and annual operation time (hours).

Results and Discussion

Full scale MBR study

A MBR plant located in southern Europe treated 300m3/day high strength wastewater from a landfill site (leachate). Due to the dry climate was the leachte wastewater characterized by a high contamination level, COD of 30.000mg/l, ammonia of 5.000mg/l and a conductivity of 50 mS/cm. As shown in Fig. 1, influent was coming into a pre-anoxic tank and mixed with recycled sludge to denitrify. The mixed liquor was then sent to an aerobic basin to nitrify the ammonia in the influent into nitrate/nitrite. After further denitrification in the post-anoxic tank, the mixed liquor was separated in the membrane tank to get effluent. The effluent was further treated in a reverse osmosis system to produce fresh water, as not water resource was available on-site.

The total submerged plate membrane area installed was 1000 m^2 and the design flux was 12.5 l/m2/h, which was lower than a MBR treating conventional municipal wastewater. The MLSS concentration in the aeration tank was maintained at around 12g/l.

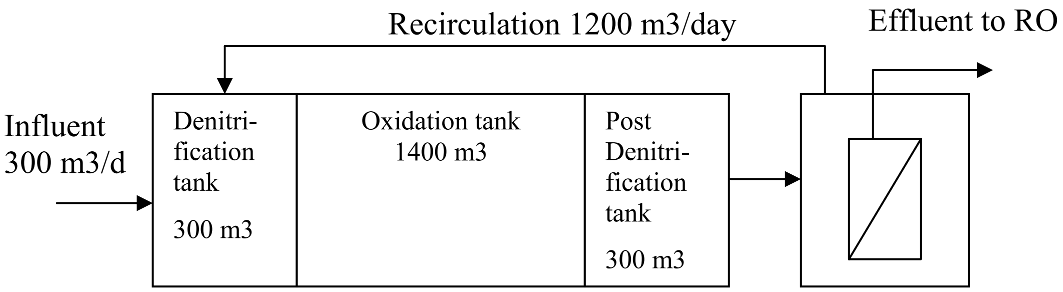

Fig. 1. Schematic of a MBR plant treating leachate wastewater from a landfill site

After start up the MBR had to be operate at much lower flux (<10 $l/m^2.h$) than designed and at high TMP, 0.3-0.4 bar. The membranes were prone to fouling and could not treat 1/3 to 1/2 of the wastewater in spite of lower than design flow (200-250m^3/day vs 300m^3/day) and frequent membrane cleanings. The high membrane fouling was probably due to the high salinity (50 mS/cm), which is about equal to that of seawater. A constant high salinity level is usually not a problem for the biological conversion process however the floc formation can be retarded resulting in increased fouling potential. The smaller MLSS size is one of the reasons why leachate wastewaters in Europe are often treated in MBR's because conventional activated sludge systems, based on Strokes law gravity separation, would require too large clarifiers. Membranes were also fouled more at high salinity conditions in lab experiments. This observation was confirmed in a municipal MBR plant, where periodic seawater intrusion was experienced (Reid, 2005). Another possibility for high fouling conditions was that the leachate might contain certain chemicals that cause fouling or that the wastewater composition made bacterial conditions less favourable resulting stress to the bacteria.

As a result of the abnormal membrane-fouling tendency, the plant was faced with a new capital investment to increase the membrane capacity 50–100% in order to prevent the landfill to fill-up with leachate wastewater. To avoid a large capital investment, the plant sought alternative solutions. Filterability tests were performed on sludge from the aeration tank and post denitrification tank to screen the best available MPE product for the sludge. Figure 2 shows the relation between the sludge filterability and MPE30 dosage. MPE30 was selected because it provided the best floc formation and the best drainage in this high salinity environment. The optimum dosage was estimated at 400 mg/L based on the total biological volume of the MBR system.

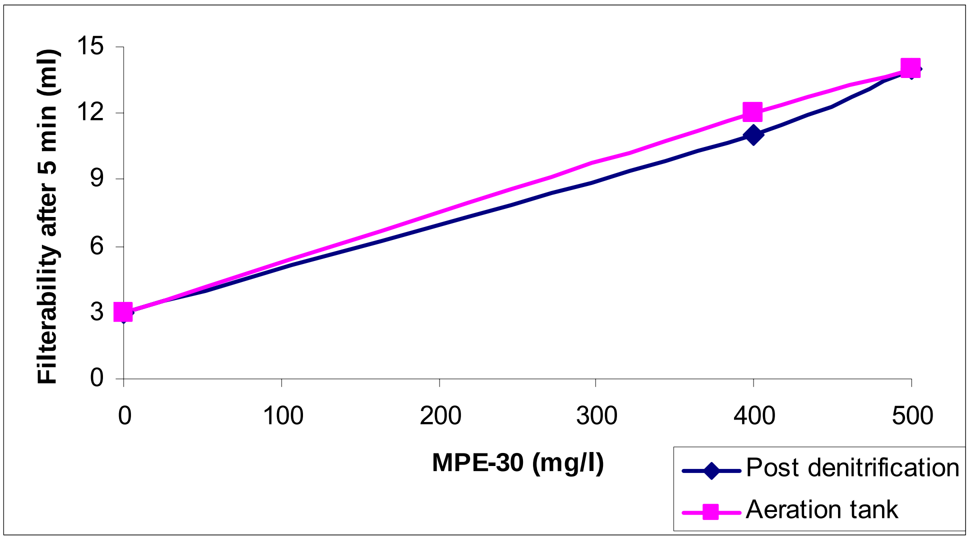

Fig 2 Sludge filterability as a function of the MPE30 dosage

A normal in-situ membrane cleaning was performed before MPE30 was added to the full-scale MBR system. Right after the dose of MPE30 (800 kg neat product into the MBR sludge recycle flow), TMP sharply decreased from 0.6 bar to 0.1 bar (Fig. 3), while flux increased simultaneously. Overall 50% more wastewater has been treated. No additional membrane cleaning other than the regular cleanings recommended by the membrane installer has been necessary. To compensate for the loss through excess sludge removal, 40 kg of MPE30 was added everyday (based on a sludge age of 20 days). After this the plant has been operated at a stable condition for 1 year.

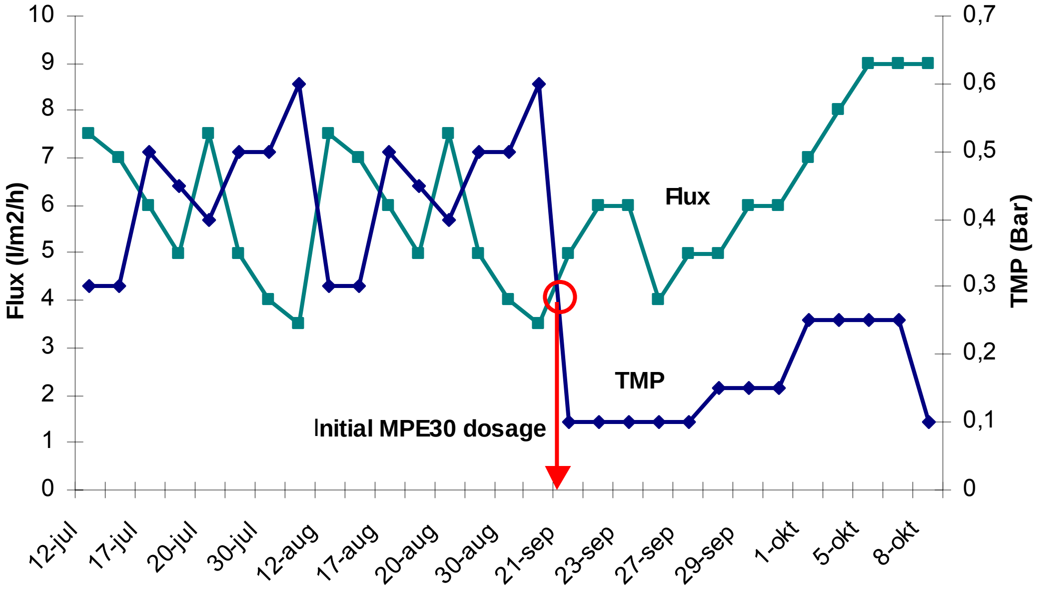

Fig. 3. Effect of MPE30 on flux and trans-membrane pressure (TMP).

As a result of using MPE30, the plant avoided membrane system expansion. Had MPE30 not been used, at least 50% more membrane would have been required, which corresponds to 500 m^2 of additional membrane. Using the identical assumptions used in above section, savings in capital was estimated to be € 75,000. Annual electricity cost savings were €10.928 and € 4.166 in membrane replacement costs (6 years life-time). If the capacity of the membrane system was increased 100%, savings should increase twice, *i.e.* capital savings of € 150,000 and annual electricity savings of € 21.856 and € 8.332 membrane replacement costs.

Conclusions

PermaCare MPE30 was effective in reducing the fouling potential in a MBR that was experiencing extremely high fouling conditions. MPE was successful in eliminating the need for capital expansion. In this example MPE30 could avoid membrane investment of € 75,000. In addition this plant could rapidly process the total wastewater flow without the need to wait for the extra hardware installation.

References

Yoon, S., Collins, J., Koppes, J., and Dave, B. (2007) Reduction of capital and operating costs of membrane bioreactor using modified polymers, IWA World Congress- Maspalomas, Grand Canaria Spain.

Collins, J., Yoon, S., Musale, D., Kong, J., Koppes, J., Sundararajan, S., Tsai, S., Hallsby, A., Cachia, P., and Kronoveter, K. (2006) Membrane performance enhancer evaluations on pilot- and full-scale membrane bioreactors, Water and Environment Journal, 20, 43-47.

Wagner, M. and Pöpel, J. (1998) Oxygen transfer and aeration efficiency-Influence of diffuser submergence, diffuser density, and blower type, Water Science & Technology, 38(3) 1-6.

Elizabeth Reid (2005) Salinity shocking and fouling amelioration in Membrane Bioreactors, PhD thesis, Cranfield University, UK.

Hwang, B., Lee W., Park, P., Lee, C., Chang, I. (2006) Effect of membrane fouling reducer on cake structure and membrane permeability in membrane bioreactor, J. Membrane Science 2006.

Highly fouling resistant ultrafiltration membranes for water and wastewater treatments

H. Susanto* and M. Ulbricht*

*Lehrstuhl für Technische Chemie II, Universität Duisburg-Essen, 45117 Essen, Germany
(E-mail: *heru.susanto@uni-due.de*; *mathias.ulbricht@uni-due.de*)
Permanent address: Department of Chemical Engineering, Universitas Diponegoro, Indonesia

Abstract Control of fouling is a critical issue to increase the competitiveness of ultrafiltration (UF) membranes for drinking water and wastewater treatments. Highly fouling resistant UF membranes synthesized by photo-graft copolymerization of a water soluble monomer, poly(ethylene glycol) methacrylate (PEGMA), onto a polyethersulfone UF membrane have been evaluated with respect to the adsorptive as well as the ultrafiltration fouling. Protein, humic substance and polysaccharide solutions were used as the model for foulants occurring in the water sources for drinking water as well as in wastewater effluents. The results show that the modified membranes exhibited a much higher fouling resistance for all foulants than the unmodified membranes. Their combined high fouling resistance and high rejection suggests that the obtained modified membranes are very promising as a new generation of thin-layer composite low fouling UF membranes for drinking water and wastewater treatment applications.

Keywords Fouling; natural organic matter; polysaccharide; protein; ultrafiltration; surface modification.

INTRODUCTION

Nowadays, along with the increasing demand of ultrafiltration (UF) for various other applications, the use of UF for drinking water and wastewater treatments has become more and more interesting (Glucina et al., 2000; Atkinson, 2006). UF not only can produce a high product (water) quality but can also reduce the cost for treatment. However, fouling − resulting in loss of performance of a membrane − is the major limitation. Consequently, it reduces the process economics and restricts a more widespread applicability of UF for water and wastewater treatments. Many studies have been devoted to overcome the problem of fouling. Basically, those studies include foulant identification/characterization, investigation of fouling mechanisms and minimizing or control of fouling. Control of fouling seems to be the most important aspect from the practical point of view. Thus, many methods have been proposed to control the fouling. Process conditions (e.g., adjusting operation conditions, pre-treatment and back-flushing) have been intensively developed in order to achieve better control of membrane fouling, but in most cases, the permeate fluxes are clearly determined by membrane itself. Therefore, it would be desirable having available high-performance membranes with a pronounced and stable resistance towards fouling by all foulants present in drinking water resources and wastewater effluents. Surface modification was then proposed as an important strategy for control of fouling (Ulbricht, 2006). Charge-based modification to reduce the fouling during natural organic matter (NOM) removal was reported (Wei, et al., 2006). However, such modification is strongly dependent on the pH and ionic content of the solution. Recently, we have started to modify a polyethersulfone (PES) membrane for UF of aquatic humic substances by a hydrophilization approach, which is not limited with respect to a general use of the obtained membranes (Susanto and Ulbricht, 2006). However, to be practically useful, the resulting membrane performance must be further improved with respect to fouling resistance, permeate flux as well as rejection. More importantly, it also needs to be evaluated with other potential foulants occurring in drinking water sources (e.g., surface water and ground water) and wastewater effluents.

During application of UF for drinking water and wastewater treatments, two big issues have been identified from past studies: NOM is considered to be primary contributor to membrane fouling in drinking water treatment (e.g., Maartens et al. (1999); Kweon and Lawler (2005); Hilal et al. (2004)), whereas soluble extracellular polymeric substances (EPS), which are mainly composed of polysaccharides, are considered as one of the important biofoulants during wastewater treatment (Rosenberger et al., 2005; Ye et al., 2005). In addition, protein, an already very well known foulant, was also reported as one of the main components in the effluents of a membrane bioreactor (Jin et al., 2004; Jarusutthirak et al., 2002; Rosenberger, 2005). Asatekin et al. (2006) used proteins, polysaccharides and NOM as models for foulants in a membrane bioreactor.

In this work, novel thin-layer polymer hydrogel composite PES-based UF membranes are evaluated for water and wastewater treatment applications. Those membranes were synthesized by photo-grafting of a monomer for a hydrogel polymer, poly(ethylene glycol) methacrylate (PEGMA), onto commercial PES UF membranes (Susanto and Ulbricht, 2007). The evaluation was done by investigation of membrane-solute interactions (adsorptive fouling) and membrane-solute-solute interactions (ultrafiltration). Myoglobin, humic acid and dextran solutions were used as the models for protein, NOM and polysaccharide components, respectively.

METHODS

Materials

Commercial PES UF membranes with a nominal molecular weight cut-off (NMWCO) of 100 kg/mol (PES-SG100) and 10 kg/mol (PES-SG10) obtained from Sartorius, Germany, were used. A PES-SG100 membrane was used as the base membrane for modification. Prior to use for experiments, the membranes were washed with ethanol by shaking at 100 rpm on a mechanical shaker for 1.5-2 hours and then equilibrated with water. Only membrane samples that had initial water permeability in the range $\pm$ 15% relative to the average values (5.71 $\pm$ 0.85 L/m^{2}hkPa) were used for modifications (cf. Susanto and Ulbricht, 2005 for membrane selection). Poly(ethylene glycol) methacrylate (PEGMA 400, the number indicating PEG molar mass in g/mol) was purchased from Polysciences Inc., Warrington, USA. N,N'-methylenbisacrylamide (MBAA), myoglobin from horse skeletal muscle (95-100% purity) and humic acid (HA) were purchased from Sigma-Aldrich Chemie GmbH, Steinheim, Germany. Potassium dihydrogen phosphate (KH_2PO_4) and disodium hydrogen phosphate dihydrate ($Na_2HPO_4.2H_2O$) were purchased from Fluka Chemie AG, Buchs, Germany. Nitrogen gas purchased from Messer Griesheim GmbH, Krefeld, Germany, was ultrahigh purity. Water purified with a Milli-Q system from Millipore was used for all experiments.

Membrane modification

A UVA Print system (Hoenle AG, Gräfelfing, Germany) equipped with a high-pressure mercury lamp with wavelengths more than 300 nm, providing homogenous illumination of up to 100 cm^2 area with an intensity of 35 $\pm$ 5 mW/cm^2 (measured with a UVA meter, Hoenle AG) was used. Circular PES membrane samples with a diameter of 25 mm were immersed into monomer solutions in a Petri dish. A second smaller glass Petri dish was used to cover the membranes and also as another deep-UV filter. Thereafter, the samples were subjected to UV irradiation for various time periods. Then, the membranes were taken out, immediately rinsed with water and then washed with excess of water to remove any unreacted monomer or physically adsorbed polymer. The washing was sequentially done at room temperature for 30 min, at 50 $\pm$ 2°C for 2 hours and again at room temperature for 30 min.

Static adsorption, ultrafiltration procedure and solute analyses

All experiments were conducted with a dead-end stirred cell filtration system. The system consisted of a filtration cell (Amicon model 8010, Millipore) connected to a reservoir (~450 or 1850 mL). It was pressurized by nitrogen. To avoid the effects of membrane compaction on the interpretation of modification and fouling data, each sample was firstly compacted by filtration of pure water at pressure of 450 kPa for at least 0.5 hours. Thereafter, the pressure was reduced to the desired pressure for water flux measurement. For static adsorption experiments, the water flux was initially measured and then a test solution (myoglobin (1 g/L; pH 7 in phosphate buffer) or humic acid (100 mg/L, pH 7.2, 1mM Ca^{2+}, conductivity 1100 mS/cm) or dextran (T10, 10 g/L in water)) was added to the cell (before use, the myoglobin as well as HA solution was pre-filtered through a 0.45 µm microfilter from Sartorius, Germany, to remove undissolved material). Thereafter, the outer membrane surface was exposed for certain time (2 h, 18 h and 3 h, for myoglobin, HA and dextran solutions, respectively) without any flux at a stirring rate of 300 rpm. Then, the test solution was removed and the membrane surface was rinsed two times by filling the cell with pure water (5 mL) and shaking it for 30 seconds. Water fluxes before and after exposing were compared. The evaluation of membrane performance was expressed in term of adsorptive fouling resistance (cf. Eq. (1)). An adsorptive fouling resistance of 1 means no adsorptive fouling was occurred. Ultrafiltration of myoglobin (1 g/L, pH 7.2) and dextran (1 g/L in water) solutions were conducted at the same transmembrane pressure (100 kPa) for all membranes, whereas ultrafiltration of humic acid solution (50 mg/L, pH 7.2, 1 mM Ca^{2+} added, conductivity 1100 µS/cm) was done at similar initial water flux (92 L/m^2h). In these experiments, the balance was connected to the PC and weight of permeate was online recorded. Profiles of permeate flux and apparent solute rejection over time were investigated. Myoglobin and humic acid concentrations were determined by measuring UV absorbance at 230 and 255 nm, respectively. The dextran concentration was analyzed by gel permeation chromatography (HP-GPC) using RI detector (cf. Susanto and Ulbricht, 2005).

$$R_{ads} = \frac{J_{ads}}{J_o} \qquad (1)$$

where R_{ads} is adsorptive fouling resistance, J_o and J_{ads} are water fluxes before and after exposing to the test solutions, respectively.

RESULTS AND DISCUSSION

First of all, it is important to note that the two modified membranes, which were evaluated in detail, are PES-g-PEGMA (PES grafted from 40 g/L of PEGMA and by using 5 min UV irradiation) and PES-g-PEGMA/MBAA (PES grafted from a solution with 40 g/L of PEGMA and 0.4 g/L MBAA and by using 4 min irradiation). The modification created thin polymer hydrogel layers on the surface of base PES UF membrane. Therefore, the modified membranes are considered as composite membranes (cf. Susanto and Ulbricht, 2007). Those modified membranes had similar rejection curve as compared with the unmodified PES membrane with NMWCO of 10 kg/mol (PES-SG10) indicating similar pore size as well membrane cut-off, but had much lower contact angle (cf. Susanto and Ulbricht, 2007). In order to consider the effects of flux-rejection trade-off during the evaluation, the PES-SG10, which had similar cut-off, was used as the reference during performance analysis.

Membrane-solute interactions (adsorptive fouling)

The adsorptive fouling resistance of the composite membranes was investigated and compared with unmodified membrane. The results are presented in Fig. 1. It is clearly observed that the composite membranes showed higher adsorptive fouling resistance than unmodified membranes meaning that water flux after adsorptive fouling was higher. For example, the modification could increase the R_{ads} caused by protein from 0.45 (for PES-SG10) to around 0.93 and the R_{ads} caused by HA could be increased from 0.67 to ~0.93. The more hydrophilic character of the surface of the composite membranes (as evidenced by their much lower contact angle) is the most probable reason for this phenomenon.

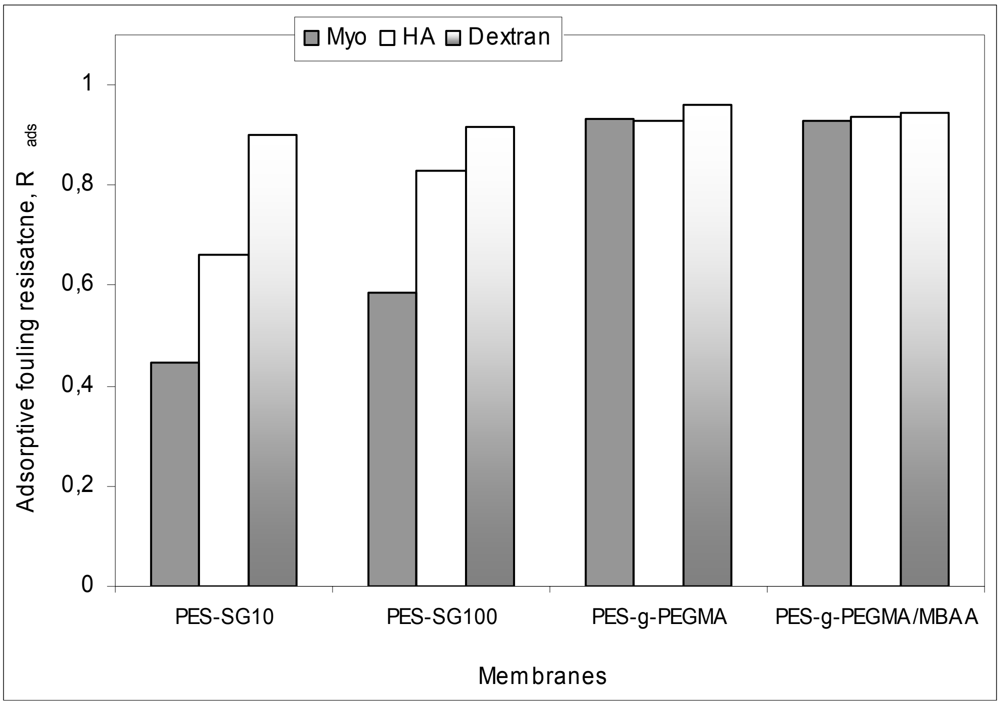

Figure 1 Adsorptive fouling resistance, R_{ads}, of unmodified and composite membranes for different test solutions (myoglobin (Myo), humic acid (HA) and dextran (T-10)). PES-SG10 and PES-SG100 are unmodified membranes with NMWCO of 10 and 100 kg/mol, respectively.

Ultrafiltration of model of drinking water and wastewater foulants

In this study, the membrane-solute-solute interactions are evaluated by ultrafiltration of the potential foulant solutions. The results are presented in term of either permeate flux/initial water flux ratio or permeate flux over the filtration time.

Interestingly, during evaluation using myoglobin both composite membranes had a much higher flux ratio than both unmodified membranes indicating that they are much less prone to fouling (Fig. 2). The unmodified membranes had a permeate flux of only ~20% (for NMWCO of 100 kg/mol) and ~30% (for NMWCO of 10 kg/mol) relative to the initial water flux, whereas both composite membranes had more than 80%. As already evoked above, this much lower fouling of composite membranes was most probably due to the much more hydrophilic character. It should be noted that the charge-based interactions should not be involved (the pH of the protein solution was at the isoelectric point). A common phenomenon during fouling study was also observed in this work. Even though membranes with larger pore size leading to higher flux had smaller flux loss in the beginning of operation, after operating of more than 10 min a more severe fouling was observed (indicated by higher flux loss relative to the water flux) than for membranes with smaller pore size (cf. PES-SG10 vs. PES-SG100). The unmodified PES membrane with NMWCO of 10 kg/mol had the highest protein rejection (~70%) whereas the unmodified PES membrane with NMWCO of 100 kg/mol had the lowest rejection (~16%). The composite membranes had slightly lower protein rejection than the 10 kg/mol PES membrane, i.e. ~58% and ~61% for PES-g-PEGMA and PES-g-

PEGMA/MBAA, respectively.

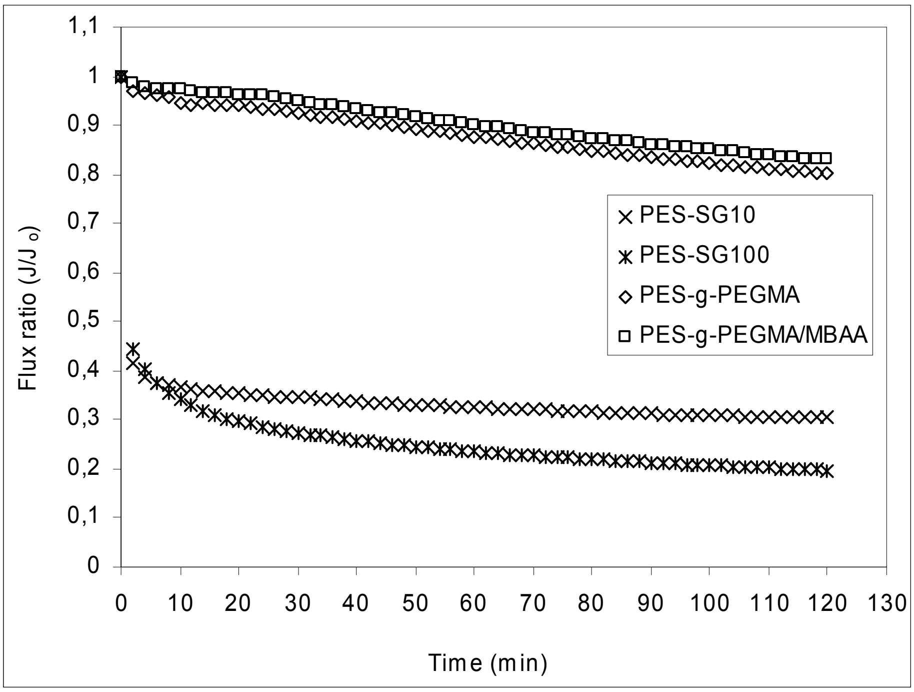

Figure 2 Flux profile as a function of time for unmodified and composite membranes during ultrafiltration of myoglobin solution (1 g/L, pH 7) at transmembrane pressure of 100 kPa. J_o is the initial water flux (L/m^{2}h). PES-SG10 and PES-SG100 are unmodified membranes with NMWCO of 10 and 100 kg/mol, respectively.

Second evaluation was done by using HA solution as the feed. Results in Fig. 3 show that both composite membranes had higher permeate flux than both unmodified membranes. The PES-g-PEGMA and PES-g-PEGMA/MBAA showed permeate fluxes approximately 80% and 72% of their initial water flux, respectively, whereas the unmodified PES membranes showed permeate flux only approximately 51% for NMWCO of 10 kg/mol and 39% for NMWCO of 100 kg/mol. Similar apparent rejections were observed, i.e. ~ 95%, ~86%, ~92% and ~92% for PES-SG10, PES-SG100, PES-g-PEGMA and PES-g-PEGMA/MBAA, respectively. It was also observed that the water fluxes after external cleaning were significantly higher for composite membranes than for unmodified membranes. External cleaning by using pure water (with shaking) could recover the initial water fluxes to approximately 90% and 86% for PES-g-PEGMA and PES-g-PEGMA/MBAA, respectively. By contrast, the same cleaning procedure for unmodified membranes yielded ~65% and ~47% recovery for PES-SG10 and PES-SG100, respectively. Then, similar to the experiments using protein, even though UF was done at similar initial water flux (~92 L/m^{2}h) in order to minimized effects of hydrodynamic conditions, the membrane with larger pore sizes showed more severe fouling (cf. PES-SG10 vs. PES-SG100). This observation agrees well with previously reported studies (Cho et al., 1999; Yuan and Zydney, 2000).

Finally, the composite membranes were evaluated by the polysaccharide foulant. The use of dextran as model for polysaccharides is supported by our previous study, which showed that dextran could significantly foul the PES UF membranes (Susanto and Ulbricht, 2005). A parallel study by Kweon and Lawler (2005) also supports this argument. As presented in Fig. 4, both composite membranes show higher relative flux ratio (permeate flux/initial water flux) than the unmodified membrane with NMWCO of 10 kg/mol. The flux ratio was increased by 10% by modification. It is important to note that both composite membranes had similar rejection of dextran T-10 compared with unmodified PES membrane with NMWCO of 10 kg/mol (all data between 74 and 82 %). Further, the relative water fluxes after external cleaning with water were higher for both composite membranes than for the unmodified membrane (i.e., 83%, 96% and 94% for PES-SG10, PES-g-PEGMA and PES-g-PEGMA, respectively).

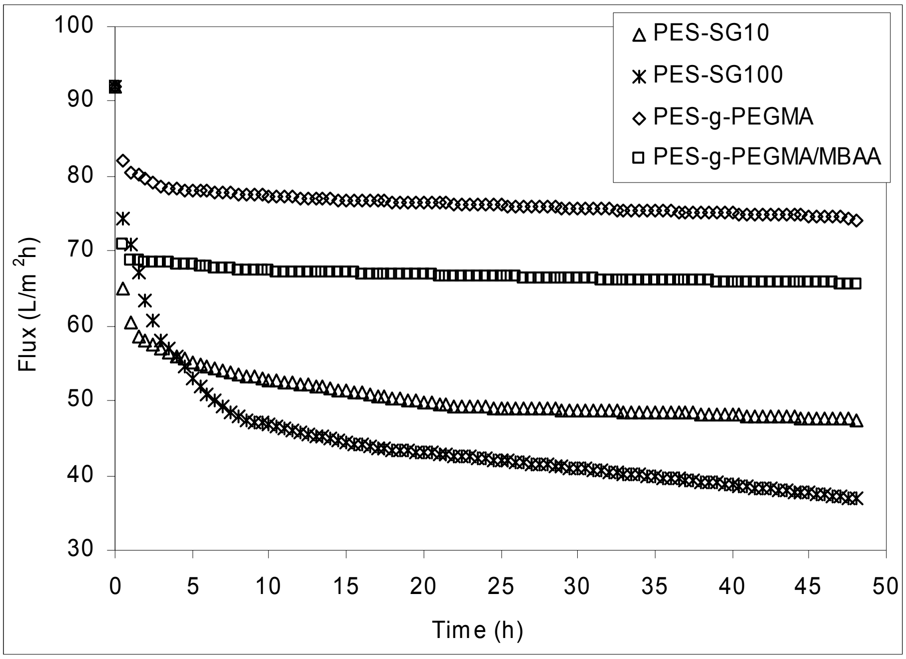

Figure 3 Flux profile as a function of time during ultrafiltration of humic acid (50 mg/L, pH 7.2, 1 mM Ca^{++}, conductivity 1100 µS/cm) at similar initial water flux (~92 L/m^2h). PES-SG10 and PES-SG100 are unmodified membranes with NMWCO of 10 and 100 kg/mol, respectively.

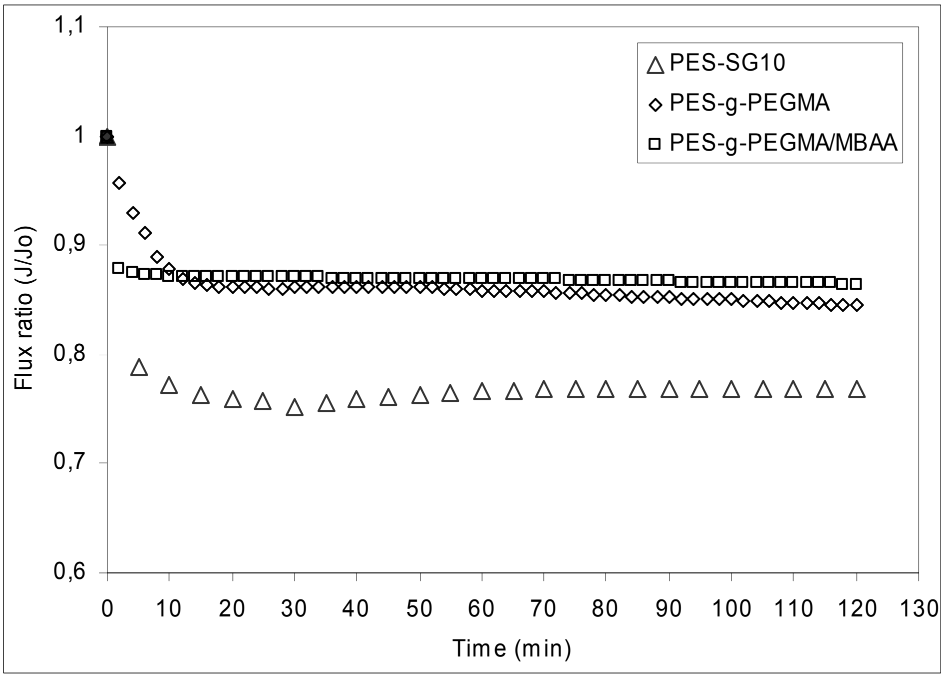

Figure 4 Flux profile as a function of time for unmodified (PES-SG10) and composite membranes during ultrafiltration of dextran T-10 (1 g/L in water) at transmembrane pressure of 100 kPa. J_o is the initial water flux (L/m^2h).

Overall, in all evaluations with different types of potential foulant solutions, both composite membranes showed significantly higher fouling resistance and water flux recovery than both unmodified membranes. This higher fouling-resistance could be attributed to the hydrogel character of grafted polymer layer, which increased the hydrophilicty of the membrane surface. Further, the differences between composite membranes without and with chemical cross-linking in the hydrogel layer (slightly higher rejection for the cross-linked at same flux –except for humic acid – and same restistance to adsorptive fouling) suggest that the internal structure of the grafted thin polymer layer could be used to "fine-tune" the membrane performance. However, a contribution of membrane pore structure to membrane-solute interactions was also clearly observed as evidenced by the fact that no composite membrane, which is absolutely inert during adsorption experiments and by the difference in fouling behaviour for membranes with different pore structure (cf. PES-SG10 vs. PES-SG100). Finally, their combined high fouling resistance and high rejection (similar to unmodified PES membrane with 10 kg/mol) suggests that the obtained modified membranes are very promising as a new generation of thin-layer composite low fouling UF membranes for drinking water and wastewater treatment applications.

CONCLUSIONS

The fouling resistance of novel thin-layer polymer hydrogel composite PES-based UF membranes, synthesized by photo-grafting of PEGMA onto commercial PES UF membranes, have been investigated for water and wastewater treatment applications. The results suggest that the resulting composite membranes had much higher adsorptive as well as ultrafiltration fouling resistance than unmodified membranes, when examined using protein, NOM and polysaccharide solutions. Much higher recovery of water flux after cleaning was other interesting result. Considering their higher fouling resistance and water flux recovery and their similar solute rejection compared to commercial unmodified PES membranes, these composite membranes should be considered as low fouling UF membrane with great potential for water and wastewater treatment applications.

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REFERENCES

Asatekin A., Menniti A., Kang S., Elimelech M, Morgenroth E. and Mayes A.M. (2006). Antifouling nanofiltration membrane for membrane bioreactors from self-assembling graft copolymers, Journal of Membrane Science, 285, 81-89.

Atkinson S. (2006). Research studies predict strong growth for MBR markets. Membrane Technology, 2006(2), 8-10.

Cho J., Amy G. and Pellegrino J. (1999). Membrane filtration of natural organic matter: initial comparisaon of rejection and flux decline characteristics with ultrafiltration and nanofiltration membranes. Water Research, 33, 2517-2526.

Glucina K., Alvarez A. and Laîné J.M. (2000). Assessment of an integrated membrane system for surface water treatment. Desalination, 132, 73-82.

Hilal N., A1-Zoubi H., Darwish N.A., Mohammad A.W. and Arabi A.M. (2004). A comprehensive review of nanofiltration membranes: Treatment, pretreatment, modelling, and atomic force microscopy. Desalination, 170, 281-308.

Jarusutthirak C. Amy G. and Croué J.-P. Fouling characteristics of wastewater effluent organic matter (EfOM) isolates on NF and UF membranes. Desalination, 145, 247-255.

Jin B., Wilén B.-M. and Lant P. (2004). Impacts of morphological, physical and chemical properties of sludge flocs on dewaterability of activated sludge. Chemical Engineering Journal, 98, 115-126.

Kweon J.H. and Lawler D.F. (2005). Investigation of membrane fouling in ultrafiltration using model organic compounds, Water Science and Technology, 51(6-7), 101-106.

Maartens A., Swart P. and Jacobs E.P. (1999). Removal of natural organic matter by ultrafiltration: characterisation, fouling and cleaning. Water Science and Technology, 40(9), 113-120.

Rosenberger S., Evenblij H., te Poele S., Wintgens T. and Laabs C. (2005). The importance of liquid phase analyses to understand fouling in membrane assisted activated sludge processes—six case studies of different European research groups. Journal of Membrane Science, 263, 113-126.

Susanto H and Ulbricht M. (2005). Influence of ultrafiltration membrane characteristics on adsorptive fouling with dextrans, Journal of Membrane Science, 266, 132-142.

Susanto H. and Ulbricht M. (2006). Performance of surface modified polyethersulfone membranes for ultrafiltration of aquatic humic substances. Desalination, 199, 384-386.

Susanto H. and Ulbricht M. (2007). Photo-grafted thin polymer hydrogel layers on PES ultrafiltration membranes: Characterization, stability and influence on separation performance. Langmuir (submitted).

Ulbricht M. (2006). Advanced functional polymer membranes. Polymer, 47, 2217-2262.

Wei X., Wang R., Li Z. and Fane A.G. (2006). Development of a novel electrophoresis-UV grafting technique to modify PES UF membranes used for NOM removal. Journal of Membrane Science, 273, 47-57.

Ye Y., Le Clech P., Chen V. and Fane A.G. (2005). Evolution of fouling during crossflow filtration of model EPS solutions. Journal of Membrane Science, 264, 190-199.

Yuan W. and Zydney A.L. (2000). Humic acid fouling during ultrafiltration, Environmental Science and Technology, 34, 5043-5050.

Impact of solid retention time on EPS and fouling tendency in membrane bioreactors

D. Al-Halbouni *, J. Hollender **, D. Tacke ***, S. Lyko ****, A. Janot ***** and W. Dott *

* Institute of Hygiene and Environmental Health, RWTH Aachen, Pauwelsstr. 30, D-52074 Aachen, Germany
(Email: djamila.al-halbouni@rwth-aachen.de; wdott@ukaachen.de)
** Eawag, Swiss Federal Institute of Aquatic Science and Technology, Überlandstr. 133, CH 8600 Dübendorf, Switzerland
(Email: juliane.hollender@eawag.ch)
*** Institute of Environmental Engineering, RWTH Aachen, Mies-van-der-Rohe-Str. 1, D-52074 Aachen, Germany
(Email: tacke@isa.rwth-aachen.de)
**** Institute of Chemical Engineering, RWTH Aachen, Turmstr. 46, D-52056 Aachen, Germany
(Email: lyko@ivt.rwth-aachen.de)
***** Erftverband, Paffendorfer Weg 42, D-50126 Bergheim, Germany
(Email: andreas.janot@erftverband.de)

Abstract Solid retention time (SRT) is mostly emphasized as a very important operating parameter impacting on fouling propensity in membrane bioreactors (MBRs). In this study the influence of SRT on fouling tendencies in two pilot MBRs was investigated. The operation parameters were identical for both plants except for the SRT which was 15 and 30 d, respectively. Slightly lower amounts of bound and soluble extracellular polymeric substances (EPS) were found in the 30 d activated sludge and supernatant compared to the SRT of 15 d. Membrane autopsies were carried out after 3 months of MBR operation with obvious decline of permeability and rise of transmembrane pressure. Analysis of EPS from those membranes showed a significantly higher amount of deposited EPS and metals on membranes and into the pores from the 15 d SRT membrane compared to 30 d SRT. These results support the conclusion that a lower SRT leads to higher fouling potential in MBRs. Bound EPS of activated sludge together with associated metals are here the main cause of fouling.

Keywords EPS; MBR; membrane fouling; SRT

INTRODUCTION

One of the most challenging issues facing further MBR development remains membrane fouling due to deposition of different materials onto or into the membrane. Some of the proposed foulants are extracellular polymeric substances (EPS), soluble microbial products (SMP), feed components, biomass itself, metal complexes and colloidal matter. Operating conditions can influence the fouling intensity and should therefore be considered together with feed and biomass characteristics as well as membrane properties. Solid retention time (SRT) is one of the very important operating parameters impacting on fouling propensity in MBRs (Le-Clech *et al.*, 2006).

However, contradictory findings have been described with regard to high versus low SRT and its effect on fouling in immersed MBRs. On the one hand, increased levels of EPS production are regarded as the reason for increased fouling rates at very low SRT. Lower fouling at extended SRT is explained by lower amounts of bound EPS rather than SMP. Such relations have been supported by Ng *et al.* (2006) who found an increased membrane fouling rate with increasing SMP and EPS concentrations at decreasing SRT. Others also reported a higher membrane fouling rate at the SRT of 10 d compared to 30 d although this fact was only related to SMP concentration but not to EPS extracted from sludge flocs (Jinsong *et al.*, 2006).

On the other hand, it has been observed that SRT had no effect on flux decline (Jarusutthirak and

Amy, 2006) or the overall fouling resistance even increased as SRT prolonged (Lee *et al.*, 2003). One reason for the higher fouling propensity at extended SRT could be the increase in mixed liquor suspended solid (MLSS) concentration and/or accumulation of non-biodegradable materials (Le-Clech *et al.*, 2006).

Although the majority of studies on the impact of SRT support the observation of reduced fouling at extended SRT it is still not clearly identified whether this fact relates to SMP, MLSS or bound EPS in the sludge or even on the membrane surface. It also depends on the particular definition of SMP and soluble or bound EPS which is highly dependent on the applied method. The methods differ at least slightly from one study to another.

In the present study a setup of pilot MBRs was established for the assessment of SRT impact on membrane fouling. The aim was to operate the MBRs for the filtration of activated sludge under completely identical conditions except for the SRT which was 15 and 30 d, respectively. These SRT were chosen as they represent realistic values that are relevant for full-scale MBR design. The analytical methods used in this study are based on a differentiation between the soluble fraction of the sludge supernatant (including SMP) and the bound EPS fraction of sludge flocs and they have already been evaluated in a long-term monitoring of a full-scale MBR (Lyko *et al.*, 2007).

METHODS

Description of the pilot plants

Two pilot plants with a bioreactor volume of 260 L each were operated as nitrification zones. The inflow to the secondary treatment of the municipal WWTP Soers (Aachen, Germany) was used as feed sludge for the pilot MBRs. After an initial phase of approximately 4 months the continuous operation mode was established. The plants contained probes for online-measurements of temperature, pressure, level and flux. The pH-value and oxygen concentration were determined manually. For maintaining the two different solid retention times of 15 d and 30 d, excess sludge was removed daily. Immersed hollow fibre modules (Puron, Germany) were used at a constant flux of 15 L m^{-2} h^{-1}. Each of the two modules consisted of three polyethersulfone (PES) fibre bundles (nominal pore size 0.05 µm) which were identically constructed and overflown in the bioreactors. The dry matter (DM) was kept constant at 12 g L^{-1}. The described two pilot plants were operated for a period of 7 months in 2005 and 3.5 months in 2006, respectively. The results presented here are from the latest study (2006); however the earlier examination (2005) revealed similar tendencies.

Sampling

Grab activated sludge samples as well as permeate were taken from the pilot MBRs and cooled during transport, storage and preparation. During a period of 3.5 months in 2006, seven separate sludge and permeate samples were taken from each MBR. Membrane autopsies were conducted once in October 2006. Three single fibres were cut from each of the two membrane modules and sealed in a plastic bag to be transported on ice to the laboratory.

EPS extraction

A method using Dowex (Sigma-Aldrich 91973) as a cationic exchange resin was used to extract EPS from activated sludge (Froelund *et al.*, 1996). Briefly, activated sludge samples were centrifuged at 30.000 x g to separate the supernatant including soluble substances from the biomass with bound EPS. Extraction of bound EPS was performed with 75 g Dowex per g dry matter for two hours stirring at 900 rpm in a beaker at 4°C. The same principle was adapted to extract EPS from membranes. For these experiments, membrane surface was first rinsed with distilled water, fibres were then cut into pieces and extracted as described above for the sludge. The ratio of Dowex to membrane surface area was 15 g to 60 – 80 cm^2. In a second step, membrane pieces were eluted in acidic or basic solution for 24 hours at ambient temperature with vigorous shaking in order to leach the remaining, more tightly bound substances including metals from the membrane pores. The ratio of cleaning solution to membrane material was approximately 10 – 15 ml for a membrane length of 140 cm (corresponds to a surface of 75 cm^2).

Analytical measurements

Dissolved organic carbon (DOC) was determined after acidification and degassing with N_2. Samples were analysed on a Dimatoc N 100 after filtration through 0.45 µm syringe filters.
The loss on ignition was determined for activated sludge according to DIN standard norm DIN EN 12879 S3.
Metal ions (Mg^{2+}, Mn^{2+}, Ca^{2+}, Fe^{3+}, Al^{3+}) were quantified by ICP-MS using a Perkin Elmer Elan DRC II. Liquid samples were acidified with 65% HNO_3 (suprapur) and 100 µL of a 1 µg/mL Rhodium stock solution were added as an internal standard to each sample. A standard mix of 30 elements (Merck 110580) was used for calibration.

RESULTS AND DISCUSSION

Membrane permeability and transmembrane pressure trends

Online measurements of flow (F) and transmembrane pressure (TMP) within the two pilot MBRs showed a significant decline of permeability (P) and an increase of TMP for both plants, thus indicating membrane fouling and a loss in membrane performance (Fig. 1 and 2).
The visible fluctuations in membrane permeability could have been caused by slight variations in the dry matter content (data not shown). On the 4th Oct 2006 a problem with the plant operation occurred and the inflow failed so that an additional drop in P was noticeable.

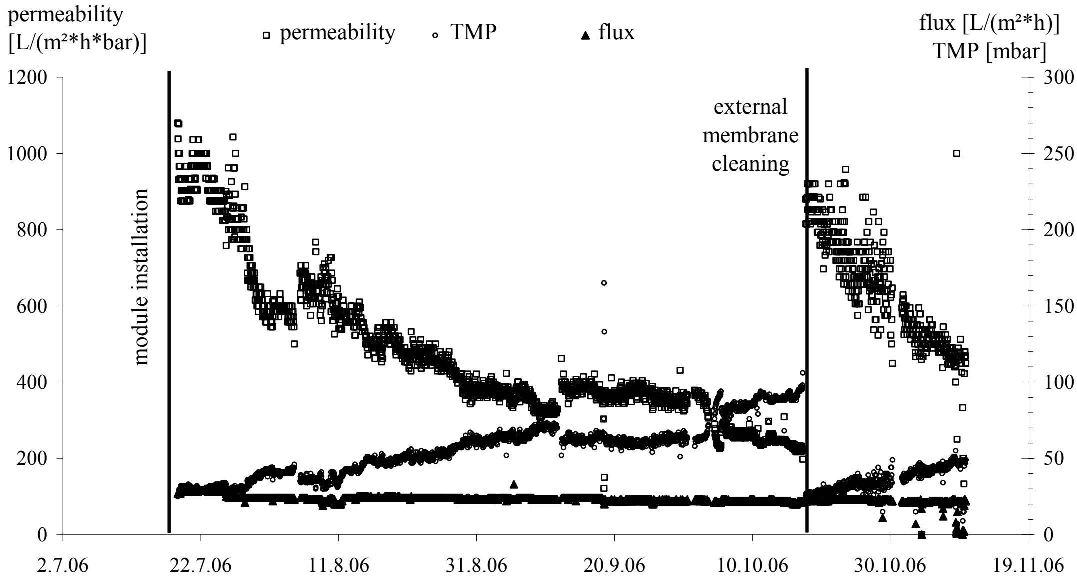

Figure 1 Recorded data from online probes within the pilot MBR of 15 d SRT.

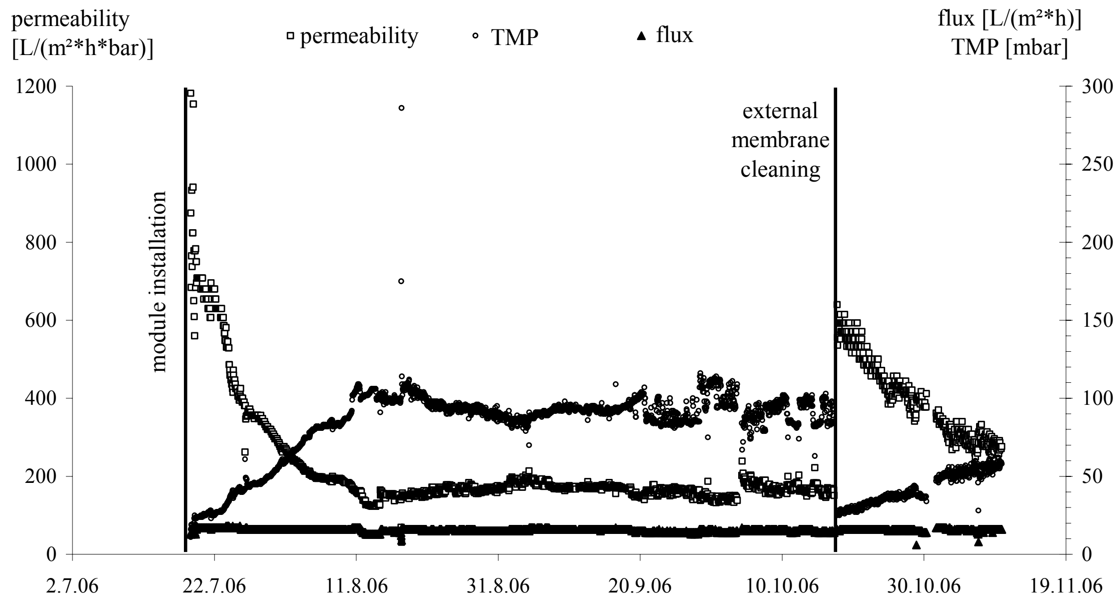

Figure 2 Recorded data from online probes within the pilot MBR of 30 d SRT.

Bound and soluble EPS in activated sludge

The amount of organic substances in the activated sludge samples from two MBRs with 15 and 30 d SRT was determined by loss on ignition. It was shown that the lower SRT leads to slightly higher amounts of organic substances (80% of dry matter) in the sludge compared to the higher SRT (77% of dry matter) (Figure 3A).

In order to examine the reason for higher organic contents in the sludge with a lower SRT, bound EPS were extracted and compared to the amount of soluble EPS. A higher amount of bound EPS was found in the activated sludge with a lower SRT (Figure 3B). The difference between the amounts of soluble EPS in the supernatants of activated sludge from the two MBRs was not that pronounced, partly because of the broader distribution of the observed DOC-concentrations in the supernatant of the MBR with 15 d SRT compared to 30 d. The mean value of DOC in the supernatant is however slightly higher in the 15 d SRT plant and is also verified by the results of the corresponding membrane permeate samples (Figure 3C).

The observed differences in DOC-concentrations of EPS in the activated sludge are not drastic but still show a tendency. One reason for that might be the chosen SRT which are close to each other in order to represent realistic values. In similar studies with very low and very high SRT (e.g. 3 d up to 60 d) the differences were more distinctive (Lee *et al.*, 2003; Liao *et al.*, 2006; Ng *et al.*, 2006).

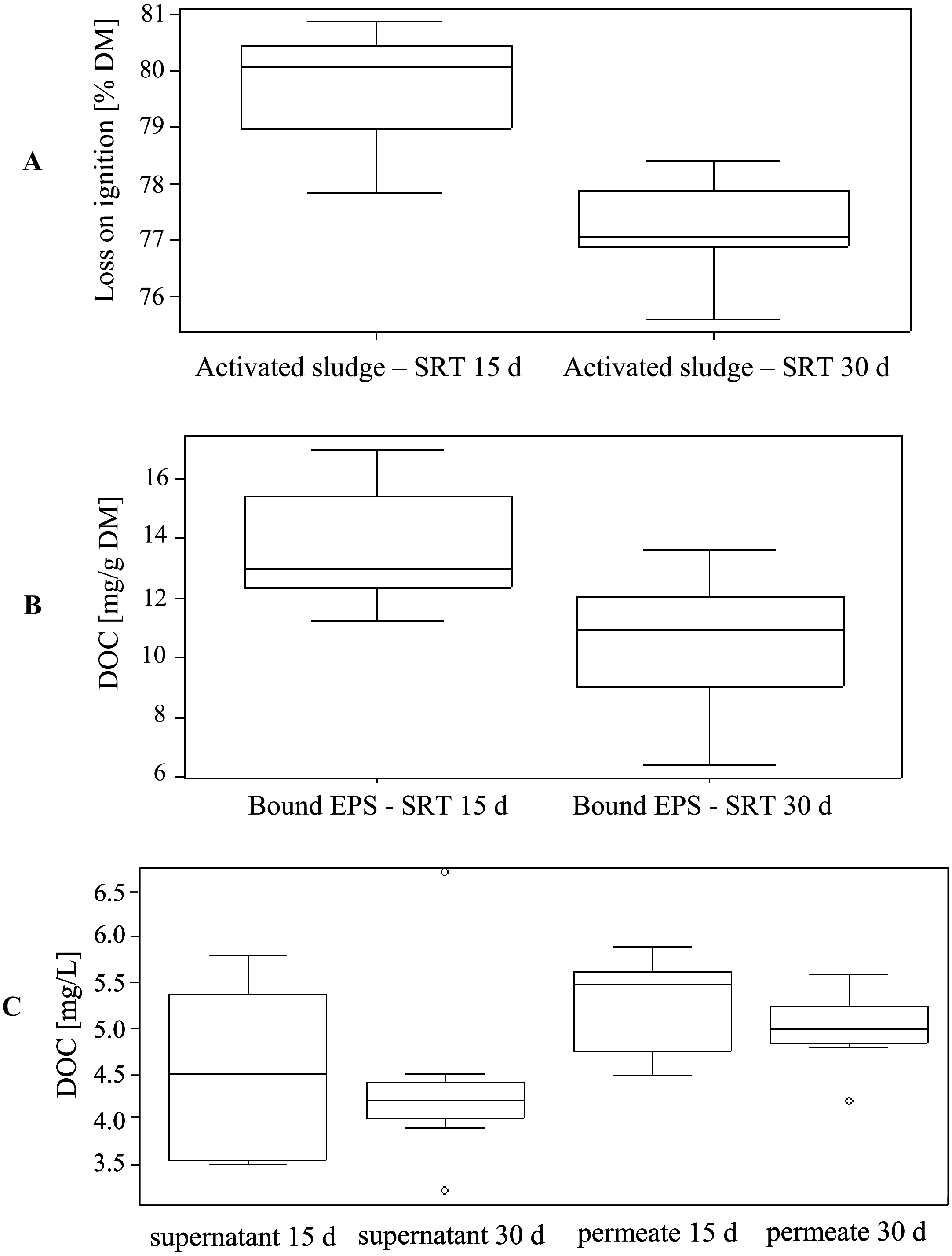

Figure 3 (A) Loss on ignition of activated sludge samples from two MBRs of 15 and 30 d SRT, respectively. (B) Bound EPS (expressed as DOC) that were extracted from activated sludge of two MBRs with 15 and 30 d SRT, respectively. (C) Soluble EPS (expressed as DOC) in activated sludge supernatant as well as in the corresponding membrane permeate of two MBRs with 15 and 30 d SRT, respectively. The shown data for A, B, C are based on 7 separate samples from each MBR.

Analysis of fouled membranes

As indicated in Fig. 1 and 2 membrane modules were removed for cleaning after severe fouling had occured. The effect of cleaning the membranes with sodium hypochlorite and citric acid is clearly shown by improved pure water permeability as well as lower TMP (Table 1).

Table 1. Effect of cleaning the membranes ex-situ by removing the whole modules from the pilot MBRs and soaking in chemicals. P and TMP were measured ex-situ by filtration of water.

Filtration of pure water.	P [L/(m²hbar)]		TMP [bar]	
	SRT = 15 d	SRT = 30 d	SRT = 15 d	SRT = 30 d
before cleaning	123	152	0.099	0.102
after cleaning[a] with NaOCl	387	287	0.033	0.051
after additional cleaning[a] with citric acid	359	369	0.044	0.044

[a] Cleaning cycle: 2 h soaking in chemical solution with 2 min filtration each 30 min.

In order to assess the amount of substances bound to the fouled membranes, autopsies were carried out before the ex-situ cleaning procedure for the whole modules began. Membranes were rinsed with distilled water first so that loosely attached sludge mass would be removed. The quantified EPS and metals therefore represent the more tightly - partially within the pores - bound substances. By using the cationic exchange resin in the first membrane extration step, a higher amount of EPS was removed from membranes of the MBR with 15 d SRT compared to 30 d (Table 2**Table**). The subsequent cleaning step involved either soaking in citric acid or in sodium hypochlorite and was effective in eluting metals from the membranes.

Table 2. Results of EPS extraction and cleaning of autopsied membranes that were removed from the modules of the pilot MBRs right before the ex-situ cleaning described in Table .

	1st step: extraction with Dowex	2nd step: elution in acidic or basic solution			
SRT [d]	EPS as DOC [mg/m^2]	Fe [mg/m^2] after elution in citric acid[b]	Fe [mg/m^2] after elution in sodium hypochlorite	Ca [mg/m^2] after elution in sodium hypochlorite	Al [mg/m^2] after elution in sodium hypochlorite
15	183.5	34.7	8.8	0.4	4.6
30	103.7	20.8	7.0	1.0	5.9

[b] No other metals were detected in the acidic wash eluate.

There is obviously a correlation between higher amounts of bound EPS in the sludge with 15 d SRT and higher amounts of EPS deposited onto and into the membranes of this particular MBR in comparison to the 30 d SRT. In addition, iron was also eluted by citric acid from membranes of the 15 d SRT plant in a higher concentration than from the comparative membranes.

From the analysis of metal ions it was shown that Ca and Mg are present in the sludge supernatant and permeate of both MBRs in the same concentrations independently of the SRT (mean values for

Ca^{2+} and Mg^{2+} in supernatant as well as permeate: 47 and 10 mg/L, respectively). These metals were not detected (or only in negligible amounts) in the eluates of fouled membranes thus confirming the minor role of this soluble fraction of sludge supernatant. If Ca^{2+} and Mg^{2+} play a role in blocking the membranes then it would more likely be within the bound EPS fraction of the sludge. However, this part of the foulants would be removed from the membranes by the first extraction step with Dowex (Table 2) which targets especially the Ca- and Mg-bound polymers. Assuming the removal of these EPS that were complexed with Ca^{2+} and Mg^{2+} in the first step, the second chemical elution step (Table 2) reveals those substances that were either not removable by the Dowex extraction or that were too tightly bound to the membrane material and pores. In the present study, iron seems to be part of such non-Dowex-extractable fractions with the ability to clogg the membranes. The existence of different fractions of EPS bound to different metal cations has recently been documented by Park and Novak (in press).

Within the bound fraction of the sludge floc iron and aluminium were the predominant metals, with higher concentrations at 30 d SRT (mean values for Fe^{3+} and Al^{3+}: 14.4 and 8.7 mg/g DM) compared to 15 d SRT (mean values for Fe^{3+} and Al^{3+}: 11.6 and 7.1 mg/g DM). Despite the higher concentration of iron in the sludge of 30 d SRT the membranes of this MBR showed a significantly lower amount of deposited iron (Table 2) compared to 15 d SRT. This shows how important the whole sludge floc, its structure and size as well as bound EPS content are for the amounts of deposited foulants on and in the membranes. The development of filamentous bacteria in contrast to non-flocculating organisms in dependence on SRT is also relevant (Massé *et al*., 2006). All these observations are additionally supported by the observed worse settleability and dewaterability of activated sludge from the MBR with 15 d SRT (by measurements of sludge volume index and capillary suction time, data not shown) and its adhesive properties with regard to the membrane surface.

CONCLUSIONS

From the comparison of two pilot MBRs with a SRT of 15 and 30 days, respectively, it was found that the amounts of soluble as well as bound EPS in the activated sludge decreased slightly as the SRT increased. The analysis of fouled membranes from both MBRs revealed a significantly higher amount of EPS and metals bound to the membranes of the 15 d SRT plant. It was concluded that effects on membrane performance by varying SRT in a range which is relevant for full-scale MBRs are not enormous; however fouling is less severe when SRT is increased.

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REFERENCES

Froelund B., Palmgren R., Keiding K. and Nielsen P. H. (1996). Extraction of extracellular polymers from activated sludge using a cation exchange resin Water Research 30(8), 1749-1758.

Jarusutthirak C. and Amy G. (2006). Role of Soluble Microbial Products (SMP) in Membrane Fouling and Flux Decline. Environmental Science and Technology 40(3), 969-974.

Jinsong Z., Chuan C. H., Jiti Z. and Fane A. G. (2006). Effect of Sludge Retention Time on Membrane Bio-Fouling Intensity in a Submerged Membrane Bioreactor. Separation Science and Technology 41(7), 1313 - 1329.

Le-Clech P., Chen V. and Fane T. A. G. (2006). Fouling in membrane bioreactors used in wastewater treatment. Journal of Membrane Science 284(1-2), 17-53.

Lee W., Kang S. and Shin H. (2003). Sludge characteristics and their contribution to microfiltration in submerged membrane bioreactors. Journal of Membrane Science 216(1-2), 217-227.

Liao, B. Q., Droppo, I. G., Leppard, G. G., Liss, S. N. (2006). Effect of solids retention time on structure and characteristics of sludge flocs in sequencing batch reactors. Water Research 40(13), 2583-2591.

Lyko S., Al-Halbouni D., Wintgens T., Janot A., Hollender J., Dott W. and Melin T. (2007). Polymeric compounds in activated sludge supernatant – characterisation and retention mechanisms at a pilot and full-scale municipal membrane bioreactor. Submitted.

Massé A., Sperandio M. and Cabassud C. (2006). Comparison of sludge characteristics and performance of a submerged membrane bioreactor and an activated sludge process at high solids retention time. Water Research 40(12). 2405-2415.

Ng H. Y., Tan T. W. and Ong S. L. (2006). Membrane Fouling of Submerged Membrane Bioreactors: Impact of Mean Cell Residence Time and the Contributing Factors. Environmental Science and Technology 40(8), 2706-2713.

Park C. and Novak J. T. (in press). Characterization of activated sludge exocellular polymers using several cation-associated extraction methods. Water Research (2007), doi:10.1016/j.watres.2007.01.031

Investigating Hydrodynamics in Submerged Hollow-Fibre Membrane Filtration Units in Municipal Wastewater Treatment using Computational Fluid Dynamics (CFD)

S. Buetehorn*, C.N. Koh*, T. Wintgens*, T. Melin*, D. Volmering** and K. Vossenkaul**

* Department of Chemical Engineering, RWTH Aachen University, Turmstr. 46, 52056 Aachen, Germany
(E-mail: buetehorn@ivt.rwth-aachen.de)
** Koch Membrane Systems GmbH, Kackertstr. 10, 52072 Aachen, Germany
(E-mail: k.vossenkaul@kochmembrane.com)

Abstract In municipal and industrial wastewater treatment, submerged membrane filtration units are used to reject suspended solids from water. Within this ultrafiltration process, air bubbles are injected into the stationary suspension in order to remove the cake layer and by that to reduce permeate flux decline. According to previous publications, the efficiency of air bubbling is depending on both bubble and fibre characteristics. Moreover, optimised process parameters like aeration rate and air bubbling frequency as well as optimised membrane module specifications like fibre diameter and gas sparging device have been identified. Within this study, the impact of fibre arrangement on the effectiveness of air bubbling is investigated by means of Computational Fluid Dynamics (CFD) simulations. For this purpose, X-ray computer tomography (CT) is used to map the instantaneous fibre displacement within a single hollow-fibre bundle. On the basis of these CT images, two different geometry models have been developed and are presented in this paper.

Keywords Air bubbling, sludge entrainment, fibre movement, cake layer removal, fouling control

INTRODUCTION

Gas bubbles injected into a stationary liquid are significantly affecting transport phenomena within the multiphase system. For this reason, gas sparging is a widely spread technique to enhance both heat and mass transfer characteristics [Farmer, 1885; Kubie, 1975]. Amongst other applications, air bubbling is used in a variety of submerged membrane processes to induce flow and to generate shear at the membrane surface in order to control fouling [Cui *et al.*, 2003]. In 1989, Yamamoto *et al.* were presenting the first study focussing on hollow-fibre membranes submerged into the biological stage of a wastewater treatment plant (submerged membrane bioreactor, sMBR). Since this pioneering work, efforts have been made to further investigate the impact of air bubbling on the overall filtration performance of the system.

According to previous publications, the bubble size and in turn the shape of the bubble are strongly influenced by the injection method, the sparger type, the gas flow rate and the size of the flow channel within this buoyancy-driven process [Cui *et al.*, 2003]. It was found that an optimum bubble size exists in the range of 2-5 mm (ellipsoidal bubbles; recognised by Zenon and Ebara). In addition, the effect of gas flow rate on the filtration performance has been intensively investigated, e.g. by measuring the deposit resistance with and without bubbling [Chang and Fane, 2000] or by the critical flux concept presented by Field *et al.* in 1995. Moreover, Guibert *et al.* [2002] suggested an intermittent aeration of membrane bundles, which leads to lower operational costs and an increased cleaning efficacy due to a superposed horizontal liquid flow (partly aerated membrane tank with differences in local mass density). An important parameter of this so-called air cycling process is the cycle frequency, which is typically set to 10 s on / 10 s off. On a larger scale, this coarse bubble aeration is inducing a liquid recirculation within the membrane tank, which has been studied by a number of research groups [Yusuf and Murray, 1993; Liu *et al.*, 2000; Shim *et al.*, 2002].

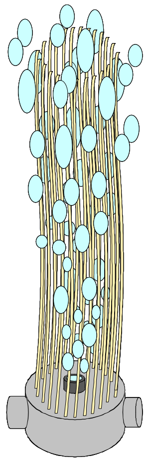
Besides surface shear transients and fluctuating liquid flow, rising air bubbles cause lateral fibre movement [Cui *et al.*, 2003]. According to Fane *et al.* [2006], mechanical fibre movement is able to significantly reduce the fouling rate. Therefore, the cleaning efficiency using thinner and by that more flexible fibres is higher compared to thicker and stiffer membranes under the same aeration conditions [Chang and Fane, 2001]. Nevertheless, the pressure drop at the lumen side is increasing and the mechanical stability is decreasing with a decreasing fibre diameter [Cui *et al.*, 2003]. This interrelationship leads to an optimum inside diameter of hollow-fibre membranes regarding a maximum productivity per unit energy [Fane *et al.*, 2002a]. Moreover, the fibre looseness within the support frame is a significant factor with respect to fouling prevention due to air bubbling [Chang and Fane, 2002], because looser fibres show a higher mobility which in turn leads to higher wall shear stresses due to fibre movement. Furthermore, the orientation of the fibre (horizontal/vertical) is of importance. While horizontal fibres are advantageous in single-phase membrane processes without gas sparging [Futselaar *et al.*, 1993], the air bubbling effectiveness using vertical oriented fibres is higher compared to horizontal fibres [Fane *et al.*, 2002b].

Complementary to the above-mentioned studies, the relationship between the efficacy of air bubbling and fibre arrangement is examined at RWTH Aachen University by means of Computational Fluid Dynamics (CFD) simulations. The objectives of these studies are (i) to model the geometry of the PURON® module provided by Koch Membrane Systems GmbH (KMS), (ii) to apply this model to different operating conditions and module configurations, and by that (iii) to investigate the influence of air flow rate and fibre arrangement on the cleaning efficiency of air bubbling.

MATERIALS AND METHODS

Materials

The PURON® module provided by KMS consists of hollow-fibre membranes arranged in bundles and submerged vertically into the activated sludge. While the lower ends of the fibres are fixed in a header, the upper ends are individually sealed and are free to move laterally. A schematic representation of a single hollow-fibre bundle and the respective technical specifications are presented in figure 1.



Membrane material	PES
Nominal pore size	0.04 µm
Fibre diameter	2.6 mm
Fibre length	1.2 m
Membrane area (single bundle)	2.2 m^2

Figure 1 Schematic representation of a hollow-fibre bundle and technical specifications [Judd, 2006; Buetehorn *et al.*, 2007]

An air nozzle consisting of an annular gas inlet is located in the centre of each bundle in order to prevent sludging. Flow channels within the cross-section of the bundle (i.e. gaps without fibres) facilitate bubble-induced sludge entrainment, whereas the specific fibre arrangement (fixed at one side only) ensures that fibrous substances are removed from the system in order to avoid clogging.

Modelling Approach

Within this research work, CFD simulations will be conducted to investigate the influence of fibre arrangement and aeration rate on the hydrodynamics within single hollow-fibre bundles. For this purpose, the complex flow pattern is to be simplified and a geometry model is to be implemented into the CFD code.

Sludge rheology. The three-phase system consisting of water, suspended solids, and gas bubbles is aimed to be modelled as two phases. While the contribution of solids to the flow characteristics is taken into account in terms of an artificial dynamic viscosity of a model liquid, the virtual properties of the gaseous phase (air) are represented accurately. The rheology of activated sludge sampled from municipal wastewater treatment plants has been extensively described in literature [Shankar and Kumar, 1994; Günder, 1999; Xing *et al.*, 2001; Cornel and Krause, 2003; Rosenberger, 2003; Moshage, 2004; Fane *et al.*, 2006; Melin, 2007]. On the basis of published data and additional experimental results, a suitable model liquid is to be identified in the upcoming period of investigations.

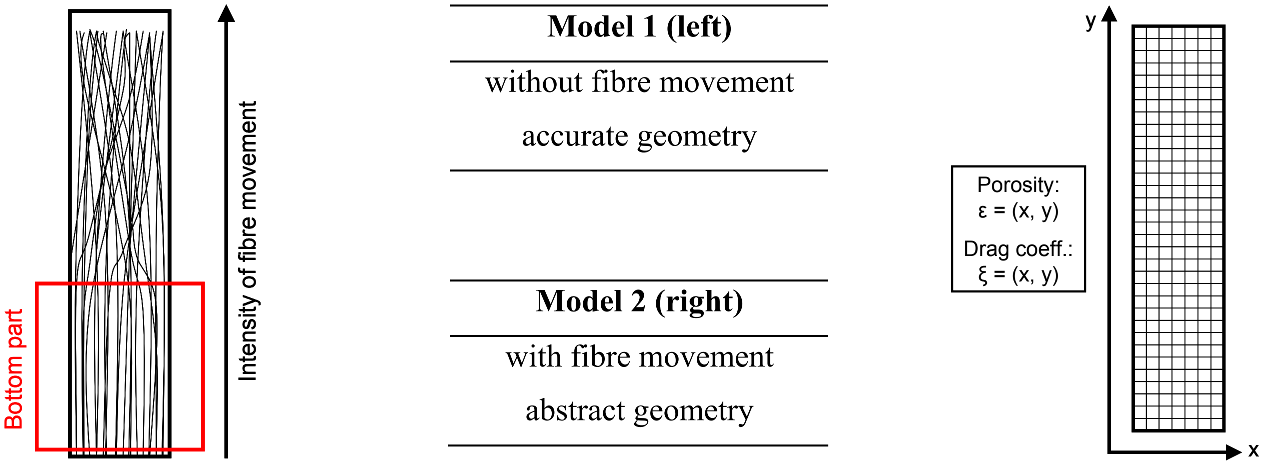


Figure 2 Modelling the geometry of hollow-fibre membrane bundles

Not moving fibres. The main focus of this study is to model the geometry of a single hollow-fibre bundle. Thus, two different modelling approaches have been identified, see figure 2. Since the lower ends of the fibres are fixed, the intensity of fibre movement in the bottom part of the bundle is lower than the intensity in the upper part. Hence, the fibre movement in the bottom part is assumed to be negligible within this first geometry model. This simplification facilitates to model the geometry accurately, i.e. to represent each single fibre out of the bundle in steady state.

Heterogeneous porous medium. In a second approach, the intensity of fibre movement is taken into account in terms of modelling the bundle as a heterogeneous porous medium. In a first stage of investigations, the geometry data attained to generate the model with not moving fibres will be converted to a stationary heterogeneous porous medium with a defined local porosity ε (x, y) and local flow resistance ξ (x, y). Subsequently, the transient character of these properties due to irregular fibre concentration within one cross-section caused by continuous fibre displacement is intended to be modelled by user defined functions (UDFs).

X-ray computer tomography (CT)

X-ray computer tomography is a non-invasive measurement technique which requires distinct difference in electron density between components to be distinguished [Chen *et al.*, 2004]. CT scanners consist of an X-ray source combined with a detector located at the opposite site of the specimen. Starting from integral values due to X-ray attenuation along the path length through the sample from many directions, local values are mathematically reconstructed within one measurement plane indicating the local composition of the sample [Natterer and Wübbeling, 2001; Bischof *et al.*, 2007]. Optionally, a three-dimensional map of the specimen can be formed via image processing on the basis of single measurement planes. For fundamentals on CT imaging, please refer to [Röntgen, 1896; Hounsfield, 1973; Mewes *et al.*, 1999; Seibert and Boone, 2005].

During the experiments, the acceleration voltage was set to 250 kV in order to improve contrast between material and ambient air (maximum acceleration voltage of the system: 420 kV). The field of view was 150 mm in diameter and was reconstructed out of an image matrix of 1024 x 1024 pixels (convolution backprojection method). The distance between two measurement planes was set to 5 mm with a collimator width ("slice thickness") of 1 mm (spatial resolution: 1 mm in height and 150 µm radial). The acquisition time per plane was 30-35 s.

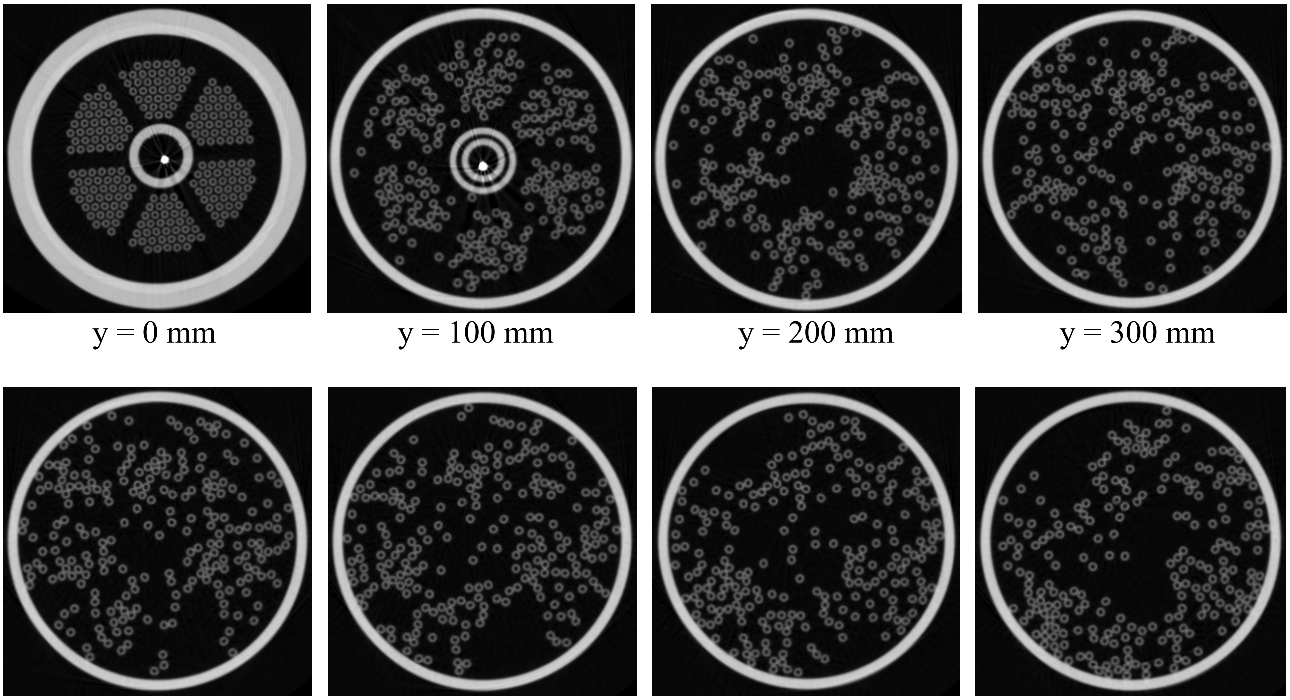
With the above-mentioned settings, a virgin PURON$^{®}$ hollow-fibre bundle of 0.75 m in length (150 planes in total) was scanned in order to map the instantaneous displacement of not moving fibres. During the measurement, the bundle was hanging in a pipe out of plastic (head first) with an inner diameter of 80 mm. The loose ends of the fibres were fixed to avoid fibre movement while the sample was rotating during the scan. Tomographical measurements were conducted at the Institute of Multiphase Processes at the Leibniz University of Hannover, Germany, with a CT device manufactured by Bio-Imaging Research, Inc. (BIR), Lincolnshire, IL.

PRELIMINARY RESULTS

Accurate description of the bundle with not moving fibres (model 1)

CT images of the cross-sections of the scanned hollow-fibre bundle are presented in figure 3 (65,536 grey scale values). The axial distance between the measurement planes is amounting to 100 mm in each case. Brightness and contrast of the images were adjusted with the software ImageJ 1.37, so that ring artefacts are masked out. These pictures are cut-outs of the measurement planes of approximately 640 x 640 pixels in size.

The first picture (y = 0 mm) shows a cross-section in the bottom part of the membrane bundle where the fibres are potted in a header. This fixing leads to a very regular fibre arrangement, i.e. groups of fibres are formed and separated by flow channels. Moreover, the gas sparger (small annulus) as well as a fixing filament (small dot) can be identified. The star-shaped artefacts in the centre of the first two images are originating from the fixing filament out of metal due to high differences in mass density. In the second picture (y = 100 mm), the fibre arrangement is less regular, although the flow channels are still observable. In addition, the gas inlet can be identified by means of an annular gap within the gas sparging device (opening at y = 20 mm). With a total length of 175 mm, the gas pipe cannot be seen at y = 200 mm and above. Nevertheless, the fibre concentration in the centre of the third picture is much lower compared to all cross-sections above. For y > 300 mm, the fibre displacement is rather stochastic. However, the fibres tend to accumulate close the wall of the pipe for y > 700 mm, presumable due to the fixing of the loose ends of the fibres in the upper part of the bundle.



y = 0 mm y = 100 mm y = 200 mm y = 300 mm

y = 400 mm y = 500 mm y = 600 mm y = 700 mm

Figure 3 CT images of the cross-sections of a single hollow-fibre bundle

Within the first modelling approach, 150 planar measurement planes have been connected to form an accurate (but steady state) three-dimensional representation of the bottom part of the hollow-fibre bundle. This geometry model will be implemented into the commercial CFD code FLUENT® in terms of an STL file describing the unstructured triangulated surface. Subsequently, an appropriate mesh will be created, e.g. a triangular mesh in combination with prism-type sub-layers close to walls if feasible. The geometry (pipe + membrane bundle) will be filled with a stationary model liquid. The annular gap of the gas sparging device is intended to be defined as the air inlet with a pre-defined gas flow rate. Since Wang *et al.* [1994] have shown that the consideration of permeate fluxes does not alter the bulk flow field (simulation of cross-flow filtration for baffled tubular channels and pulsatile flow), permeate suction is not taken into account within this study. The preliminary settings of the CFD software have been identified within initial calculations and are summarised in table 1.

Table 1. Preliminary settings of the commercial CFD code FLUENT®.

multi-phase model	Volume of Fluid (VOF) [Hirt and Nichols, 1981]
turbulence model	Reynolds Normalization Group (RNG) k-ε [Yakhot and Orszag, 1986]
model liquid	polysaccharide solution (to be identified)
mesh type	hexahedron mesh + prism-type sub-layers (if feasible)

Description of the bundle as a heterogeneous porous medium (model 2)

The accurate map of fibre displacement described in the previous section is aimed to be modelled as a heterogeneous porous medium, firstly with steady state local porosity ε (x, y) and local flow resistance ξ (x, y). Therefore, a number of cross-sections (e.g. cross-sections with an axial distance of 5 cm) will be separated into planar elements of either annular or rectangular type as schematically shown in figure 4.

Figure 4 Different concepts for the separation of cross-sections (left: annular, right: rectangular)

In a first step, the permeate channels of the membranes (i.e. the insides of the hollow-fibre annulus shown in each picture) will be supplementary filled with circles in order to distinguish between feed channel (black coloured) and membrane materials respectively permeate channels (white coloured). Subsequently, one of the grids shown in figure 4 will be defined within MATLAB® and the fractions of white coloured and black coloured areas will be automatically calculated. According to the area fraction of the flow channel, the local porosity is to be defined for each planar element of the cross-sections. Upon completion, the spatial porosity will be interpolated out of the stepwise two-dimensional porosity distributions within each measurement plane. As a last step, validation experiments will be conducted in order to correlate the average porosity within one volumetric element (expressed as the amount of fibres) with the respective local flow resistance. Thus, an appropriate experimental setup consisting of identical pipes filled with a varying number of fibres will be used. During these experiments, the pressure loss of a representative model liquid (e.g. a polysaccharide solution) flowing through the columns will be measured as a function of porosity.

CONCLUSIONS AND OUTLOOK

The design of submerged filtration units consisting of hollow-fibre membranes is a significant factor with respect to hydrodynamic conditions during operation. Complementary to various publications focussing on bubble-induced sludge entrainment and fibre movement, the overall objective of this study is to investigate the impact of fibre arrangement on the cleaning efficiency of air bubbling by means of CFD simulations.

Prior to the simulations, a literature review on the rheology of activated sludge was carried out and the results of several research groups were compared with each other. It was consistently found the dynamic viscosity of the activated sludge is decreasing as the shear rate is increasing (shear thinning characteristics) and the MLSS is decreasing. Nevertheless, these experiments have been conducted using different sludge characterisation protocols (e.g. different measurement systems, temperature, and test duration), so that a direct comparison of results is difficult. For this reason, a test series is to be performed at RWTH Aachen University in order to establish a rheological model which will be subsequently implemented into the CFD code.

Furthermore, CT images were generated in order to (i) model the geometry with not moving fibres by implementing a three-dimensional map of a single hollow-fibre bundle into the CFD code, and to (ii) define a heterogeneous porous medium out of these images. Since the realisation of the latter approach is rather complex, the transient character of the porous medium is neglected within the first stage of investigations. Nevertheless, fibre movement is aimed to be taken into account by defining a transient heterogeneous porous medium with time-dependent local porosity ε (x, y, t) and local flow resistance ξ (x, y, t). Besides optical methods for direct observation, nuclear magnetic resonance (NMR) imaging [Chen *et al.*, 2004] is taken into consideration for model validation in terms of tracking bubble-induced fibre movement.

To conclude, efforts will be made in the upcoming period of the project to further understand the relationship between fibre arrangement and bubble induced sludge entrainment in this field of membrane application. For this purpose, standard and recently developed hollow-fibre membrane modules manufactured and kindly provided by KMS will be investigated, so that the findings out this research work will be directly fed into the development of the PURON® system.

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REFERENCES

Bischof J.C., Mahr B., Choi J.H., Behling M. and Mewes D. (2007). Use of X-ray tomography to Map Crystalline and Amorphous Phases in Frozen Biomaterials. In: Annals of Biomedical Engineering, 35 (2) (2007) 292-304.

Buetehorn S., Koh C.N., Wintgens T, Melin T., Volmering D. and Vossenkaul K. (2007). Investigating the Impact of Membrane Pore Size Distribution on the Performance in Activated Sludge Filtration and the Susceptibility to Fouling. In: Proceedings: Aachener Membran Kolloquium, 11 (2007) 371-381.

Chang S. and Fane A.G. (2000). Characteristics of microfiltration of suspensions with inter-fibre two-phase flow. In: Journal of Chemical Technology and Biotechnology, 75 (2000) 533-540.

Chang S. and Fane A.G. (2001). The effect of fibre diameter on filtration and flux distribution – relevance to submerged hollow fibre modules. In: Journal of Membrane Science, 184 (2) (2001) 221-231.

Chang S. and Fane A.G. (2002). Filtration of biomass with lab-scale submerged hollow fibre membrane module: effect of operational conditions and module configuration. In: Journal of Chemical Technology and Biotechnology, 77 (2002) 1030-1038.

Chang S., Fane A.G. and Vigneswaran S. (2002a). Modelling and optimisation of submerged hollow fibre membrane modules. In: American Institute of Chemical Engineers Journal, 48 (10) (2002) 2203-2212.

Chang S., Fane A.G. and Vigneswaran S. (2002b). Experimental assessment of filtration of biomass with model axial and transverse fibres. In: Chemical Engineering Journal, 87 (2002) 121-127.

Chen V., Li H. and Fane A.G. (2004). Non-invasive observation of synthetic membrane processes – a review of methods. In: Journal of Membrane Science, 241 (2004) 23-44.

Cornel P. and Krause S. (2003). Aufbereitung von Industrie- und Prozessabwasser mit Membranverfahren und Membranbelebungsverfahren. In: Proceedings: Aachener Tagung Siedlungswasserwirtschaft und Verfahrenstechnik, 5 (2003) A14.1-A14.22.

Cui Z.F., Chang S. and Fane A.G. (2003). The use of gas bubbling to enhance membrane processes. In: Journal of Membrane Science, 221 (2003) 1-35.

Farmer M.G. (1885). Apparatus for refining copper by electricity. US Patent, 322 170 (1885).

Field R.W., Wu D., Howell J.A. and Gupta B.B. (1995). Critical flux concept for microfiltration fouling. In: Journal of Membrane Science, 100 (1995) 259-272.

Futselaar H., Zoontjes R.J.C., Reith T. and Rácz I.G. (1993). Economic comparison of transverse and longitudinal flow hollow fiber membrane modules for reverse osmosis and ultrafiltration. In: Desalination, 90 (1993) 345-361.

Günder B. (1999). Das Membranbelebungsverfahren in der kommunalen Abwasserbehandlung. PhD thesis, Forschungs- und Entwicklungsinstitut für Industrie- und Siedlungswasserwirtschaft sowie Abfallwirtschaft e.V., Stuttgart, Germany.

Guibert D., Ben Aim R., Rabie H. and Cote P. (2002). Aeration performance of immersed hollow-fibre membranes in a bentonite suspension. In: Desalination, 148 (2002) 395-400.

Hirt C.W. and Nichols B.D. (1981). Volume of Fluid (VOF) Method for the Dynamics of Free Boundaries. In: Journal of Scientific Physics, 39 (1981) 201-225.

Hounsfield G.N. (1973). Computerized transverse axial scanning (tomography): Part I. Description of system. In: British Journal of Radiology, 46 (1973) 1016-1022.

Judd S. (2006). The MBR Book - Principles and Applications of Membrane Bioreactors for Water and Wastewater Treatment. ELSEVIER, Amsterdam.

Kubie J. (1975). Bubble induced heat transfer in two-phase gas-liquid flow. In: International Journal of Heat and Mass Transfer, 18 (1975) 537-551.

Liu R., Huang X., Wang C., Chen L. and Qian Y. (2000). Study on hydraulic characteristics in a submerged membrane bioreactor process. In: Process Biochemistry, 36 (2000) 249-254.

Melin T. (2007). Strategien zur Foulingkontrolle bei Membranbelebungsanlagen in der kommunalen Abwasserbehandlung. Final Report AZ IV-9-042 198, Ministerium für Umwelt und Naturschutz, Landwirtschaft und Verbraucherschutz (MUNLV) des Landes Nordrhein-Westfalen, Düsseldorf, Germany.

Moshage U. (2004). Rheologie kommunaler Klärschlämme – Messmethoden und Praxisrelevanz. PhD thesis, Institut für Siedlungswasserwirtschaft, Technische Universität Braunschweig.

Natterer F. and Wübbeling F. (2001). Mathematical Methods in Image Reconstruction. In: Society for Industrial and Applied Mathematics, Philadelphia, 2001.

Petritsch G., Reinecke N. and Mewes D. (1999). Visualization Techniques in Process Engineering. Wiley-VCH, Weinheim.

Röntgen W.K. (189). Eine neue Art von Strahlen. In: Sitzungsberichte der Würzburger Physik.-medic. Gesellschaft, (1896) 1-12.

Rosenberger S. (2003). Charakterisierung von belebtem Schlamm in Membranbelebungsreaktoren zur Abwasserreinigung. PhD thesis, Institut für Verfahrenstechnik, Technische Universität Berlin.

Seibert J.A. and Boone J.M. (2005). X-ray Imaging Physics for Nuclear Medicine Technologists. Part 2: X-Ray Interactions and Image Formation. In: Journal of Nuclear Medicine Technology, 33 (1) (2005) 3-18.

Shankar P.N. and Kumar M. (1994). Experimental determination of the kinematic viscosity of glycerol water mixtures. In: Proceedings: Mathematical and Physical Sciences, 444 (1994) 573-581.

Shim J.K., Yoo I.K. and Lee Y.M. (2002). Design and operation considerations for wastewater treatment using flat submerged membrane bioreactor. In: Process Biochemistry, 38 (2002) 279-285.

Wang Y., Howell J.A., Field R.W. and Wu D. (1994). Simulation of cross-flow filtration for baffled tubular channels and pulsatile flow. In: Journal of Membrane Science, 95 (3) (1994) 243-258.

Wicaksana F., Fane A.G. and Chen V. (2006). Fibre movement induced by bubbling using submerged hollow-fibre membranes. In: Journal of Membrane Science, 271 (2006) 186-195.

Xing C.H., Qian Y., Wen X.H., Wu W.Z. and Sun D. (2001). Physical and biological characteristics of a tangential-flow MBR for municipal wastewater treatment. In: Journal of Membrane Science, 191 (2001) 31-42.

Yakhot V. and Orszag S. (1986). Renormalized Group Analysis of Turbulence. In: Journal of Scientific Computing, 1 (1) (1986) 3-51.

Yamamoto K., Hiasa M., Mahmood T. and Matsuo T. (1989). Direct solid-liquid separation using hollow fiber membrane in an activated sludge aeration tank. In: Water Science and Technology, 21 (1989) 43-54.

Yusuf C. and Murray M.Y. (1993). Prediction of liquid circulation velocity in airlift reactors with biological media. In: Chemical Engineering Progress, 6 (1993), 38-45

Model-based optimisation of the biological performance of a sidestream MBR

I. Nopens*, G. Sin*, T. Jiang*, L. d'Antonio*, S. Stama*, J. Zhao* and P.A. Vanrolleghem*,**

* BIOMATH, Department of Applied Mathematics, Biometrics and Process Control, Ghent University, Coupure Links 653, 9000 Gent, Belgium (E-mail: ingmar.nopens@ugent.be)
** modelEAU, , Département Génie Civil, Pavillon Pouliot, Université Laval, Québec G1K 7P4, QC, Canada

Abstract A model-based optimisation of the operation in view of the biological performance in terms of nitrogen (N) and phosphorus (P) removal of a pilot-scale side-stream MBR has been performed by means of a two-tier scenario analysis. The methodology uses two different scenario analyses to simulate the effect of three degrees of freedom in the MBR system: (1) DO set-point in the aerobic reactor, (2) sludge residence time and (3) internal recirculation rate. The scenarios are simulated using a calibrated ASM2d MBR model. Effluent quality, in terms of nitrate, ammonia and phosphate, is used to select the best scenario. It proved to be a compromise between nitrogen and phosphorus removal as these are linked. A 42% reduction in ammonium and a 32% reduction in nitrate concentration were achieved. Phosphate removal is partly sacrificed (39% increase) compared to the standard operation.

Keywords
ASM2d; modelling, MBR, nutrient removal; optimisation; scenario analysis

INTRODUCTION

Constraints on the effluent quality demanded from wastewater treatment plants (WWTPs) are becoming more stringent in view of the implementation of the EU Water Framework Directive. To meet these constraints, the performance of the currently used biological nutrient removing technologies needs to be improved considerably. In the case of membrane bioreactors (MBR), the effluent suspended solids criterion is not an issue as it is removed to a great extent by the membrane. However, the effluent nitrogen and phosphorus criteria are usually the most challenging to comply with. These criteria are mostly met by developing an optimal configuration and operational strategy for the MBR system under study.

The objective of this study is to improve the MBR performance in terms of effluent quality using a systematic approach relying on a model-based optimization methodology. During the last two decades mathematical models have been developed for the conventional activated sludge process, resulting in a suite of models known as the Activated Sludge Models (ASM) (Henze et al., 2000). These models have been successfully applied for various purposes ranging from capacity evaluation to optimization of operation and controller development for activated sludge systems (Demuynck et al., 1994; Hvala et al., 2001; Artan et al., 2002; Sin et al., 2004; Corominas et al., 2006). These models can also be applied for the description of the biological processes in MBR systems, since the underlying biological processes are similar (Jiang et al., 2005). One advantage of using a model-based approach is that the number of scenarios that can be evaluated increases considerably, e.g. from 5-6 scenarios typically tested in experimental approaches to thousands as tested using models (e.g. Sin et al., 2004).

The model-based optimization methodology used in this study is adopted from Sin et al. (2004) and is shown in Fig 1. In this contribution, we evaluate the methodology for improving the effluent quality of a pilot-scale side stream MBR performing COD, N and P removal. The methodology is based on running a multitude of scenarios to simulate the effect of different operational parameters (in the jargon of optimization, *degrees of freedom*) on the MBR performance. In this optimization study, we focused on the following operational parameters: DO set-point (DOsp) in the aerobic phase, the sludge residence time (SRT) and the internal recirculation rate (Qint) of sludge from the aerobic/anoxic to the anaerobic compartment. The ASM2d model was chosen to describe the biological processes occurring in the pilot-scale MBR. In what follows, the pilot-scale MBR system under study is described, the calibrated model is introduced and the optimisation methodology is illustrated. Subsequently, the results of the optimisation exercise are shown and discussed.

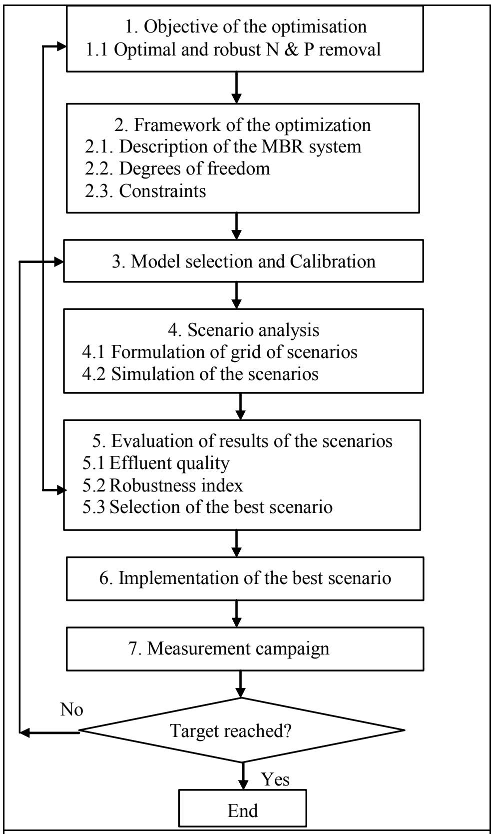

Figure 1 A systematic methodology for the model-based optimisation of MBR systems (after Sin et al., 2004)

MATERIALS AND METHODS

Pilot-scale MBR

The pilot-scale MBR consists of an anaerobic compartment (8 l), an aerobic/anoxic compartment (17 l) and a membrane loop (3.8 l) and is operated following a UCT type configuration treating a synthetic influent with a composition mimicking a domestic wastewater. The MBR, automated for data acquisition and control, is operated in a 40 min aerobic/anoxic cycle mode. Each cycle consists of 17 min aerobic and 23 min anoxic conditions. During the first 11 minutes of the anoxic phase, the sludge is mixed within the aerobic/anoxic compartment. During the last 12 minutes of the anoxic phase, the sludge from the aerobic/anoxic compartment is recycled to the anaerobic compartment since it then contains the lowest concentration of nitrate. Biomass separation is achieved by a tubular PVDF membrane module with a surface area of 0.17 m^2 (Norit, XF, The Netherlands) and the fouling is controlled by a periodical backwash operating at a 475 seconds cycle mode. Each cycle, consisting of 450 seconds of filtration and 18 seconds of backwashing, is followed by a short 7 seconds relaxation period during which the permeate pump is stopped. A detailed description of the MBR is given in Jiang et al. (2007).

ASM2d MBR model

The lab-scale MBR was modelled using ASM2d (Henze *et al.*,2000). All modelling and simulations were performed using WEST® (MOSTforWater, NV, Kortrijk, Belgium) a powerful modelling and simulation software platform designed for the modelling of WWTP (Vanhooren *et al.*, 2003).
The ASM2d MBR model was calibrated using averaged long-term (3 months) daily monitoring data (NH_4, NO_3, PO_4 and TSS) and data of a detailed measurement campaign (NH_4-N, NO_3-N and PO_4-P measurements in the anaerobic and aerobic/anoxic compartments) of one cycle (40’). The discussion of this calibration is beyond the scope of this contribution. Results of the calibrated model are shown in Figure 2, merely for the reader to asses the model quality. For details on the entire calibration process, the reader is referred to Jiang et al. (2007).

Optimization methodology

The optimization methodology is shown in Figure 1. The first step is the definition of the objective of the optimisation study that will serve as the main criterion in selecting the optimal operational strategy for the system. The second step defines the framework of the optimisation and included the description of the system, and the definition of the degrees of freedom and constraints of the system. This is followed by the model selection and calibration step to obtain a realistic model of the MBR system. In the fourth step, different levels of scenarios based on the above degrees of freedom and constraints are formulated and simulated using the calibrated model. In the following step (5), all simulated scenarios are thoroughly evaluated using effluent quality. A best (or optimal) scenario is selected that meets the defined objective(s). This scenario is then to be implemented in the system (step 6) and evaluated after three sludge ages (for the changes to take effect and a transient to fade out) by both monitoring the effluent quality of the lab-scale MBR on a daily basis and also by performing a detailed measurement campaign, i.e. step 7. The methodology can be repeated if the target has not been reached, starting from step 3.

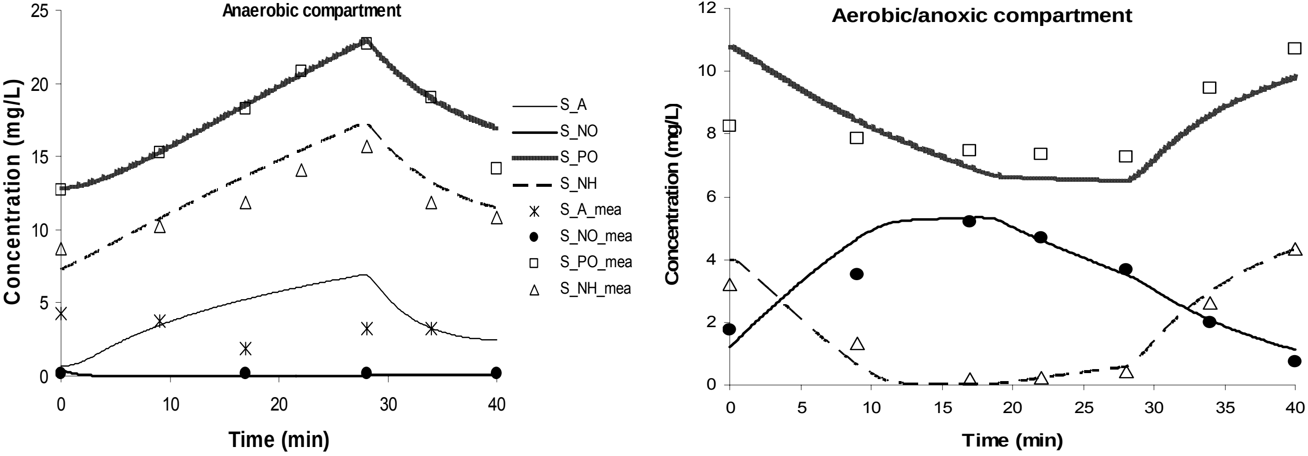

Figure 2 Results of the calibration with the comparison of the simulation and measurements in the anaerobic and aerobic/anoxic compartment during 1 cycle (Jiang et al., 2007)

Scenario analysis

The calibrated model is used in a scenario analysis to evaluate the effect of the three chosen operational parameters of the MBR: the DOsp in the aerobic phase, the sludge residence time (SRT) and the internal recirculation rate (Qint) (see Table 1). For a thorough analysis of individual and combined effects of parameters on the performance of the MBR, three levels of scenario analysis can be adopted: (1) one parameter is varied, while the other two parameters are fixed to the reference values (2) two parameters are co-varied, while one parameter is fixed, and (3) all three parameters are co-varied at the same time (grid design). So far, only the first 2 levels have been applied in this work. The considered parameter ranges are shown in Table 1. All scenarios are simulated for three times the corresponding SRT. This is needed to ensure stable operation and, hence, a reliable comparison between different scenarios.

The simulation results from the scenarios were evaluated using the effluent quality for COD, N and P removal. The parameter range considered in the second level of scenarios was decided based on the interpretation of the first level results (see below). The modelling and simulation platform WEST® that was used provides a scenario analysis module.

Table 1. Levels of scenarios to simulate the single and simultaneous effect of three degrees freedom on the MBR performance

Levels	SRT (days)	DOsp (mg/l)	Qint (m^3/d)	Total no. scenarios
1	[5, 10, 15, 17.55(1), 20, 25, 30, 45, 60]	[0.25, 0.5, 0.75, 1, 1.25, 1.5, 1.75, 2(1), 2.25, 2.5, 2.75, 3, 3.25, 3.5, 3.75, 4, 4.25, 4.5, 4.75, 5]	[0.264, 0.423, 0.864(1), 1.728, 2.592, 3.456, 4.32]	36
2	[7.5, 10, 12.5, 15, 17.55(1), 20, 22.5, 25]	[0.25, 0.5, 0.75, 1, 1.25, 1.5, 1.75, 2(1)]	[0.264, 0.523, 0.781, 1.040, 1.229, 1.557, 1.816, 2.075, 2.333, 2.592]	224

(1) Reference values

RESULTS

First level of scenario analysis

The results of the first level scenario analysis are shown in Figures 2-4. In total 36 scenarios were simulated.

Increasing the internal recirculation rate (Qint) results in a decrease of the effluent nitrate concentration. However, this happens at the expense of phosphorus removal, which deteriorates slightly. It is important to note that beyond a certain point (i.e. 1.728 m^3/l) further increasing of Qint did not result in a further improvement of nitrate removal. This may be due to a limited amount of readily biodegradable COD which is not sufficient to denitrify the remaining nitrate in the anaerobic tank. Increasing Qint did not have a significant effect on the COD and ammonia removal efficiency.

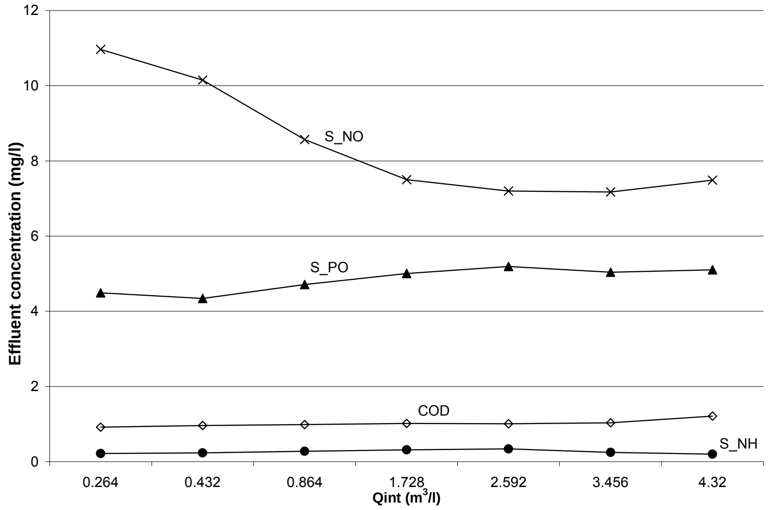

Figure 3 Effluent phosphorus, nitrate, ammonia and COD concentrations for level 1 scenario analysis with varying Qint (SRT=17.5d and DOsp=2 mg/l)

Figure 4 reveals that an increasing DOsp slightly increases the effluent concentrations of nitrate and phosphorus, whereas the efficiency of COD and ammonia removal remain almost unchanged at a satisfactory level. A DO-sp below 0.5 causes a large increase particularly in the effluent concentration for ammonia, indicating washout of nitrifiers. This is not surprising since it is known that the affinity constant of nitrifiers for oxygen is quite high, meaning that lower oxygen levels will decrease the observed growth rate of nitrifiers.

The DOsp is observed to affect the phosphorus removal in two distinctive ways. First, when DO is too low (e.g. 0.25 mg/l), biological phosphorus removal collapses because poly-phosphate accumulating organisms (PAOs) are unable to fully take up the released phosphorus in the aerobic phase, since the oxygen level is controlled below or close to the oxygen affinity constant of PAOs (leading to almost 50% decrease in the P-uptake rate and washout of PAOs). Second, the optimal P-removal is achieved at a DOsp equal to 0.5 mg/l, which also corresponds to the minimum nitrate concentration achieved in the MBR. Higher DOsp values lead to a gradual decrease in P-removal caused by the well-known nitrate effect. Also nitrate levels gradually increase as increased oxygen hampers the extent of simultaneous nitrification and denitrification (SND) in the system. Overall, the extent of phosphorus removal depends on the level of nitrate in the MBR (see Figure 4).

Figure 5 shows that increased SRTs result in a decreased P- and COD-removal. However, both ammonia and nitrate removal improved. The latter is understandable since an increasing SRT leads to a higher biomass concentration in the system. This eventually leads to a higher anoxic endogenous respiration and, hence, more nitrate removal.

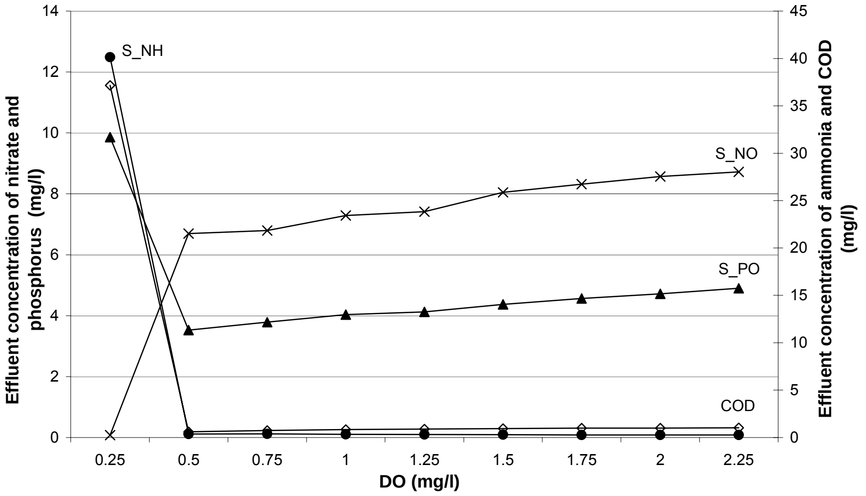

Figure 4 Effluent phosphorus, nitrate, ammonia and COD concentrations for level 1 scenario analysis with varying DOsp (SRT=17.5d and Qint=0.864m^3/d)

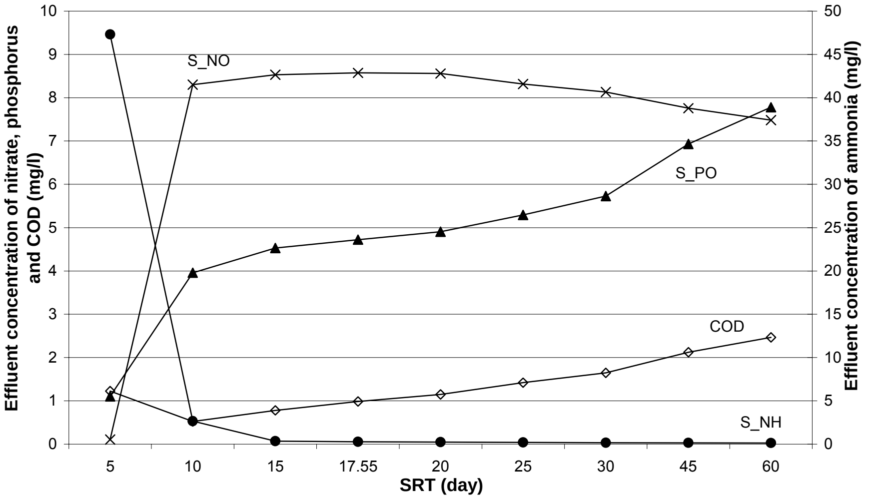

Figure 5 Effluent phosphorus, nitrate, ammonia and COD concentrations for level 1 scenario analysis with varying SRT (DO=2mg/l and Qint=0.864m^3/d)

Concerning the SRT effect on P-removal, one also notices that increasing SRT beyond 25d has a strong negative effect (note the increase in slope beyond 25d). This negative effect of high SRTs stems from the P-release and P-uptake mechanism, which depends in a non-linear way on the concentration of the intracellular storage products, polyphosphate and PHA in PAOs. Under fixed influent organic loading, an increase in the SRT has a dilution effect on the intracellular products. Subsequently, this leads to a reduction in the P-release and P-uptake rates of PAOs, with as a result bad P-removal (Smolders et al., 1995). Finally, it should be noted that the SRT of 10d is a point of minimum and maximum for COD and nitrate concentration respectively. Ammonia and nitrate concentrations largely increase and decrease respectively with SRT values lower than 10d mainly due to wash-out effects on the nitrifiers and PAOs.

Second level of scenario analysis

The results of the second level scenario analysis are shown in Figure 6. In total 224 scenarios were simulated. The advantage of the second level scenario analysis is that these trends can be evaluated quantitatively at different settings of another degree of freedom. Based on the fact that most of the scenarios analysed resulted in high COD and ammonia removal efficiencies, the discussion is limited to the phosphorus and nitrate effluent concentration plots only. The same trends as in the first level scenario analysis can be observed.

DISCUSSION

Some general elements are observed from the detailed analysis of the two levels of scenario analysis:

- **Qint:** Increasing the recirculation flow rate decreases the effluent concentration of nitrate. This is because a high internal recirculation recycles more nitrate back to the anaerobic tank from the aerobic/anoxic tank, which in turn enhances overall nitrate removal. As mentioned above, this enhanced nitrate removal happens at the expense of phosphorus removal, due to the increased consumption of influent VFAs by the denitrifying heterotrophs.
- **DOsp:** Increasing the DOsp is only efficient for ammonia removal, while it has a negative effect on both phosphorus and nitrate removal because the simultaneous nitrification and denitrification decreases in the aerobic tank.
- **SRT:** Too low SRTs are bad for P and N removal as it causes washout of nitrifiers and PAOs from the system. High SRTs are good for the ammonium removal efficiency up to a certain point (15 days in this study) where ammonia removal efficiency is no longer affected by an increased SRT. A high SRT is observed to elevate the COD effluent concentration, probably due to an increased decay of biomass (as SRT increases) and the increased level of hydrolysis products being produced and can end up in the effluent. As mentioned earlier, too high SRTs cause a decrease in the efficiency of phosphorus removal, mainly because of the inverse relationship between SRT and the dynamics of P-storage and P-release (Smolders et al., 1995).

Determining the optimal scenario for nutrient removal

Based on the previous discussion, it can be concluded that (1) DOsp should be kept low, (2) SRT should be intermediate and (3) Qint should be intermediate as well. Under given conditions and constraints (e.g. fixed organic loading), it seems impossible to find a scenario (among 260) that provides a phosphorus removal comparable to the reference case without drastically sacrificing on the N removal. Having this in mind, the best scenario was chosen as the one providing the following criteria: (i) complete nitrification (NH4 < 0.2 mgN/l) and (ii) optimal denitrification and P-removal (equal weight for the effluent nitrate and phosphate). This led to the selection of the scenario given in Table 2. It results in a 42% decrease in ammonium and a 32% reduction in nitrate concentration. However, P-removal is partly sacrificed (39% increase).

The optimisation suggests to maximise simultaneous nitrification and denitrification (SND) in the system as a means to optimise P and N removal. For our particular system, lowering the oxygen set-point appears to provide this objective (under the optimal SRT and Qint). This observation is very similar to the conclusions of the model-based optimisation of other systems, particularly with low organic COD/P ratios (Sin et al., 2004, Corominas et al., 2006). However, in traditional activated sludge systems with gravity settling, low oxygen is known to be a common cause of filamentous bulking and bad settling (among many other factors), and hence it is often avoided (Sin et al, 2006; Comas et al., 2006). For the MBR systems, we anticipate that this should not be a concern since the solid-liquid separation is performed with ultrafiltration. Another drawback of low DOsp is possible accumulation of nitrite. This was not included in the model and can not be verified at this point. It will be ignored at this stage. The obtained optimal scenario will now be implemented and tested in the pilot scale MBR and evaluated.

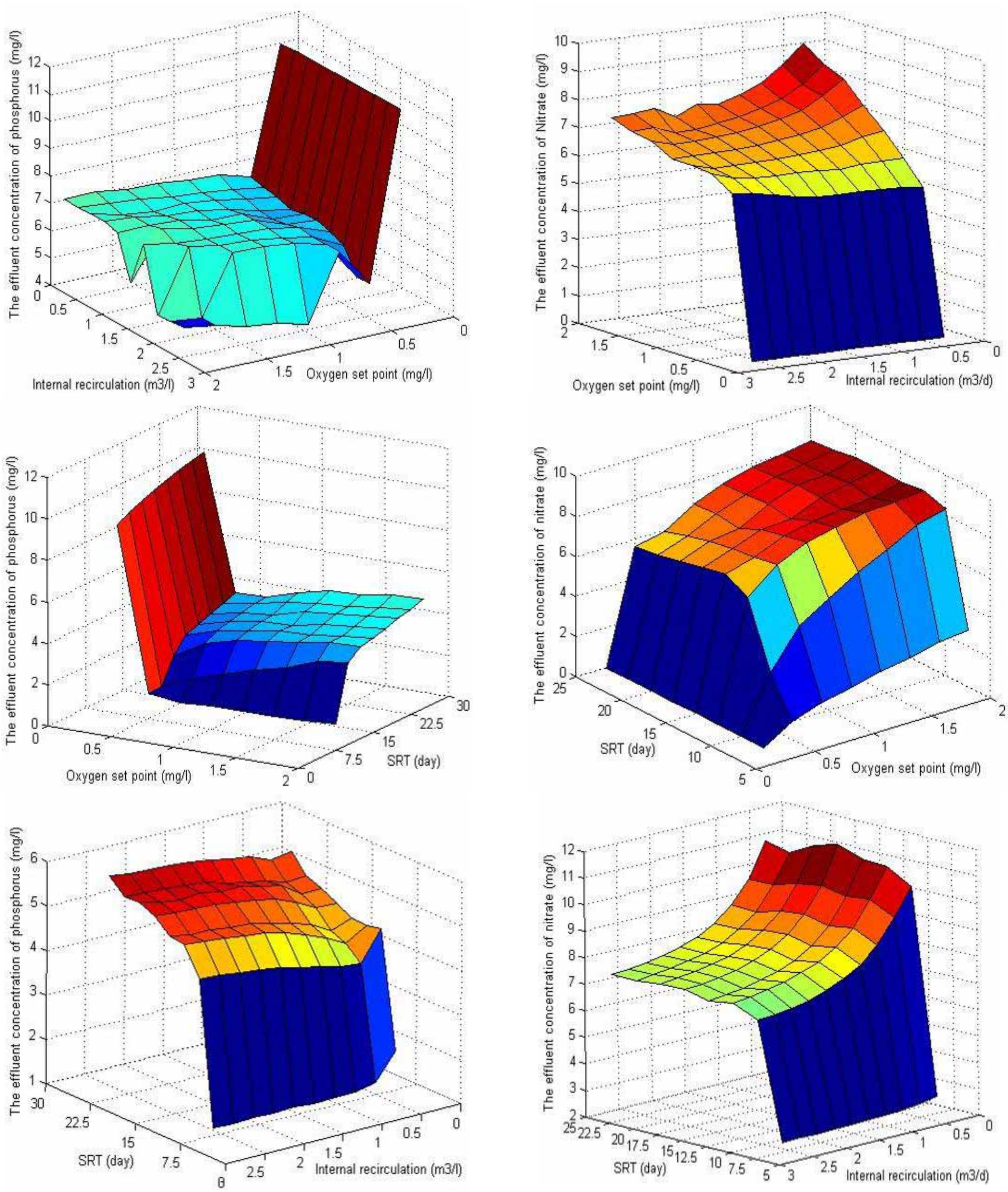

Figure 6 Effluent phosphorous (left) and nitrate (right) concentrations for the second level scenario analysis: fixed SRT (top), fixed Qint (middle), fixed DOsp (bottom)

Table 2. Optimal operational strategy obtained from the model-based optimisation methodology

	SRT d	Qint m^3/d	DO mg/l	Nitrate mg/l	Phosphate mg/l	Ammonia mg/l	COD mg/l
Reference	17.55	0.864	2.00	8.57	4.72	0.28	0.98
Best scenario	17.55	1.04	0.75	5.84	6.58	0.16	2.20

CONCLUSIONS

The improvement of nutrient removal of a side stream MBR was investigated using a model-based approach. Individual as well as combined impacts of three degrees of freedom (DOsp in aerobic reactor, Qint and SRT) were investigated using scenario analysis. It was found that:

- Too low DO causes washout of nitrifiers and PAOs, while increasing DOsp too much negatively influenced both nitrate and phosphate removal
- Low SRTs result in washout of nitrifiers whereas too high values had a negative effect on phosphate removal
- High Qint has a positive effect on nitrate removal, but deteriorates P-removal. However, too high values do not have a significant effect on N and P removal.

The optimal scenario was found as a compromise between N and P removal, which suggested a 32% decrease in nitrate concentration and a 42% decrease in ammonia concentration. Phosphate, however, had to be sacrificed to achieve this high nitrogen removal performance (39% increase).

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REFERENCES

Artan, N., Wilderer, P., Orhon, D., Tasli, R. and Morgenroth, E. (2002). Model evaluation and optimisation of nutrient removal potential for sequencing batch reactors. Wat. Sci. Tech., 28(4), 423-432.

Comas, J., Rodriguez-Roda, I., Poch, M., Gernaey, K.V., Rosen, C., and Jeppsson, U. (2006) Extension of the IWA/COST simulation benchmark to include expert reasoning for system performance evaluation. Wat. Sci.Tech. 53(4-5), 331-339.

Corominas, L., Sin, G., Puig, S., Traore, A., Balaguer, M., Colprim, J. and Vanrolleghem, P.A. (2006). Model-based evaluation of an on-line control strategy for SBRs based on OUR and ORP measurements. Wat. Sci. Tech., 53(4-5), 161-169.

Demuynck, C., Vanrolleghem, P.A., Mingneau, C., Liessens, J. and Verstraete, W. (1994). NDBEPR process optimisation in SBRs: Reduction of external carbon source and oxygen supply. Wat. Sci. Tech., 30(4), 169-179.

Henze, M., Gujer, W., Mino, T. and van Loosdrecht, M.C.M. (2000). Activated Sludge Models ASM1, ASM2, ASM2d and ASM3. IWA Scientific and Technical Report 9 IWA. London.

Hvala, N., Zec, M., Ros, M. and Strmcnik, S. (2001). Design of a sequencing batch reactor sequence with an input load partition in a simulation-based experimental environment. Water Env. Res., 73, 146-153.

Jiang, T., Liu, X., Kennedy, M.D., Schippers, J.C. and Vanrolleghem, P.A. (2005). Calibrating a side-stream membrane bioreactor using Activated Sludge Model No. 1. Wat. Sci. Tech., 52(10-11), 359-367.

Jiang, T., Sin, G., Spanjers, H., Nopens, I., Kennedy, M.D., van der Meer, W.G.J., Futselaar, H., Amy, G. and Vanrolleghem, P.A. (2007) Comparison of the activated sludge modelling approach between membrane bioreactor and conventional activated sludge processes. Wat. Res., (submitted).

Jiang, T. (2007). Characterisation and modelling of soluble microbial products in membrane bioreactors. PhD thesis, Ghent University, Belgium.

Sin, G., Insel, G., Lee, D. and Vanrolleghem, P.A. (2004). Optimal but robust N and P removal in SBRs: A model-based systematic study of operation scenarios. Wat. Sci. Tech., 50(10), 97-105.

Sin G., Villez, K., and Vanrolleghem, P.A. (2006) Application of a model-based optimisation methodology for nutrient removing SBRs leads to falsification of the model. Wat. Sci.Tech. 53 (4-5), 95-103.

Smolders, GJF, Klop, JM, Van Loosdrecht, MCM and Heijnen, JJ (1995) A metabolic model of the biological phosphorus removal process; effect of the sludge retention time. Biotech. Bioeng. 48:222–233

Vanhooren, H., Meirlaen, J., Amerlinck, Y., Claeys, F., Vangheluwe, H. and Vanrolleghem, P.A. (2003) WEST: Modelling biological wastewater treatment. J. Hydroinformatics, 5, 27-50.

Membrane Bioreactor Aeration: Investigation of the velocity flow pattern

D. Tacke*, J. Pinnekamp*, H. Prieske**, M. Kraume**

* Institute of Environmental Engineering, RWTH Aachen University, Germany
(E-mail: tacke@isa.rwth-aachen.de, pinnekamp@isa.rwth-aachen.de)
** Department of Chemical Engineering, Technische Universität Berlin, Germany
(E-mail: helmut.prieske@tu-berlin.de, matthias.kraume@tu-berlin.de)

Abstract Results of investigations concerning membrane bioreactor aeration are presented which were carried out at the Institute of Environmental Engineering of RWTH Aachen University (ISA) in cooperation with the Department of Chemical Engineering of the Technische Universität Berlin. In the field of industrial and municipal wastewater treatment the use of membrane bioreactors (MBR) is of increasing interest especially due to the high requirements on effluent quality nowadays. The design of aeration systems is a very important aspect of MBR development because it influences both cost of operation and filtration flux. The ISA has carried out tests concerning the velocity flow pattern in flat sheet membrane modules (developed by the A3 water solutions GmbH) to identify the effects of different aeration systems, aeration intensities and module constructions. The Department of Chemical Engineering is currently using the results obtained from the ADV to calibrate a numerical model which simulates two phase water and gas flow within an aerated membrane module. Optical investigations concerning the bubble distribution gives a better understanding of the flow conditions in MBR. Developing a numerical tool for membrane module optimization concerning the hydrodynamics is the aim of the investigation of membrane bioreactor aeration.

Keywords municipal wastewater treatment, membrane bioreactor, aeration, velocity flow pattern

INTRODUCTION

In the field of industrial and municipal wastewater treatment the use of membrane bioreactors (MBR) is increasing especially due to the high requirement on effluent quality nowadays. In Germany there are at present seven municipal wastewater treatment plants with a total capacity of about 120,000 inhabitants applying MBR technology (Pinnekamp, 2004). The high potential of this technology, especially the high effluent quality, will result in a significant increase of these figures, if the reduction of operating costs and the solution of nowadays technical teething problems can be managed. While MBR is an established technology for waste water treatment, efficient fouling reduction by air scour remains a problem. With regard to the cost of operation, approximately 70% of the energy input usually spent on the aeration (Kraume and Bracklow, 2003). Krause and Cornel (2005) reported that for an MBR with flat sheet membranes an energy input of about 1 kWh was necessary per 1 m³ of treated waste water. Around 0.2 kWh was used for oxygen supply of the activated sludge and 0.7 kWh for the deposit reduction by air scour. Concerning operation costs design of aeration systems is a very important aspect of MBR development because it influences both operating costs and filtration flux. To use the aeration more efficiently the MBR was designed as an airlift loop reactor. Here, the reactor volume is divided into a downcomer and an aerated riser section. The different gas holdups cause a different hydrostatic pressure in both regions which initiates a circulation velocity.

The Institute of Environmental Engineering of RWTH Aachen University (ISA) has carried out tests in water and glycerol (34 % by volume glycerol) concerning the velocity flow pattern in a test plant and a pilot plant equipped with flat sheet membrane modules. Streaming conditions were measured

within a selected part of the cross-section using Acoustic Doppler Velocimetry (ADV, Nortek Inc.). From the measured velocities the contribution of the injected air inside the reactor can be derived because the uprising bubbles cause the water flow.
The Department of Chemical Engineering of the Technische Universität Berlin is currently using the results obtained from the ADV to calibrate a numerical model which simulates two phase water and gas flow within an aerated membrane module. Furthermore experimental investigations were carried out in a quasi 2 dimensional model to analyse the bubble distribution optical by video imaging. Numerical simulations were used to perform parameter studies by varying geometrical values or operating conditions (e.g. channel width, bubble diameter). The next step to a systematically optimization of aeration will be the use of numerical methods which can reduce time and effort for construction and evaluation of several gradually improved physical models.

The main objectives of the investigations were to analyze the flow inside a MBR and to achieve a homogeneous bubble distribution over the whole cross-section of the membrane module together with minimal energy input. To analyze flow conditions within a membrane module flow velocities can be considered. An uneven distribution of the air flow at the bottom of a module leads to considerably different local velocities within the channels between the membrane plates. An inhomogeneous distribution of uprising air might lead to so called dead zones with no or very low flow velocities within the membrane module which effects sludge deposition (layer cake) on the membrane surface. In these zones filtration flux declines or becomes impossible. Therefore higher cross flow velocities are advantageous as they limit the deposition on the membrane surfaces (Bérubé and Lei, 2006; Ueda et al., 1997; Vera et al., 2000).
The aim was to investigate the flow behaviour inside a MBR as well as to identify the effects of different aeration systems, which means different positions of the aeration tubes, aeration intensities and module construction.

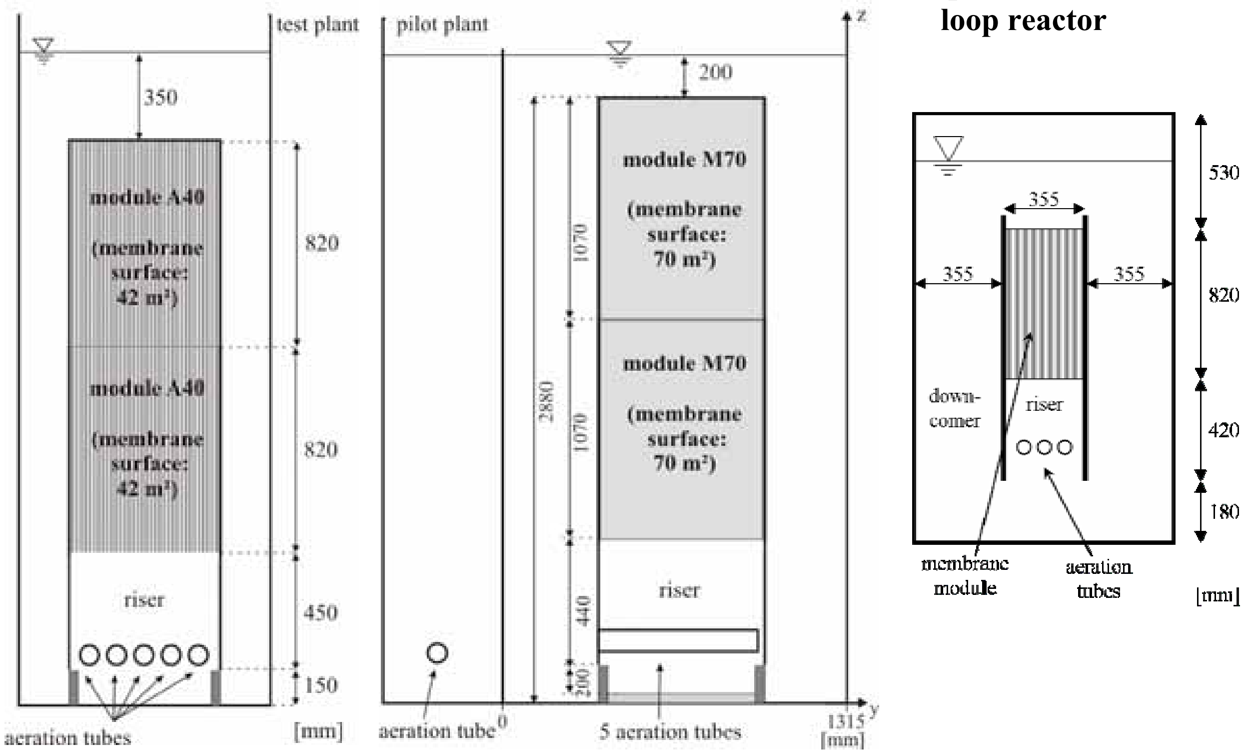
MATERIALS AND METHODS

Experimental setup

For experimental investigations at the ISA, a test plant and a pilot plant with a volume of approximately 2 m³ and 8 m³ are employed (Figure 6a, b). Because of envisaged tests with activated sludge the pilot plant is equipped with a collection basin. Commercial flat sheet membrane modules (A3 Water Solutions GmbH) with surface areas of about 42 m² up to 140 m² are located in the riser equipped with aeration tubes (ENVICON). The tests were carried out using the module types A40 and a newer developed M70 with a distance between the membrane sheets of 7 mm. Streaming conditions were measured within a selected part of the cross-section above one or two membrane modules arranged on top of each other (double deck) of the membrane modules. Measurement was also carried out in the downcomer between the membrane module and the reactor wall in the pilot plant. Measurement inside the module is not possible with ADV. Therefore streaming conditions inside the module must be estimated from the velocities close to the top of the module. The experimental velocity values are averaged over 1 minute with an acquisition rate of 5 or 25 Hz.
The experiments in the test plant were conducted with water and glycerol (34 % by volume glycerol). Glycerol was used to simulate viscosities at about 3 mPa s which is comparable to the viscosity of typical activated sludge for MBR with suspended solids concentration of about 12 g/L. In the pilot plant up to now streaming conditions were measured in water.

The experimental investigations at the Department of Chemical Engineering of the Technische Universität Berlin were carried out with water and air in a quasi 2 dimensional loop reactor shown schematically in Figure 6c with 2.1 m height, 1.2 m width and 0.1 m depth. To simulate the displacement and the wall effects of a flat sheet membrane module, several plates of acrylic glass with a thickness of 5 mm were arranged with a fixed distance of 7 mm (model of membrane plates).

Three tubular aerators (ENVICON) were employed for the aeration with small bubbles, which have an average diameter of about 2 - 3 mm. The whole set-up represents a quasi 2 d model of a typical MBR. In transferring the geometry from 3 d to 2 d the relation between the area of the riser and the downcomer was kept constant. The global gas holdup was determined by optical analysis of the increased volume in comparison to the unaerated condition.



a) test plant **b) pilot plant** **c) quasi 2 dimensional loop reactor**

Figure 6 Test plant (a) and pilot plant (b) used for investigation of the velocity flow pattern equipped with different A3 membrane modules (A40, M70); quasi 2dimensional loop reactor (c)

Numerical method

Numerical simulations were applied for parameter studies in which geometrical dimensions or operating conditions were varied. The commercial CFD code ANSYS-CFX10 was used to simulate the two phase flow with an Eulerian-Eulerian approach. To consider the momentum transfer between the continuous liquid phase and the dispersed gas phase, the Grace Drag Model was chosen which includes the effect of bubble deformation and gas holdup on the resistance coefficient. For simplification, only a fixed bubble size was assumed in the stationary simulations on a hexahedral mesh with about 35,000 cells. This domain represents one half of the quasi 2d reactor. To reduce the computational effort, a fixed degassing condition on the top of the reactor was used. Turbulence effects were taken into account with the Shear Stress Transport (SST) Model for the continuous phase and a zero-equation model for the dispersed phase.

RESULTS AND DISCUSSION

Intensity of aeration and module construction

One focus of the tests was to identify a proper intensity of aeration. Measurements at different aeration intensities from 0.15 $Nm^3/(m^2\,h)$ up to 1.2 $Nm^3/(m^2\,h)$ concerning the membrane surface area have been carried out. Results of the maximum velocities measured above the modules are shown in Figure . Measurements were operated at different heights above the module – concerning the streaming conditions inside the module, measurement as near as possible above the module was most interesting. Because the ADV probe needs minimum distance to boundaries of about 50 mm measurement was carried out 60 and 70 mm above the module. In Figure it can be seen that there is not a linear increase of velocities for higher aeration rates. Interacting factors limiting an increase in velocity are membrane plate distance, increasing gas content and increase of the shear resistance due to a higher streaming pressure.

The highest velocities above one single module A40 were measured at an aeration flow rate of 0.8 Nm³/(m² h). Concurrently the most even distribution of injected air could be established. In the case of two membrane modules (double deck) with one riser the measured velocities are similar to those of a single module. For a single membrane module A40 an aeration of 0.8 Nm³/(m² h) was discovered to assure good operating conditions. If the membrane surface is doubled without increasing aeration strength, the specific aeration rate related to the membrane area is halved down to 0.4 Nm³/(m² h). The velocity flow pattern was as well investigated for the newer M70 membrane module double deck configuration. According to the A3 GmbH lower aeration rates from 0.15 up to 0.32 Nm³/(m² h) have been investigated. The values of the A40 and the M70 double deck configuration seem to result in one curve (Figure).

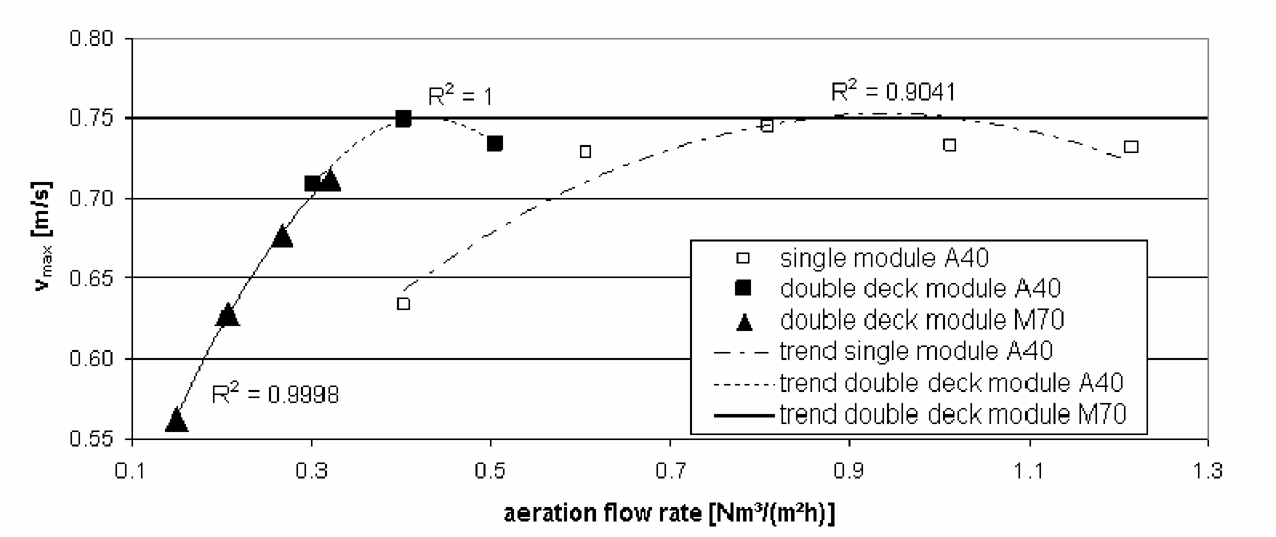


Figure 2 Relation between aeration flow rate and maximum flow velocities measured above different membrane modules (A40, M70)

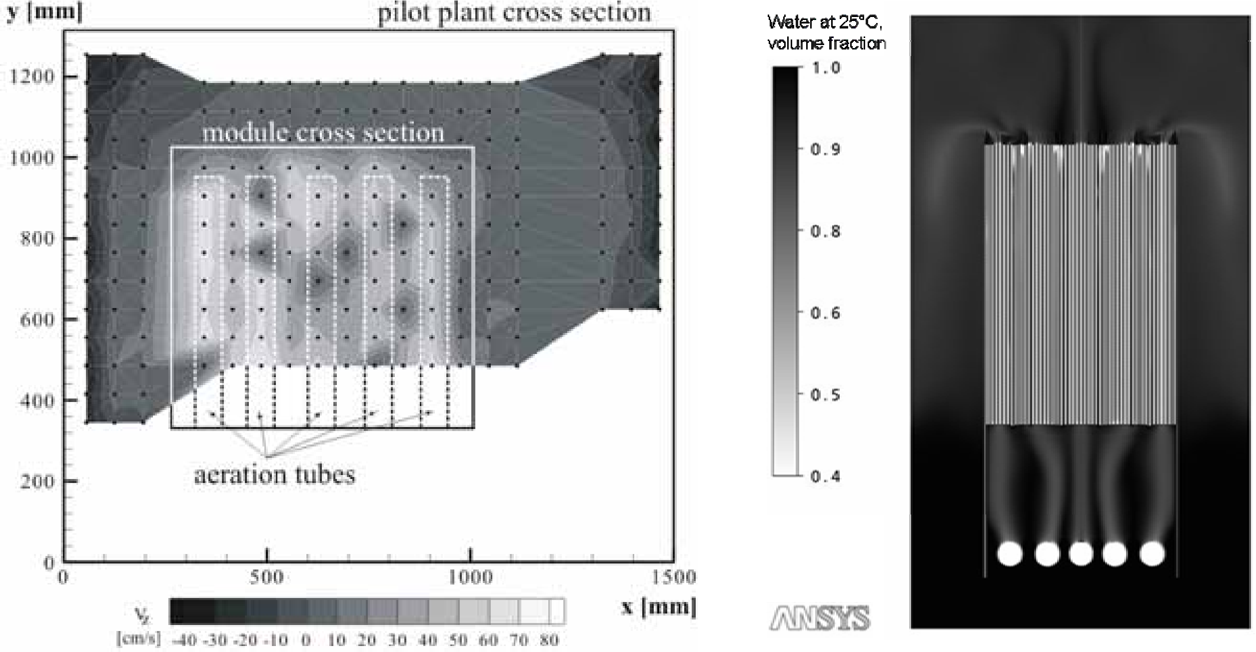
The first tests for measuring the flow pattern were carried out in water. Next was to determine whether and how there is an influence of viscosity on the streaming conditions inside a membrane module. Two effects could be identified by tests carried out with glycerol (34 % by volume glycerol): Firstly, there is a reduction of the velocities in vertical direction of about 30 %, and secondly, distribution of the flow pattern is smoother with less streaming peaks (Tacke, 2005a, b). Verification of this result will be done by measurement in activated sludge.

The Department of Chemical Engineering of the Technische Universität Berlin simulates the two-phase flow (water and gas) within an aerated membrane module using the results from the velocity measurements of the RWTH Aachen University. Numerical simulations allow a detailed view into the flow field, especially inside the membrane module where ADV cannot be used. Qualitatively the same phenomena as in the experimental investigations can be determined, but the velocity values are higher by approximately 15 % (Prieske et al., 2006). Additionally, the results of the simulation do not show the flattening in the increase of vertical velocities with rising flow rate.

Flow behaviour inside the riser

The aeration system consists of a riser and five aeration tubes. The number of aeration tubes and the orientation of the tubes are varied. As a result, the number of aeration tubes obviously has a large effect on the streaming pattern measured above the module (Tacke et al., 2006). If less than five aeration tubes are applied – as suggested by the A3 GmbH – there are backflows inside the module, and the position of the tubes below the module can precisely be seen from the streaming pattern measured above the module (Tacke et al., 2006). If the riser contains the suggested five aeration tubes there are no backflows inside the module. Although the position of the aeration tubes can be estimated from the velocity flow pattern as shown in Figure a. This effect could be noticed for both the A40 and the M70 membrane module.

A homogeneous bubble distribution over the entire cross section of the riser requires the flow approaching the tubes to be already homogeneous. Therefore, it was necessary to arrange the aerators outside the recirculation zone at the bottom of the riser. According to Prieske et al. (2006) especially around the tubular aerator closest to the riser wall the flow field is influenced by the direction change of the flow from the downcomer into the riser section. Furthermore the CFD simulation (Figure b) shows the same effect that could be seen from the measurement above the membrane module: an uneven distribution of air inside the module (Figure b). It suggests itself that the parallel orientation of the aeration tubes intensifies this effect. However tests with parallel and perpendicular orientation of the aeration tubes concerning membrane alignment showed no effect on the flow pattern (Tacke, 2005a).



a) velocity flow pattern above M70 (measurement)

b) contribution of the air inside A40 (CFD) (Prieske, 2006)

Figure 3 Influence of the aeration system on the velocity flow pattern as well as on the contribution of the air

Applied in activated sludge, the membrane plates that receive a weaker aeration will have more problems with sludge deposition on the surface and therefore the flux of an uneven aerated membrane module will decline. Hence an even distribution of aeration below the module is important for a well operating membrane module.

Flow behaviour inside the downcomer

In an airlift loop configuration the uprising air in the riser causes a circulation flow inside the whole reactor. Thus there has to be an opponent flow direction in the downcomer of the MBR. At the ISA measurement was carried out in the downcomer of the reactor to investigate the phenomena which reduce the circulation velocity. The positions of the ADV probe are marked with black dots.

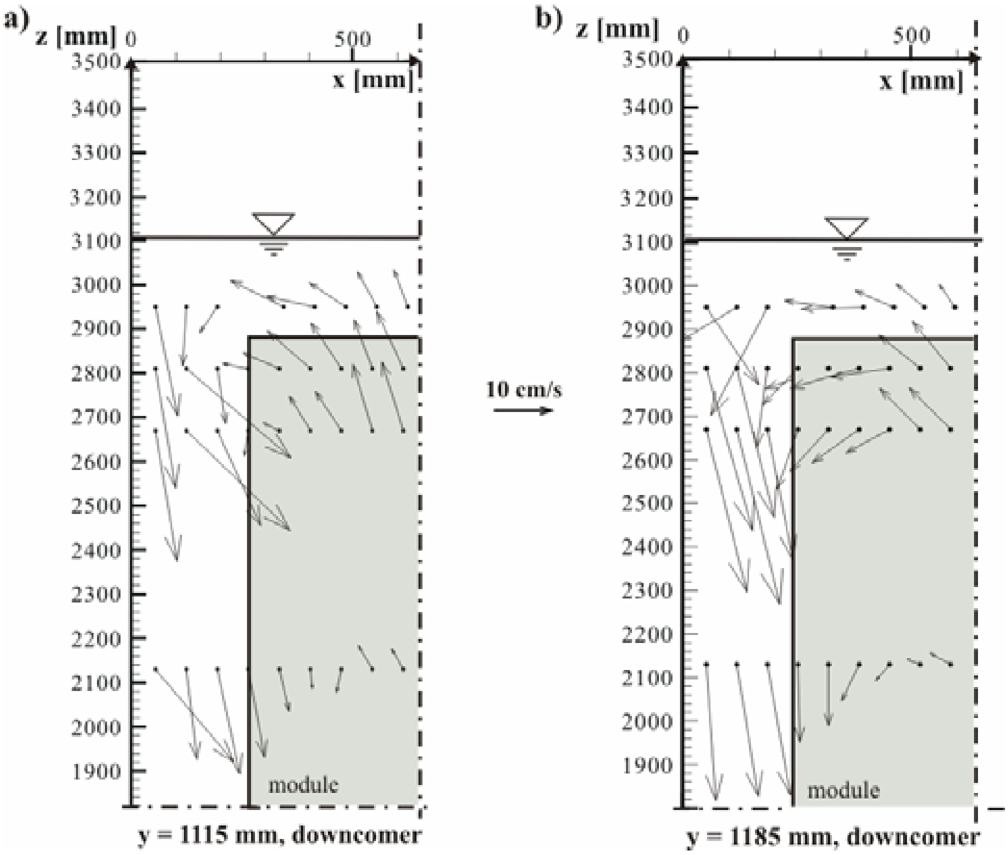


Figure 4 Velocity flow pattern in the downcomer of the pilot plant; at a distance of 130 mm to the membrane module wall (a) and at a distance of 130 mm to the reactor wall (b)

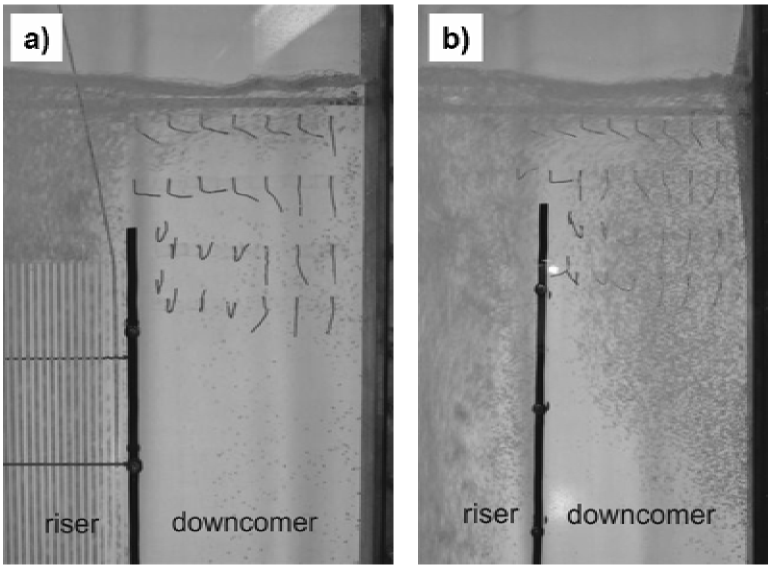


Figure 5 Visualization of the flow direction in the loop reactor with (a) and without (b) a membrane module

In Figure (a, b) the results in two sectional views in the downcomer are illustrated. Both views show a downwards orientated flow (x = 0 up to ~300 mm) as expected. In the case of Figure b the position of the sectional view is near to the reactor wall and shows higher and more distinctive downwards orientated velocities as the results nearer to the membrane module wall (Figure a). Furthermore it can be seen an unexpected uprising flow between the membrane module and the reactor wall. The experimental investigations at the Department of Chemical Engineering of the Technische Universität Berlin in the quasi 2 dimensional loop reactor shows results nearly comparable to the velocity measurement at the ISA. With the help of filaments attached on the wall inside the reactor the direction of the liquid flow can be estimated optically. Beside the effect of the recirculation flow inside the downcomer it is obvious that the membrane module in the riser effects less bubbles and a reduction of flow in the downcomer.

CONCLUSIONS

An uneven distribution of uprising air within a membrane module leads to dead zones with no or very low flow velocities which effects sludge deposit on membrane surface and filtration flux decline. Therefore the flow conditions in membrane bioreactors are an important object of investigation. The comparison with an airlift loop reactor (Chisti et al., 1988) gives a good basis but has to be modified because of the huge influence of the membrane module in the riser.

In this project the velocity flow pattern in a membrane bioreactor has been investigated with several tools: velocity measurement by Acoustic Doppler Velocimetry, optical observation in a model and numerical simulation. According to this for an even distribution of air several aspects have to be considered:

- Higher aeration intensity will not guarantee a smoother flow pattern or higher velocities within a module. For a newly developed module velocity measurement can be a helpful tool to choose the right aeration rate.
- An even distribution of aeration below a module is important to avoid backstreams inside a module and to achieve even distribution of air inside a module.
- The use of two membrane modules on top of each other (double deck) is an important way to minimize the specific power input for the aeration.
- The circulation velocity is reduced due to the resistance of the membrane module. Uprising velocities in the riser as well as downwards orientated velocities in the downcomer are reduced which effects less bubbles dragged into the downcomer.
- Inside the downcomer there is something like a “recirculation flow” which reduces the circulation velocity in the whole reactor. Particularly in the zone near the wall of the membrane module bubbles rise up.

Further investigations concerning flow conditions within a membrane module in activated sludge have to be done to consider the influence of a suspension on the flow pattern and, as a very important fact, to test the relation between aeration system and filtration flux. There will be tests in a bigger reactor concerning the influence of the cross section of the downcomer and the presence of several modules. In this case double deck membrane modules are put in one reactor to vary streaming conditions by minimizing or enlarging the distance between the modules respectively the reactor walls.

ACKNOWLEDGEMENTS

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REFERENCES

Bérubé P.R., Lei E. (2006). The effect of hydrodynamic conditions and system configurations on the permeate flux in a submerged hollow fiber membrane system. Journal of Membrane Science, Vol. 271, pp. 29-37

Chist M.Y. (1989). Airlift Bioreactors. Elsevier Applied Science, London, ISBN 1-85166-320-7

Chisti M.Y., Halard B., Moo-Young M. (1988). Liquid circulation in airlift reactors. Chemical Engineering Science, Vol. 43. No. 3, pp. 451-457.

Kraume M., Bracklow U. (2003). Das Membranbelebungsverfahren in der kommunalen Abwasserbehandlung – Betriebserfahrungen und Bemessungsansätze in Deutschland. (Membrane technology in the municipal waste water treatment operating experience and design rules in Germany) Tagungsband 5. Aachener Tagung: Membrantechnik in der Wasseraufbereitung und Abwasserbehandlung, T. Melin, J. Pinnekamp. Aachen. Ü2-1 – Ü2-20, ISBN 3-921955-28-9

Krause S., Cornel P. (2005). Untersuchungen zum Energiebedarf von Membranbelebungsreaktoren unter besonderer Berücksichtigung der Belüftungssysteme (Investigations of the energy demand of MBRs especially with regard to the aeration system). Tagungsband 6. Aachener Tagung: Membrantechnik in der Wasseraufbereitung und Abwasserbehandlung, A18, ISBN 3-86130-775-8.

Pinnekamp J., Friedrich H. (2004): Membrane Technology for Wastewater Treatment. Ministry for Environment and Nature Conservation, Agriculture and Consumer Protection of the federal state North Rhine Westphalia, ISBN 978-3-939377-01-6

Prieske H., Tacke D., Baumgarten S., Pinnekamp J., Drews A., Kraume M. (2006). Aeration of a Membrane Bio Reactor in an Airlift loop configuration. 5th European Meeting on Chemical Industry and environment (EMChIE 2006), Vienna, Austria, 03.05.-05.05.2006

Tacke D. (2005a). Untersuchung der Strömungsverhältnisse in getauchten Modulsystemen als Werkzeug bei der Modulentwicklung (Investigation of Velocity Flow Pattern in Submerged Membrane Bioreactor Systems as a Tool for Module Developement). IWW-Kolloquium, Mülheim, 12.05.2005

Tacke D. (2005b). Untersuchungen zur Moduldurch- und Modulumströmung als Werkzeug bei der Modulentwicklung. Membrantechnik (Investigation of Velocity Flow Pattern as a Tool for Module Developement). Tagungsband 6. Aachener Tagung: Membrantechnik in der Wasseraufbereitung und Abwasserbehandlung, T. Melin, J. Pinnekamp. Aachen. P9-1 – P9-12, ISBN 3-86130-775-8

Tacke D., Pinnekamp J., Prieske H., Kraume M. (2006). Improvement of Membrane Bioreactor Aeration. Environmental Science and Technology 2006, American Science Press, United States of America, ISBN 0-9768853-6-0

Ueda T., Hata K., Kikuoka Y., Seino O., (1997). Effects of aeration on suction pressure in a submerged membrane bioreactor, Water Research. Vol. 55, Issue. 3, pp. 489-494.

Vera L., Delgado S., Elmaleh S. (2000). Dimensionless number for the steady-state flux of cross-flow microfiltration and ultrafiltration with gas sparging. Chemical Engineering Science, Vol. 55, pp. 3419-3428.

Hydrodynamic CFD simulation of a two-phase flow in a single tube of an ultrafiltration membrane for a side-stream membrane bioreactor

N. Rios*, I. Nopens* and P. Vanrolleghem*

*BIOMATH, Department of Applied Mathematics, Biometrics and Process Control. Coupure Links 653, B-9000, Ghent, Belgium
(E-mail: nriosrat@biomath.ugent.be)

Abstract Membrane fouling is the main drawback of full-scale application of membrane bioreactors (MBRs) that have restricted its commercialization due to reduction of productivity, increased maintenance and operational cost. Literature has shown that the hydrodynamics near the membrane surface play an important role on fouling. This work aims at implementing a CFD model that describes a two phase (sludge + air) flow. Moreover, shear stress distributions near the membrane surface were determined for different air velocities. This allows for better quantification of shear and coupling with filtration models.

Keywords:
Computational fluid dynamics – Ultrafiltration – Fouling – Side-stream membrane bioreactor – Modelling

INTRODUCTION

Membrane bioreactors (MBRs) combine biological treatment with membrane separation technology. There are two types of membrane bioreactors, one has the membrane inside the bioreactor, commonly called immersed membrane bioreactor (iMBR); and the other has the membrane in series with the bioreactor, named side-stream membrane bioreactor (sMBR). This work focuses on the last type: the sMBR configuration, that uses a tubular ultrafiltration membrane (X-FLOW) manufactured by Norit (The Netherlands). A common problem encountered with MBR's is the fouling of the membrane resulting in frequent cleaning and even membrane replacement that increases the operational costs considerably. The membrane fouling happens due to three principal factors (Mulder 1998): 1) Membrane material, 2) Feed water quality: the bulk organic type and concentration, the solution conditions (pH, ionic strength), microorganisms, inorganic and organic substances (EPS); 3) Operating conditions: transmembrane pressure (TMP), cross-flow velocity and temperature. In terms of operation, three aspects are considered to be important with regard to the fouling of the membrane: particle size distribution (PSD), biological parameters and process hydrodynamics (figure 1). Due to the complexity of the MBR process, the modelling exercise is broken down in the development of several sub-models. For the hydrodynamic process it is important to account for the interaction between the biology and the membrane. Therefore, a model is being developed by means of computational fluid dynamics (CFD) using Fluent© (Ansys Inc.). The CFD model should consider several factors, among which: the geometry of the membrane, the boundary conditions (pressure, cross flow velocity and gas flow rate), the feed concentration and physical properties of the sludge like density, surface tension and viscosity.

To reduce the fouling on the membrane air is often introduced in the sludge flow to create a gas-liquid two-phase cross-flow (Cui et al. 2003). This is done for the following reasons: 1) increase the permeate flux; 2) improve membrane rejection characteristics (reduction of fouling) due to the early transition from laminar to turbulent flow (Ghosh and Cui 1999), 3) increase the surface shear stress to remove foulants that are already attached and 4) increase the mass transfer between the cake layer and the bulk region. In literature it has been observed that the slug flow pattern is the most effective as well as the least energy consuming process in ultrafiltration membranes (Bellara et al. 1996; Cui et al. 1997; Smith and Cui 2004). Other studies suggest to operate the system in laminar rather than in turbulent regime to decrease the energy consumption (Ndinisa et al. 2005).

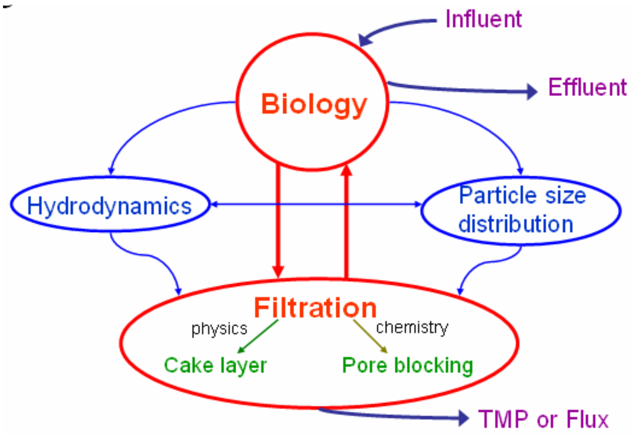

Figure 1 Proposed integral mechanistic model for MBR systems

Numerical models have been developed using the Volume of Fluid (VOF) approach to track down the gas-liquid interface in the slug flow. Because the tube is usually in a vertical position it is possible to simulate it in 2D and assume it to be axisymmetric. Using CFX, which is a commercial CFD software with the VOF approach, Ndinisa et al. (2005) used a laminar flow model and found that the VOF model yields good results for the velocity profile in the bubble but it does not work properly in the wake region due to the eddy formation. Therefore, they complement the VOF with a two fluid Eulerian model to overcome the problem. Also it is recommended to select a fine grid (0.25 mm) to capture the velocity profile with high accuracy. Other researches used Fluent with the VOF model and the geometric reconstruction scheme to capture the shape of the bubble and the shear stress in the surface of the membrane. They use the RNG k-ε turbulent model and link the shear stress with the mass transfer coefficient to determine the permeate flux (Smith et al. 2005; Taha and Cui 2002; Taha et al. 2006; Taha and Cui 2006a; Taha and Cui 2006b). It is important to mention that these models only apply to one single bubble and not for the whole length of the tube. Therefore, it is not considered that in a tube containing several Taylor bubbles relative acceleration and hence coalescence occurs. The objective of this paper is to simulate a single membrane tube of 1 m of length and 5.2 mm of internal diameter and observe the behaviour of the developed Taylor bubbles as well as the shear stress experienced near the membrane surface.

MATERIALS AND METHODS

Hydrodynamic model

This work focuses on a gas-liquid flow (air + sludge). It is important to consider five characteristics of the flow before developing a CFD multiphase model: 1) The flow pattern: there are four specific flows: bubbly, slug, churn and annular flow; 2) the hold-up of each phase and their relative velocity; 3) physical properties of the phases; 4) pressure drop along the tube and 5) flow rates (Coulson and Richardson 2002).

Activated sludge exhibits a non-Newtonian fluid behaviour (Hasar et al. 2004; Mori et al. 2006; Pollice et al. 2007; Rosenberger et al. 2006; Seyssiecq et al. 2003). Therefore, a Herschel-Bulkley modified using Papanastasiou’s adaptation model (equation (1)) is used to model the apparent viscosity. For inclusion in a CFD code and to enable the description of the rheological behaviour at different total suspended solids (TSS) concentrations the parameters of the modified Herschel-Bulkley model have been expressed as a function of TSS (equation (2)) (Rios et al. 2007).

$$\tau = \tau_0\left(1 - e^{-m\dot{\gamma}}\right) + k\dot{\gamma}^n$$

(1)

$$\tau = \left[0.00686 \cdot \mathrm{TSS}^{2.08413}\right]\left(1 - e^{-0.03355 \cdot \dot{\gamma}}\right) + \left[0.00001 \cdot \mathrm{TSS} + 0.00028\right]\dot{\gamma}^{1.46224} \quad (2)$$

Where n is the flow behaviour index, k is the fluid consistency index, τ is the shear stress (Pa) and $\dot{\gamma}$ is the shear rate (s^{-1}). The density of the sludge (ρ_L) is defined by (Jiang 2007):

$$\rho_L = 1000 + 0.2\,TSS \quad (3)$$

Because the sludge is a non-Newtonian fluid it is necessary to modify the Reynolds number for flow of liquids in pipes using the Metzner and Reed Reynolds number (Re_{MR}) (Coulson and Richardson 2002):

$$\mathrm{Re}_{MR} = 8\left(\frac{n}{6n+2}\right)^n \frac{\rho_L u_{SL}^{2-n} d^n}{k} = 358 \quad (4)$$

Where d is the internal diameter of the tubular membrane and u_{SL} is the superficial liquid velocity. The value of the Reynolds number in this case (358) clearly shows that the flow is laminar. Therefore, the CFD simulation is carried out using the laminar model. The physical properties of the sludge were calculated at a TSS of 10 g/l, being the average value of the loop just before entering the membrane. A summary of the parameters of the lab-scale MBR used at Biomath is presented in table 1. The air properties are obtained from tables at 15°C and the surface tension value for the sludge-air mixture is similar to that of water-air.

Table 1 Lab-scale MBR parameters at 15°C

Parameter name		Units	Value
Pipe diameter	d	m	0.0052
Superficial liquid velocity	u_{SL}	m/s	0.5
Superficial gas velocity	u_{SG}	m/s	0.5
Liquid density	ρ_L	kg/m^3	1002
Gas density	ρ_G	kg/m^3	1.22845
Liquid viscosity	μ_L	Pa s	0.0088
Gas viscosity	μ_G	Pa s	1.76291x10^{-5}
Surface tension	σ	N/m	0.07342
Flow behaviour index	n	-	1.46224
Fluid consistency index	k	-	0.00038

Unfortunately, the membrane tubes of the lab-scale sMBR are not transparent. Therefore, it is not possible to visually determine the flow pattern appearing inside the tubes. Also velocity profiles, typically determined using optical methods such as PIV (Particle Image Velocimetry) or LDV (Laser Doppler Velocimetry) cannot be used to measure the velocity profile across the membrane.

Based on that, it is necessary to determine the flow patterns by means of the velocity ratio (r), which is the relation between the superficial gas and liquid velocity (Vera et al. 2000):

$$r = \frac{u_{SG}}{u_{SG} + u_{SL}}$$
(5)

Table 2 Flow patterns depending of the velocity ratio (Vera et al. 2000)

Flow pattern	r
Bubbly flow	$0 < r \leq 0.25$
Slug flow	$0.25 < r \leq 0.9$
Churn flow	$0.9 < r < 1$
Annular flow	$r \approx 1$

In this study, both gas and liquid velocities were 0.5 m/s, hence, the velocity ratio equals 0.5, and based on table 2, it can be assumed that the flow pattern is slug flow. However, it should be noted that this analysis is rather simple and does not take into account different operational characteristics, such as inclination of the membrane, density, surface tension and viscosity of the fluids. Therefore, a rigorous analysis needs to be made, e.g. following the scheme for flow pattern determination proposed by Crowe (2005). The latter confirmed the appearance of a slug flow (result not shown).

Model geometry

The membrane module in the Biomath MBR contains twelve membrane tubes made of polyvinylidene fluoride (PVDF). It has a length of 1 m and an internal diameter of 5.2 mm. A two-dimensional axisymmetic geometry was built in Gambit® v2.2. To properly capture the shape of the bubble it is necessary to have a fine grid and an even fined grid close to the wall. The latter is required to properly describe the shear stress close to the membrane. A growth function was used to build the grid with these properties. The grid cells close to the wall have a size of 10 µm. A grid of 26 x 2960 was constructed (Figure 2).

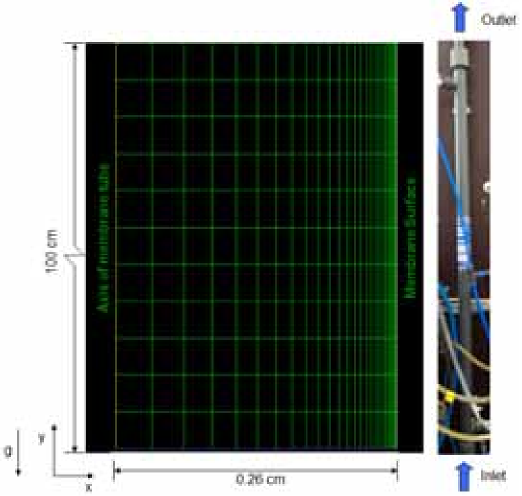

Figure 2 Membrane mesh

CFD model

A two-phase (sludge + air) flow model was implemented in Fluent® 6.3 using the Eulerian model which is a multi-fluid approach. This implies that each phase has a set of equations that needs to be solved and the phases are linked together by their respective volume fractions. This approach is known as the Euler-Euler approach. The VOF has been developed for immiscible fluids where the position of the interface between the fluids is of utmost importance. There is a single set of momentum equations for each fluid and a volume fraction for each phase. This model is used for slug flow, stratified flow and free-surface flows. The governing equations for the two fluids are defined in tensor notation as follows (Taha and Cui 2002):

Continuity equation (mass conservation)

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \vec{v}) = 0 \tag{6}$$

Momentum equation for VOF (Navier-Stokes)

$$\frac{\partial}{\partial t}(\rho \vec{v}) + \nabla \cdot (\rho \vec{v} \vec{v}) = -\nabla p + \nabla \cdot \left[\mu\left(\nabla \vec{v} + \nabla \vec{v}^{T}\right)\right] + \rho \vec{g} + \vec{F} \tag{7}$$

Volume fraction equation VOF (interface liquid-gas)

$$\frac{\partial \alpha_G}{\partial t} + \vec{v} \cdot \nabla \alpha_G = 0 \tag{8}$$

$$\alpha_G + \alpha_L = 1 \tag{9}$$

Surface tension (continuum surface tension model CSF)

$$p = \sigma \kappa \tag{10}$$

Where ρ is the density of the fluid, v is the velocity vector, μ is the fluid viscosity, g is the acceleration due to gravity, p is the pressure, κ is the surface curvature, σ is the surface tension and α_G and α_L are the volume fraction of gas and liquid respectively.

The selected solver uses the explicit interpolation scheme with geometric reconstruct discretization for VOF which is recommended for transient unsteady simulations. The geometry reconstruct discretization is a scheme for surface tracking (bubble interface). To solve the momentum transport equation the 2nd order upwind scheme is used which decreases the numerical diffusion but at the same time reduces the stability of the solution compared to cases using the 1st order upwind scheme. For pressure-velocity coupling the PISO (pressure implicit splitting of operators) scheme is used for faster convergence (Taha and Cui 2004).

For non-Newtonian fluids in a laminar flow inside a pipe, the shear stress can be computed by the Metzner and Reed correlation between the friction factor and the Reynolds number (Coulson and Richardson 1999):

$$\tau = \frac{8 \rho_L u_{SL}^2}{\mathrm{Re}_{MR}} \tag{11}$$

For this study, a value of 5.6 Pa was found.

RESULTS

Preliminary simulations were performed using constant physical properties both for air and sludge. Figure 3 shows the contours of the volume of air for the inlet and outlet of the membrane in the case of 0.5 m/s of air. In the inlet (approximately 5 cm of length) it can be observed that there is a mixture between air and sludge because both phases are entering together with the same volumetric fraction as was mentioned previously (same velocity). In the outlet one can observe that the bubbles have the specific Taylor bubble shape (bullet shape) with a round nose and a cylindrical body. Also, it is possible to observe that the bubbles have different sizes. This is due to the fact that bubbles accelerate and coalesce with a preceding bubble. This phenomenon occurs because the wake generates acceleration on the previous bubble. If the wake would be larger (i.e. with more space between consecutive bubbles) this acceleration could be avoided and all bubbles will move with a constant ascension velocity and have a similar size (Kawaji et al 1997).

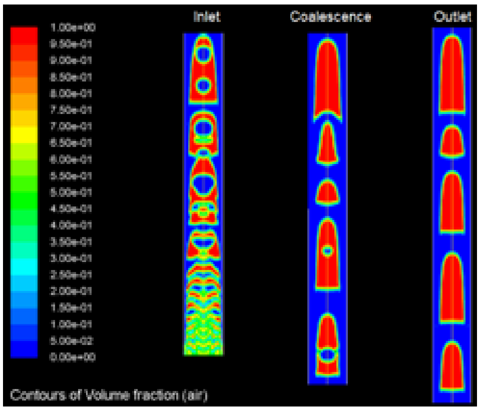

Figure 3 Contours of the volume fraction of air for a velocity of 0.5 m/s for sludge and air

Simulations were also carried out by maintaining the sludge velocity constant (0.5 m/s) and varying the air velocity from 0 to 1 m/s. The contours of the volume fraction of air at the outlet and the shear stresses near the membrane surface are presented in figure 4. It can be observed that the Taylor bubbles that develop (when $\mu_G > 0\ m/s$) differ in shape and length. At 1 m/s the slug flow starts to transform to a churn flow (although $r = 0.33$ which is well below 0.9; see table 2).

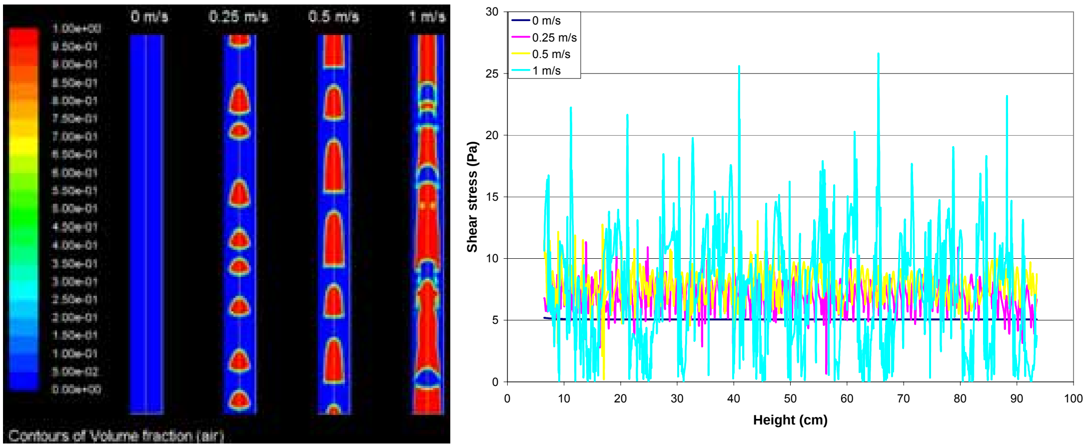

Figure 4 Contours of the volume fraction of air at the outlet (left) and profiles of shear stress near the membrane surface (right) for different air velocities (0, 0.25, 0.5 and 1 m/s) and a constant sludge velocity of 0.5 m/s

It is also possible to observe that the shear rate oscillates due to the continuous passing of bubbles close to the membrane surface. This means that at a certain location of the membrane, a variety of shear stresses occurs at a certain frequency. In order to interpret this in a better way a histogram, which shows the frequency of the occurrence of a certain shear stress at any place of the membrane (figure 5), can be used. The histograms show two things: 1) the range of shear stresses the membrane surface is expressed to and 2) the frequency or the time of this exposure.

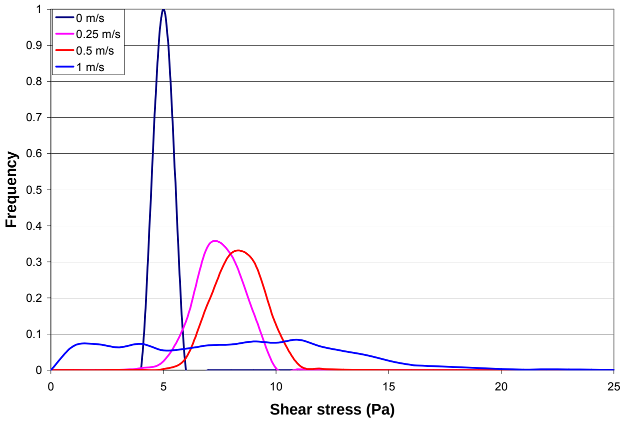

Figure 5 Histogram of the shear stress for different air velocities

It is possible to observe that when the air velocity is 0 m/s, the value of the shear stress is close to 5 Pa, which is close to the value predicted by equation (11). When the air velocity increases to 0.25 m/s, the shape of the histogram changes (wider) and shifts to higher shear stresses (around 7 Pa). At an air velocity of 0.5 m/s, the shape does not alter too much, but an additional shift to higher stresses occurs (around 8 Pa). At an air velocity of 1 m/s, the histogram changes drastically as it is smeared out over the entire shear stress range. This means that having a slug flow increases the mass transfer coefficient from the membrane surface to the bulk region. In a next phase, these distributions should be linked with a filtration model in order to quantitatively investigate its impact on the fouling process.

CONCLUSIONS

A model of the two-phase flow (sludge + air) inside a single membrane tube was implemented in Fluent® by means of the Volume of Fluid (VOF) approach. An axisymmetric geometry in 2-D for a single membrane tube with a fine mesh to capture the shape of the bubbles and the shear stress near the membrane surface was used. Preliminary simulations using constant physical properties of the sludge were performed and resulted in a slug flow. Simulation with varying air velocities (0 to 1 m/s) were performed and the shear stress the membrane surface is exposed to was quantified and presented in terms of histograms. It was found that both the shape and the shear stress range changed significantly with increasing air velocities.

ACKNOWLEDGMENTS

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REFERENCES

Bellara, S. R., Cui, Z. F., and Pepper, D. S. (1996). Gas sparging to enhance permeate flux in ultrafiltration using hollow fibre membranes. *Journal of Membrane Science*, 121(2), 175-184.

Coulson, J. M., and Richardson, J. F. (1999). Chemical Engineering. Butterworth - Heinemann. UK.

Coulson, J. M., and Richardson, J. F. (2002). Chemical Engineering. Butterworth - Heinemann. UK

Crowe, C. (2005). Multiphase Flow Handbook. CRC. USA.

Cui, Z. F., Bellara, S. R., and Homewood, P. (1997). Airlift crossflow membrane filtration - a feasibility study with dextran ultrafiltration. *Journal of Membrane Science*, 128(1), 83-91.

Cui, Z. F., Chang, S., and Fane, A. G. (2003). The use of gas bubbling to enhance membrane processes. *Journal of Membrane Science*, 221(1-2), 1-35.

Ghosh, R., and Cui, Z. F. (1999). Mass transfer in gas-sparged ultrafiltration: Upward slug flow in tubular membranes. *Journal of Membrane Science*, 162(1-2), 91-102.

Hasar, H., Kinaci, C., Unlu, A., Togrul, H., and Ipek, U. (2004). Rheological properties of activated sludge in a sMBR. *Biochemical Engineering Journal*, 20(1), 1-6.

Jiang, T. (2007). Characterization and Modelling of Soluble Microbial Products in Membrane Bioreactors., PhD thesis, Biomath, Ghent University, Belgium.

Mori, M., Seyssiecq, I., and Roche, N. (2006). Rheological measurements of sewage sludge for various solids concentrations and geometry. *Process Biochemistry*, 41(7), 1656-1662.

Mulder, M. (1998). Basic Principles of Membrane Technology. Springer. Germany.

Ndinisa, N. V., Wiley, D. E., and Fletcher, D. F. (2005). Computational fluid dynamics simulations of Taylor bubbles in tubular membranes model validation and application to laminar flow systems. *Chemical Engineering Research and Design*, 83(1 A), 40-49.

Pollice, A., Giordano, C., Laera, G., Saturno, D., and Mininni, G. (2007). Rheology of sludge in a complete retention membrane bioreactor. *Environmental Technolgy*, 27, 723-732.

Rios, N., Nopens, I., Jiang, T., De Schepper, V., Jiang, T., Verstraete, W., and Vanrolleghem, P. A. (2007). A rheological model for activated sludge in a Side-Stream MBR." *Proceedings for the international conference on membranes for water and wastewater treatment, May 15 - 17, 2007, Harrogate, UK.*

Rosenberger, R., Kubin, K., and Kraume, M. (2006). Rheology of activated sludge in membrane bioreactors. *Engineering in Life Sciences*, 2(9), 269-275.

Seyssiecq, I., Ferrasse, J. H., and Roche, N. (2003). State-of-the-art: Rheological characterisation of wastewater treatment sludge. *Biochemical Engineering Journal*, 16(1), 41-56.

Smith, S. R., and Cui, Z. F. (2004). Gas-slug enhanced hollow fibre ultrafiltration - An experimental study. *Journal of Membrane Science*, 242(1-2), 117-128.

Smith, S. R., Taha, T., and Cui, Z. F. (2005). Using an improved 1D boundary layer model with CFD for flux prediction in gas-sparged tubular membrane ultrafiltration. *Water Science and Technology*, 51(6-7), 69-76.

Taha, T., and Cui, Z. F. (2004). Hydrodynamics of slug flow inside capillaries. *Chemical Engineering Science*, 59(6), 1181-1190.

Taha, T., Cheong, W. L., Field, R. W., and Cui, Z. F. (2006). Gas-sparged ultrafiltration using horizontal and inclined tubular membranes-A CFD study. *Journal of Membrane Science*, 279(1-2), 487-494.

Taha, T., and Cui, Z. F. (2002). CFD modelling of gas-sparged ultrafiltration in tubular membranes. *Journal of Membrane Science*, 210(1), 13-27.

Taha, T., and Cui, Z. F. (2006a). CFD modelling of slug flow in vertical tubes. *Chemical Engineering Science*, 61(2), 676-687.

Taha, T., and Cui, Z. F. (2006b). CFD modelling of slug flow inside square capillaries. *Chemical Engineering Science*, 61(2), 665-675.

Vera, L., Villarroel, R., Delgado, S., and Elmaleh, S. (2000). Enhancing microfiltration through an inorganic tubular membrane by gas sparging. *Journal of Membrane Science*, 165(1), 47-57.

Evaluation of Membrane Bioreactor Performance via Residence Time Distribution Analysis: Effects of Membrane Configuration

M. W. D. Brannock*, Benjamin Küchle**, Y. Wang*, G. Leslie*

* UNESCO Centre for Membrane Science and Technology, University of New South Wales, Sydney, 2052, Australia
(E-mail: m.brannock@unsw.edu.au, y.wang@unsw.edu.au, g.leslie@unsw.edu.au)
** Institut für Energie und Umweltverfahrenstechnik/Wassertechnik, Universität Duisburg-Essen, Duisburg, 47048, Germany
(E-mail: benjamin.kuechle@uni-duisburg-essen.de)

Abstract The membrane bioreactor (MBR) represents the “state of the art” for the treatment of municipal wastewater. The design and optimisation of MBR units requires knowledge of the biokinetics, fouling potential and mixing. Although the mixing within an MBR system is of critical importance to the performance, MBRs are mainly designed on the basis of biokinetics and fouling potential of the treatment system but not on the hydrodynamics. Consequently, the mixing within the MBR process has been an insufficiently understood aspect of MBR design. One method to characterise the mixing is based on the concept of residence time distribution (RTD). In this work, tracer studies using lithium chloride were performed to acquire RTD profiles of two full-scale MBR systems with different membrane (flat sheet and hollow fibre) and bioreactor configurations (see Figure 1 & Table 1). The authors are not aware of any other published tracer studies results of full-scale MBRs. Analysis of the RTD profiles indicated that the hollow fibre MBR was closer to completely mixed conditions due to a higher mixing power to volume ratio. The flat sheet membrane filtration vessels require a larger volume due to the larger size of modules while the bioreactor of the flat sheet MBR required a larger volume due to the design (simultaneous nitrification denitrification). The mixing energy per volume of permeate used by the hollow fibre membrane vessel was lower than that of flat sheet module MBR (see Table 5). Mixing is a key design consideration for wastewater processes, therefore the MBR RTDs obtained will provide an important tool for the validation of the hydrodynamic models of MBRs for use in design. Currently, many designers estimate the RTD using compartmental (network-of-reactors) modelling while making assumptions on the flow regime in each reactor. Compartmental models of each MBR examined were developed and presented in this paper for comparison against the tracer study results. Compartmental modelling is unable to predict energy input requirements and, as highlighted in this paper, does not always predict the MBR RTD successfully. This emphasises the need for more fundamental modelling of MBR hydrodynamics; of which computational fluid dynamics modelling (CFD) provides.

Keywords
RTD, residence time distribution, MBR, membrane bioreactor, CFD, computational fluid dynamics

INTRODUCTION

The design and optimisation of MBR units requires knowledge of the biokinetics, fouling potential and mixing. Although the mixing within an MBR system is of critical importance to the performance of the system, MBRs are mainly designed on the basis of the biokinetics and fouling potential of the treatment system but not on the hydrodynamics. Consequently, the mixing within the MBR process has been an insufficiently understood aspect of MBR design.

The performance of a reactor is largely influenced by the retention time of a reactant in the reactor vessel (Young, 1991). The retention time of the reactant is determined by the mixing in the reactor (Pena *et al.*, 2006). Many researchers have noted the importance of mixing in achieving efficient conversion of reactants (Levenspiel, 1999; Pena *et al.*, 2006; Potier *et al.*, 2005). One method to characterise the mixing is based on the concept of residence time distributions (RTD). The degree of mixing energy input and reactor/membrane configuration affects the output response (or RTD) which describes the type of mixing occurring in the system.

In this work, tracer studies using lithium chloride were performed to acquire RTD profiles of two large scale MBR systems with different membrane (flat sheet and hollow fibre) and bioreactor configurations (see Figure 1). As far as the authors are aware, prior to this paper, the results of tracer studies of full-scale MBRs have not been published in literature; although Wang *et al.* (2007) recently presented tracer study results of two MBR pilot plants. Analysis of the RTD profiles indicated that the hollow fibre MBR was closer to completely mixed conditions due to the higher power to volume ratio. The flat sheet membrane filtration vessels requires a larger volume due to the larger size of modules while the bioreactor of the flat sheet MBR also required a larger volume due to the design (simultaneous nitrification denitrification). The energy (per volume of permeate) required for the mixing of the hollow fibre membrane vessel was lower than that of flat sheet module MBR (see Table 5). Consequently, hollow fibre module was more energy efficient than flat sheet module.

Mixing is a key design consideration for wastewater processes, therefore the MBR RTDs obtained will provide an important tool for the validation of the hydrodynamic models of MBRs for use in design. Currently, many designers estimate the RTD using compartmental (network of reactors) modelling while making assumptions on the flow regime in each reactor. Consequently, due to their wide-spread use, compartmental models of each MBR examined were developed and presented in this paper for comparison against the tracer study results. Compartmental modelling is unable to predict energy input requirements and, as highlighted in this paper, does not always approximate the MBR RTD successfully. This emphasises the need for more complex and fundamental modelling of MBR hydrodynamics; both of which computational fluid dynamics modelling (CFD) provides (Olivet *et al.*, 2005; Potier *et al.*, 2005).

METHODS

Site Description

Two full-scale MBRs located in Australia were examined for this work, one having hollow fibre membranes and the other having flat sheet membranes. Site 1 is a flat sheet (FS) membrane MBR. It is the primary STP for the local township and provides recycled water for the surrounding region. The plant, which consists to process streams in parallel, is sized to treat an Average Dry Weather Flow of 3.4 ML/d (1.7 ML/d each stream) and is designed for nutrient removal via simultaneous nitrification/denitrification (SND). See Figure 1 for a diagram of the process stream examined; the Site 1 (FS) MBR is comprised one of the two parallel process streams. Tables 2 and 3 give an overview of the biological parameters during the tracer study. Site 2 is a hollow fibre (HF) membrane MBR and operates at large sewage treatment plant (STP). It receives primary treated sewage from the STP and produces recycled water for the site and local area. The plant is sized to treat 2ML/d of influent and is designed for nutrient removal so it possesses an anoxic zone, aerobic zone and an internal recycle (see Figure 1).

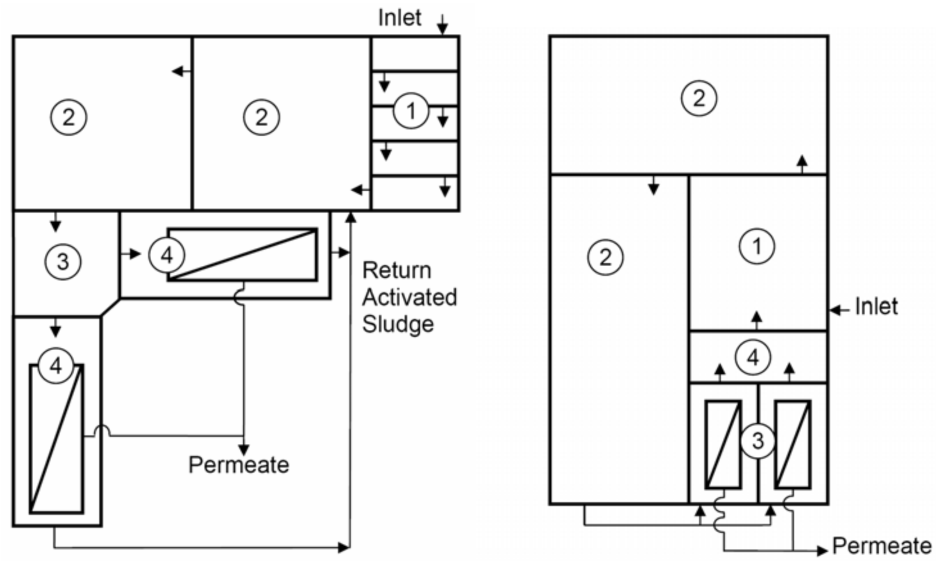

Figure 1 Overview of the process setup: Site 1 (FS) MBR (*left*) – 1. Bioselector, 2. Swing aerobic & anoxic zones, 3. Aerobic zone, 4. Membrane filtration vessel; Site 2 (HF) MBR (*right*) – 1. Anoxic zone, 2. Aerobic zones, 3. Membrane filtration zone, 4. De-aeration zone.

Table 1 Operating process parameters of the two MBRs during each trial.

Parameters	Units	Site 1 (FS) MBR	Site 2 (HF) MBR
Average Influent Flowrate	Ml/d	1.1	1.1
Total Volume of Bioreactor Vessels	m^3	852	435
Total Volume of Membrane Filtration Vessels	m^3	392	36
MLSS	g/L	11.3	5.0
Membrane Type	-	Flat sheet	Hollow fibre
Net Membrane Flux	$L/m^2/hr$	11.8	29.0
Mixed Liquor Return Flowrate	m^3/hr	461	433
Sludge Age	days	16.6	9.9
Air Flowrate into Bioreactor	Nm^3/hr	109	419
Air Flowrate into Membrane Filtration Vessel	Nm^3/hr	992	918

Table 2 Average feed characteristics during each trial.

Site	COD	BOD_5	NH_3-N	SS	TDS	Alkalinity	Temp	pH
	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L $CaCO_3$	^{o}C	-
Site 1 (FS) MBR	608	260	55.0	284	1175	384	24.4	7.3
Site 2 (HF) MBR	482	200	33.0	325	749	247	22.1	6.9

Table 3 Effluent characteristics during each trial.

Site	COD	BOD_5	NH_3-N	NO_X-N	SS	TDS	Alkalinity	Temp	pH
	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L $CaCO_3$	^{o}C	-
Site 1 (FS) MBR	48.8	2.0	0.7	1.5	1.5	960	112	25.6	7.5
Site 2 (HF) MBR	29.0	4.5	0.1	16.1	1.0	786	78	23.3	7.2

Experimental Procedure

The tracer studies were carried out using a pulse input of lithium chloride delivered at the MBR inlet (post-screening) with the tracer response being measured in the permeate and other relevant sample points. Lithium chloride is commonly used for tracer studies of wastewater processes due to its inert nature (Pena *et al.*; 2006, Potier *et al.*, 2005; Smith *et al.*, 1996). The amount of tracer used corresponded to a bulk concentration of 1.5 mg Li^+/L (i.e. mass of lithium is divided by volume of the MBR). This ensured that the tracer response is much greater than the detection limit of theanalysis technique. The Li^+ concentration was measured using ICP-AES (Inductively Couple Plasma – Atomic Emmission Spectrophotometry) and had a detection limit of 0.008mg Li^+/L

Dosing solutions were prepared with concentrations of 40-60g Li^+/L and a maximum dosage volume of 25L; this ensured a small dosage volume, low dosage time yet at a small density difference between the dosage solution and mixed liquor. The dosage solution was pumped in at approximately 75L/min over 20 seconds into the inlet stream. For Site 2 (HF) MBR this is 0.02% of the HRT and for Site 1 (FS) MBR is 0.004% of the HRT. This enabled effectively instantaneous delivery of the tracer. To obtain reproducible results at both sites, the tracer studies were undertaken with as many constant process parameters possible. The intermittent influent flow and the switching on/off of the aeration were still experienced. The tracer studies commenced at exactly the same time of the day. Sampling was undertaken for four hydraulic residence times ensuring close to 100% tracer recovery.

Modelling

Compartmental modelling has been used for comparison against the experimentally determined RTDs. Compartmental modelling, as described by Levenspiel (1999) utilises a network of the idealised flow extremes of plug flow and completely mixed flow arranged in different configurations. This method of approximating the mixing is used by process simulation tools such as BioWIN (COST, 2002). The common approach is to assume that each individual vessel is assumed to be completely mixed. In this work, completely mixed vessels with the same volume and inlet/outlet flowrates as the real system were utilised; similar to the approached used by process simulation tools (COST, 2002).

RESULTS AND DISCUSSION

Residence Time Distribution

The tracer study methodology employed obtained reproducible results (see normalised RTD curves shown in Figure 2), and almost complete recovery of tracer (Table 4). Both MBRs appear to have an overall mixing behaviour close to complete mixing, although the Site 1 (FS) MBR appears to be not as close. It also shows bulges in the RTD curve which are due to the diurnal nature of the influent; the bulge occurs every 24hours while the permeate flowrate is at its lowest. This is due to average flow used in RTD derivation, allowing RTD comparison. It does however produce artefacts in the RTD curve with highly variable flows; Site 1 (FS) MBR perrmeate flow ranged 20-180% of the mean, while Site 2 (HF) MBR permeate flow ranged 60-140% of the mean

Burrows *et al.* (1999) and Thirumurthi (1969) have used various relationships between the hydraulic residence time (T), peak time (t_p) and the mean residence time (t_m) to quantitatively assess the RTD curves. These include the plug flow index (t_p/T), "dead" zone index (t_m/T) and the short-circuiting index ($1-t_p/t_m$). These indicate that the Site 1 (FS) MBR has a lower volume of dead zones (although this is sensitive to errors in flowrate and volume measurement) and has a marginally lower amount of short circuiting. The Site 2 (HF) MBR however is shown to be closer to completely mixed; due to the greater mixing energy input per unit volume (see Table 5). Smith *et al*

(1993) has noted that these indices are not as accurate measure of mixing characteristics as the dispersion number (or inverse of the Peclet number, Pe_r). The Peclet numbers indicate that both MBRs have “high degree of dispersion”, according to Levenspiel’s (1999) criteria, and therefore deviate greatly from plug flow. The Peclet number and number of completely mixed tanks in series (*n*) confirm that the Site 2 (HF) MBR is closer to completely mixed conditions.

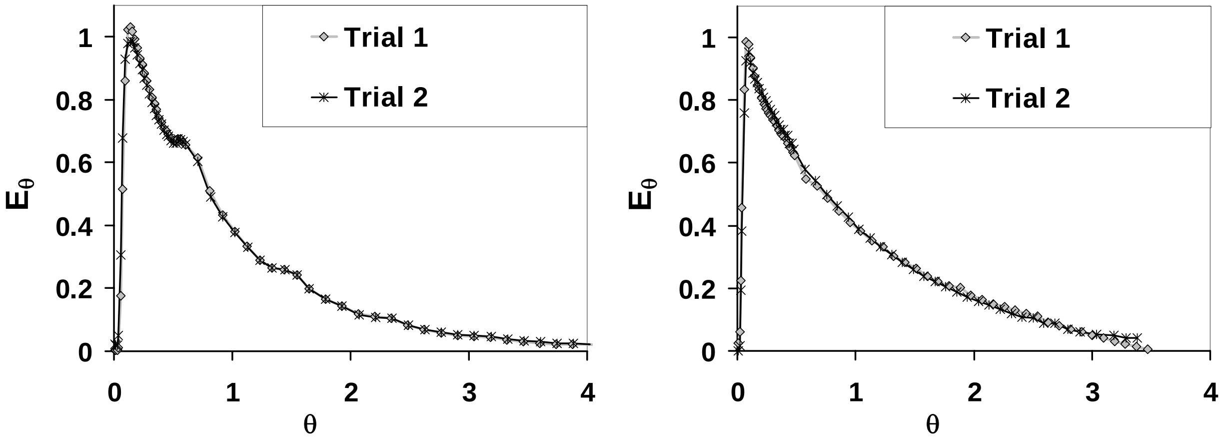

Figure 2 Dimensionless RTD – Site 1 (FS) MBR (*left*) and Site 2 (HF) MBR (*right*).

Table 4 Quantitative RTD properties

Site	Trial	Rec. [%]	T [h]	t_m [h]	σ_θ^2 [-]	s_θ^3 [-]	t_p [h]	t_p/T [-]	t_m/T [-]	$(1-t_p/t_m)$ [-]	Pe_r [-]	n [-]
Site 1 (FS) MBR	1	82.7%	10.3	8.93	0.591	0.351	0.67	0.065	0.86	0.925	1.83	1.32
	2	96.0%	10.3	9.07	0.603	0.398	0.83	0.081	0.88	0.908	1.75	1.35
Site 2 (HF) MBR	1	99.7%	27.5	28.9	0.682	0.701	3.33	0.121	1.05	0.885	1.28	1.46
	2	99.5%	27.5	28.7	0.679	0.676	3.92	0.142	1.04	0.863	1.29	1.44

The degree of mixing within the membrane filtration vessels was also assessed through measurement of tracer response at an alternative location in the tank. Any large difference between tracer response in permeate and other point would indicate a deviation from complete mixing. The largest difference between the curves has been found to be 0.4% of the hydraulic retention time as can be seen in Figure 3. This indicates that the mixing energy from aeration and recirculated sludge being sufficiently close to complete mixing.

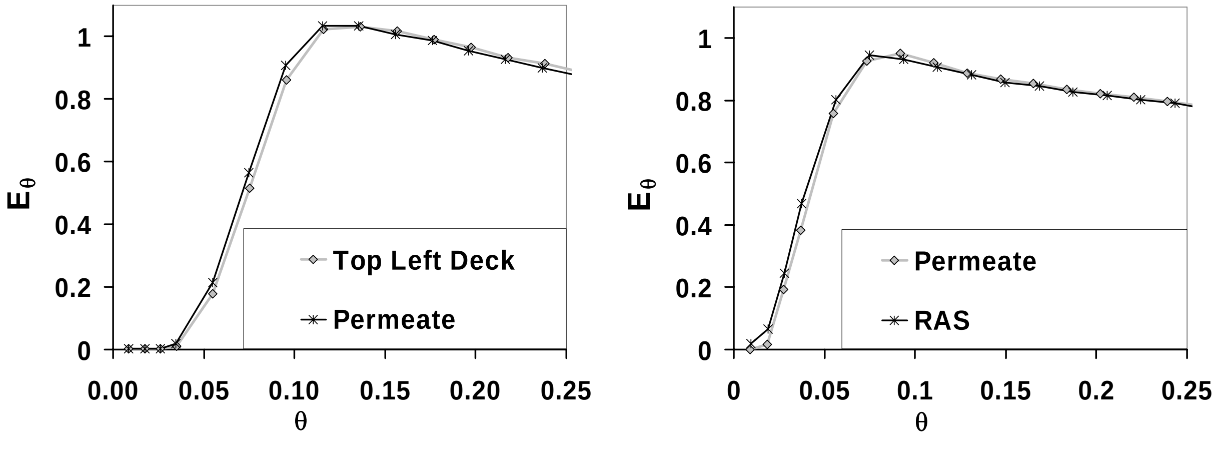

Figure 3 Comparison of tracer response– Site 1 (FS) MBR (*left*) & Site 2 (HF) MBR (*right*).

Mixing Energy

The energy usage for mixing was determined for each MBR and is presented in Table 5. The drivers of mixing are considered to be mechanical mixing, bioreactor aeration, membrane vessel aeration and recirculation pumping. The energy usage was calculated on the basis of motor runtimes and motor speeds. For both MBRs the calculated total energy usage closely matched the energy drawn from the power grid (e.g. Site 2 (HF) MBR calculated power usage was 42.9kW while the power drawn was 45.0kW).

The Site 1 (FS) MBR bioreactor required higher mechanical mixer power input; likely due to the much larger reactor volume involved. However, it did not require as much mixing induced via aeration. Both of these factors are likely due to the SND design. The total mixing power (with or without recirculation pumps) required for the bioreactor is roughly the same for each MBR even though the Site 1 (FS) MBR has a much larger bioreactor volume. The notable difference is the amount of power required for the mixing of the membrane filtration vessel where Site 1 (FS) MBR receives more than double the power input for the aeration blowers.

The total specific power input in terms of volume is much lower for Site 1 (FS) MBR, although the energy usage per volume of permeate is significantly higher (see Table 5). This may be due to the extra treatment volume required by the Site 1 (FS) MBR to handle the larger variations in flows or that SND requires larger reactor volumes due to lower overall reactions rates. It may also be due to the extra mixing energy requirements of the flat sheet membranes. The Site 1 (FS) MBR requires over twice the membrane blower energy per volume of permeate produced than the Site 2 (HF) MBR.

Wang *et al.* (2007) used another tool for comparison of energy usage is the velocity gradient (G). This not only takes into account the power input (P) and reactor volume (V), but also the viscosity of the liquid (μ) (see Equation 1 below). The viscosity of the liquid was determined using the correlation presented by Judd (2006). Depending on the design of the MBR (mixing energy distribution and vessel configuration) higher overall velocity gradient may move the reactor closer to complete mixing. The overall velocity gradient for the Site 2 (HF) MBR 1.84 times greater than for the Site 1 (FS) MBR.

$$G = \left(\frac{P}{\mu V} \right)^{0.5} \qquad \text{Equation (1)}$$

Table 5 MBR Power Input

Parameters	Units	Site 1 (FS) MBR	Site 2 (HF) MBR
Power - Mixer	kW	7.1	2.2
Power - Bioreactor Blower	kW	3.3	8.5
Power - Membrane Vessel Blower	kW	29.5	13.8
Power - Recirculation Pump	kW	16.0	18.5
Power - Total	kW	55.8	42.9
Total Specific Power Input – Volume Basis	W/m^3	44.8	91.2
Total Specific Power Input – Flowrate Basis	kWh/m^3	1.23	0.939
Memb. Blower Spec. Power Input – Volume Basis	W/m^3	75.2	235
Memb. Blower Spec. Power Input – Flowrate Basis	kWh/m^3	0.651	0.301
Velocity Gradient (G)	s^{-1}	116.0	213.5

Compartmental Modelling

The compartmental modelling undertaken comprised a network of complete mixed vessels. The compartmental modelling which provided the best agreement with experimental data had a virtual vessel assigned for each physical vessel (i.e. no grouping together of vessels). The Site 1 (FS) MBR model had 11 vessels which had an overlap between experimental and model RTD of 93.8%, whereas the next best model of 7 vessels (the vessels in the bioselecter, as shown in Figure 1, being grouped together) had an overlap of 86.3%. The Site 2 (HF) MBR model had 6 vessels for the best fit model with an overlap between RTDs of 96.7%. Figure 4 demonstrates the very good fit between compartmental model and the Site 2 (HF) MBR RTD. It also shows that compartmental modelling has difficulty predicting the position of the peak for the Site 1 (FS) MBR RTD; this is due to the complex flow in the bioreactor as complete mixing of the membrane filtration vessel is confirmed for both sites. As the compartmental modelling was performed under steady state conditions it cannot predict the effect of transient behaviour such as the diurnal flow as represented by the bulges in the RTD.

The above comparison between model and experimental RTD highlights the potential disadvantages of the *ad hoc* usage of the compartmental modelling methods which biokinetic modelling tools such as BioWIN take advantage of (COST, 2002). Specifically, the designer must make broad assumptions on the mixing regime within reactor vessels which may not always provide a realistic answer, especially for more complex flows. Fundamental hydrodynamics modelling tools such as CFD do not make these assumptions and can assist in the understanding and optimisation of processes as noted by Leclerc *et al.* (2000) and Olivet *et al.* (2005).

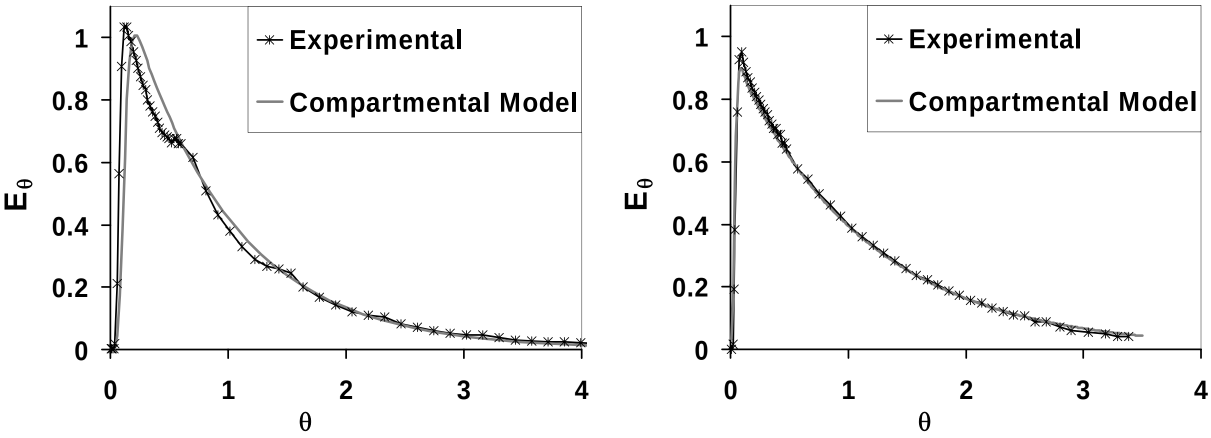

Figure 4 Comparison between tracer response RTD and the compartmental model RTD curves – Site 1 (FS) MBR (*left*) and Site 2 (HF) MBR (*right*).

CONCLUSIONS

The experimental methodology employed provided reproducible results with high recovery of tracer. The results show that different MBR designs, both with respect to the bioreactor and membrane filtration vessel, have differing effects on the RTD. The higher power input per unit volume of reactor at Site 2 (HF) MBR volume creates more completely mixed conditions. However, in terms of energy usage per unit volume of permeate, the Site 1 (FS) MBR has higher requirements. This is likely due to a number of factors, such as larger reactor volume (due to the lower reaction rates of SND and larger membrane modules) and the flat sheet panel configuration of the Site 1 (FS) MBR requiring more mixing energy. In fact, the flat sheet panels more than twice the membrane blower mixing energy per volume of permeate than the hollow fibre modules of the Site 2 (HF) MBR.

It has been shown that the deficiency of compartmental modelling is that it is unable to predict the behaviour of complex flows observed in the bioreactor vessels of the MBR. In addition to this, there is no possibility for compartmental modelling either to predict energy usage or optimise its use. This is where application of CFD to the design of MBRs would be of a great advantage. CFD is able to predict the hydrodynamics and energy usage using the fundamental equations of fluid dynamics. It does not require assumptions with regards to reactor vessel hydrodynamics to be made. The RTDs presented here will also form part of the validation of a computational fluid dynamics model of an MBR.

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REFERENCES

Burrows, L.J., Stokes, A.J., West, J.R., Forster, C.F. and Martin, A.D. (1999) Evaluation of different analytical methods for tracer studies in aeration lanes of activated sludge plants. Water Research 33(2), 367-374.

COST (2002) The COST 'simulation benchmark': description and simulator manual, p. 154, European Co-operation in the field of Scientific and Technical Research.

Judd, S. and Judd, C. (2006) The MBR book : principles and applications of membrane bioreactors in water and wastewater treatment, Elsevier, Amsterdam ; Boston ; London.

Leclerc, J.P., Claudel, S., Lintz, H.G., Potier, O. and Antoine, B. (2000) Theoretical interpretation of residence-time distribution measurements in industrial processes. Oil & Gas Science and Technology-Revue De L Institut Francais Du Petrole 55(2), 159-169.

Levenspiel, O. (1999) Chemical reaction engineering, Wiley, New York.

Olivet, D., Valls, J., Gordillo, M.A., Freixo, A. and Sanchez, A. (2005) Application of residence time distribution technique to the study of the hydrodynamic behaviour of a full-scale wastewater treatment plant plug-flow bioreactor. Journal of Chemical Technology and Biotechnology 80(4), 425-432.

Pena, M.R., Mara, D.D. and Avella, G.P. (2006) Dispersion and treatment performance analysis of an UASB reactor under different hydraulic loading rates. Water Research 40(3), 445-452.

Potier, O., Leclerc, J.P. and Pons, M.N. (2005) Influence of geometrical and operational parameters on the axial dispersion in an aerated channel reactor. Water Research 39(18), 4454-4462.

Smith, L.C., Elliot, D.J. and James, A. (1993) Characterisation of mixing patterns in an anaerobic digester by means of tracer curve analysis. Ecological Modelling 69(3-4), 267-285.

Smith, L.C., Elliot, D.J. and James, A. (1996) Mixing in upflow anaerobic filters and its influence on performance and scale-up. Water Research 30(12), 3061-3073.

Thirumurthi, D. (1969) A break-through in the tracer studies of sedimentation tanks. J. Water Pollution Control Federation 41, 405-418.

Wang, Y. (2007) Evaluation of membrane bioreactor performance via residence time distribution: effects of membrane configuration and mixing, IWA Harrogate, UK.

Young, J.C. (1991) Factors affecting the design and performance of upflow anaerobic filters. Water Science and Technology 24(8), 133-155.

Flow structures in MBR-tanks

J. Saalbach* and M. Hunze**

* FlowConcept GmbH, Vahrenwalder Str. 7, 30165 Hannover, Germany,
(E-mail: saalbach@flow-concept.de)
** FlowConcept GmbH, Vahrenwalder Str. 7, 30165 Hannover, Germany,
(E-mail: hunze@flow-concept.de)

Abstract In this paper a CFD model is presented that allows for an approximation of the flow field within MBR-tanks. The technique is demonstrated by means of two examples considering both hollow-fibre and plate-membrane modules. The flow field is computed and a comparison between measured and computed flow velocities shows the good quality of the numerical model.

Keywords CFD-modelling; flow structures; MBR-tank

INTRODUCTION

Membrane technologies are used in a variety of applications today in both industrial and municipal wastewater treatment. Through the operation of membrane bioreactors already in use in several existing plants, knowledge has been gained for the planning process of new MBR-tanks. Since CFD simulations are already used to analyse and optimize processes in traditional water treatment plants a study was conducted to show the potential of CFD in combination with MBR technologies in respect to both planning and operation. Investigations on the hydraulic conditions in a filtration tank can be used to optimize:

- - the position of membrane modules and module design,
- - the position and type of aeration elements as well as the air-loading,
- - the geometry and design of the filtration tank itself.

When using CFD to model an MBR-plant a variety of obstacles have to be tackled. Phenomena surrounding membranes take place on a micro-scale requiring high mesh resolutions which result in a large element number even for the investigation of very small areas. For modelling a whole MBR-plant a macro-scale approximation has to be used in order to keep the element number and computation effort within certain limits. Therefore, an appropriate modelling technique has to be applied to transfer phenomena from one scale to the other.

In this study the possibility of modelling membrane modules as zones of porous media is investigated. The idea is to regard a whole membrane module as one zone with macroscopically identical characteristics. This way, single membranes – hollow-fibres or sheets - do not have to be modelled individually. Effects on the flow field caused by the area normally occupied by a membrane module are transferred to a zone of porous media. The flow resistance values have to be calibrated according to the type of membranes used in the membrane module in order to produce a corresponding flow field. In this study two types of membrane modules are modelled. Hollow-fibre membranes are flexible whereas plate-membranes remain at a fixed position within a module. The latter pose a challenge because the flow field inside a plate membrane should be unaffected by the flow field around it. At the same time, it has to be permeable to allow a flux across the membrane, resulting in a significant modelling challenge.

Additionally the influence of aeration which is linked to the use of membrane technology is considered. As a result the flow field largely depends on the amount and position of the aeration elements. The effects of aeration on the flow field will be taken into account in the model. In this article the modelling procedure will be demonstrated on the basis of two systems.

METHODS

An MBR-tank can be considered as a multiphase system consisting of the interacting phases water, air, and activated sludge. For modelling work the software package Fluent (Fluent Inc., 2006) is used. It is extended by self-developed approaches. The Algebraic-Slip-Mixture-model (Manninen et al., 1996) is used for the description of the movement of water and air resulting in a two-phase-fluid with volume fractions for the corresponding phases. On the basis of this, a 3-dimensional flow field is computed.

Processes involving aeration tend to be highly turbulent thus requiring a suitable turbulence model. In this study a modification of the *k-ε* model, the *rng-k-ε* model (Orszag et al., 1996) is used. The turbulent kinetic energy *k* and the turbulent dissipation rate *ε* are used to calculate the turbulent viscosity η_t.

The activated sludge is modelled as a substance concentration in the water phase which accounts for density and viscosity impacts on the fluid as well as the settling process of the sludge. Activated sludge transport is carried out by the advection-diffusion-equation.

The membrane modules are modelled as zones of porous media. This assumption is suitable for considering the influence the modules have on the flow field, i.e. the flow resistance. Resistance values for all three spatial directions are associated with the porous zones and can be altered to match the values of the modules under investigation. Resistance values have to be determined on the basis of velocity measurements within real MBR-tanks under realistic system conditions.

RESULTS AND DISCUSSION

The MBR-tank of WWTP Sobelgra

The first system under investigation is an MBR-tank of WWTP Sobelgra (Belgium). A top view sketch is provided in figure 1. The tank has eight membrane modules (KOCH Membrane Systems) installed 2.2 m above the bottom. Around each module walls are installed thus water will mainly have a vertical flow direction. Height of the modules is 1.8 m and the water level is at 4.2 m.

The 3-dimensional computer model is provided in figure 2. The green boxes indicate zones of porous media modelling the eight hollow-fibre membrane modules. Walls surrounding the modules are depicted in black. Below the modules aeration elements are installed which are depicted in blue. At the bottom of the tank, pipes with drill holes serve as inlets. The modelled inlets can be seen depicted as little blue squares at the bottom of the tank in figure 2.

In the state which is simulated, only four of the membrane modules are operational thus a permeate flux only occurs in modules 5-8, the four modules to the right in figure 2. A permeate flux of 26 m^3/h is considered for the four modules combined. Accordingly 10% of the total air-load is used for aeration below modules 1 to 4 and 90% is used for aeration below modules 5-8. A total air-load of 29 m^3/min is considered.

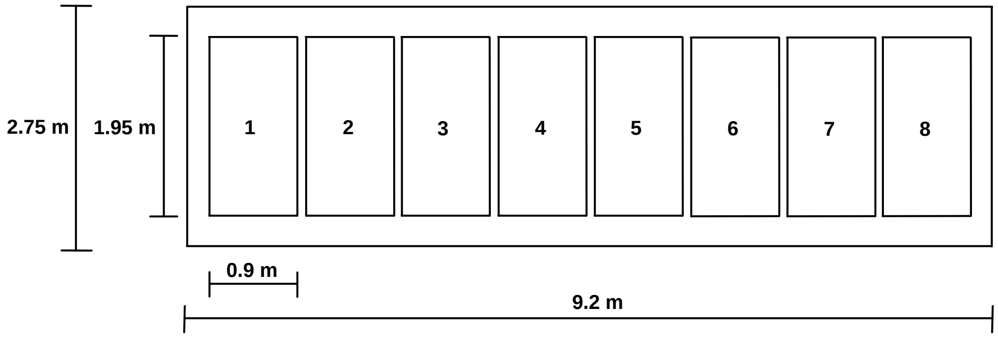


Figure 1 Top view sketch of the MBR-tank of WWTP Sobelgra (Belgium).

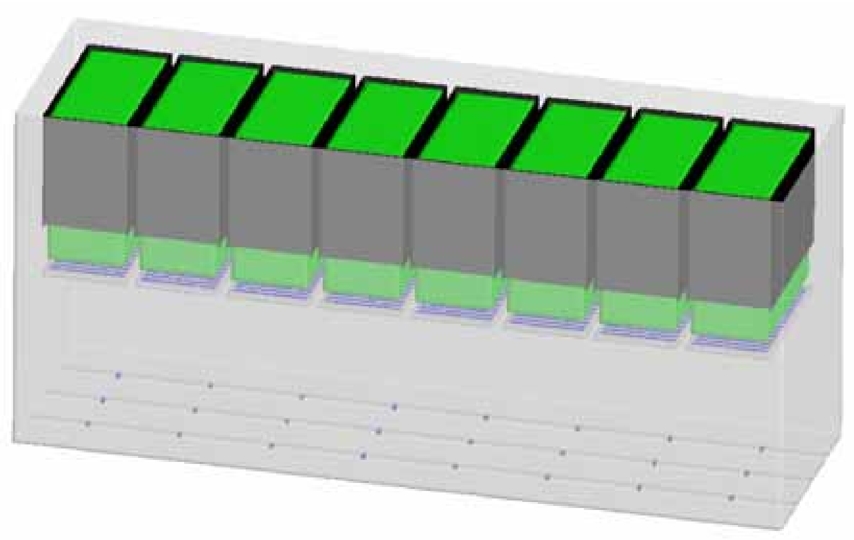


Figure 2 Computer model of the MBR-tank of WWTP Sobelgra (Belgium).

Simulation results after t = 700 s are presented in figures 3 to 5 which give a detailed insight of the system conditions. Clearly aeration has a significant influence on the flow field. The velocities below the aerators are generally lower than in the upper zone. This can be seen in the contour plot of velocity magnitude (figure 3) and the vector plot (figure 4) where the arrow length denotes velocity magnitude. The presented cross sections are located in the middle of the tank.

Comparing the velocities to the air-phase distribution (figure 5) the influence of aeration on the flow field can be seen. A general upward movement above the aerators can be seen in the vector plot. The associated downward movement takes place at the very left and right side of the tank, but also in the area of module 4. The buoyancy due to aeration is less than the suction applied by module 5. Therefore in the area of module 4 although aerated from below a downward movement occurs.

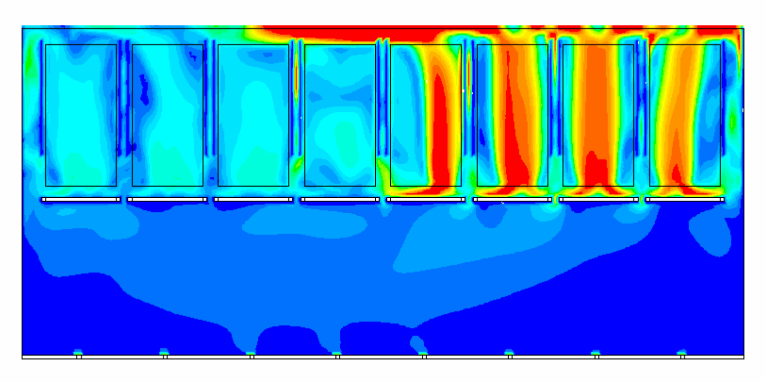


Figure 3 Velocity magnitude [m/s]; scaled from 0.0 [blue] – 0.5 [red], WWTP Sobelgra.

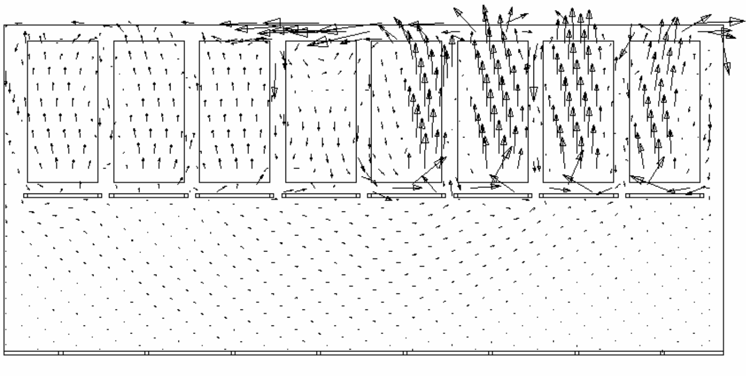


Figure 4 Velocity magnitude vectors, WWTP Sobelgra.

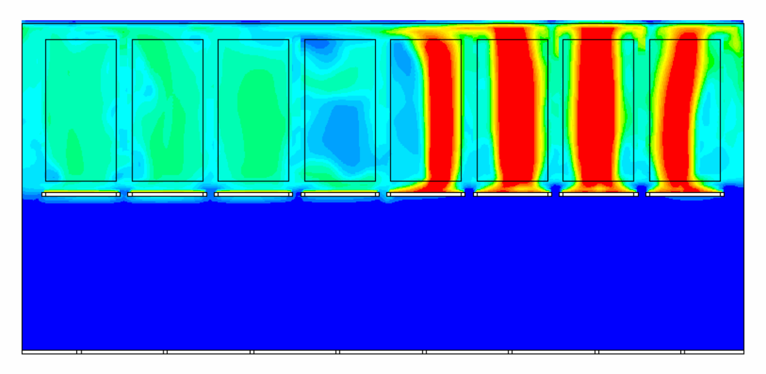


Figure 5 Air-phase distribution by volume fraction; scaled from 0.0 [blue] – 0.3 [red], WWTP Sobelgra.

Validation of the model assumptions is done by comparing velocity measurements to computed values. Figure 6 presents time series of measured and computed velocities over a period of 120 s. Due to the highly unsteady nature of aerated systems a complete matching of the curves is not expected. However, the overall range and fluctuation of the x- and z-velocities show good accordance. The computed y-velocities, vertical velocities, differ significantly from the measured ones. Overall the simulated values are slightly larger than those measured. This may be due to a difference in the air-load between measured and simulated state because a direct determination of the air-load during the measured state was not possible. Additionally the period of 120s considered for comparison are chosen arbitrarily from the simulation results. For a different period of time vertical velocities may differ from those used in figure 6 significantly. The impact of aeration is the dominating factor on the vertical velocities causing it to be highly unsteady.

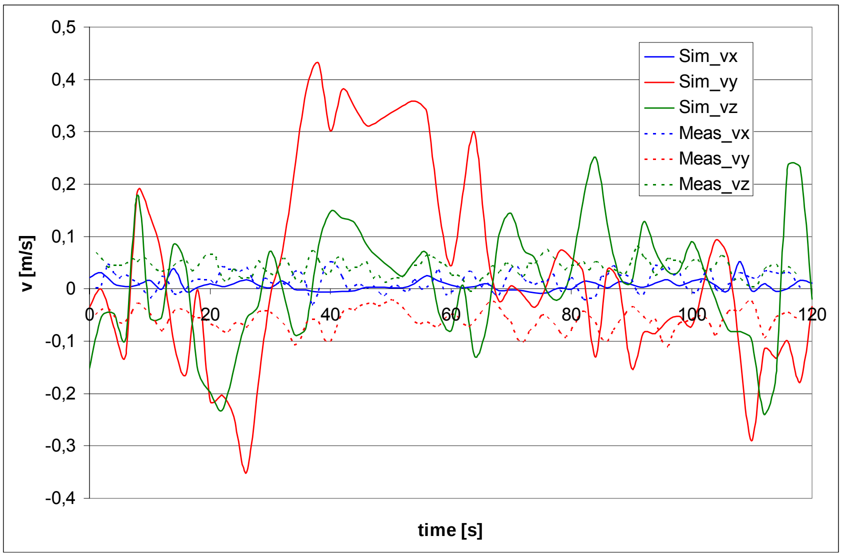
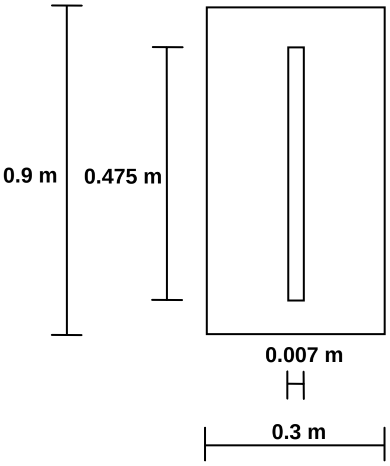


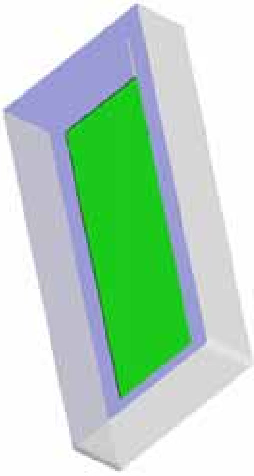
Figure 6 Comparison between measured and computed velocity values, WWTP Sobelgra.

The MBR-tank of WWTP Heenvliet

The second system under investigation is an MBR-tank of WWTP Heenvliet (The Netherlands). Modules consisting of plate-membranes (Toray Membrane) are installed in this tank. In order to develop an approach for modelling plate-membrane modules for use within a CFD-model, first a single plate-membrane model is developed.

Figure 7 shows a top view sketch of the model setup and figures 8 and 9 show the computer model. A modelled plate membrane comprises of two porous zones acting as cover sheets with a fluid zone in between. The simulation setup can be seen in figure 9 with the flow direction as depicted. Porous zones, modelling the membrane are depicted in green. A negative pressure of 1 bar is applied in order to obtain a permeate flux. The height of the membrane is 1.57 m and the water level is at 2.0 m. In order to calibrate the model resistance values for the porous zones were chosen to match a permeate flux of 1000 l /(m^2 h bar) (Judd, 2006).





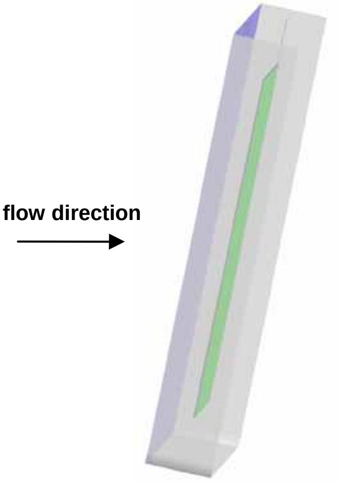


Figure 7 Top view sketch of a plate membrane model.

Figure 8 Computer model of a plate membrane.

Figure 9 Computer model of a plate membrane; side view.

Results on the basis of the calibrated model are presented in figures 10 to 14. The flow field, figures 10 and 11, is influenced by the resistance posed by the plate. The water is mainly directed around the plate (figure 11) and only to a small extend sucked through the membrane.

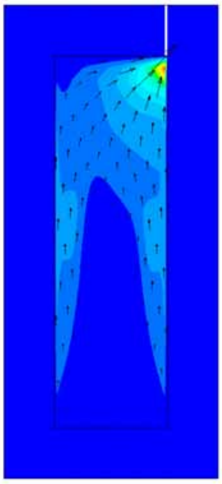


Figure 10 Velocity magnitude [m/s]; scaled from 0.0 [blue] – 1.0 [red].

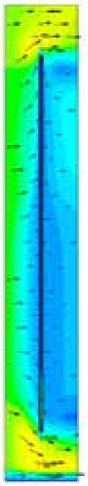


Figure 11 Velocity magnitude [m/s]; scaled from 0.0 [blue] – 0.002 [red].

Calculated pressure distributions can be seen in figures 12 to 14. Clearly a large pressure difference between inside the plate membrane and the outside water is calculated (figure 12). Outside the membrane, the pressure distribution builds up according to the flow field. The water has to move around the membrane (figure 11) with the highest velocities right next to the plate. Accordingly the pressure distribution is characterized by the dynamic pressure with the highest values next to the membrane (figure 13). Inside the membrane a pressure gradient builds up from top to bottom (figure 14). The lowest pressure can be seen where the permeate withdrawal takes place, i.e. in the top right corner of the plate-membrane (figure 14). Further down the membrane higher pressure values can be observed. All pressure values are relative values.

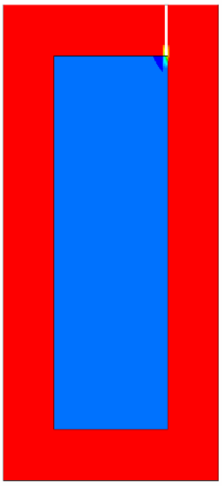


Figure 12 Pressure distribution [N/m^2]; scaled from -80000.0 [blue] – 0.0 [red]

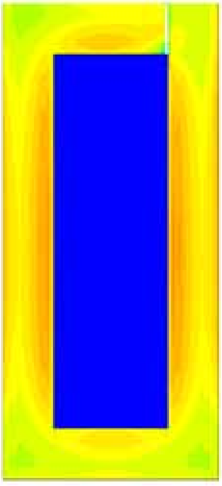


Figure 13 Pressure distribution [N/m^2]; scaled from -0.01 [blue] – 0.01 [red]

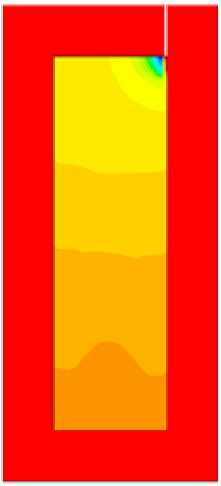


Figure 14 Pressure distribution [N/m^2]; scaled from -77000.0 [blue] –

-74000.0 [red]

CONCLUSIONS

This paper presents a 3-dimensional numerical simulation model which allows a realistic description of the flow structure in MBR-tanks. Computation of the flow field is carried out by the Algebraic-Slip-Mixture-model. Transport of activated sludge is taken into account the model. In order to be able to investigate an entire MBR-tank, membrane modules are modelled as a whole, combining single membranes. These modules are taken as zones of porous media with resistance values according to the type of membrane module installed. Resistance values are determined on the basis of velocity measurements. Two examples demonstrating the modelling process are presented. The results obtained, show that the modelling technique used is suitable for investigations on flow structures in MBR-tanks.

LITERATURE

FLUENT (2006). User-manual. Fluent, Inc., Lebanon, USA.

Judd, S. (2006). The MBR Book: Principles and Applications of Membrane Bioreactors for Water and Wastewater Treatment. Elsevier Science.

Manninen, M., Taivassalo, V. and Kallio, S. (1996). On the mixture model for multiphase flow. Technical report, Technical Research Centre of Finland, Espoo.

Orszag, S.A., Staroselsky, I., Flannery, W.S., Zhang, Y. Introduction to renormalization group modelling of turbulence. Simulation and Modelling of Turbulent Flows. Oxford University Press, 1996.

Application of Submerged Membranes for the Treatment of Spent Filter backwash Water

F. Saravia*, C. Zwiener* and F.H. Frimmel*

*Engler-Bunte-Institut, Water Chemistry, Universität Karlsruhe, Engler-Bunte-Ring 1, 76131, Karlsruhe, Germany (*florencia.saravia@ebi-wasser.uka.de*, *christian.zwiener@ebi-wasser.uka.de*, fritzt.frimmel@ebi-wasser.uka.de)

Abstract The application of membrane filtration in the treatment of spent filter backwash water (SFBW) permits efficient removal of microorganisms, suspended particles and organic substances, depending on the used membrane molecular weight cut-off. However, flux decline, due to deposits and adsorption of substances (salts, colloids, organics, particles, microorganism, etc.) tends to limit the use of membranes.

Characterization of SFBW samples from different waterworks showed that three major factors contribute to the SFBW properties: the raw water itself, the time interval of sand filter operation and additional treatment steps. The main differences between SFBW samples were found principally in DOC, TOC and turbidity. Experiments with submerged membranes (lab and pilot modules) showed that there was a clear correlation between feed DOC concentration and the flux decline: when the DOC-concentration increases, the flux decline increases. Additionally the presence of calcium led to not only an important flux decline but to high adsorption of NOM on membrane surface. Filtration of SFBW revealed that the flux decline is much higher than by filtration of model solutions. More aspects such as TOC, size and type of particles should be taken into consideration, in order to get a better understanding of the fouling processes on submerged membranes.

Keywords Calcium; NOM; particles; SFBW; submerged membranes; UF

INTRODUCTION

The treatment of spent filter backwash water (SFBW) depends on the treatment characteristics of water works, the water source quality and the culture of the country. In some cases SFBW is recycled for drinking water treatment using different pre-treatment techniques. In other plants SFBW is discharged to the sewage system with or without treatment. This practice can contaminate the environment and is cost intensive due to regulations of restricted waste water disposal. Furthermore, it does not save the water resources, and is therefore not sustainable, especially in countries with limited water resources. Recycling of SFBW sustains environmental protection and saves water resources. However, when recycling is performed improperly, it may increase the risk of re-introducing contaminants into the treatment process (Arora et al., 2001, Cornwell and Macphee, 2001, Adin et al., 2002).

Membrane technologies can offer efficient removal of suspended particles, organic substances and pathogens depending on the molecular weight cut-off or pore size on the used membranes. One of the principal limitations of membrane processes is flux decline and blocking of the membranes by fouling. Interactions (adsorption, pore plugging, precipitation, cake formation, etc.) between membrane and water components cause flux decline and variation of the membrane's rejection properties (Schäfer et al., 2002).

In this work, typical SFBW from drinking water treatment plants was firstly characterized in terms of NOM (natural organic matter), cations and particles. Secondly, different membrane filtration experiments of SFBW and model solutions were carried out in order to analyze the effect of these parameters on the filtration performance and fouling formation.

MATERIALS AND METHODS

Spent Filter Backwash Water Samples

SFBW Samples were collected from four different drinking water treatment plants, corresponding to different water sources and treatment processes. Table 1 describes the drinking water plants concerned, further referred to as A, B, C and D.

All samples were conserved by 3-5ºC after collection. The following parameters were measured: cations and anions concentration, LC- UV / OCD (Liquid chromatography with UV absorption and Organic Carbon Detection) TOC, DOC, Turbidity, SAK_{254}, conductivity. SFBW settling properties were also tested.

Table 1. Characteristics of the treatment plants sampled

Drinking water plant	A	B	C	D
Raw water	Groundwater	Surface water (River)	Surface water (Lake)	Surface and Groundwater
Raw water DOC (mg/L),	0.8 -1.1	2.6 (av.)	0.9 – 1.4	2-3 (Surface water)
Drinking water DOC (mg/L), av.	0.96	0.9	0.9 – 1.4	0.6 – 1.1
Filter media	High rate sand filter	Anthracite/sand filter	High rate sand filter	Anthracite/sand filter
Backwash	after 5 days	after 4 – 5 days	every 6 - 7 days	after 3 – 4 days
Portion of treated (drinking) water used for backwash (in %)	1-3	5-6	no data	0,8 –1

Bench-scale membrane filtration

A bench-scale immersed membrane module was designed to perform short-time preliminary tests, which could then at a later time be correlated to results of the pilot module described in section c. A scheme of the bench scale module is shown in Figure 1. The module operates under similar conditions as the pilot module, i.e. under vacuum whilst introducing air bubbles at the bottom of the membrane.

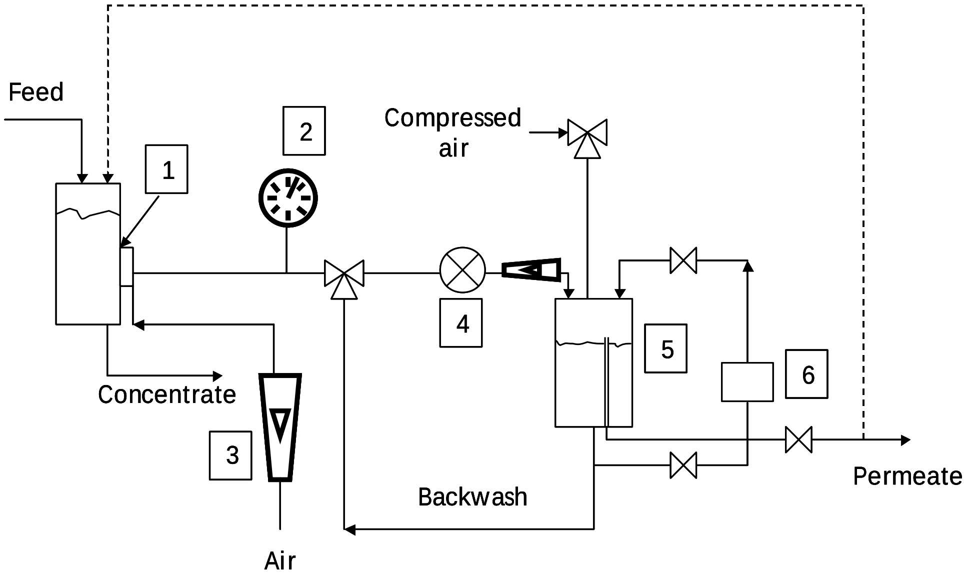


Figure 1 Bench scale immersed membrane (1- submerged membrane, 2- manometer, 3- air flow meter, 4- gear pump, 5- backwash reservoir, 6- particle counter)

The membrane used in the bench-scale experiments consists of hydrophilic polyethersulfone with an absolute pore size of 0.1 µm, manufactured by Pall.

The influence of DOC-concentration and cations on membrane filtration was investigated. To evaluate the effect of DOC, 5 solutions with low ion content and different DOC concentrations ranging from 1.5 to 15 mg/l were prepared using 0.45 µm filtered NOM from lake Hohloh (Black Forest, Germany) diluted with demineralized water. The effect of cations on the elimination of DOC was investigated using solutions of $CaCl_2$ and $FeCl_3$ at a constant initial DOC concentration of 5.2 mg/l.

Experiments were done at a temperature of 20ºC ± 1ºC and a pH of 7.5 ± 0.2 .

Membrane filtration with the pilot module

Experiments were carried out with an immersed ultrafiltration module (ZeeWeed ®, Zenon Environmental Inc., Canada). The UF hollow fiber module has an outside-in configuration and is immersed in the feed-water tank (Figure 2). The membrane has a nominal pore size of 0.035µm .

The membranes operate under partial vacuum created by a gear pump. Air is introduced continuously at the bottom of the membrane module. The air bubbling produces a moderate shear stress, which generates back transport of filtered colloidal particles from the membrane surfaces (Shimizu et al., 1996). Air bubbles also creates turbulence, which scrubs and cleans the outside of the membrane fibers (Best et al., 2000)

Each 30 minutes, the membrane was backwashed with permeate, during 30 seconds. The combination of air bubbling and backwash cause a partial removal of the foulants accumulated on the membrane surface.

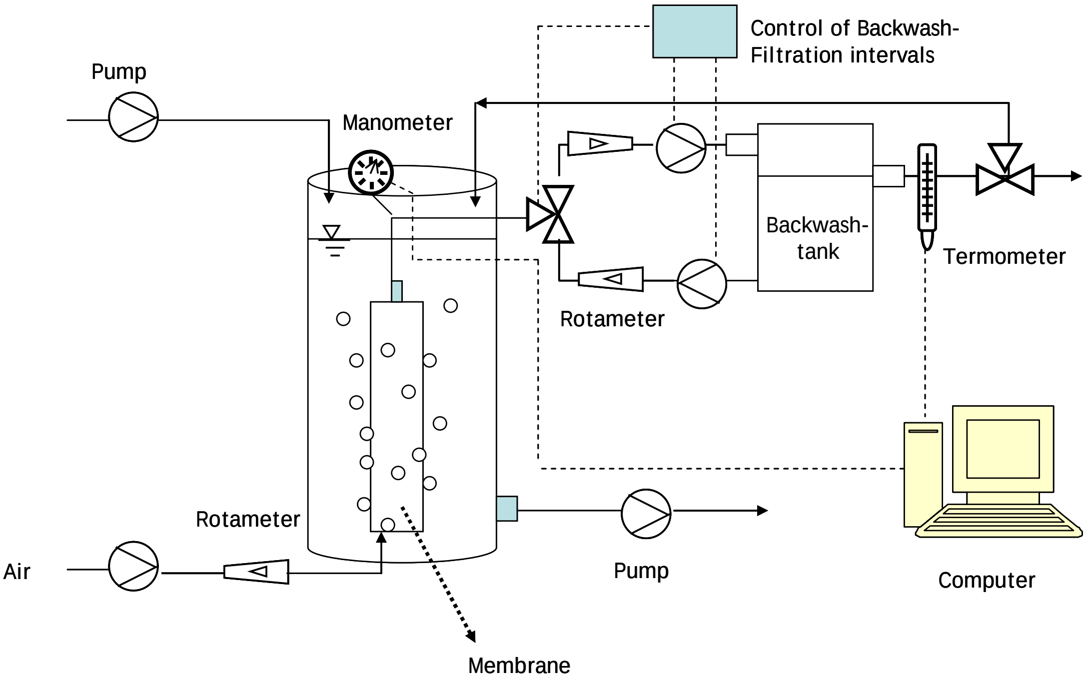


Figure 2 Schematic flow diagram for the ultrafiltration of SFBW

In between experiments, the membrane was cleaned with NaClO (150-200 ppm) during at least 5 hours and if necessary with citric acid (pH = 3) during at least 5 hours. Citric acid was used to eliminate iron and manganese flocs deposited on the membrane surface and within the membrane pores. Each experiment was conducted over a period of 20 days with a flux of 60 L/h (64.5 L/hm^2).

RESULTS AND DISCUSSION

Characterization of SFBW

Measurements of SFBW properties are summarized in Table 2. The quality of SFBW principally depended on: raw water source and quality, treatment process and sand filter type and operation. Due to the wide variation, treatment of SFBW can only be described in general and should as a consequence be reconsidered for each individual case.

Table 2 Characteristics of SFBW samples.

Drinking water plant	pH	Conductivity (µS/cm)	DOC (mg/L)	TOC (mg/L)	SAK $_{254}$ 1/cm	Turbidity NTU
A	7.5	655	1.0	2.9	2.2	1039
B	7.9	516	1.6	2.9	5.5	12,4
C	7.2	456	4.6	29.5	5.0	1142
D	7.8	432	1.7	20.2	4.0	245

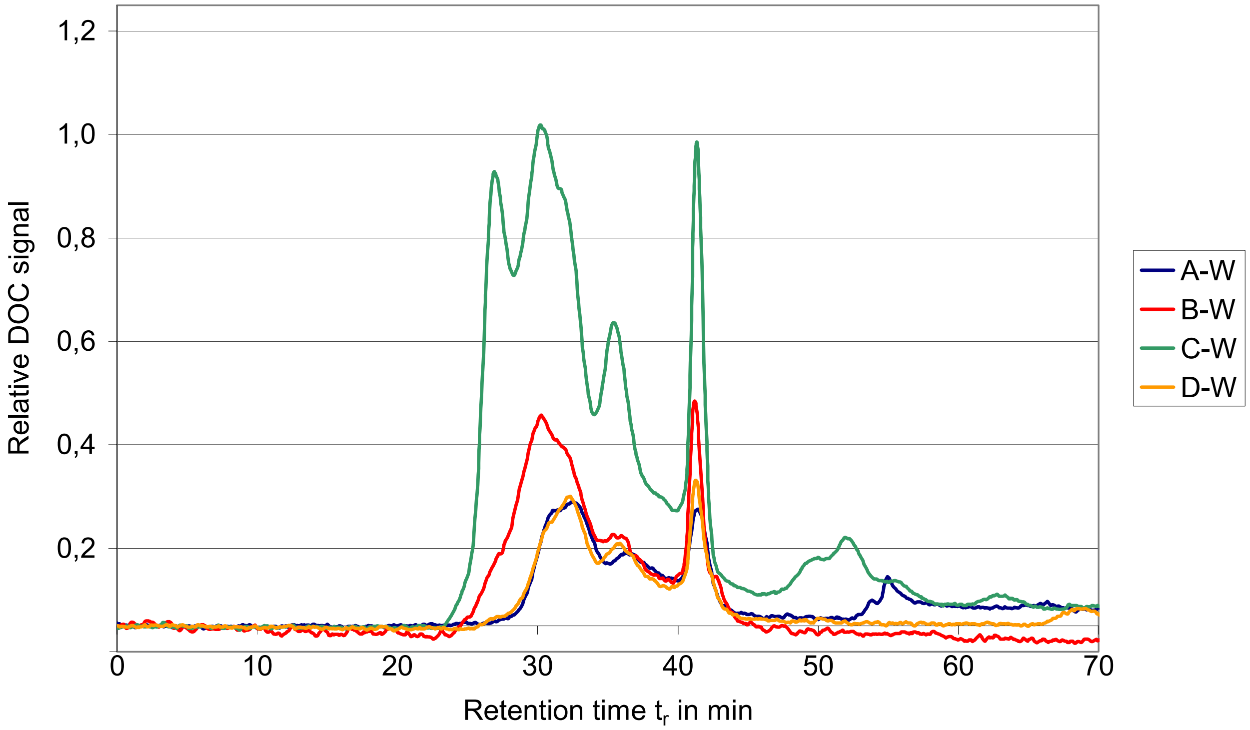


Figure 3 LC-OCD of SFBW samples

LC- UV / OCD measurements are shown in figure 3. Samples contained the same DOC-fractions in different concentrations with a considerable fraction of humic substances. The SFBW from plant C showed an additional high molecular mass fraction with a retention time (t_r) of 27 minutes. (Huber and Frimmel, 1996) and a low molecular fraction with a retention time between 47 and 58 minutes probably corresponding to by-products derived from microorganisms or biodegradation.

The settling experiments of the SFBW samples proved that within one hour turbidity, Fe and Mn concentrations were reduced up to 10 % of the original values. As a consequence settling could be considered to pre-treat SFBW and minimize the amount of flocs and particles before membrane filtration in a simple and cheap way.

Bench-scale membrane filtration

A first group of experiments studied the effect of DOC concentration on filtration by immersed membranes. The brown-water solutions applied had a DOC concentration of 1.5, 2.2, 5.2, 10.2 and 15.3 mg/L, respectively. Flux decline increased with higher DOC concentration (Figure 4). Organic rejection, measured as DOC concentration in permeate divided by DOC concentration of raw water, was generally between 0-5 %.

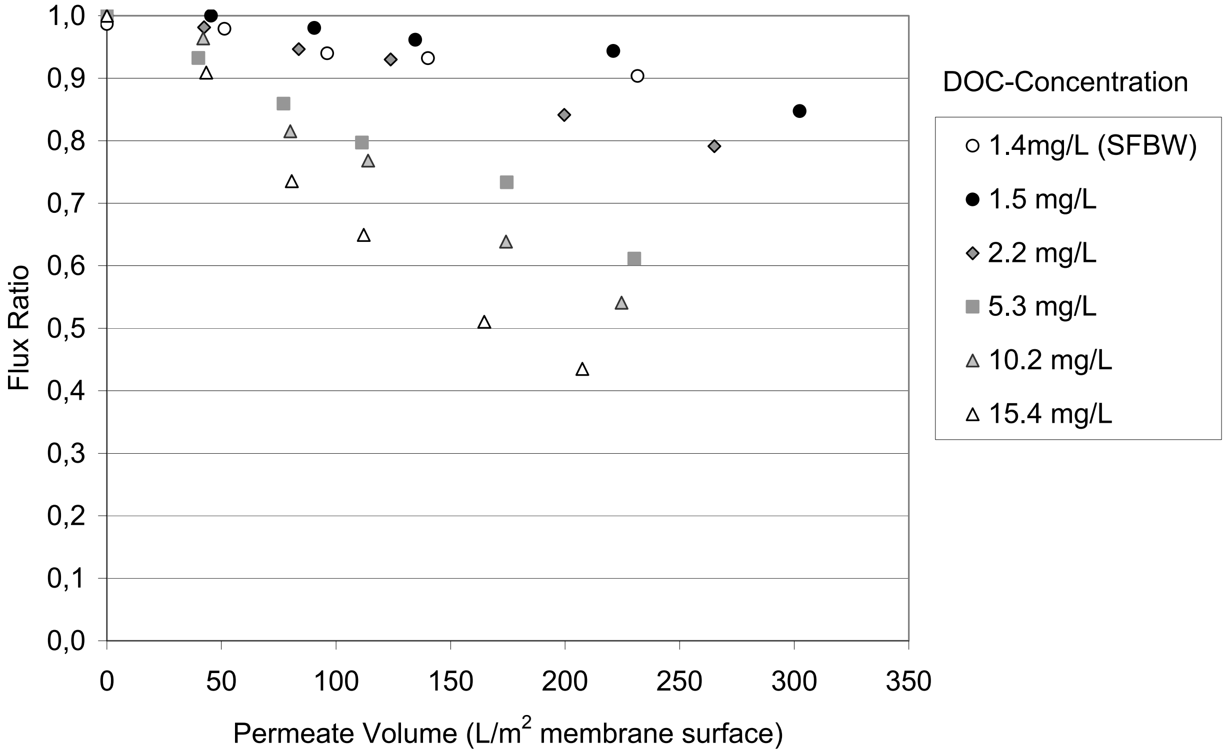


Figure 4 Effect of DOC-concentration by membrane filtration (bench-scale)

Alternately, SFBW of plant A was filtered and compared to the results of the lake Hohloh solutions in Figure 4. The SFBW sample had a DOC of 1.2 mg/L. Flux decline was similar to the one of a solution of initial DOC in the same concentration and pH. This demonstrates the strong effect of DOC concentration on membrane fouling and flux decline. The effect of particle size must also be considered. In this case the particle size was in average 20 µm, big enough to impede pore plugging. In another set of experiments the effect of cations (calcium and iron) on DOC-rejection and flux decline was studied. Humic substances act as a natural complexant in the environment. In presence of calcium, there is expected that NOM-metal complexes and aggregates are formed. In presence of higher iron concentrations, there could be also iron (III) hydroxide precipitation and flocculation. Flux measurements are shown in figure 5. All experiments showed a high flux decline, due to metal complex formation and agglomeration. The presence of iron produced a higher flux decline in comparison with calcium. Formation of iron hydroxide could be verified, since the measurement of iron and calcium concentrations with ICP-OES (not shown here), showed a significant difference between permeate and feed. For iron up to 50% less and no significant difference for calcium (between 0-2%) was found in the permeate. Pore plugging and corresponding fouling could be expected due to formation of micro-flocs with dimensions close to the membrane pore size (Best et al., 2000).

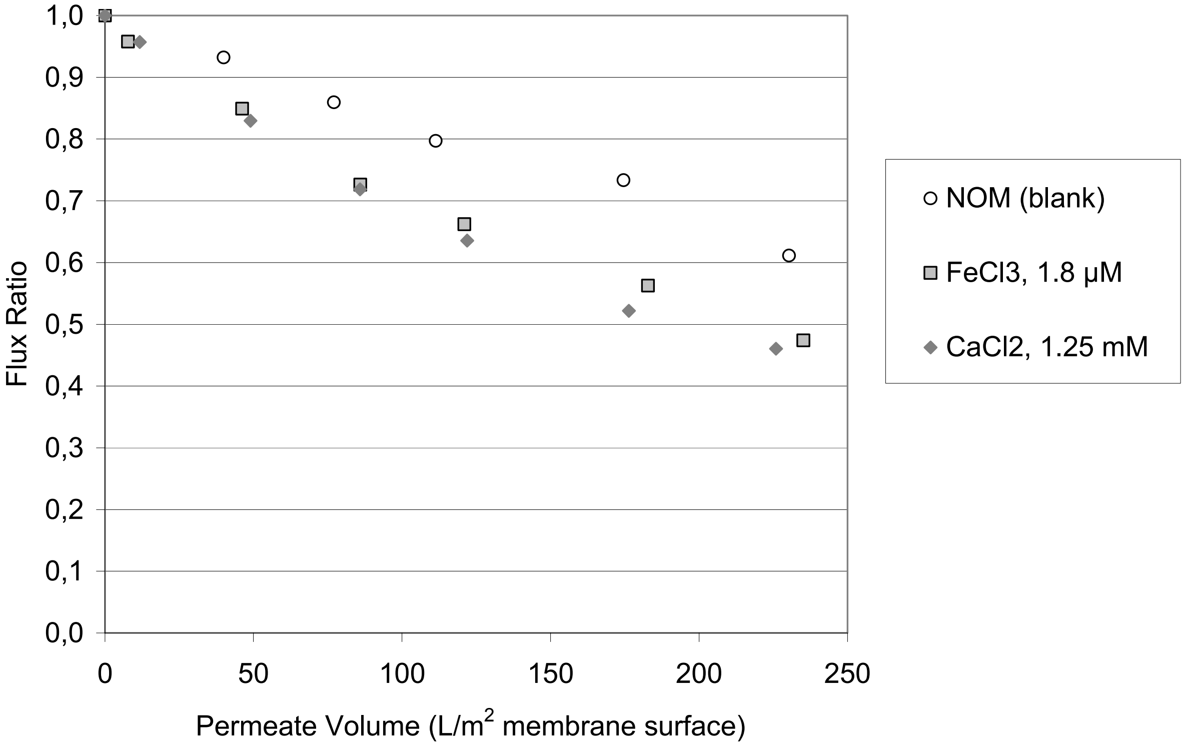


Figure 5 Flux ratio over filtration volume of the 0,1µm membrane, as a function of solution with different calcium and iron concentrations (DOC= 5.2 mg/L ± 0.3 mg/L, pH=7,5, Temp. = 20ºC)

Membrane filtration with the immersed membrane module (Zenon)

Experiments were carried out in periods of 20 days. The membrane module was backwashed according to the protocol given above but there was no special cleaning with chemicals. Figure 6 shows the flux of the immersed module when filtering different solutions. In order to allow correlation to the bench-scale experiments, solutions were prepared in the same manner as described in section “Bench scale membrane filtration”.

Experiments with the pilot module confirmed the importance of DOC-concentration and the presence of calcium-ions in the flux decline (see fig.6). There was, as in the bench-scale tests, a clear correlation between feed DOC concentration and the flux decline: when the DOC-concentration increases, the flux decline increases (Saravia, et al. 2006).

Experiments with added Ca^{2+} ions showed a much higher flux decline compared to those without Ca^{2+} addition (see fig. 6). It is suggested that the formation and aggregation of calcium complexes with organic matter cause an increase of fouling and a higher flux decline by microfiltration, generally due to pore plugging.

Experiments have clearly shown that DOC-concentration and calcium ions are important contributors to membrane fouling and flux decline. However, filtration of SFBW taken from a water plant revealed that the flux decline is much higher than after filtration of model solutions. SFBW usually has high iron contents and a considerable portion of microorganisms. The iron forms quite strong complexes with NOM, which can plug membrane pores and adsorb on the membrane surface, reducing in this way membrane performance. The presence of microorganisms and algae contributes to the formation of biofouling.

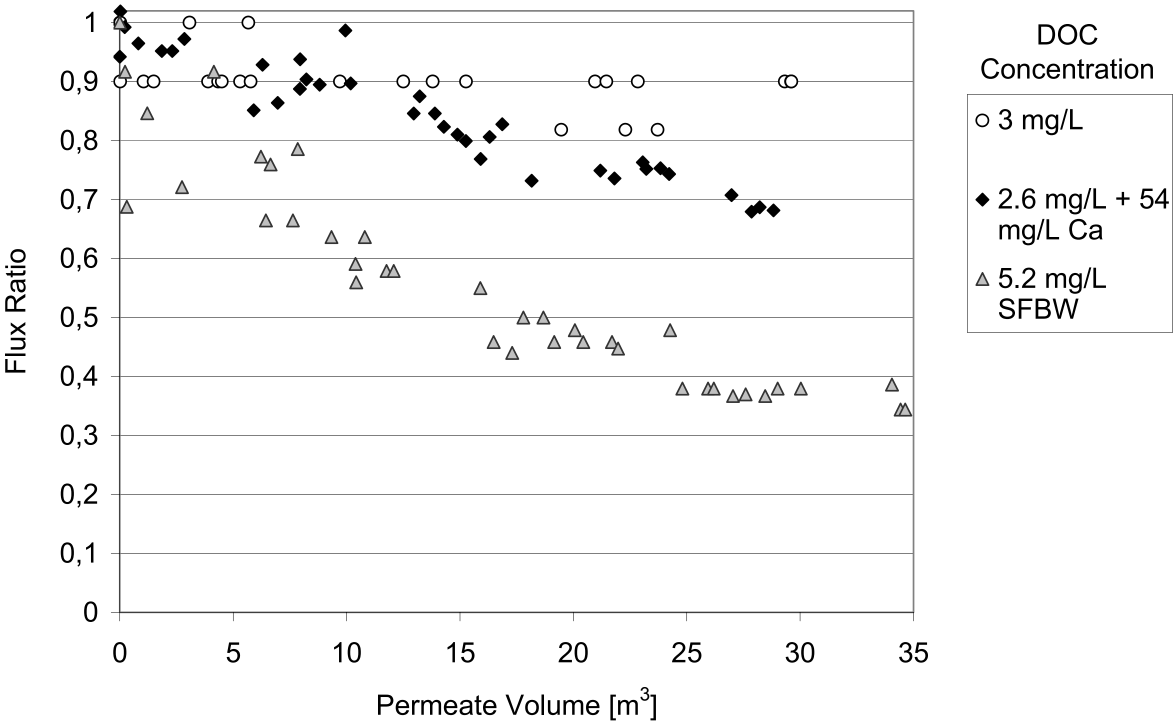


Figure 6 Flux ratio over filtrate volume for the filtration with the immersed module

Membrane cleaning with chemicals was done between experiments. After experiments with NOM-rich water the cleaning was performed using NaOH and NaClO. The use of NaClO permitted the recovery of the initial flux. After the filtration of SFBW, the membrane was covered with deposits of iron and manganese flocs and was cleaned first using citric acid in order to eliminate iron-flocs and iron complexes. After citric acid cleaning the initial flux was only partly recovered (only metal complexes and flocs were dissolved). Only with a cleaning with sodium hypochlorite solution (150-200 ppm) the entire initial flux was recovered.

CONCLUSIONS

The quality of SFBW principally relates to raw water quality, kind of water treatment and sand-filter operation. Due to the high variation in quality, the design of a SFBW treatment for production of drinking water can only be considered in general terms. Separate studies are necessary to match the specific situation of individual raw waters and treatment steps.

The treatment of filter backwash water with membrane technologies is promising and offers secure rejection of microorganisms, bacteria, iron and manganese flocs and other suspended materials. However, membrane fouling and as a consequence flux decline can be a severe drawback of membrane application.

Bench-scale and pilot-scale experiments revealed that natural organic matter plays an important role in the flux decline in membrane filtration. Cation concentrations (calcium and iron) also have an influence on flux decline because of the formation of NOM-metals and aggregates: the presence of calcium and iron was observed to slightly increase the DOC rejection at a higher flux decline with respect to flux decline in solutions with low ion concentrations.

Pilot-scale experiments were carried out during a period of 20 days without chemical cleaning. During this period of time, the pilot module was operated at 64.5 L/hm^2. A high flux decline could be observed in experiments with SFBW in comparison with model solution with similar DOC concentration. Consequently, more factors must be accounted in order to better understand the fouling process on submerged membranes and to more efficiently apply membrane separation in the treatment of SFBW.

The application of optimized membrane technology in backwash water treatment will help to run water supply in an economic way and to protect aquatic systems.

REFERENCES

Adin, A.; Dean, L; Bonner, F.; Nasser, A.; Huberman, Z. (2002): Characterization and destabilization of spent filter backwash water particles, Water Sciences and technology: Water Supply 2(2) 115-122.

Arora, H., Di Giovanni, G. and Lechevallier, M. (2001): Spent filter backwash water contaminants and treatment strategies, Journal AWWA 93(5) 100-113.

Best, G.; Singh, M.; Mourato, D.; Chang Y.J. (2000): Application of immersed ultrafiltration membranes for organic removal and disinfection by-Product Reduction, Water Supply 1(5-6) 221-231.

Cornwell, D.A.and Macphee, M.J. (2001): Effects of spent filter backwash water, Journal AWWA 93(4) 153-162.

Huber, S.A.; Frimmel, F.H. (1996): Size exclusion Chromatographie with organic carbon detection (LC-OCD): a fast and reliable method for the characterization of hydrophilic organic matter in natural waters, Vom Wasser, 86 227-290.

Saravia, F., Zwiener, C. and Frimmel, F.H.F. (2006): Interactions between membrane surface, dissolved organic substances and ions in submerged membrane filtration, Desalination 192, 280-287.

Schäfer, A. I.; Schwicker, U., Fischer, M.M., Fane A.G.; Waite, T.D.(2000): Microfiltration of colloids and natural organic matter, Journal of Membrane Science 171 151-172.

Shimizu, Y.; Okuno Y-I; Uryu, K.; Ohtsubo, S.; Watanabe, A. (1996): Filtration characteristics of hollow fiber microfiltration membranes used in membrane bioreactor for domestic wastewater treatment, Water Research 30(10) 2385-2392.

Treatment of Landfill leachate with Reverse Osmosis Membrane Technologies

Fangyue Li*

*Technical University Hamburg Harburg, Institute for Water Resources and Water Supply, Schwarzenbergstr. 95 E, D-21073 Hamburg, Germany
(Email: Li.fangyue@tu-harburg.de)

Abstract In this paper, a leachate purification system equipped with open channel spiral wound modules was investigated. In Phase I, effluent from an activated sludge process followed by a coagulation/sedimentation process was fed into the landfill leachate purification unit. An average permeate flux of 4.86 $l/m^2.h$ was achieved during Phase I. After around two week's operation, permeate flux dropped dramatically from 6.5 $l/m^2.h$ to 2.93 $l/m^2.h$, showing the occurrence of severe membrane bio-fouling.

In Phase II, raw leachate was fed directly into the reverse osmosis leachate treatment system and an average flux of 7.8 $l/m^2.h$ was maintained during the observation period. An average recovery rate of 70% was achieved. Dissolved solids were eliminated by 98.2%. Reduction rate of COD was greater than 99.5% and COD was always lower than 15 mg/l in permeate. More than 98% NH_4-N was reduced. Elimination rate of chloride was more than 99%. Concentrations of heavy metals and chloride in permeate were lower than the German landfill leachate discharging standard. After effective membrane cleaning, a negligible permeate flux drop was observed in phase II.

Keywords Disc Tube module (DT module); Landfill leachate treatment; Open channel module; Reverse osmosis; Spiral wound module; Tubular module

INTRODUCTION

Landfill leachate is heavily loaded wastewater with different types of organic and inorganic contaminants and represents a major risk with respect to contamination of natural water resources (Christensen *et al.*, 1992). Treatment of Municipal Solid Waste (MSW) landfill leachate can be a very complex process if low discharge values have to be obtained. Quality and quantity of leachate vary with landfill age and climate conditions. High concentrations of COD, BOD_5, heavy metals, NH_4-N, low BOD_5/COD ratio and the lack of nutrients in the methanogenic phase have restricted the application of biological treatment processes. Furthermore, biologically treated leachate still has relatively high concentration of COD and halogenated compounds (AOX) that need to be eliminated to minimize the negative influences on the natural water cycles. Due to the development of modern high rejection reverse osmosis membranes to retain both organic and inorganic contaminants at high rejection rate and properly designed membrane modules, more and more leachate treatment plants have selected reverse osmosis technologies for landfill leachate purification. It has been proven that purification of landfill leachate demands the use of open channel reverse osmosis membrane modules (Peters, 2001).

CHARACTERISTIC OF LANDFILL LEACHATE

Corresponding to the decomposition process of MSW disposed in landfill, leachate undergoes five different development phases in the landfill reactor (Figure 1). These development phases include the first aerobic phase (I), the acid fermentation phase (II), the intermediate anaerobic phase (III), the methanogenic phase (IV) and the second aerobic phase (V) (Christensen *et al.*, 1992). It might take several hundreds years for landfill to reach its final stable phase. Therefore, treatment of landfill leachate can last for a very long time after wastes are disposed. In each development phase, quantity and quality of landfill leachate show very different physical and chemical characteristics. Generally speaking, landfill leachate consists of 80-95% inorganic components and 5-20% organic components (Rautenbach, *et al.*, 1996). The inorganic substances include HCO_{3^-}, Cl^-, SO_4^{2-}, Na^+, K^+ and Ca^{2+}. The organic contents vary dramatically according to different influencing parameters and can be roughly classified into three main groups (Cossu *et al.*, 2007): low molecular organic acids and alcohols, humic substances and fulvic acid-like materials. Low molecular organic acids mainly consist of fatty acids and can be easily degraded through biological processes. In acid fermentation phase these substances cover more than 90% of total TOC. Humic substances are derived from cellulose and lignin with a high molecular weight and are rather stable organics. These substances represent 0.5% to 5% of leachate organic contents. Fulvic acid-like materials with medium to high molecular weight are predominant in methanogenic leachate and are difficult to be degraded biologically. Besides these organics, several other organic substances such as benzene, phenols and AOX have been detected in the methanogenic leachate. It was also reported that hazardous compounds such as polyaromatic hydrocarbons (PAH), polychlorinated biphenyls (PCB) are in landfill leachate (Schwarzenbeck *et al.*, 2004).

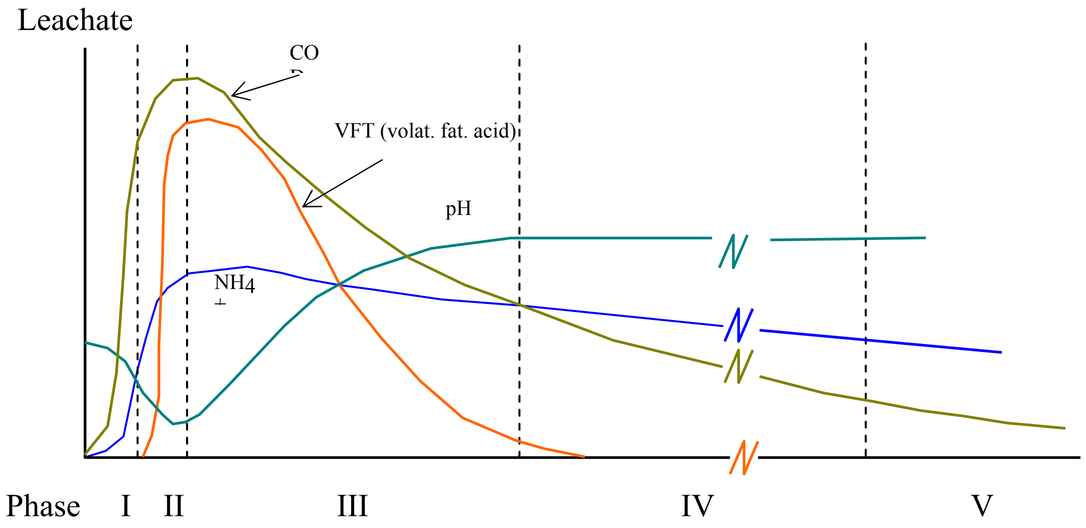


Figure 1 Development of leachate composition in a landfill (Christensen, 1992).

DEVELOPMENT OF REVERSE OSMOSIS TECHNOLOGIES FOR LANDFILL LEACHATE TREATMENT

The frequently used processes for landfill leachate treatment include discharging to sewage treatment plants, on site biological treatment including anaerobic and aerobic treatment, on site physical and chemical treatment including flocculation/precipitation, membrane separation processes, activated carbon adsorption, chemical oxidation, ammonia stripping, evaporation, leachate re-circulation and reeds bed etc. It is not difficult to reach the limiting standard for BOD_5 and NH_4-N through well designed biological treatment processes with nitrification and de-nitrification. But large amount of non-biodegradable organic substances cannot be eliminated below the discharging standard by a single biological process. Although the landfill leachate discharging standard regarding inorganic salts and trace organic substances are not clearly defined in Germany, these substances have negative impacts on the environment. This is due to the fact that introducing inorganic components and trace organics to the natural water cycles at even very low concentrations can lead to bioaccumulation in ecological system. Taking chloride for example, the average chloride concentration in landfill leachate is 2150 mg/l (Christensen *et al.*, 1992). While, biological treatment processes have no effect on its concentration reduction. However, a discharging limit of 250 mg/l for chloride is required in some federal states of Germany, which means processes such as reverse osmosis shall be applied to meet this requirement.

Baumgarten *et al.* (1996) had reported that high rejection reverse osmosis can retain both dissolved solids and metals to a range from 88% to 97%. York *et al.* (1999) had pointed out that the elimination rates may sometimes reach to 99%. Due to above mentioned leachate characteristics, the development of high rejection reverse osmosis membranes, high recovery rate up to 95%, and the properly designed membrane modules, treatment of landfill leachate with reverse osmosis technologies has shown an increasing trend in the past 25 years worldwide. Leachate treatment system equipped with reverse osmosis membrane modules can be operated and monitored continuously by an electronic control system. Reverse osmosis membranes are flexible against changes of leachate quality and quantity, because increasing and decreasing of purification capacity can be easily done by adding and taking away membrane modules (Peters, 1998). Reverse osmosis leachate treatment system can be operated at a pressure range from 40 to 60 bar. The pressure applied in the high pressure reverse osmosis system can be as high as 200 bar. Although reverse osmosis technology has superior removal rate for both dissolved organic and inorganic substances, due to high levels of suspended solids, colloids, dissolved organic matters, metal oxides, bacteria and their metabolites in landfill leachate, fouling of membranes occurred inevitably after certain period of operation, which often has detrimental effect on the performance of membranes. When salts concentrations exceed their solubility on the feed side, precipitation of salts on the reverse osmosis membrane surface happens, which leads to the decline of permeate flux, the increase of trans-membrane pressure and demands frequent chemical membrane cleaning. Both fouling and scaling have apparent reduction on membrane performance. Due to these characteristics, purification of landfill leachate with reverse osmosis technologies demands the use of open channel membrane modules.

Among the membrane modules, tubular module is the least susceptible to fouling and easiest to clean. The landfill of Utingen in Switzerland was the first to purify landfill leachate with tubular reverse osmosis in industrial scale in 1984 (Linde *et al.*, 1995). Leachate treatment plant in VAM landfill and SOW landfill in Netherlands were also equipped with tubular reverse osmosis in 1986 and 1987, respectively. Tubular reverse osmosis used for the purification of landfill leachate was often operated at pressure range of 40 to 60 bar. A total recovery rate of 54% was achieved in the VAM leachate treatment plant. The rejection rate of the COD and NH_4-N in VAM leachate purification plant is greater than 90% (Linde *et al.*, 1995).

The Disk Tube (DT) module has been firstly applied for seawater and brackish water desalination since 1982. The leachate treatment plant in Schwabach, Germany was the first landfill leachate treatment plant installed with DT modules with a single leachate stage at a pressure of 65 bar in 1988 (Anonymous, 2007). Leachate is pre-filtrated through a sand filter and a cartridge filter, which are aimed to eliminate suspended solids larger than 10 µm. The pH of the raw leachate is adjusted by H_2SO_4 to a range of 6.0 to 6.5 to minimize scaling penitential. Pre-filtration is designed to reduce the fouling index of the feed water and make sure that the feed quality is sufficiently good for the operation of DT system. The typical reverse osmosis leachate treatment performance installed with DT modules is present in Table 1.

Table 1 Typical plant performance of a DT reverse osmosis leachate purification system

Parameter	Leachate	Permeate	Rejection
pH	7.7	6.8	-
El. conductivity (dS/m)	17.25	0.382	97.8%
COD (mg/l)	1797	15	99.2%
NH_4-N (mg/l)	366	9.8	97.3%
Cl^- (mg/l)	2830	48.4	98.3%
Na^+ (mg/l)	4180	55.9	98.7%
Heavy metals (mg/l)	0.25	<0.005	>98%

MATERIALS AND METHODS

Raw landfill leachate was collected from a German landfill and stored in a leachate collection tank before subsequent use. The landfill leachate was immediately characterized after sample was taken on site. The membrane modules used in this experiment are open channel spiral wound modules. The ultrasonic welded membrane envelopes are made of two flat membranes, between which an internal fabric fleece is used. Feed side spacers separate the membrane envelopes with a channel height of 3 mm, which guarantees fluids with high content of dissolved solids, can be processed without operation problem. An open channel formed on the feed side ensures high feed flow rate, which is needed for high cross flow velocity over the membrane surface and, therefore, is able to tolerate high dissolved solids and high turbidity and achieve a greater resistance to scaling and fouling. Suspended particulates deposited on the membrane surface can be flushed away in the opened channel effectively. A simplified flow diagram of the reverse osmosis leachate treatment unit used in this study is presented in Figure 2.

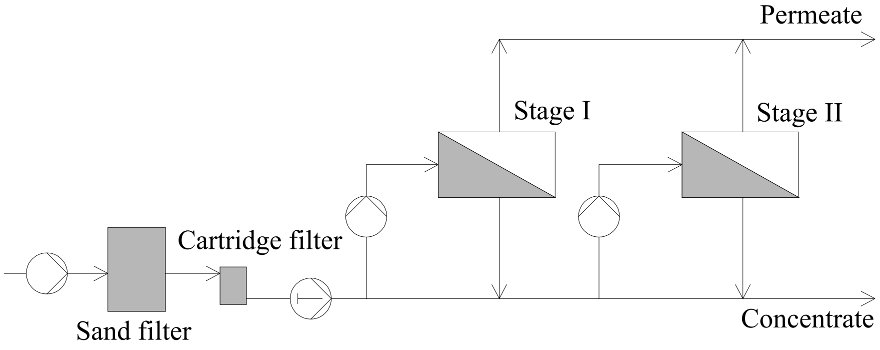


Figure 2 A simplified flow diagram of the reverse osmosis leachate treatment system

This leachate treatment unit has a total volumetric flow rate of 1750 l/h. Raw leachate was fed from an equalization tank to a sand filter. Sand filter was used to remove suspended particles larger than 50µm in size. Sand filter can be plugged quickly and required frequent backwashing. Followed the sand filter, H_2SO_4 was dosed into the leachate to maintain a pH value between 6.0-6.5, which aimed to increase the solubility of the inorganic salts. Cartridge filter was applied for further removal of the suspended solids larger than 10µm. Cartridge filter will be replaced once the pressure drop reaches to 2 bars. A feed and bleed system was chosen for the leachate purification plant. Thin film composite reverse osmosis (FT30) supplied by FILMTECH was chosen for the open channel spiral wound modules. In stage I, three modules with a total membrane area of 76.8 m^2 were connected in series. Pressurized leachate was fed into stage I and flowed axially to the membrane modules.

Portion of the concentrate from stage I was re-circulated and the rest was bled with the pre-treated leachate to stage II. Stage II also consisted of three modules joined in series with a membrane area of 76.8 m^2. Permeate was collected and stored in a collected tank. Both stages were equipped with a booster pump, which was employed to maintain a cross flow rate of $8m^3/h$. This leachate treatment system was operated at a constant flux rate. Over the operating time, the feed pressure was regulated to compensate the fluctuation of feed water temperature, salinity and permeate flux decline.

RESULTS AND DISCUSSION

The physical and chemical compositions of the landfill leachate are shown in Table 2. The measured pH varied from 7.8 to 8.0 with an average value of 7.9. The temperature of the raw leachate was in the range from 8 to 26 °C. The electrical conductivity changed with the variation of precipitation, evaporation and practice of re-infiltration of leachate concentrate to landfill body. During the observation period, it varied from 13.5 to 21.1dS/m with a value of 17.7dS/m on average. COD concentration of the raw leachate ranged from 2700 to 3500mg/l. The measured NH_4-N concentration during the test period varied from 800 to 1200 mg/l. Observed ammonia nitrogen concentration in our study was quite high compared with the values reported by Christensen, *et al.* (1992). This was due to the fact that rejected NH_4-N in the concentrate from a reverse osmosis leachate treatment system was re-infiltrated to the landfill body. Concentration of chloride in our study ranged from 2200 to 3500 mg/l. Based on the observed pH, COD, NH_4-N values and landfill age, the leachate used in our research was in its methnogenic phase and its biodegradability was low. Low biodegradability indicates that it is better treated with physicochemical techniques such as reverse osmosis technology rather than with biological processes.

Table 2 Compositions of the landfill leachate

Parameter	Leachate
pH	7.8-8.0
Temperature (°C)	8-26
El. Conductivity (dS/m)	13.5-21.1
COD (mg/l)	2700-3500
NH_4-N (mg/l)	800-1200
Cl^- (mg/l)	2200-3500

Variation of the permeate flux

Membrane filtration performance was characterized by the fluctuation of permeate flux in this study. Activated sludge process followed by a coagulation/sedimentation process was applied as leachate pre-treatment step in Phase I. Effluent from the pre-treatment step was fed into the leachate treatment unit. An average permeate flux of 4.86 $l/m^2.h$ and an average recovery rate of 53.4% were achieved during Phase I (Figure 2). The trans-membrane pressure difference was maintained at 20 bar initially and increased to around 40 bar before membrane cleaning. After around two week's operation, permeate flux dropped dramatically from 6.5 $l/m^2.h$ to 2.93 $l/m^2.h$ (variation of leachate composition was not considered), showing the occurrence of severe membrane fouling. Application of membrane chemical cleaning and rising of trans-membrane pressure difference up to 68 bar did not recover permeate flux despite larger net driving force. This clearly indicated a permanent loss of membrane permeability. Fast declining of membrane permeate flux in phase I was obviously caused by membrane bio-fouling. It is commonly believed that membrane fouling coupled with permeate flux decline is caused mainly by the soluble microbial product (SMP). SMP is defined as soluble cellular components released during cell lysis and then diffuse through the cell membrane. SMP consists of polysaccharides, proteins and some other compounds like DNA derivates (Laspidou *et al.,* 2002). Coagulation/sedimentation process can not eliminate the SMP effectively from the secondary clarifier effluent and SMP can pass through sand filter and cartridge filter and enter the reverse osmosis modules. Under high pressure, high cross flow, SMP formed a thin film layer on membrane surface, which was difficult to be removed by chemical cleaning. Unsatisfied results obtained during Phase I further proved that it is better to treat landfill leachate under methnogenic phase through reverse osmosis technologies directly.

Due to severe membrane bio fouling, the spiral wound modules used in this study were replaced and the leachate pre-treatment process was discarded in Phase II. Raw leachate was fed directly into the reverse osmosis leachate treatment system. After restarting operation, an average flux of 7.8 l/m^2.h was maintained during the observation period. Similar to Phase I, trans-membrane pressure difference increased from 20 bar initially to 40 bar before membrane cleaning. As it is shown in Figure 2, permeate flux achieved in Phase II was kept in a range from 6.5 to 8.14 l/m^2.h, which was 30% more than that obtained in Phase I. Compared with the trans-membrane pressure applied in Phase I, average trans-membrane pressure difference in Phase II decreased by around 30%. Both trans-membrane pressure and permeate flux were kept in a constant level and no significant permeate flux decline was observed in Phase II. During the observation period, an average recovery rate of 70% was achieved. Concentrate generated from the leachate stage represented nearly 30% of the total leachate volume. To increase the total recovery rate of this leachate treatment plant, concentrate stream needs to be further purified through a high pressure reverse osmosis stage in the future.

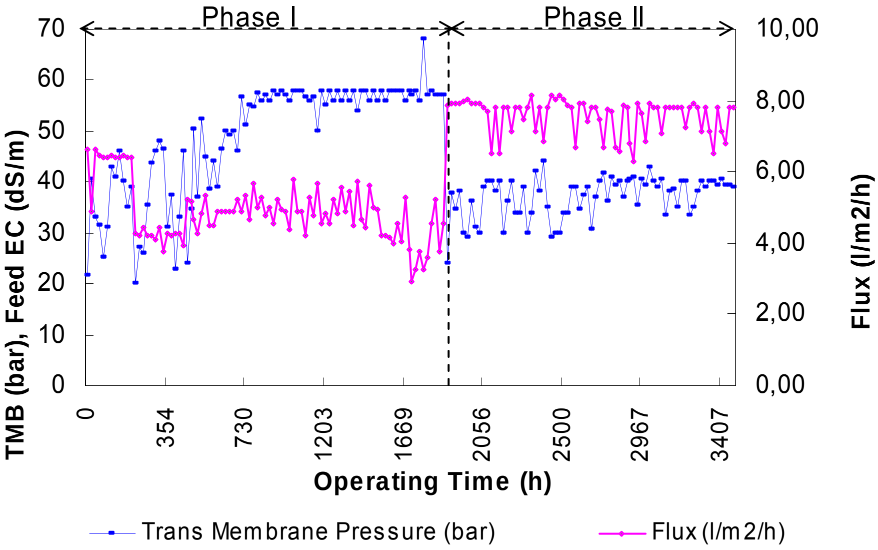


Figure 3 Variation of permeate flux and trans-membrane pressure difference with operating time

Reduction of the pollutants

The reduction of organic and inorganic compounds was assessed on the basis of reduction in the contaminants represented by electrical conductivity, COD, NH_4-N, heavy metals and chlorides. Except the electrical conductivity, reductions of pollutants were not analyzed in Phase I. Reduction rate of electrical conductivity can be regarded as the reduction rate of dissolved solids. During the observation period, an average dissolved solids reduction rate of 98.2% was achieved (Figure 3). Compared with the results from Baumgarten *et al.* (1996), no significant reduction of leachate electrical conductivity due to the application of biological pre-treatment was observed in our study. Reduction rate of COD was greater than 99.5% in Phase II and the permeate COD was always lower than 15 mg/l. More than 98% NH_4-N was reduced during the observation time. NH_4-N in permeate was sometimes slightly higher than the discharging limit due to a higher NH_4-N concentration in the feed. Since NH_4-N concentration in permeate didn’t constantly meet the German landfill leachate discharging standard, a permeate stage reverse osmosis unit shall be added in the future.

Concentrations of heavy metals in permeate (not shown) were also lower than the discharging standard issued in Germany. Elimination rate of chloride was more than 99% and chloride concentration in permeate was lower than the discharge level in Germany. Major pollutants reduction rates are summarized in Table 3.

Table 3 Major pollutants reduction rates

Parameter	Leachate	Permeate	Rejection
pH	7.9	6.8	-
El. Conductivity (dS/m)	16.5	0.38	98.2%
COD (mg/l)	3100	15	>99.5%
NH_4-N (mg/l)	1000	11.27	98.9%
Cl^- (mg/l)	2850	23.16	99.2.3%

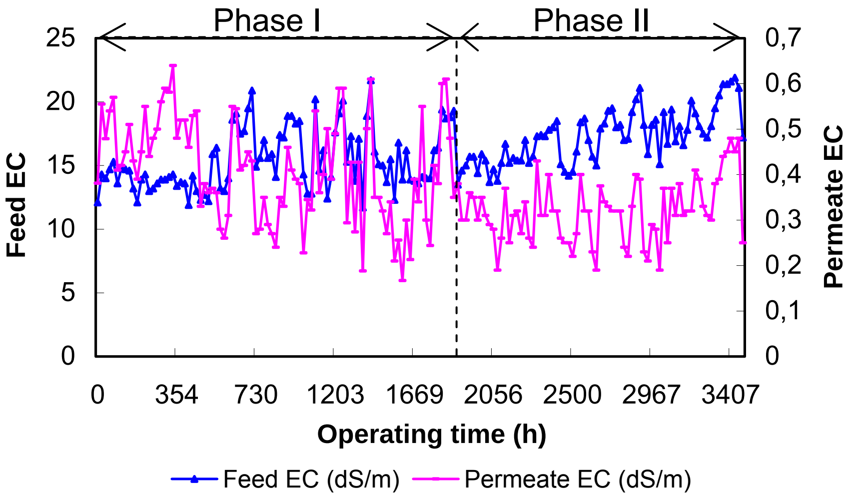


Figure 3 Variation of feed and permeate electrical conductivity with operating time

Membrane cleaning

After around 90 hours continuous operation, permeate flux decreased from 8.14 l/m^{2}h to 6.5 l/m^{2}h in Phase II, indicating the occurrence of membrane fouling. Membrane fouling was caused by the depositions of suspended and dissolved solids on the external membrane surface. Since the concentration factor achieved in Phase II was less than 4, declining of permeate flux was caused by bio fouling rather than scaling. Therefore only 1% sodium hypochloride alkaline agent was applied at trans-membrane pressure difference of 5 bar through the modules for membrane cleaning. After 10 minutes soaking, 30 l alkaline diluted by 600 l permeate was circulated in the membrane modules until the cleaner temperature raises to 40 °C. Elevated temperature was effective to break down and removed the bio fouling substances. Before and after each membrane cleaning cycle, permeate was used to rinse the membrane surface. After effective membrane cleaning with alkaline agent, a negligible permeate flux drop was observed. During the observation period, no acid cleaner was used for membrane cleaning.

CONCLUSIONS

Due to the development of high rejection reverse osmosis membranes to reject both organic and inorganic contaminants at rejection rate up to 99% and the properly designed membrane modules, reverse osmosis appears to be an interesting technology for landfill leachate treatment. A landfill leachate purification system equipped with the open channel spiral wound modules was studied.

During Phase I, effluent from the activated sludge followed by coagulation/sedimentation process was fed into the landfill leachate purification unit. An average permeate flux of 4.86 l/m^2.h and an average recovery rate of 53.4% were achieved during Phase I. After around two week's operation, permeate flux dropped dramatically from 6.5 l/m^2.h to 2.93 l/m^2.h (variation of leachate composition was not considered), showing the occurrence of severe membrane fouling.

Due to severe membrane bio fouling caused by SMP, the spiral wound modules were replaced and the biological pre-treatment followed by coagulation/sedimentation was discarded in Phase II. Raw leachate was fed directly into the reverse osmosis leachate treatment system in Phase II. After restarting operation, an average flux of 7.8 l/m^2.h was maintained at an initial trans-membrane pressure difference of 20 bar, increasing to 40 bar before membrane cleaning. Permeate flux obtained in Phase II was around 30% higher than that obtained in Phase I. In comparison with Phase I, the average trans-membrane pressure difference applied in Phase II decreased by around 30%. Poor filtration results obtained in Phase I further proved that it is better to treat landfill leachate under methnogenic phase through reverse osmosis technologies. An average recovery rate of 70% was achieved in Phase II.

During the observation period, dissolved solids were eliminated by 98.2%. Reduction rate of COD was greater than 99.5% and the permeate COD was always lower than 15 mg/l. More than 98% NH_4-N was reduced. Elimination rate of chloride was more than 99%. Concentrations of heavy metals and chloride in permeate were lower than the German leachate discharging standard.

After effective membrane cleaning with NaClO alkaline agent, a negligible permeate flux drop was observed. During the whole observation period, no acid cleaner was used for membrane cleaning.

REFERENCES

Anonymous (2007). Reference list of landfill leachate treatment with DT module technology. Accessed in 2007 at http://www.deponie-stief.de.

Baumgarten, G. and Seyfried, C. F. (1996). Experiences and new developments in biological pre-treatment and physical post treatment of landfill leachate. Wat. Sci. Tech., 34 (7-8), 445–453.

Christensen, T. H., Cossu, R. and Stegmann, R. (Eds.) (1992). Landfilling of Waste Leachate, Elsevier Applied Science, London (1992).

Cossu, R. (2007). Treatment of MSW Landfill Leachate by electrochemical oxidation. Accessed in 2007 at http://www.sardiniasymposium.it.

Laspidou, C. S. and Rittmann, B. E. (2002). A unified theory for extracellular polymeric substances, soluble microbial products, and active and inert biomass. Wat. Res., 36, 2711-2720.

Linde, K., Jönssone, A. and Wimmerstedt, R. (1995). Treatment of three types of landfill leachate with reverse osmosis. Desalination, 101 (1995), 21-30.

Peters, T. A. (1998). Purification of landfill leachate with membrane filtration. Filtration & Separation. Jan/Feb 1998, 33-36.

Peters, T. A. (2001), High advanced open channel membrane desalination. Desalination, 134 (2001), 213-219.

Rautenbach, R. and Linn, T. (1996). High-pressure reverse osmosis and nano-filtration, a "zero discharge" process combination for the treatment of waste water with severe fouling/scaling potential. Desalination, 105 (1996), 63-70.

Schwarzenbeck, N., Leonhard, K. and Wilderer, P. A. (2004). Treatment of landfill leachate - high tech or low tech? case study. Wat. Sci. Tech., 48 (11), 277–284.

York, R. J., Thiel, R. S. and Beaudry, E. G. (1999). Full-scale experience of direct osmosis concentration applied to leachate management, Proceedings Sardinia 99, Seventh International Landfill Symposium, Italy, Oct 1999, pp. 359.

Submerged membranes in wastewater lagoons: a new approach to wastewater reuse in agriculture

K. Teschner*, W. Hegemann*, C.F. Gökcay** and O. Komesli**

* Department of Water Quality Control, University of Technology Berlin, Straße des 17. Juni 135, 10623 Berlin, D (E-mail: Katharina.Teschner@TU-Berlin.de)
** Department of Environmental Engineering, Middle East Technical University, 06531 Ankara, TR (E-mail: cfgokcay@metu.edu.tr; okankomesli@yahoo.com)

Abstract Wastewater reuse is one major approach to solving the growing shortage of fresh water worldwide. Already applied in various countries different treatment technologies are used to fulfil the hygienic requirements defined by national or international laws and guidelines. In many rural areas in hot and arid regions wastewater treatment is often neglected due to the lack of investment capital. To grow agricultural crops, often recycled water of poor quality is used for irrigation.
Wastewater lagoons are easy to built and maintain at low costs. Membrane technology is an advanced technical process producing an effluent of high microbiological quality. In a hybrid system lagoons and membranes are brought together to supply safe irrigation water for dry climates.

Keywords Irrigation, membrane filtration, reuse, wastewater lagoon, wastewater

INTRODUCTION

Reuse of wastewater is expanding worldwide. Valuable drinking water for agricultural irrigation can be substituted by treated wastewater. To avoid health risks for farmers and consumers disinfection should be included in the wastewater treatment process to remove pathogenic micro-organisms. Possibilities range from chlorination, ozonisation, use of UV radiation to membrane filtration. Applying chlorine to wastewater can prove hazardous as remaining compounds can form carcinogenic by-products. UV radiation and ozonisation require prior conventional wastewater treatment and are therefore costly in investment and operating costs. Membrane filtration is the mechanical separation of liquids and solids. There is no formation of by-products and investment costs have dropped significantly over the past decade.

Membranes are used worldwide in conventional wastewater treatment. Usually they are applied as MBR (membrane bioreactor) systems in wastewater treatment plants. Due to dropping prices and a comparatively small footprint in their favour, there has been a steady development over the past decade. But membrane filtration is an old technology which can be applied in various fields. Depending on the permeation mechanism (pore size or diffusion) and aggregate condition (liquid or gaseous) membranes can among others be used in liquid/ liquid separation for reverse osmosis (desalinisation). Filtration ranges from nanofiltration to microfiltration. Low-molecular substances can pass as permeate whereas colloids and partly macro-molecules are retained. The filtration process needs a transmembrane pressure difference, which is either achieved by hydrostatic pressure or vacuum pumps. Problems of the conventional dead end flow, like clogging of the membrane surface due to filter cakes, have been reduced by using cross flow filtration, which keeps the covering layer at a constant level.

Wastewater lagoons are simple systems for efficient wastewater treatment applied worldwide. They are easy to construct and to maintain. Their investment and running costs are low compared to conventional wastewater treatment plants. Depending on their size, disinfection can be achieved. Long retention times cause faecal coliform die-off. Lagoon effluent is used for restricted, but also for unrestricted irrigation. But current lagoon design uses large footprints. Naturally aerated lagoons are shallow and have large surfaces. This leads to massive evaporation losses.

In a new hybrid system membrane modules are submerged in wastewater lagoons (figure 1) slightly altering the MBR process. By adding the submerged membranes, the lagoon will automatically be turned into an technically aerated lagoon as constant aeration is used for flushing the membrane surface. The footprint of wastewater lagoons can be reduced as the biomass is retained in the lagoon. The addition of membrane units allows biomass concentrations up to 15 g/L compared to around 3 g/L in the conventional activated sludge process. Evaporation losses can be cut down by minimising the lagoon surface. Safe irrigation water is produced by the installation of the desinfection step.

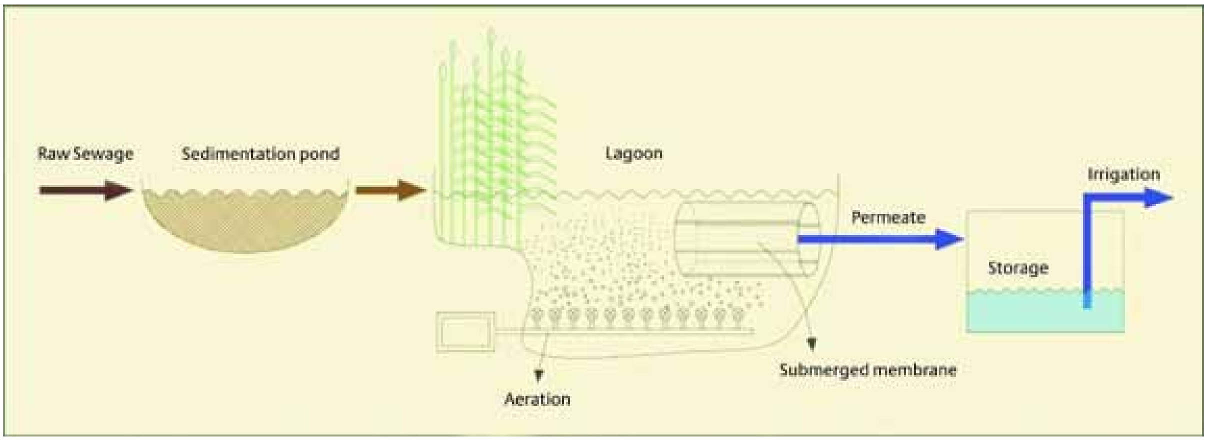


Figure 1. Application of submerged membranes in wastewater lagoons

METHODS AND MATERIALS

In cooperation with the Middle East Technical University (METU) in Ankara, Turkey, the University of Technology Berlin (TUB) is investigating the application of submerged membranes in wastewater lagoons. Research aims are the performance of the hybrid system regarding removal of COD and BOD as well as nutrients and pathogens. Pilot plants are in operation in Germany and in Turkey. Wildberg (Germany) is situated in a continental temperate climate zone, Ankara (Turkey) in the Central Anatolian Highland which has a continental steppe (semiarid) climate. Annual precipitation in both cases is less than 500 mm. There is a difference in temperature patterns, Ankara is characterized by hot summers and extremely cold winters.

Results of the project will lead to design criteria for membrane lagoons adjusted to different climatic conditions. High water temperatures have a positive effect on membrane filtration. One focus is nitrogen. In the German wastewater lagoon nitrification should be enhanced to reduce the amount of ammonia going into the surface water. In the Turkish plant nitrification should be supressed to keep ammonia in the irrigation water in order to safe artificial fertiliser.

Pilot Plant Germany

Two test units are installed on site on the wastewater lagoon Wildberg, Germany, which is composed of four separate lagoons serving about 750 inhabitants producing about 70 m³/d of wastewater. It was designed according to the German design recommendations given by the DWA (1989) and was installed in the mid 1990ies, when a large number of new treatment facilities throughout former East Germany were constructed. The influent has to pass a screen. The first two lagoons are technically aerated.

Both membrane units installed are ultrafiltration plate modules with a pore size of 0.035 µm containing 6 m² membrane surface each. These are standard units which are normally used for single houses. They are operated on a gros flux of 6 L/m²h, filtration is driven by vacuum pumps.

One module is submerged in the second lagoon (volume of 700 m³), the other one is installed in a 1 m³ container, also being fed from the second lagoon (figure 2). The additional tank of smaller volume was necessary in order to increase the biomass concentration. The lagoon module is operated intermittedly with 100 sec operation and 100 sec relaxation cycles at a flow of 36 L/h giving a net flux of 3 L/m²h. This comparitively low rate was chosen, because of the biomass structure in the lagoon. The biomass concentration is very low around 1 g/L. This is a problem, as small biomass particles cannot easily be removed from the membrane surface by flushing air and suspended activated sludge flakes. Hydraulic retention time in the lagoon is infinite due to the small size of the membrane unit.

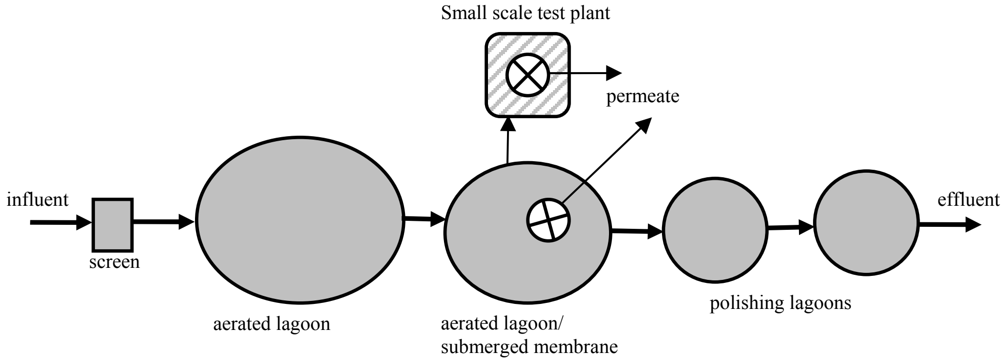


Figure 2. Schematic diagram of the pilot plant Wildberg (Germany)

The container was initially filled and operated on lagoon water, but the biomass was slow growing apart from the rapid built up of an algae population. This proved difficult for the membrane and biomass concentration never increased to more than 3 g/L. In a second step activated sludge from a local conventional wastewater treatment plant was used as a start up in the container containing a biomass concentration of about 7 g/L. The container module is also is operated intermittedly, but at a higher net flux of 5 L/m²h with 10 min operation and 2 min relaxation cycles at a flow of 36 L/h. Hydraulic retention time in the container is about one day. The container pilot plant is controlled by level sensors operating the influent pump submerged in the second lagoon.

Usually in Germany effluent of wastewater treatment plants is discharged into surface waters where remaining pollutants are diluted. Compared to COD and BOD where the input is limited – 150 mg/L COD and 40 mg/L BOD for plants up to 1000 inhabitants – there is no limit on nitrogen. However ammonia and nitrate lead to eutrophication. If nitrification does not take place in the treatment plant it is shifted into the stream or river causing oxygen depletion.

Pilot Plant Turkey

The METU used to operate a decentralised wastewater lagoon plant before the campus was connected to the central Ankara treatment plant more than a decade ago. Unfortunately for the project the disused lagoons have a volume of about 3000 m³ and were too big to accommodate the pilot plant. Therefore the former pretreatment facility with a total volume of 110 m³ was chosen to be rebuilt as ‘lagoon’. The pilot plant (figure 3) is serving about 1000 inhabitants.

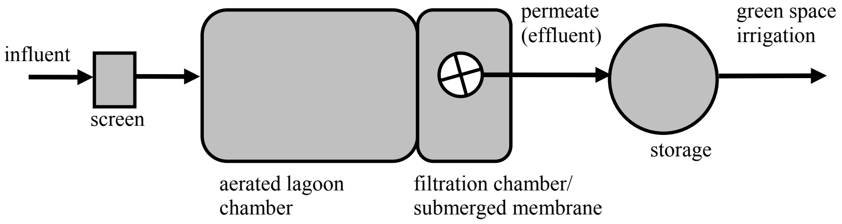


Figure 3. Schematic diagram of the full-scale pilot plant Ankara (Turkey)

About 200 m³/d are taken from the main sewer. A side stream of wastewater from the academic village and from dormitories is pumped from the sewer line to the plant. The incoming wastewater is screened as coarse particles in the influent can damage the membranes. Screenings larger than 3 mm are separated and discharged as solid waste. The influent goes into an aerated lagoon compartment without further pretreatment. The lagoon chamber is separated from the filter chamber by a wall. Both chambers are connected by five screw operated valves. To ensure circulation in the system, also a recirculation pump is installed, which pumps sludge from the filter chamber back to the lagoon tank. Filtration is driven by a vacuum pump. The permeate is collected and stored in a closed reservoir before being used for irrigation of green spaces.

The submerged membrane unit is a rotating plate membrane with a pore size of 0.038 µm. Along with the coarse aeration from the flushing blower, the rotation of 2.5 rpm creates a cross flow across the membrane surface. The total surface area is 540 m². The vacuum pump is operated intermittedly with 8 min operation and 2 min relaxation. The flow was first adjusted to 7.5 L/h, giving a flux of around 10 L/m²h. In the past year the flow has been reduced to 5 L/h. Hydraulic retention time is 18 to 22 h. Excess sludge from the filter chamber is removed irregularly as needed.

DISCUSSION AND RESULTS

Both pilot plants have been in operation for nearly two years now. It has been proven that the hybrid system works efficiently. With growing biomass concentration the ‘sludge’ in the lagoon does not settle well as also decribed by Ben Aim and Semmens (2002), because of the variety of bacteria and micro-organisms. They do not any longer depend on the ability to settle as their residence is defined by the sludge residence time. Flux decline over operation time has occurred in both pilot plants. Chemical cleaning was necessary to restore the membranes.

Optimisation of the operating cycle in future should lead to the longest possible intervalls between cleaning. Generally there still is the need for more understanding of the very complex relationsships and interactions in the membrane process: composition of the wastewater, particles and their distribution, the characteristics of bio-solids, retention time are only some among many factors to be dealt with.

Results of the pilot plant Wildberg

Compared to the effluent quality of the Wildberg lagoon system a much better quality is achieved by the added submerged membranes. The influent COD concentration ranges at a high level between 1500 to 2000 mg/L, which is not unusual for the rural north-eastern region in Germany because of low water consumption. According to German requirements the effluent of the lagoon plant is not to exceed a COD of 150 mg/L. But especially in wintertime the efficiency of lagoons can drop as the activity of micro-organisms slows down due to low temperatures. This is also the case in the Wildberg plant. In the lagoon effluent often more than 200 mg/L are measured, whereas COD in both permeates does not exceed 100 mg/L. COD elimination by the membranes averages 94 % compared to an average of about 82 % in the lagoon (figure 4).

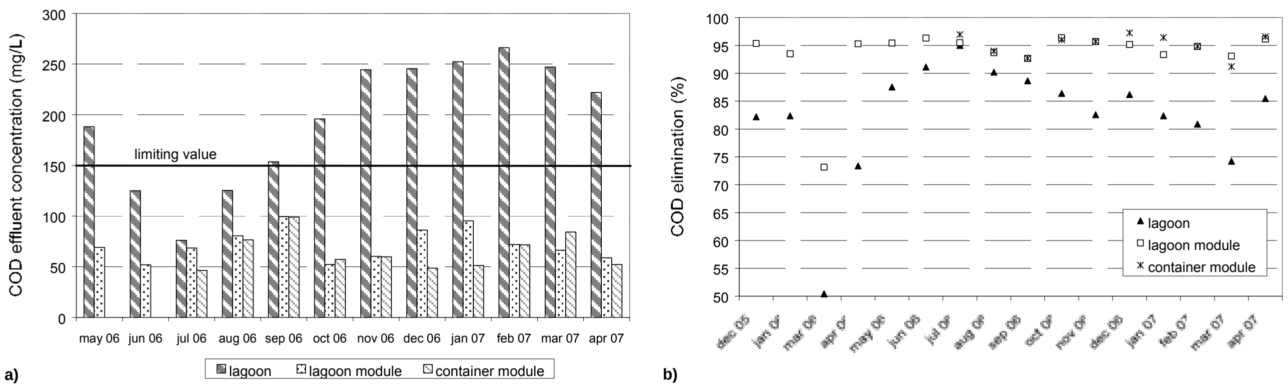


Figure 4. (a) Effluent COD concentration, monthly average value (b) COD elimination, monthly average value, pilot plant Wildberg

In the influent BOD_5 concentrations between 500 to 800 mg/L were measured. BOD_5 in the effluent should not exceed 40 mg/L. With membrane filtration the effluent BOD_5 could be reduced to concentrations less than 10 mg/L. BOD_5 in the lagoon effluent was around 40-70 mg/L. This is often due to high algae concentrations. Usually samples are not filtered prior to measurements.

The results for ammonia also show a better performance of the membrane units. Nitrification is higher in summer and continues into wintertime, when it has already stopped in the lagoon (figure 5). This can not be achieved by conventional wastewater lagoons as nitrifiers are normally washed out and can not regrow quickly in cold temperatures. Ben Aim and Semmens (2002) describe as general feature of MBRs that slow growing micro-organisms are retained. This is also the case for submerged membranes in lagoons.

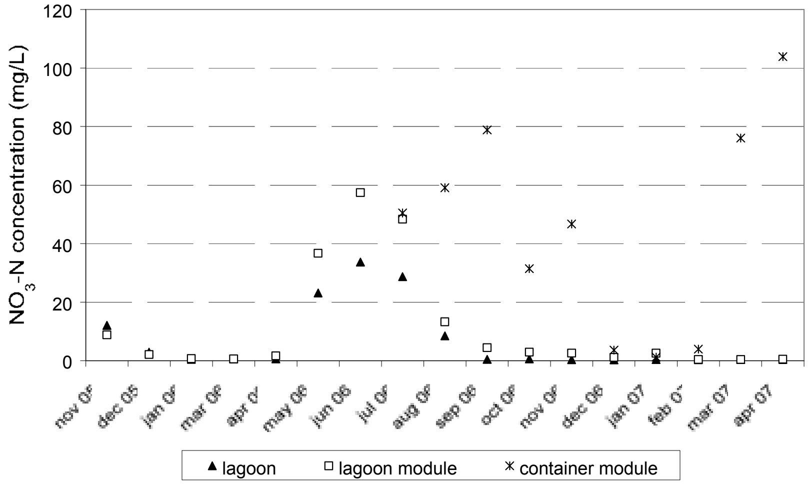


Figure 5. Nitrate concentration in effluent and permeates, pilot plant Wildberg

During the investigation the focus shifted towards removal of pathogens. Annual precipitation has been decreasing considerably in the region of Berlin over recent years, making wastewater reuse also a topic in Germany. A first step is the support of the local water balance. In this case pathogen removal is not necessary. But demands for irrigation water are growing. In general effluent of lagoons in warm climates with a hydraulic retention time of 20 days would be suitable for unrestricted irrigation according to the old WHO guidelines (1989). The new WHO guidelines (2006) do not formulate limiting values, but give a calculation of DALYs (Disability-adjusted Life years) with 10^{-6} DALY loss pppy (per person per year) being the tolerable additional disease burden. This new system is not easily palpable for the practioners, that is why a 'Guide to the Guidelines' seems appropriate (Mara, 2006).

Even though there is a 'natural' disinfection in wastewater lagoons due to temperature, hydraulic residence time, algal toxins, sedimentation and sunlight as Davies-Collies (2006) describes it, it can not compare to the results by membrane filtration. In autumn 2006 faecal coliforms were measured over a period of seven weeks according to the 76/160/EWG method used for the monitoring of bathing waters. Lagoon effluent regularly contained 110000 faecal coliforms/100 mL, whereas the count in both permeates were less than 30 faecal coliforms/ 100 mL. The applied method cannot detect less than 30 counts/ 100 mL. Therefore it can be assumed that the actual count was even lower. Surface waters containing no more than 100 faecal coliforms/ 100 mL are regarded safe to swim in according to the EEC bathing water directive (1976).

Results of the pilot plant Ankara

One aim of the operation of this pilot plant was the supression of nitrification and reduction of carbon only. Initially this was thought to be achieved by applying a very high flux to reduce hydraulic retention time as well as sludge age. This way nitrifying bacteria would not have had the opportunity to establish. But the flux is limited by the performance of the membrane and could not be increased to more than 10 L/m²h. Then the oxygen input was reduced to less than 0.5 mg/L in the lagoon chamber to deprive the nitrifiers of adequate supply. This kind of operation had implications on the COD removal. COD in the influent averages around 480 mg/L, there are very few peaks close to 1000 mg/L. In the effluent it averages around 16 mg/L. When the oxygen input was lowered, COD effluent concentration went up to more than 20 mg/L (figure 6).

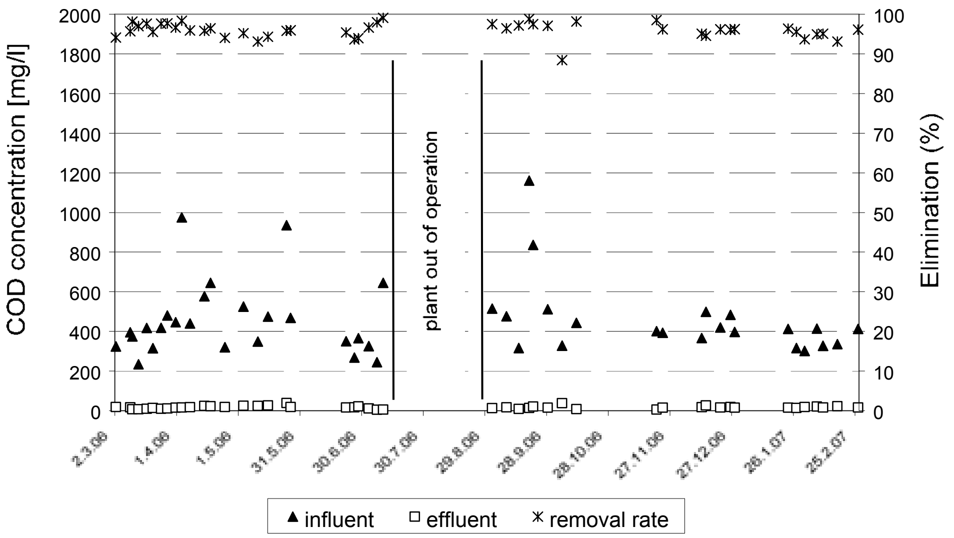


Figure 6. COD concentration and elimination, pilot plant Ankara

Results from the pilot plant show that nitrification cannot not be suppressed in the system. In practise the sludge age was infinitely long and nitrifiers were always present. Nitrification could only be reduced by reduction of the aeration in the lagoon chamber from 2.0 mg/L to 0.5 mg/L. Partial nitrification led to ammonia and nitrate concentrations in the effluent which can be assimilated by the crops (figure 7). Crop requirements of nitrogen are very complex and vary, depending on the type of plant and the specific growing stage during the vegetative period. The efficient management of crops, soil and wastewater effluent application rates can stimulate growth and prevent the effects of nitrate pollution and overfertilisation (Vazquez-Monteil et al., 1995). The lawn at the METU Teknopark throve well with irrigation from the pilot plant effluent.

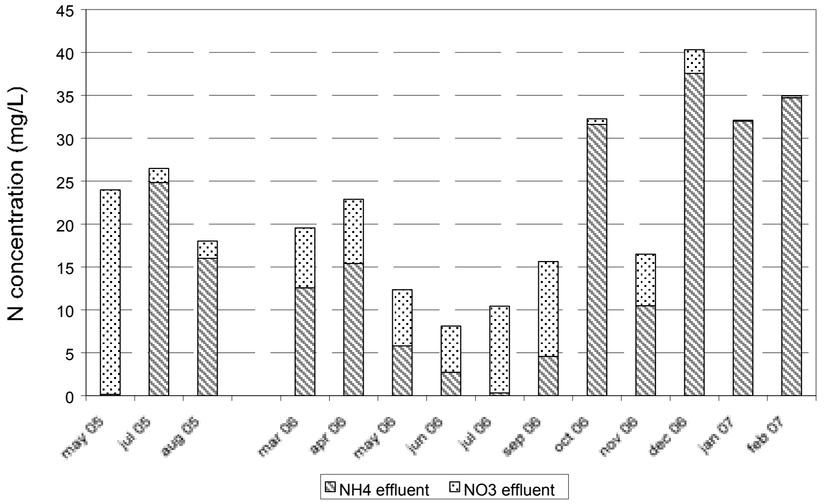


Figure 7. Nitrogen in effluent, available for plant uptake (NH_4-N + NO_3-N concentration), pilot plant Ankara

The design of a separate filter chamber, though preferred by membrane manufacturers because of easier accessability to the units in case of maintenance and chemical cleaning, has proven a disadvantage. Problems occur recirculating the contents of the filter chamber to the main lagoon. Biomass concentration is about twice as high in the filter chamber (figure 8) and the use of a recirculation pump adds up in operational costs (energy consumption). Former studies showed the possibility to increase the biomass concentration to about 30 g/L, but recommended not to exceed 15 g/L because of the dramatic decrease of oxygen transfer concentrations (Günder, Krauth, 1999). Biomass concentration should be limited to 15 g/L in warmer summertime and should be reduced during cold water temperatures in winter with increasing viscosity.

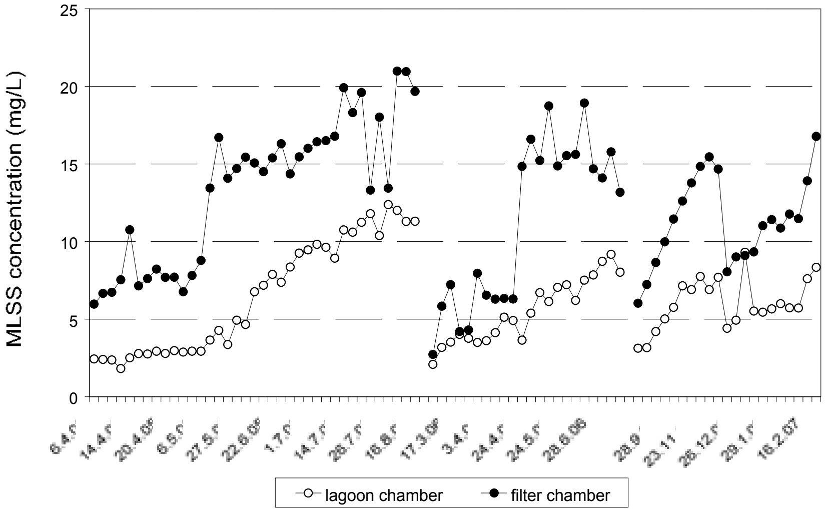


Figure 8. Development of biomass concentration, pilot plant Ankara

Biomass is also important for pathogen removal. Many viruses are smaller than the pores of ultra- or microfiltration membranes. Using the very small norovirus as well as coliphage as an alternative index for virus, which were both smaller than the applied microfiltation membrane, Oota et al.

(2005) measured efficient norovirus and coliphage removal. They reason, that in both cases the virus attached themselves to activated sludge particles and therefore cannot pass the membrane.

In the pilot plant faecal coliforms were chosen as indicators for pathogen removal. In 1989 the WHO recommended no more than one human intestinal nematode egg per litre and no more than 1000 faecal coliform bacteria per 100 mL as microbiological quality for unrestricted irrigation. Usually the count in the pilot plant was zero or less than 5 faecal coliforms per 100 mL. If the count went slightly up this was due to recontamination of the effluent pipe, which then had to be disinfected. If the count went up further it indicated a leakage problem in the system which had to be fixed. Still the monitored pathogen removal allows unrestricted reuse for agricultural irrigation and is well suited for irrigation of the lawns of the METU Teknopark.

Development of design criteria

One of the main aims of the project is the development of design criteria for membrane supported lagoons. The systems is planned for up to 1000 inhabitants (between 100-200 m^3/d depending on the water consumption) to be applied locally in rural areas.

Generally the MBR process applies a sludge age of 20-25 days and a hydraulic retention time of a few hours. The retention of biomass including bacteria and nematode eggs depends on the pore size of the membrane. In a conventional wastwater lagoon slow growing micro-organisms are washed out. Retention times in lagoons can vary according to their design. Mara (2000) calculates a hydraulic retention time of about 20 days in warm climates for the removal of faecal coliforms and helminth eggs. The hydraulic design of the hybrid system is planned for a retention time of two days giving an extra day as buffer. The plant should consist of a settling lagoon followed by a second lagoon, where the membrane units are submerged, preferably along an accessible bridge allowing maintenance and removal for cleaning. The effluent is stored in a covered storage tank. The membranes should be submerged in the lagoon and not be installed in a filter chamber as the circulation of biomass is a problem in a 2-chamber-setup. Also the air of the flushing blower cannot be used for lagoon aeration which adds up in investment costs for a conventional aeration system. Flushing aeration moves the lagoon surface making it harder for insects to breed, as they prefer still waters.

If a lower effluent quality is required for some applications (i.e. car washing, toilet flushing, fire protection and fire fighting systems) it is also possible to pass a side stream through the membrane to be used for unrestricted irrigation (agriculture, horticulture, urban recreational and open space watering, residential garden watering), or clothes washing (which is included in reuse purposes in Australia).

CONCLUSION

A hybrid system of submerged membrane units in wastewater lagoons works well for producing an effluent suitable for unrestricted irrigation. Despite the technical procedure the system can be applied locally in decentralised wastewater treatment. The combination creates many synergy effects. Lagoon surface and volume can be reduced as the biomass concentration is increased. This minimizes evaporation losses. Operation with partial nitrification helps saving artificial fertiliser. Flushing air for the membranes is used to provide oxygen needed by the micro-organisms. This saves investment and running costs. Renewable energy can cover the operational demand. Cutting down costs and keeping the membrane units as simple as possible allows implementation also in developing countries.

ACKNOWLEDGMENTS

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REFERENCES

Ben Aim, R.M. and Semmens, M.J. (2002). Membrane bioreactors for wastewater treatment and reuse: a success story. Water Science Technology, 47(1), 1-5.

Davies-Colley, R. (2006). Pond disinfection. In: Pond Treatment Technology, A. Shilton (ed.), London: IWA Publishing, pp.100-136.

European Economic Community (EEC) (1976). Council Directive of 8 December 1975 concerning the quality of bathing water, 76/160/EEC. Official Journal of the European Community.

Grundsätze für Bemessung, Bau und Betrieb von Abwasserteichen für kommunales Abwasser (Manual for design, construction and operation of wastewater lagoons for municipal wastewater) (1989). DWA Arbeitsblatt A 201, Hennef.

Günder, B. and Krauth, K. (1999). Replacement of secondary clarification by membrane separation-results with tubular, plate and hollow fibre modules. Water Science Technology, 40(4-5), 311-320.

Mara, D.D. (2000). The production of microbiological safe effluents for wastewater reuse in the Middle East and North Africa. Water, Air and Soil Pollution, 123, 595-603.

Mara, D.D. (2006). http://www.personal.leeds.ac.uk/~cen6ddm/Reuse/Guide2Guidelines.pdf, informal publication (draft)

Oota, S., Murakami, T., Takemura, K. and Noto, K. (2005). Evaluation of MBR effluent characteristics for reuse purposes. Water Science Technology, 51(6-7), 441-446.

Vazquez-Montiel, O., Horan, N.J. and Mara, D.D. (1995). Effects of nitrogen application rates using treated wastewaters, on nitrogen uptake and crop yield based on pot trials with maize and soyabean. Water Research, 29(8), 1945-1949

World Health Organisation (WHO) (1989). Health Guidelines for the Use of Wastewater in Agriculture and Aquaculture. Technical. Rep. Series No. 778, Geneva, Switzerland.

World Health Organisation (WHO) (2006). Guidelines for the Safe Use of Wastewater, Exceta and Greywater, Volume 2: Wastewater Use in Agriculture. Geneva, Switzerland.

Evaluation of innovative operation concept for flat sheet MBR filtration system

L. Weinrich*, A. Grélot**

*A3 Water Solutions, Magdeburger Str. 16a, 45881 Gelsenkirschen, Germany
(E-mail: lutz-weinrich@a3-gmbh.com)
** Anjou Recherche, Chemin de la Digue, BP 76, 78603 Maisons-Laffitte Cedex, France
(E-mail: aurelie.grelot@veolia.com)

Abstract
One of the most limiting factors for the extension and acceptance of MBR filtration systems for municipal and industrial wastewater is the impact of membrane fouling to maintenance, operation and cleaning efforts. One field of action in the European Research Project “AMEDEUS” is the development and testing of MBR module concepts with innovative fouling-prevention technology of 3 European module manufacturers.

This article deals with the performances of the flat-sheet modules by A3 Water Solutions GmbH in double-deck configuration evaluated over 10 months in Anjou Recherche under typical biological operation conditions for MBR systems (MLSS=10 g/l; SRT=25 days). By using a double-deck configuration, it is possible to operate with a net flux of 25.5 l/m².h at 20°C, a membrane air flow rate of 0.20 Nm³/h.$m^2_{\text{of membrane}}$ to achieve a stable permeability of around 500-600 l/m².h.bar. Additionally, it was observed that it is possible to recover the membrane performance after biofouling during operation without an intensive cleaning and to maintain stable permeability during peak flows.

The evaluated concepts for equipping and operating MBR systems will be applied to several full-scale plants that are constructed by A3 Water Solutions GmbH.

Keywords
Aeration, Cleaning, Flat sheet membrane, Fouling, Membrane bioreactor

INTRODUCTION

Membrane separation processes have been developed to replace the secondary clarifiers traditionally used in activated sludge treatment systems since the early 70s (Smith *et al.*,1969). MBR systems feature many advantages over conventional processes with their highly improved effluent quality, increased organic loading, reduced footprint occupation and sludge production (Tazi-Pain *et al.*, 2001; Judd, 2006).

The first generation of membrane bioreactors were operated with organic or inorganic tubular membranes placed in external recirculation loops. Immersed bioreactors have been developed in the 80s following the idea of Yamamoto *et al.* (1989) in order to simplify the use of these systems and to reduce operating costs. In this configuration, the membranes are directly immersed in the tank containing the biological sludge and the treated permeate is extracted (Côté *et al.,* 1998). Many different membrane filtration systems using flat sheet, hollow fiber or tubular membranes with micro- or ultrafiltration membrane have been developed for the biomass separation during the last decade. However, in most cases, membrane fouling remains the most serious problem affecting system performance and limiting the widespread application of MBRs. Indeed fouling leads to a decline in permeate flux, requiring more frequent membrane cleaning leading and more frequent replacement, which then increases operation costs.

A3 Water Solutions GmbH, as a supplier of MBR plant technology, developed a flat sheet module concept for the use in MBR applications. Its simple modular design allows stacking the modules, easy utilisation and good filtration performances. The present article describes trials that were accomplished in Year 2006 / 2007 in collaboration between A3 Water Solutions GmbH and Anjou Recherche, the Research Center of Veolia Water, within the framework of the European project "AMEDEUS".

Throughout the study, the biological operating conditions were kept constant and the attention was focused in optimizing the hydraulic operating conditions and in the cleaning strategy. The trials cover three aspects of the novel flat-sheet MBR filtration system:

- Optimization of the hydraulic performances,
- Membrane performances after biological stress and during peak flows,
- Impact of the double deck configuration, 2 modules stacked on top of each other.

MATERIAL & METHODS

The A3 Water Solutions GmbH concept for MBR filtration modules is based upon the arrangement of several flat-sheet membrane cushions to a block module as shown in Figure 1. The space between the plates is previously selected, to ensure maximum filtration efficiency. Aeration system operating with medium-size bubbles is installed below the filtration module. The resulting turbulence in the gas-liquid mixture ascending through the spaces between the membrane plates enables to detach the deposits. The plates are fixed by a coated housing that seals the membrane flat sheet, and also allows on the filtrate suck-off through filtrate channels. Filtration occurs from outside to inside of the plates. The membrane material is PVDF with an average pore size of 0.2 µm. The clean water permeability is above 1.500 l/m².h.bar.

Because of the modular concept, arranging of a double- or even triple-decker is very simple. The membrane area of A3´s standardized modules is 3, 6, 20 and 70 m². Additionally, all other module sizes are possible by altering the bag size or quantity according to the customers' requirements. In this application, two 70 m² modules are arranged as a double-deck, with an aeration module at the bottom, consisting of membrane tube aerators. Every module is equipped with its own filtrate pipe, in order to enable the evaluation of the impact of double-deck configuration.

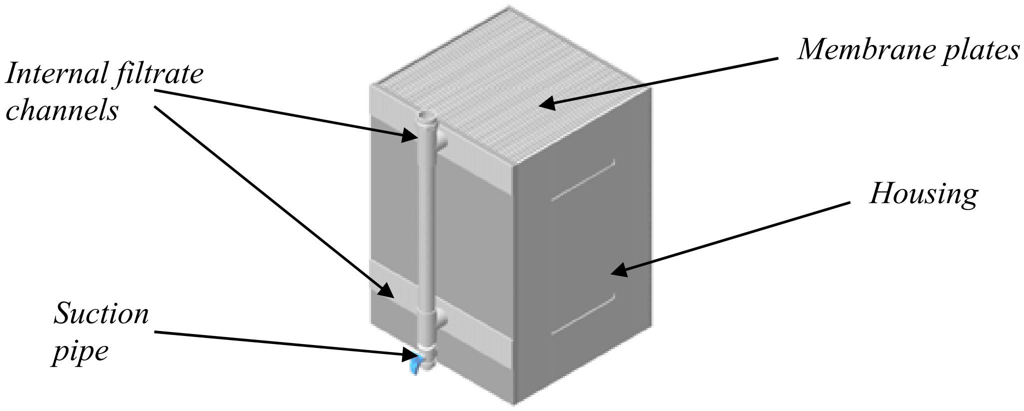

Figure 1 A3 module

Anjou Recherche has built a flexible pilot platform as shown in Figure 2 that was operated from May 2006 to April 2007 to evaluate the A3 technology. The pilot is fed by municipal waste water from the town of Maisons Laffitte by a pump after screening through a 1mm drum screen. The mean feed water characteristics are given in Table 1.

Table 1 Mean feed water quality (around 150 Samples)

Parameter	COD (mg/l)	N_t (mg/l)	$N\text{-}NH_4^+$ (mg/l)	TSS (mg/l)	pH
Municipal waste water	520	57	39	169	7.7

The pilot is composed of a biological tank (1.6m^3 of sludge volume) and a membrane tank (2.6 m^3 of sludge volume) as shown in Figure 3. The wastewater arrives at the bottom of the biological tank in which the nitrification and the denitrification occur. This tank is intermittently aerated and agitated with an impeller to ensure that sufficiently high oxygen concentrations can be reached for biological processes. Mixed liquor is circulated with a pump from the biological tank to the filtration tank. The filtration tank consists of an aerated tank in which two A3 water solutions modules are immersed. A pump is used to extract the permeate water from the membrane. The permeate water is collected in a storage tank and can be partly re-circulated to the biological reactor or in the membrane tank. The concentrated mixed liquor is returned to the biological tank. The biological operating conditions are kept constant during all the trials in order to consider only the impact of the hydraulic operating conditions. The chosen biological operating conditions are the typical conditions of a treatment for membrane activated sludge processes. The mean biological operating conditions are given in Table 2.

Figure 2 Pilot platform

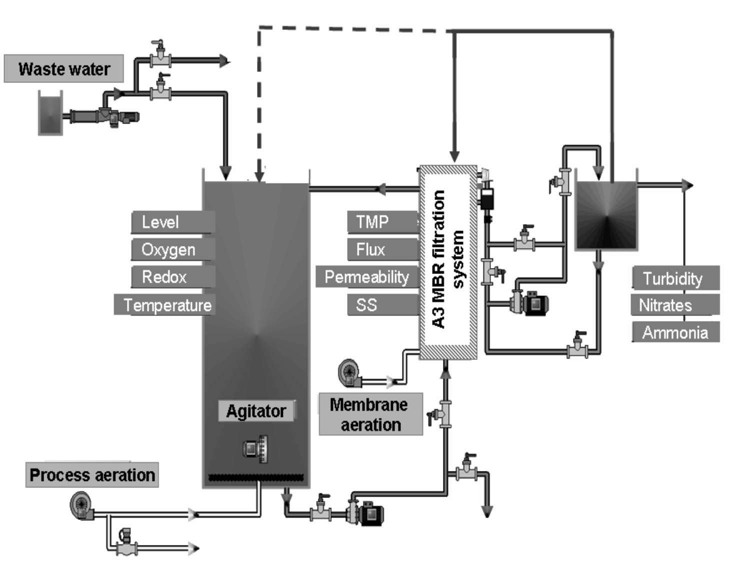

Figure 3 Pilot plant design

Table 2 Mean biological operating conditions

Parameter	Design	Average
SRT (days)	25	28
Volumetric loading rate (kg COD.m^{-3}.d^{-1})	1.3	1.2
F/ M ratio (kgCOD.kg MLSS^{-1}.d^{-1})	0.1	0.11
Sludge concentration evaluated in total (g/L)	10	10.5
Sludge concentration measured in the biological tank (g/L)	8.7	9.1
HRT (h)	8.4	11.5

The hydraulic performances of the membrane are monitored automatically and the pilot is also equipped with a data acquisition system. This allows the membrane performances (transmembrane pressure, filtration flow rate, temperature), the quality of the permeate (ammonia and nitrate concentration) and the characteristics of the sludge (suspended solids, redox, oxygen concentration) to be continuously recorded.

In parallel, the raw water and permeate analyses were performed every day to evaluate the treatment performances of the pilot unit. The characteristics of the sludge are also analyzed regularly in order to see if the fouling of the membrane is due to some biological stress. The sludge characteristics and the frequency of the measures are given in the Table 3.

Table 3 Mean sludge characteristics

Parameter	Average	Minimum	Maximum	Samples
Sludge concentration in biological tank (g/l)	9.1	3	14	148
COD of the sludge supernatant (mg/l)	46.2	10	189	147
D50 (µm)	27.5	24	33.2	25
Viscosity (Pa.s)	0.008	0.004	0.018	37
CST (s)	12	6.7	61	40
Specific resistance (10^{12} m^{-1}.kg^{-1})	10.6	5	24	27
Polysaccharides concentration (mg/l)	6.4	0	23	32
Proteins concentration (mg/l)	18.2	9	32	28

RESULTS AND DISCUSSION

Biological treatment

The trials started in June 2006 for a duration of ten months. The quality of the treated water during the six first months is given in Table 4.

The biological treatment was highly efficient for the whole trials period: COD removal was always greater than 94%, the total suspended solids were totally removed with a very low turbidity in the treated water (< 0.1 NTU) and coliform concentration in the permeate was only on average 18 nb/100ml corresponding to a bacteria removal of 7 log.

The average total nitrogen removal is only ca. 55% since the start of the trials due to the excessive membrane aeration for the quantity of available activated sludge volume. The nitrogen removal can be improved on a real station. This pilot design was chosen in order to fit to all module sizes that will be tested in the framework of the AMEDEUS project. Indeed, the other European manufacturers plan to deliver modules with a smaller membrane area.

Table 4 Quality of the treated water

Parameter	Average	Minimum	Maximum	Samples number	Mean removal rate
[COD] (mg/L)	16.4	4	69	151	96.5%
[Nt] (mg/L)	24.9	12	79	138	56.3%
Total coliforms (nb/100mL)	18	0	110	10	7.2 log
Thermotolerant coliforms (nb/100mL)	1.5	0	6	10	6.7 log

Filtration system

The initial clean water membrane permeability was measured around 1600 L/m^2.bar at 20°C with fluxes between 15 and 40 L/h.m^2 at 20°C.

In the first phase of the trials, the filtration conditions were optimized for the modules. Initially, the pilot operated with standard conditions: an instantaneous flux of 13 l/m^2.h at 20°C; Filtration / Relaxation cycles of 8 min / 2 min and a membrane air flow rate of 0.34 Nm^3/h.m$^2_{\text{of membrane}}$. The net flux was increased step by step from 10 to 26 l/m^2.h at 20°C by 3-5 L/m^2.h increments at 20°C. The permeability decreased quite fast from 1.000 to 800 l/m^2.h.bar at 20°C due to the evolution of the sludge quality and stabilized around 550 l/m^2.h.bar at 20°C with a net flux of 26.4 l/m^2.h at 20°C as shown in Figure 4. A higher flux was not achievable without a rapid decrease of the permeability.

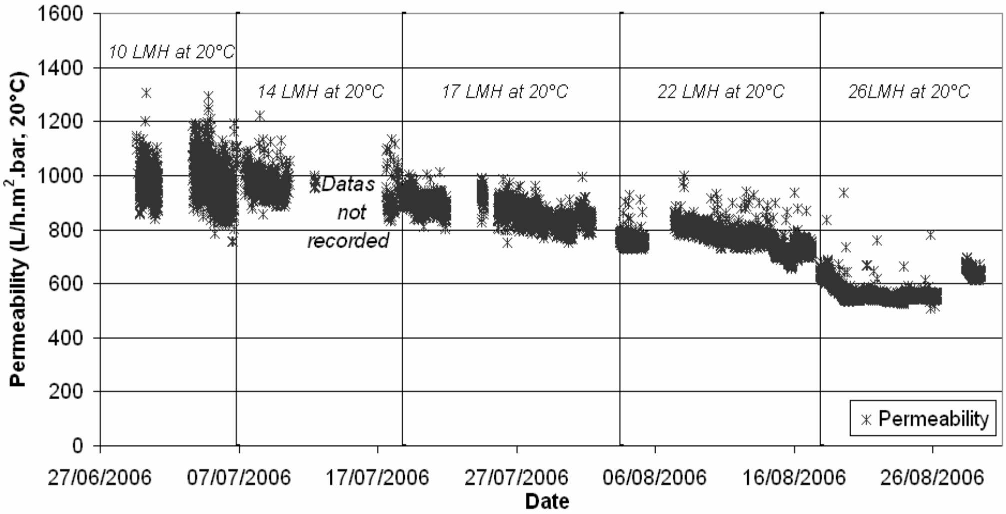

Figure 4 Optimization of the filtration flux

The cleaning strategy was then optimized to enable to reach a stable permeability with a net flux of 25.5 L/h.m^2 with a lower membrane air flow rate of only 0.20 Nm^3/h.m$^2_{\text{of membrane}}$ as shown in Figure 5.

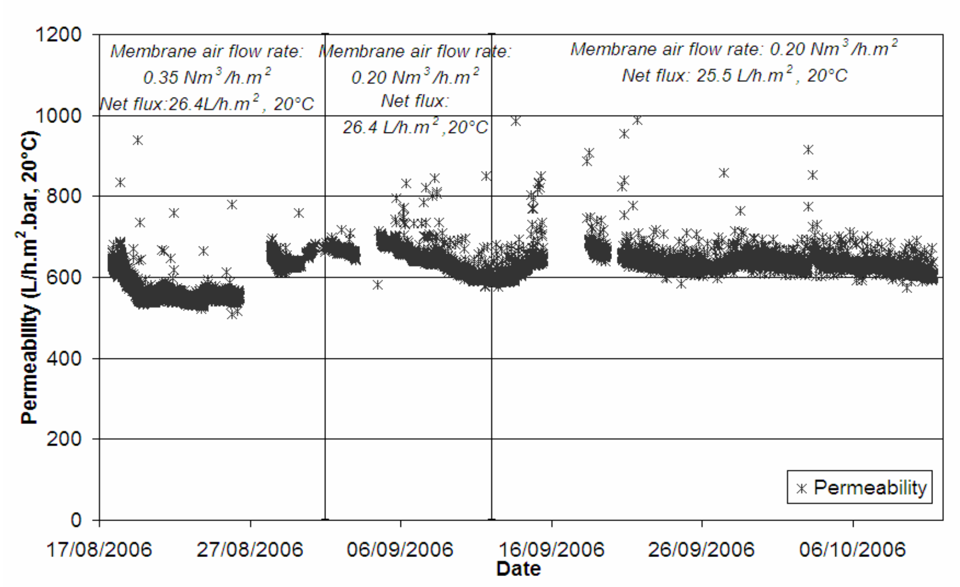

Figure 5 Optimization of the membrane cleaning strategy

Following this period, some biological disruptions (intense foaming) occurred that caused a permeability drop to 250-300 L/h.m^2.bar at 20°C. With an adapted cleaning strategy and without an intensive cleaning, the membrane permeability was successfully recovered with the same net flux and same membrane air flow rate as shown in Figure 6. The fouling mechanisms due to the changes in sludge quality are not yet clearly identified and more investigations are needed.

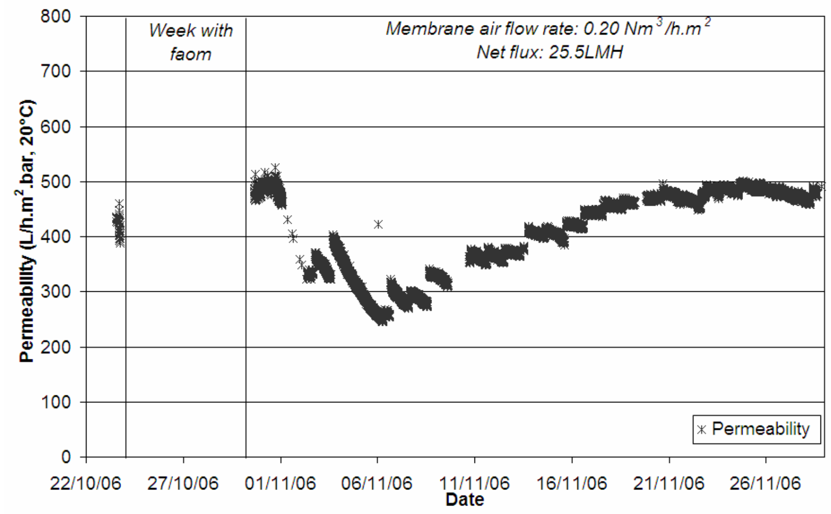

Figure 6 Recovery of the membrane permeability

One more field of interest was to see if the fouling-stabilized system can handle peak flows without a drop of the permeability. To investigate this, peak flows were programmed to occur twice a day during two hours (between 9:00 and 11:00 and between 15:00 and 17:00). The instantaneous filtration flux was increased up to 1.5 times the initial filtration flux. At the beginning (zones 1 and 3), only the filtration flux was increased and then the feed flow rate was increased proportionally to the filtration flux (zones 2 and 4). The membrane permeability evolution is shown in the Figure 7.

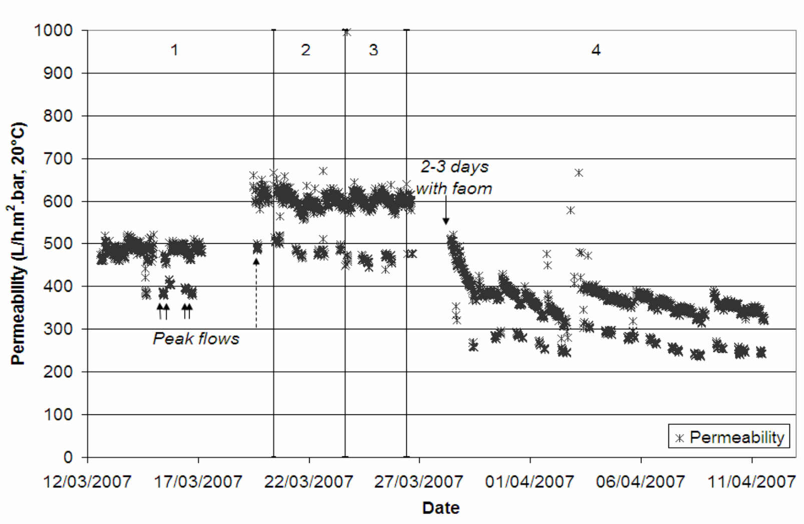

Figure 7 Peak flows

During the peak flows, a decrease of the permeability was observed but the same membrane performance was recovered after the peak flows. No loss of permeability was observed over the first two weeks of peak flow test. On 26-29 March 2007, foam appeared in the pilot and some sludge was lost causing a decrease of the suspended solids in the process from 13 to 5 g/l. Because of the biological stress due to the formation of foam and the loss of sludge, the membrane permeability decreased from 600 to 350-400 L/h.m^2.bar at 20°C. In spite of that, the operating conditions and peak flows were kept constant. With these conditions, the membrane permeability stabilized slowly with the recovery of a better sludge quality. So, it was concluded that the process can support peak flows of 2h at a 1.5 higher filtration flux twice a day.

Throughout the trials, the permeability of each module (top and bottom) was measured once per week as shown in Figure 8. Both modules seem to follow a similar fouling pattern. This observation correlated also for the evolution of the critical flux of each module.

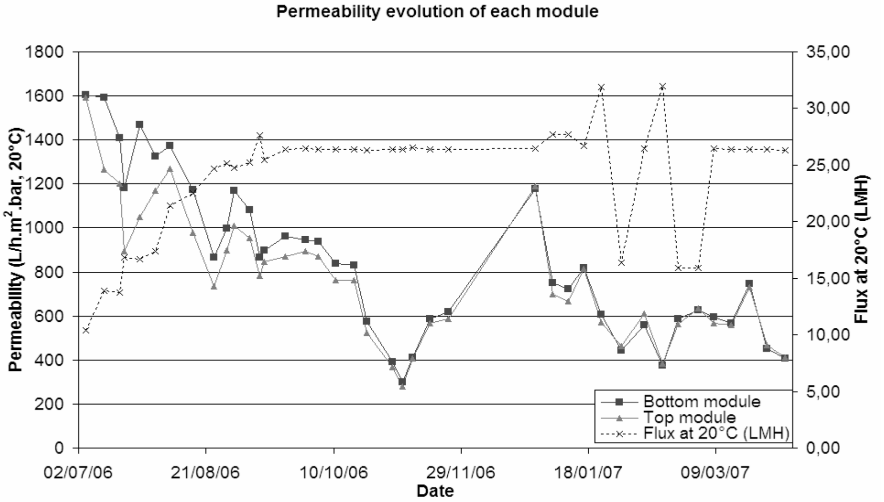

Figure 8 Evolution of the permeability of each module

FULL-SCALE PLANTS EPERIMENTS

A3 Water Solutions GmbH, as a constructor and deliverer of turn-key MBR plants, is involved in the system management and survey of the MBR in Xanten shown in Figure 9. The innovative operation concept will be validated at full scale on the Xanten plant.

Location: Xanten – Vynen, Germany
Input: Municipal wastewater
Size: 2.000 p.e.
Membrane area: 2.080 m^2

Figure 9 MBR container unit in Xanten

CONCLUSIONS

The flat sheet MBR technology of A3 water solutions could be continuously operated over 10 months in Anjou Recherche in order to perform the assessment of a novel operation concept. The biological operating conditions were kept constant during all the trials. The optimum net flux found was 25.5 L/h.m^2 at 20°C for a relatively low membrane air flow rate, that is 0.2 Nm^3/h.m$^2_{\text{of membrane}}$ corresponding to a permeability around 500-600 L/h.m^2.bar at 20°C. Good filtration performance could be maintained even in exceptional circumstances (after fouling due to biological stress, peak flows) with low operation expenses. The permeability of each module followed a similar evolution. The double-deck configuration did not seem to have a detrimental effect on the fouling of both modules.

After completion of the trials in Anjou Recherche, the study will be continued by A3 water solutions in pilot and full scale plants to further develop the novel MBR filtration system. Next step can be the test of a triple- or even quadruple deck in order to further decrease the specific air demand. Long-term performances need to be verified with these configurations.

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REFERENCES

Côté P., Buisson H., Praderie M. (1998). Immersed membranes activated sludge process applied to the treatment of municipal wastewater. Wat. Sci. Tech., 38 (4-5), 437-442.

Judd, S. (2006). The MBR Book: Principles and Applications of Membrane Bioreactors in Water and Wastewater Treatment. Elsevier, Oxford.

Smith J. C., Di Gregorio, Talcott R. M. and D. (1969). The use of membranes for activated sludge separation. 24th Annual Purdue Industrial Waste Conference, 1300-1310.

Tazi-Pain A., Schrotter J.-C., David N., Buisson H. (2001). First year of operation of the first full scale sequenced aeration MBR for municipal wastewater treatment in Perthes-en Gâtinais. Proceedings of engineering with membranes, 1, 43-48.

Yamamoto K., Hiasa M., Mahmood T., Masuo T. (1989). Direct solid-liquid separation using hollow fiber membrane in an activated-sludge aeration tank. Wat. Sci. Tech., 21, 43-44.

Experience with a newly developed submerged membrane system for MBR application

S. Krause* and U. Meyer-Blumenroth*

* Microdyn-Nadir GmbH, Rheingaustr. 190-196, 65203 Wiesbaden, Germany
(E-mail: s.krause@microdyn-nadir.de)

Abstract Due to the small footprint and superior effluent quality, the number of membrane bioreactors (MBR) used in wastewater treatment is increasing. There are more than 400 MBR applications in Europe (Lesjean and Huisjes, 2007). Microdyn-Nadir has developed a new submerged membrane system for MBR applications (BIO-CEL®). The overall goal of the development is the operation of MBR systems with low investment and operational costs (e.g. manpower, energy demand and chemical demand). The core piece of the newly developed module is the back-flushable flat membrane sheet. The back-flush ability is achieved by a lamination of PES membrane and polyester drainage layer. The challenge was to ensure a sufficient adhesion strength between the two layers and at the same time maintaining high water permeability. First results show an excellent permeability in the range of 350 – 500L/m²/h/bar in tap water. In activated sludge peak flows up to 35 L/m²/h were reached.

Keywords: flat sheet membrane, MBR, hydraulic flow, materials

INTRODUCTION

Membrane bioreactors (MBR) combine the activated sludge process (ASP) for wastewater treatment with biomass separation from the mixed liquor by ultra- or microfiltration membranes.

Usually the membranes are submerged directly into the activated sludge and the treated wastewater (permeate) is sucked by vacuum or by gravity flow. Due to the high fouling potential of the biomass, the fouling of the membranes needs to be controlled. In submerged systems the fouling is controlled by aerators installed underneath the membrane module which generates a “crossflow” (crossflow aeration). Often coarse bubble aeration acts as the source of scour at the membrane surface, but fine bubble aerators are also used.

The mixed liquor suspended solids (MLSS) concentration can be increased because no solids or bacteria can pass the membranes (the pore size is in the range of 0.05 – 0.5 µm) and the process is independent from sedimentation. MBR are usually operated at MLSS concentrations of about 10 g/L. Thus, the reactor volume, compared to the conventional activated sludge process (CASP), is reduced so that higher volumetric loads are feasible.

Advantages of the process are the superior effluent quality (which can be directly reused or, if purification is required, easily conditioned compared to conventional systems) characterized by the complete solids and bacteria removal, the small footprint of the plant due to more compact aeration tanks, the absence of a final sedimentation tank and the modular construction. The challenge in operating MBRs is the control of membrane fouling and operational expenses.

Membrane fouling is caused by the deposition of biosolids, colloidal species, scalants or macromolecular species on the membrane surface [Judd, 2006] which leads to a flux and permeability decline. The fouling is difficult to describe because of the heterogeneity of the activated sludge. Factors such as the biomass characteristics, extracellular polymeric substances (EPS), pore size, surface characteristics and material of the membrane, the module construction and the operating conditions affect the fouling rate [e.g. Chang et al., 2002]. In submerged modules blocking and braiding of the modules are observed [e.g. Voßenkaul, 2005] which reduces the active membrane surface. Braiding is caused by hairs and/or long fibers (e.g. additive cellulose fibers); this is more common in hollow fiber modules. The hairs or fibers loop around the membrane and are not discharged out of the module. Insufficient upward flow affects the sludge transport out of the module [Voßenkaul, 2005] which will cause the modules to be blocked. This results in frequent membrane cleanings which affect the cost-efficiency of the membrane (membranes are out of operation and therefore additional membrane surface must be hold out) and the environment is impacted by chemicals (e.g. sodium hypochlorite NaOCl).

The goal of developing a new module therefore was to design a module that provided a high membrane surface area, prevented braiding and sludging, and has an efficient crossflow aeration system. Overall the development focused on an economic new submerged membrane system. This paper will discuss the development steps of the new Bio-Cel® Module (materials, manufacture, construction etc) and on the operation results.

MATERIALS AND METHODS OF MODULE DEVELOPMENT

The scope for a new module construction is to use the advantages of the established systems without using the disadvantages. For the implementation of the membranes into the process, they are fixed in elements and housed in technical units, so-called modules. Membrane modules for biomass separation should exhibit the following properties [Stephenson et al., 2000, ATV-DVWK, 2002, Judd 2006]:

- Chemically, thermal and mechanically resistant materials
- back flushable membrane sheets where in-situ cleaning may also be applied
- high packing density and low specific weight
- obtain a low flow resistance
- prevention of braiding
- no dead zones where sludge can accumulate
- prevention of blocking at the membrane edge
- modularization, retrofit
- efficient crossflow aeration inducing a high degree of turbulence at the feed side to reduce fouling
- low energy requirements per unit volume of treated water

Materials

The choice of materials was important during development, as the materials must be chemical, thermal and mechanical resistant, easy to handle and economic (e.g. Judd, 2006). Mainly polymeric membranes are used, but some ceramic membranes are also used. Polymeric membranes are fabricated to have a high surface porosity and a narrow pore size distribution. Only a limited number of materials are suitable for membranes in MBR applications, being polyvinylidene difluoride (PVDF), polyether sulphone (PES), polyethylene (PE) and polypropylene (PP). A hydrophilic polyether sulphone (PES) ultra filtration membrane was chosen (Figure 1) because of the low biofouling characteristics. The membrane support and the drainage are made of polyester. The housing is manufactured of stainless steel and the piping from polyvinyl chloride (PVC).

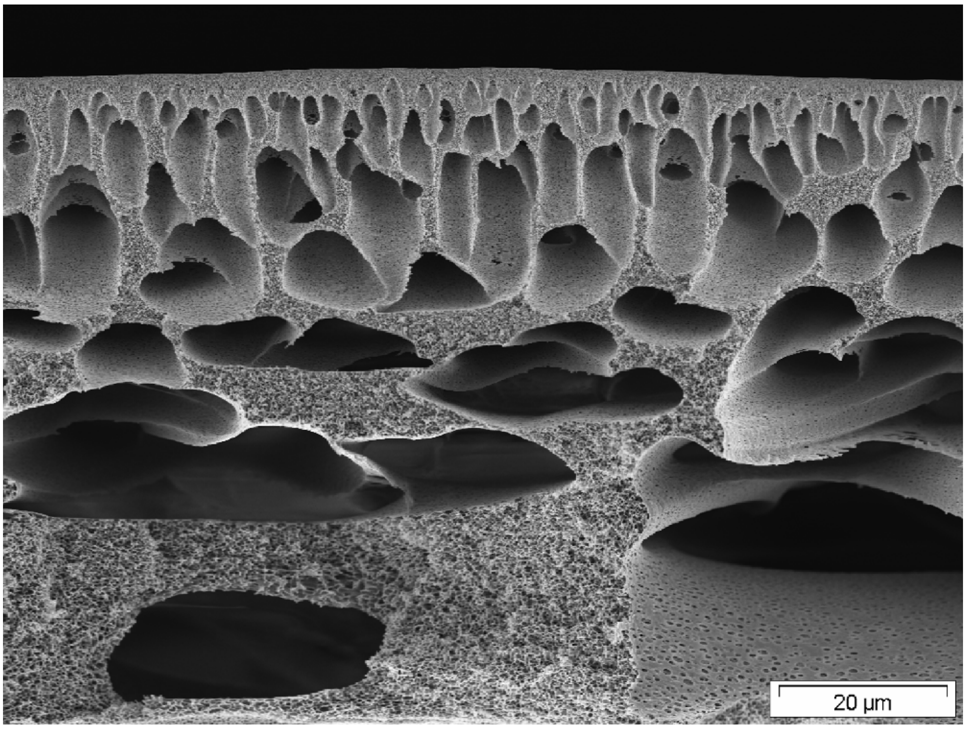

Figure 1 PES membrane

Achievement of back-flushable flat membrane sheets

The core piece of the newly developed module is the back-flushable flat membrane sheet. The back-flush ability is achieved by a lamination of PES membrane and a polyester drainage layer (Figure 2). The challenge was to ensure sufficient adhesion strength between the two layers and at the same time maintaining high water permeability. This also allows for efficient in-situ cleaning by backwashing with chemicals. The 2 mm thin self supporting membrane sheets give you a high packing density and at the same time a low specific weight because there is no plate supporting system required.

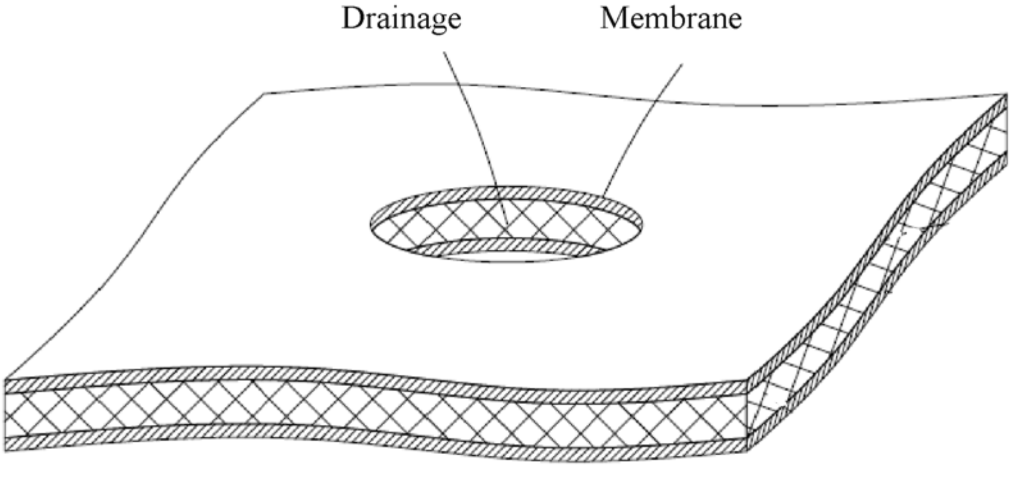

Figure 2 newly developed flexible, self supporting membrane sheet

Obtaining low flow resistance

The pressure loss inside the membrane must be low in order to obtain a low flow resistance. This is especially important for (chemical) backwashing in order to clean all of the membrane pores. Therefore a new drainage was developed with the permeate suction drawn from the middle of the membrane instead of the top or side of the membrane.

Module construction

Flat membrane sheets were used in order to prevent braiding of the membranes because hairs or fibres can not loop around them. The top and bottom of the module is open in order to prevent sludge deposits at the bottom. The hydraulic optimization of the module eliminated dead zones where sludge can accumulate. A frameless module construction is possible because the newly developed membrane sheets are self supporting. The velocity at the membrane edge is sufficient in order to avoid sludge deposits because there are guiding plates (baffles) in the module that keep a gap of about 1 cm to the membranes. The total weight of the module is reduced due to the self supporting sheets requiring no supporting plates.
The developed module is depicted in Figure 2 (Type BIO-CEL® BC100-100; 100m² membrane surface).

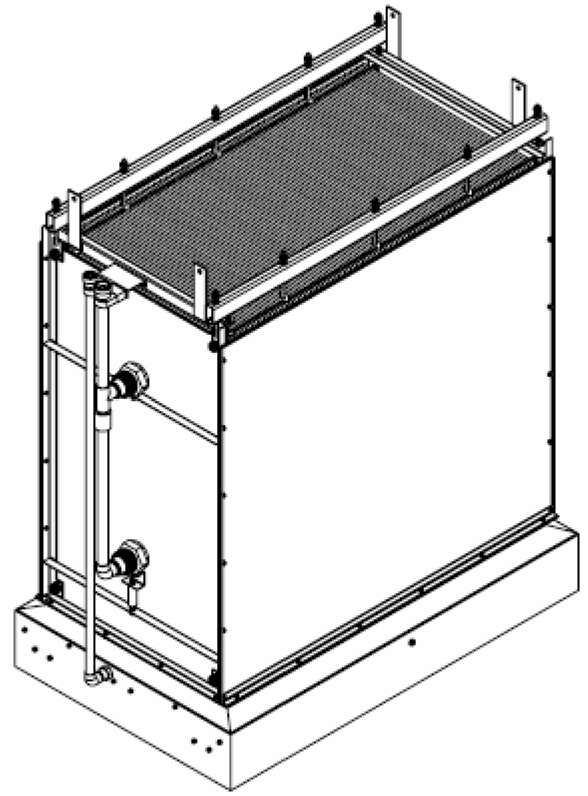

Figure 3 Bio-Cel® BC100-100 Module (100m²)

Efficient crossflow aeration

To determine the efficiency of the crossflow aeration the velocity flow was measured. Different bubble diameters (fine bubble up to coarse bubble system) were investigated. The measurements were performed using a “Marsh-Mc. Birney 2000 Flomate” flow meter. The velocity of tap water was measured first in order to judge the bubble diameter and to see the dispersion. Afterwards the viscosity changes were made using carboxymethyl cellulose (CMC) (Tylose H100000 YP2-DEAT 043780), because of the transparency of CMC-Water in order to see the dispersion of the bubbles. In the last step measurements in activated sludge were performed. The airflow rate was from 0.3 to

0.7m³ per m² membrane surface. The goal was the evaluation of an optimized dispersion and maximal flow velocity along the membranes.

PILOT PLANTS

Different pilot plants were built in order to test the module. A laboratory plant has been operating with synthetic wastewater (Table 1) since July 2005 (Plant #1).

Table 1 Composition of synthetic wastewater

Molasses ($C_6H_{12}O_6$)	50 g/L
Urea (H_2NCONH_2)	4.6 g/L
Magnesium sulphate ($MgSO_4$ x $7H_2O$)	4.1 g/L
Calcium chloride ($CaCl_2$ x $2H_2O$)	0.2 g/L
Potassium di-hydrogene phosphate (KH_2PO_4)	1.1 g/L
Ferric (III)chloride ($FeCl_3$)	0.04 g/L

The total Volume is about 3.5 m³. This plant consists of a de-nitrification tank (0.8 m³), nitrification tank (1.2 m³) and 5 filtration tanks in parallel (each about 0.3 m³). The filtration tanks are equipped with 10 m² membrane modules each.

Another plant (Plant #2) is operating with real wastewater conditions at a municipal wastewater treatment plant since September 2006. The total volume is about 12 m³ (de-nitrification tank 4.2 m³; nitrification tank 7.8 m³). In this application the membranes (50 m² surface area) are submerged directly into the nitrification tank. In Table 1 the key parameters of both pilot plants are given.

Table 2 parameters of the pilot plants

Parameter	Plant #1	Plant #2
Volume	3.5 m³	12.0 m³
COD Influent	675 mg/L	420 mg/L
MLSS	10 g/L	12 g/L
Sludge load	0.11 kgCOD/kgMLSS/d	0.04 kgCOD/kgMLSS/d
Volume load	1.00 kgCOD/m³/d	0.44 kgCOD/m³/d

RESULTS

Crossflow Induction:

Underneath the membranes the module is equipped with an aeration system for crossflow induction. The aeration system was optimized by measuring the flow velocity. The best performing system used 1 mm holes. Using this system the distribution of the air bubbles is almost constant. Water velocities directly underneath the membranes of about 0.30 m/s were measured in tap water. The relative velocity of the air bubbles are about 0.3 m/s (Krause, 2005), the air bubbles move in tap water at almost 0.6 m/s alongside the membranes.

In a highly viscous CMC-water solution the fluid velocity was reduced to 0.28m/s at 20mPas and to 0.23 m/s at 25 mPas. When increasing the viscosity the fluid flow decreases. Measurements were taken up to 100 mPas, the fluid velocity reduced to about 0.10 m/s. In activated sludge (MLSS = 10g/L) the average velocity was about 0.27 m/s.

Filtration performance and maximal peak flow:
The permeability of the BIO-CEL® system in clean water is in the range of 350-500 L/m²/h/bar which is comparable to other submerged ultrafiltration modules (van der Roest et al., 2003). During filtration in activated sludge the permeability decreases as the TMP increases and the fouling layer on the membrane surface grows. In Figure 4 the permeability of a 4 month time span is shown of Plant #2, during this time the membranes were not chemically cleaned. The average flux during this time span was about 13 L/m²/h. The permeability decreased from about 350L/m²/h/bar in October 2006 to > 200 L/m²/h/bar in March 2007. Although the plant was operated during the winter, no chemical cleaning was necessary.

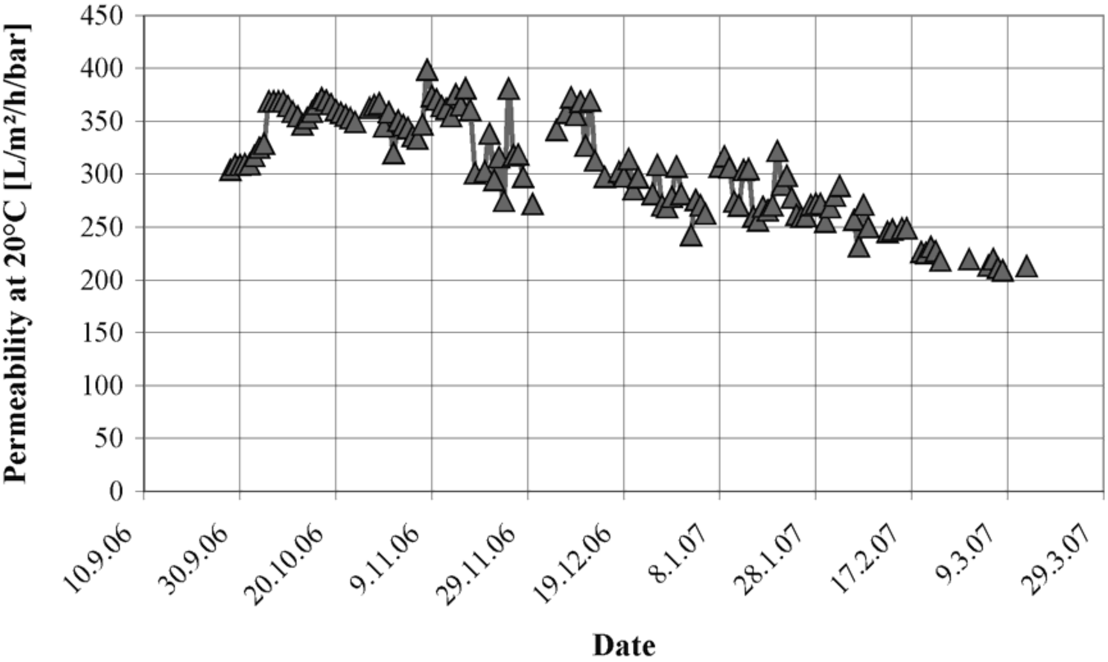

Figure 4 Permeability at Plant #2 (municipal wastewater)

In Plant #1 the operation with activated sludge shows a maximum flux of 35 L/m²/h. A permanent flux of 20 L/m²/h was obtained within a period of several weeks. A higher flux will result in more frequent chemical cleanings. The new module's filtration performance overall has more than satisfying result.

Chemical cleaning:
Cleaning of the module was tested with in-situ cleaning using chemically enhanced backwashing and in a chemical bath. For maintenance cleaning a chemically enhanced backwashing can be used (e.g. using acids and/or hydrogen peroxide). This cleaning should be performed regularly in order to minimize the bio-film growth. For a more intensive and greater effect, a chemical in-situ bath inside the filtration tank is recommended. After the activated sludge is removed the tank is filled with water and chemicals (e.g. acids and/or sodium hypochloride) for 3-5 hours. The permeability will normally return to the initial permeability.
Altogether the following different cleaning steps can be performed:

- backwashing with permeate (usually automatic every few minutes in-situ)
- chemically enhanced backwashing or maintenance cleaning (full automatic)
- intensive cleaning (chemical bath)

Nutrient removal:

The nutrient removal is excellent due to the SS free effluent. The COD removal in Plant #1 is about 97 %, in Plant #2 (real municipal wastewater) about 96 %. In Figure 5 the COD removal of both plants are depicted, showing the loading rate vs. removal rate.

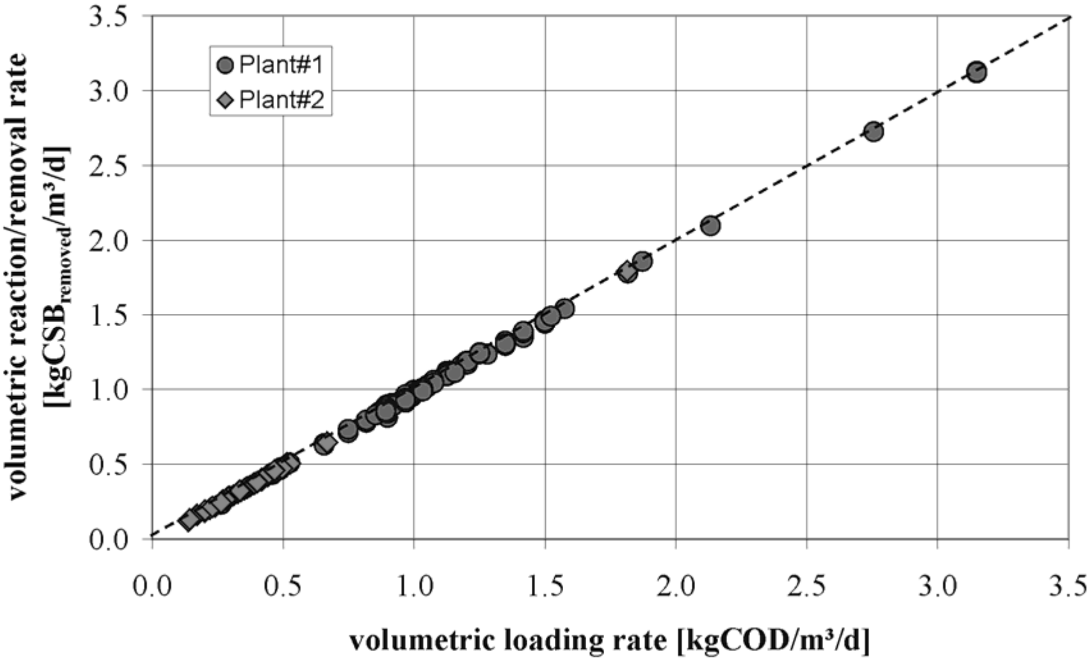

Figure 5 COD removal of both pilot plants

Energy demand:

In MBR applications the energy consumptions are the aeration systems for both the oxygen supply and for the crossflow generation. As the alpha factor reduces at increasing MLSS (Cornel et al., 2003), additional energy is required for the oxygen supply in comparison to CASP. Most of the energy is required for crossflow generation (Krause, 2005). Factors affecting the energy demand are the depth of submergence, the aeration rate and the flux. The specific airflow rate of the Bio-Cel® modules is in the range of 0.5–0.8m³N per m² membrane per hour which is comparable to other MBR modules. The depth of submergence is very shallow as the module measures only 1.35 m in height. Depending on the flux and installation the energy demand for crossflow induction is in normally in the range of 0.2–0.5 kWh/m³.

CONCLUSIONS AND SUMMARY

The experience in recent years shows that the MBR applications are proven and can be applied both in municipal and industrial wastewater treatments. The performance of a successful design and operation is possible. Optimizing the module’s construction is important because sludging, braiding and membrane blocking can occur during operation. The optimization of the energy demand of MBRs and the improvement of the fouling control (maximum utilization of the used air) are also required. In addition the frequency of chemical cleanings must be reduced because they impact the environment and require a high degree of man-power.

To address all of these challenges a new module which combines the advantages of different module designs was developed (Bio-Cel®). A flexible, self supporting flat sheet membrane was generated by laminating the membrane to the drainage layer. With this lamination no plate or other supporting elements are necessary. The membrane sheet is back-flushable which integrates the advantages of a hollow fiber system. Braiding is not possible due to it being a flat sheet membrane. The drainage layer was optimized for a low flow resistance.

Velocity measurements have shown an excellent crossflow generation regarding bubble-distribution and liquid velocity. The filtration performance had maximum fluxes up to 35 L/m²/h. Different pilot tests have shown that the membranes need less chemical cleaning at lower fluxes (e.g. no chemical cleaning for more than currently 180 days of operation at fluxes < 13 L/m²/h in municipal application; still running). Therefore the design of the membrane surface should take into account the maximum flux as well as the average flux of the membranes.

If cleaning is required chemical backwashing and/or an intensive chemical bath can be used for membrane regeneration.

REFERENCES

ATV-DVWK, (2002). Aufbereitung von Industrieabwasser und Prozesswasser mit Membranverfahren und Membranbelebungsverfahren – Teil 2: aerobe Membranbelebungsverfahren, KA Wasserwirtschaft, Abwasser, Abfall; 49 No. 11

Chang, I.-S.; Le Clech, P.; Jefferson, B.; Judd, S. (2002). Membrane Fouling in Membrane Bioreactors for Wastewater Treatment, Journal of Environmental Engineering, Vol. 128, No. 11, pp 1018-1029

Cornel, P.; Wagner, M.; Krause, S. (2003). Investigation of oxygen transfer rates in full scale membrane bioreactors, Wat.Sci.Tech., 47 (11) pp 313-319

Judd, S. (2006). The MBR Book. Elsevier, UK. ISBN-10:1-85-617481-6

Krause, S. (2005). Untersuchungen zum Energiebedarf von Membranbelebungsanlagen. PhD thesis, Department of Civil Engineering, Technische Universität Darmstadt, ISBN 3-932518-62-4

Lesjean, B. and Huisjes, E.H. (2007). Survey of European MBR Market, trends and perspectives. The 4th international Membrane Conference, Proceedings, Harrogate, UK

Stephenson, T.; Judd, S.; Jefferson, B.; Brindle, K. (2000). Membrane Bioreactors for Wastewater Treatment, IWA Publishing, London, ISBN 1900222078

Van der Roest, H. F.; Lawrence, D. P.; van Bentem, A. G. N.(2002). Membrane Bioreactors for municipal wastewater treatment, IWA Publishing, London, ISBN 1843390116

Voßenkaul, K. (2005). Grundlagen des Membranbelebungsverfahrens, Proceedings DWA-DVGW Membrantage, 21.-23. Juni 2005, Osnabrück, Germany

Comparison of aerobic and anaerobic MBRs for wastewater treatment

N. Martín García, A. Soarez, M. Pidou, J.N. Lester , S. Judd and B.Jefferson

School of Water Sciences, Cranfield University, Cranfield, Bedfordshire MK43 0AL, UK
(E-mail: n.martingarcia@cranfield.ac.uk; B.Jefferson@cranfield.ac.uk)

Abstract Aerobic MBRs have been widely employed for the treatment of domestic wastewaters while anaerobic treatment coupled to membrane filtration has found successful application in the treatment of high strength industrial wastewaters achieving high COD removals and methane productions which could compensate the costs of heating the reactor. Although for low strength wastewaters not much benefit from biogas could be expected the uncouplement of solid and hydraulic retention times and absence of aeration make AnMBRs a potential technology for the treatment of domestic sewage. In this paper the feasibility of AnMBRs in the treatment of low strength wastewaters has been studied by comparing their biological and membrane performance with aerobic MBRs through literature research data. The energy requirements associated to biological and membrane operation have been estimated and compared for different wastewater strengths.

Keywords: Anaerobic,, Aerobic, Membrane Bioreactors, Energy.

INTRODUCTION

Membrane bioreactors represent a specific subset of bioreactor technology where the membrane replaces alternative means of solids retention such as a gravity sedimentation tank. The membrane acts as a true barrier such that it will retain all material larger than the size of the pores offered by the membrane. Typical membrane pore sizes range between 0.01 and 0.4 μm although shortly after start up a dynamic layer develops on the membrane surface effectively reducing the pore size by up to an order of magnitude. Consequently, the process will retain all solids, micro organisms and a large percentage of colloids found within the bioreactor. The impact of including a membrane is a complete uncoupling of the hydraulic and sludge retention times providing greater operational flexibility and the potential to intensify the process. Commonly this is observed as an increase in biomass concentration up to 40 g/L although more a typical mixed liquid concentration in an MBR is 8-15 g/L due to aeration and membrane fouling. More detailed investigations into the nature of MBR bioreactors reveals that the microbial community acts similarly to that of a long sludge age activated sludge system irrespective of the operating conditions (Jefferson, 2005).

The advantages and disadvantages of MBRs are often quoted (Judd, 2006) but perhaps the key ones are lower effluent concentrations, lower sludge production (advantages) and membrane fouling and high energy demands (disadvantages). The advantages have meant that aerobic MBRs are becoming more widely adopted when either tight effluent consents have to be meet, a small footprint is required, robust disinfection is required or the water is to be reused (Judd, 2006). The capital and energy costs associated with the process have currently limited the technology for more mainstream applications. However, by considering anaerobic MBRs the potential to reduce energy demand is extensive. A balance is then required to ascertain the role such a technology can play in modern process flow sheets for wastewater treatment. The current papers presents a model study to ascertain the energy balance in anaerobic MBRs to provide insights into the key factors influencing the efficacy of using such technology for mainstream wastewater treatment.

MODEL DEVELOPMENT

The influence of the main operational parameters and wastewater characteristics on biological and membrane performance of both aerobic and anaerobic MBRs was directly compared from literature data. This information was employed for the assessment of the energy requirements of both systems. Biological and membrane aeration together with permeate pumping are the main energy components of submerged aerobic MBRs (Judd, 2006). Anaerobic MBRs require membrane gas scouring and permeate pumping energy inputs as well as biogas recycling in order to mix the reactor content. Chen and Hashimoto kinetic model for the anaerobic treatment of organic wastes applied to AnMBRs in order to estimate the amount of methane generated for different influent wastewater strengths and temperatures so that the energy recovered could be included in the energy balance.

Anaerobic MBR model for low strength wastewaters

Although biological processes are often modelled through Monod kinetics in which the specific microbial growth rate μ (d $^{-1}$) depends on the concentration of limiting substrate in the reactor, Contois kinetic equation which includes both substrate and biomass concentrations was employed to represent the behaviour of AnMBRs. The anaerobic model developed by Chen and Hashimoto (1978) is based on the application of the biomass and substrate mass balances and Contois kinetics in order to express the effluent quality as function of the operational parameters and the influent substrate concentration. Additionally, as substrate degradation is linked to methane generation the model parameters can also be estimated from biogas production at different operational conditions. In the steady state the biomass and substrate mass balances could be expressed according to the following expressions;

$$\frac{dX}{dt} = \mu \cdot X - \frac{X}{SRT} \quad [1] \qquad \frac{dS}{dt} = -r + \frac{S_0 - S_{eff}}{HRT} - \frac{S}{SRT} \quad [2]$$

Where X would represent biomass concentration in g COD l^{-1}, S_0 and S_{eff} the influent total and membrane effluent COD (g COD l^{-1}). HRT and SRT are the hydraulic and sludge retention times respectively expressed in days and S would represent the soluble non-VFA COD present in the bioreactor which is rejected by the membrane and wasted together with the biomass. As the SRT is greater than HRT the last term in equation 2 can be considered negligible compared to the others. The volumetric substrate utilization rate r (g l^{-1} d^{-1}) can also be related to the specific biomass growth rate and yield Y (g COD_X g COD_S^{-1}) through the equation;

$$r = \frac{\mu \cdot X}{Y} \quad [3]$$

Although different offsets between membrane effluent VFA and COD have been found for the anaerobic treatment of domestic wastewaters (Chu, 2005; Hu, 2006) it will be assumed that the biomass grows in relation to effluent concentration level according to Contois kinetics:

$$\mu = \frac{\mu_{\max} \cdot S_{eff}}{\beta \cdot X + S_{eff}} \quad [4]$$

Where μ_{max}(d^{-1}) represents the maximum specific biomass growth rate and β is a dimensionless parameter. Rearranging equations 1 to 4 the effluent substrate concentration in steady state will be;

$$\frac{S_{eff}}{S_0} = \frac{K}{(\mu_{\max} - SRT^{-1}) \cdot HRT + K} \quad [5]$$

In equation 5, K represents the product of β and Y. If we consider B as the volume of methane produced per gram of COD added to the digester and B_0 as the maximum amount of methane that could be generated from the complete utilization of it, their difference (B-B_0) will represent the amount of biodegradable substrate in the bioreactor, which has been assumed to be the same as in the membrane effluent $S_{eff.}$ Therefore equation 5 could also be expressed in terms of methane production Q_{CH4} (l d^{-1});

$$Q_{CH4} = \frac{Q \cdot S_0 \cdot B_0}{1 + K \cdot \left[(\mu_{\max} - SRT^{-1}) \cdot HRT\right]^{-1}} - Q \cdot S_{CH4} \quad [6]$$

Where S_{CH4} represents the amount of methane dissolved in the permeate expressed in normal litres of methane per litre of water which was calculated for different temperatures according to Henry's Law (Tchobanoglous,2003) assuming a headspace pressure in the AnMBRs of 1 bar and a methane content in the biogas of 70 %. The parameters in Equation 5, K and μ_{max} were optimized from the total COD removal efficiency data reported by Chu (2005) for temperatures between 11 and 25 C. Similarly, methane production data of an AnMBR treating synthetic wastewater at 35 C given by Hu (2006) was employed for the assessment of K an Bo from equation 6.In order to represent the dependence of the specific growth rate with temperature their values were also fitted to an Arrhenius type equation resulting in the following expression;

$$\mu_{\max} = 4.2832 \cdot 10^{10} \cdot e^{\frac{-7801}{T}} \quad [7]$$

As a result, values of 0.0035 and 0.235 for K and B_0 were selected for the assessment of the effluent quality and methane production in AnMBRs.

Energy assessment of aerobic and anaerobic MBRs

Energy requirements of aerobic MBRs were calculated according to the model presented in the recently published MBR book (Judd, 2006). The oxygen requirements were obtained from the COD balance and the air flow to the membrane diffusers $Q_{AIR,MEM}$ (N m^{3} $h^{-}1$) estimated from the specific aeration demand (m^{3} air m^{-2} $h^{-}1$) correlations of full scale MBRs for flat sheet and hollow fibre membrane modules (equations 8 and 9 respectively):

$$SAD_m = 0.0044 \cdot J_{net} + 0.708 \quad [8] \qquad SAD_m = 0.0052 \cdot J_{net} + 0.326 \quad [9]$$

Where J_{net} represents the net flux in lmh. It was assumed that coarse and fine bubble diffusers are used for biological and membrane aeration respectively and that the alpha factor depends on the mixed liquor suspended solids according to Gunder (2001). The biological air flow $Q_{AIR,BIO}$ (N m^{3} $h^{-}1$) was calculated taking into account the oxygen transferred by membrane aeration in order to meet the remaining oxygen requirements. The energy associated to membrane and biological aeration was obtained from power consumption of a blower delivering $Q_{AIR,MEM}$ and $Q_{AIR,BIO}$ at the static pressure of the liquid column in the membrane tank. The energy required for permeate pumping was calculated directly from the membrane operation data as the product of the permeate flow and transmembrane pressure taking into account a pump efficiency of 60 %.

In the case of anaerobic MBRs the operational costs associated to permeate pumping were obtained from membrane operational data. Operational costs were calculated whenever biogas was used to enhance permeability (Hu 2006, Kayawake, 1991) due to membrane scouring. In the studies in which membrane fouling was controlled by relaxation or permeate back flush (Chu, 2005, Lee, 2001, Wen, 1999) the net flux was taken into account for the assessment of the specific energy requirements. The energy for mixing the bioreactor content was assumed to be 8 W per cubic metre of reactor (Tchobanoglous, 2003).The energy content of biogas was calculated taking into account the heat of combustion of methane.

RESULTS AND DISCUSSION

Biological performance

The influence of organic loading rate on the COD removal efficiency and specific methane production of AnMBRs treating different types of wastewater is shown in Figure 1. High strength wastewaters coming from food processing industries or animal wastes show higher removal efficiencies than low strength wastewaters probably due to their higher anaerobic biodegradability (Liao, 2006). However a common trend in which high effluent qualities are linked to low methane productions, and vice versa is observed across all studies.

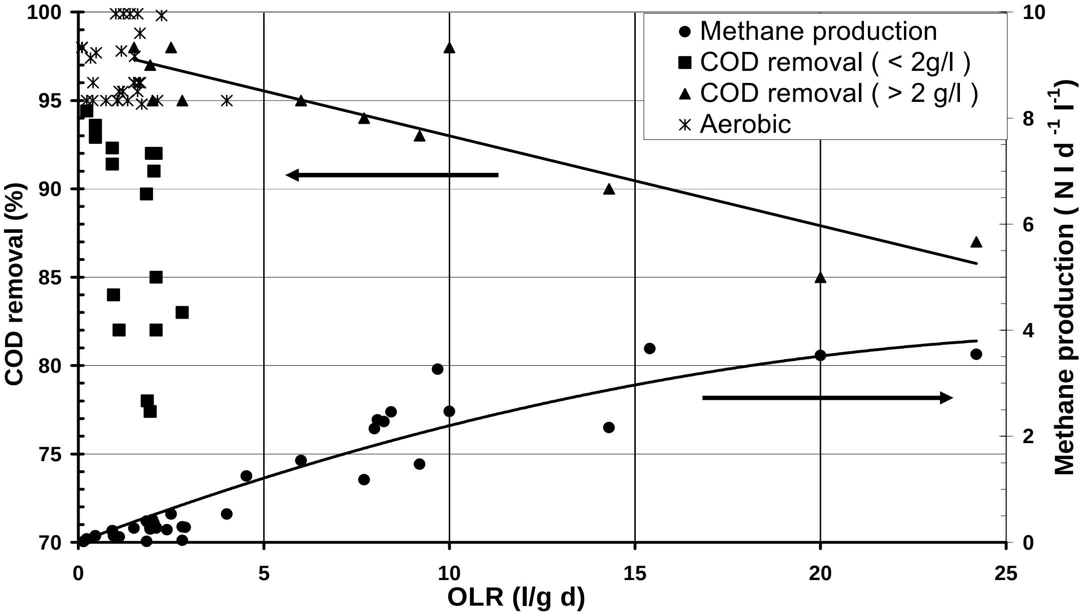

Figure 1 Influence of OLR on COD removal and methane production in AnMBRs.

The biological performance of aerobic and anaerobic MBRs treating diluted wastewaters is also presented in Figure 1. Higher COD removals are achieved for aerobic systems irrespectively of the HRT even when the systems are operated with sludge retention times as high as 150 days (Chu, 2005, Wen 1999). Such high SRT are necessary in the anaerobic systems in order to attain an acceptable level of active micro organisms and effluent qualities. For example Cadi (1994) reported a 78 % COD removal in an AnMBR treating soluble starch at 6 h HRT and 45 days SRT .Under similar experimental conditions Hu (2006) showed a higher COD removal of 90 % with a SRT of 150 days using glucose as substrate. Despite such high SRT both temperature and HRT also seem to affect their performance. For instance Chu (2005) showed a decrease on COD removal from over 95 % to 80 % with a change in temperature from 25 to 11 ºC at an HRT of 5.7 h. In the same temperature range Wen (1999) reported a 9% and 35 % decrease on COD removal based on the effluent and bioreactor dissolved COD respectively showing the importance of membrane rejection of soluble compounds in AnMBRs. A study developed in Cranfield University (Fawehinmi, 2004) showed how after acclimatisation effluent COD below 90 mg l^{-1} could be obtained in an anaerobic MBR operated with complete retention of solids and a HRT of 6 h at temperatures around 12 C.

Figure 2 represent the theoretical influence of HRT, SRT and temperature given by equation 5 on the effluent quality of AnMBRs together with literature data from different studies. Higher COD removals are obtained as HRT and temperatures (represented by different curves) increase. The number on each curve represents the SRT, which influence seems more important at lower than at higher temperatures For instance, a 95 % COD removal could be achieved for a HRT of 5 hours at 25 C while 20 hours will be necessary for a temperature of 10 C considering a SRT of 150 days in both cases. Similarly, while for a temperature of 15 C HRT should be increased 1 hour in order to achieve 90 % COD removal when SRT is decreased from 150 to 50 days, for 10 C, the HRT should be increased almost 3 hours

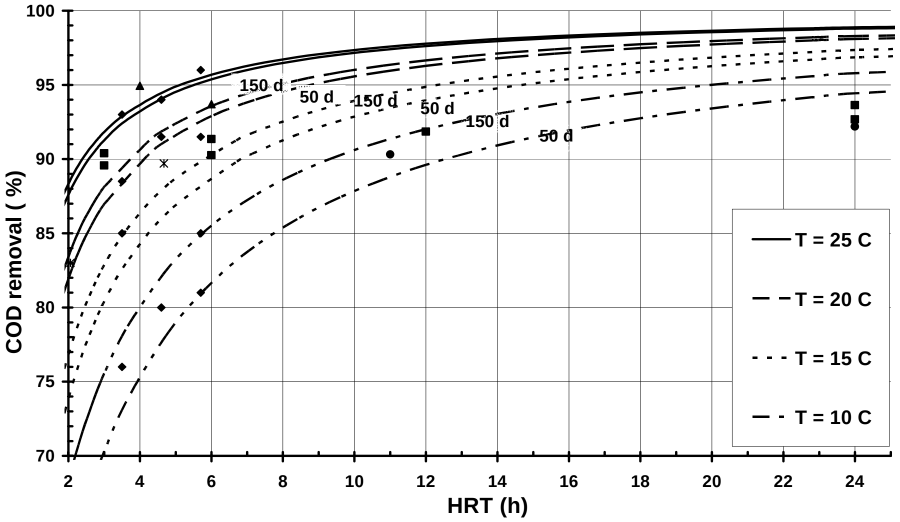

Figure 2 COD removal in anaerobic MBRs (Equation 5). ■ Hu (2006) , ▲ Wen (1999) , ◊ Chu ●Cadi (1994).

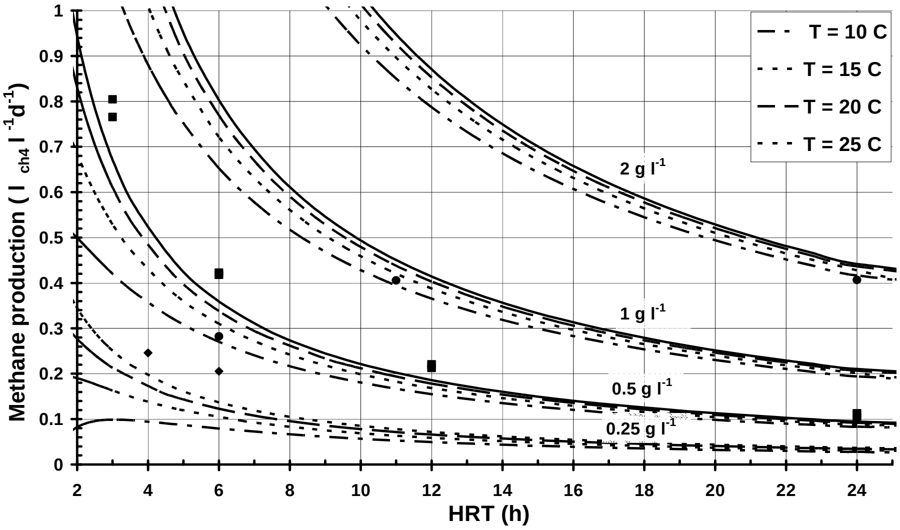

Figure 3 Methane production in anaerobic MBRs (Equation 6). ■ Hu (2006) , ▲ Wen (1999) , ◊ Chu ●Cadi (1994).

The impact of HRT, temperature and wastewater strength (Equation 6) on methane production is represented in Figure 3.The negative influence between effluent quality and energy observed across different studies and shown in Figure 1 seems to be well represented by the model as methane production increases with decreasing HRT. However, higher temperatures and influent COD enhance methane production. For instance, for a HRT of 4 hours methane production is doubled when temperature increases form 10 to 25 C and it is almost 3 times higher when influent COD increases from 0.25 to 0.5 g l^1 at the higher temperature and the same HRT. Although significant deviations can be found between the theoretical values given by equations 5 and 6 and the literature data due to differences in wastewater characteristics, reactor configurations and membrane rejections Chen & Hashimoto represented most of the trends observed.

Membrane performance

Sustainable fluxes had been attained in AnMBRs through several methods achieving different permeability levels at the expense of increasing energy requirements or decreasing permeate productivity. Amongst the highest fluxes (120 lmh) were those reported by Elmaleh (1997) applying a TMP of 0.5 bars and 25 Pa shear stress in an AnMBR fed with acetate at mesophilic conditions. Also for sidestream configuration, Beaubien (1996) showed a constant permeability of 50 lmh/bar independent from solid concentration and crossflow velocity at low TMP. Although for the higher TMP range no further increase in permeability was observed, the flux showed a linear increase with crossflow velocity. Although less investigated than for aerobic MBRs, biogas membrane scouring has also shown to be an effective method of achieving sustainable fluxes in anaerobic systems. For instance Hu (2006) employed 3 m^3 $h^{-\ 1}$ of biogas per squared metre of membrane surface at a flux of 8 lmh resulting in a stable TMP of 0.4 bars. Permeabilties of 40 lmh bar^{-1} were reported by Imasaka (1993) with 0.1 µm ceramic membranes for the same gas rate membrane area ratio but with a cross flow velocity across the membrane module of 0.2 m s^{-1} and periodic backwash of 30 second every half an hour. Overall the permeability is much lower than for aerobic MBRs which for full scale domestic wastewater treatment plants is between 150 -250 lmh bar^{-1} (Judd, 2006) even when the amount of gas provided to the membrane is 4 times higher (Equation 9). Also for submerged membranes, permeabilities of around 10 lmh bar^{-1} were reported by Chu (2005) and Wen (1999) after two weeks of intermittent 3-4 min on and 1.5 min off operation showing the lower efficiency of physical cleaning by itself in controlling permeate flux.

Higher fouling propensity of anaerobic sludges has already been reported and explained by the higher levels of colloidal matter encountered in AnMBRs as compared to aerobic systems (Judd, 2006, Fawehimni, 2004). For instance, Lejean (2005) showed a linear increase in fouling rate with polysaccharides concentration between 3 and 15 mg l^{-1} in an aerobic MBR fed with municipal wastewater while Harada (1994) reported maximum soluble carbohydrates levels of 80 mg l^{-1} (synthetic 5.5 g l^{-1}) and Aquino (2006) 19-35 mg l^{-1} (synthetic 0.45 g l^{-1}). Several studies (Imasaka, 1993, Lee, 2001) in which the resistance in series model has been applied to study fouling in AnMBRs have shown how the main cause of decreased permeability is the formation of a cake layer on the membrane surface where this colloidal matter accumulates. For example, Chu (2005) reported an average polysaccharides and proteins content in the sludge attached to the membrane surface of 12 and 6.4 mg/g VSS respectively while only 5.25 and 1.1 mg/g VSS was found in the granular sludge.

Energy requirements

The different energy components of both aerobic and anaerobic submerged MBRs treating different wastewaters were calculated and represented in figures 4 and 5 respectively. The horizontal lines in Figure 5 indicate the amount of energy required to heat the reactor up to 35 ºC for influent wastewater temperatures of 15, 20 and 25 C, taking into account that 50 % of the energy could be recovered by heating the influent with the permeate. Figure 5 shows how the energy required to heat the reactor can be completely recovered from biogas utilization at wastewater strengths above 5 g l^{-1}.

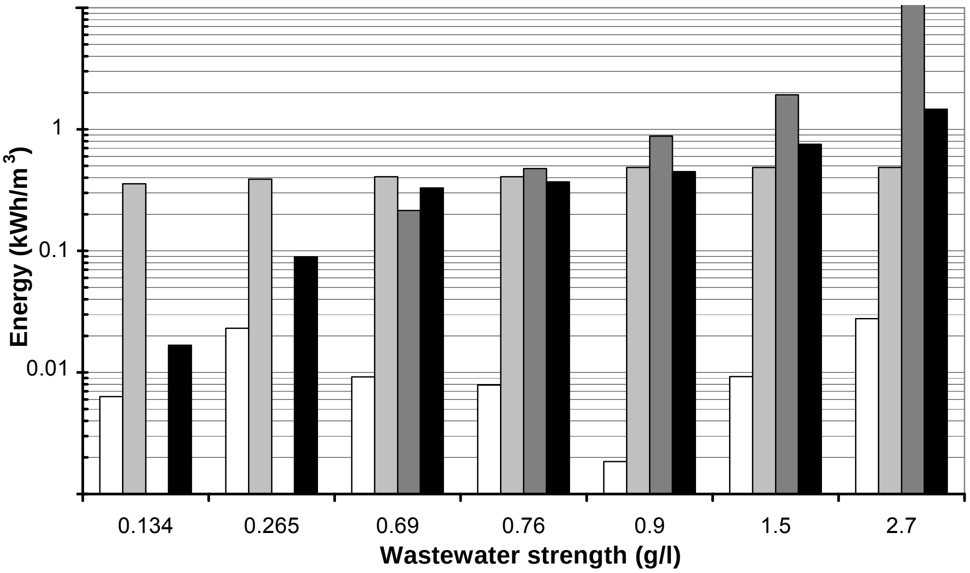

Figure 4 Energy requirements distribution for aerobic submerged MBRs. Permeate pumping ☐Membrane scouring ▨ Biological aeration ▨ Methane (electricity)■

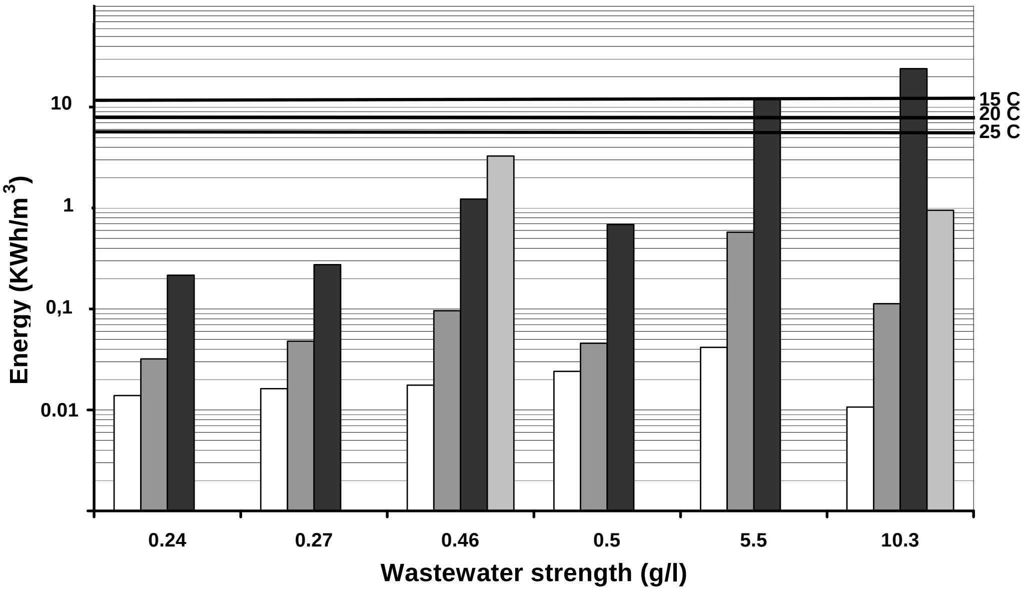

Figure 5 Energy requirements distribution for An submerged MBRs. Permeate pumping☐ Mixing ▨ Membrane scouring ▨ Methane (heat 5)■

Submerged aerobic MBRs require an average of 0.5 kWh·m^{-3} for membrane aeration and increasing biological aeration requirements that result in a power consumptions going from 0.2 to 2 kWh·m^{-3} as wastewaters strengths increase from 0.7 to 1.5 g COD·L^{-1} (Figure 4). Together with the energy requirements of submerged aerobic MBRs, the energy that could be recovered from biogas (as electrical power) has also been represented in Figure 4. This was calculated according to equation 6 assuming conversion efficiency to electrical power of 25 %, a temperature of 20 C and SRT of 150 days. The HRT was estimated from equation 5 so that the same effluent quality as in each of the aerobic studies was achieved. As Figure 4 shows, not only the energy required for biological aeration could be saved, but increasing amounts of methane providing between 0.09 to 0.75 KWh m^{-3} could be produced as wastewater strength increases between 0.2 g l^{-} and 1.5 g l^{-1} at 20 C. However, as already pointed out in the previous section, the only experimental data regarding the use of biogas for membrane scouring, shows how higher volumetric flows are required in anaerobic systems in order to achieve sustainable fluxes. Therefore the energy requirements for membrane scouring in anaerobic MBRs could be between two and six times the ones of aerobic MBRs compensating or even exceeding the biological aeration savings.

CONCLUSIONS

Aerobic MBRs are able to maintain high COD removals over a wide range of organic loading rates, while for anaerobic MBRs both temperature and hydraulic loading have a significant influence on the biological performance even if the system is operated with complete retention of solids. Membrane seems to play an important role in COD removal, as rejections between 30 and 80% have been observed in different studies.

Because of the higher colloidal concentration levels found in AnMBRs, their permeabilties are lower than for their aerobic counterpart even when side stream and submerged membranes are operated at high cross flow velocities and gas flow rates respectively. Within AnMBRs the higher permeabilties have been obtained by applying high crossflow velocities in sidestream configuration, followed by gas membrane scouring and relaxation in submerged systems.

The energy requirements of aerobic and anaerobic MBRs depend on the wastewater strength and membrane operation. It has been shown that the energy required to heat the bioreactor up to mesophilic conditions can only be recovered by methane utilisation at a wastewater strength of around 5 g l^{-1}.For domestic wastewaters at ambient temperatures the energy balance between aerobic and anaerobic MBRs is directly related to membrane operation. Increasing benefits from AnMBRs could be obtained from both absence of aeration and methane production as wastewater strength increases as long as the energy requirements to achieve at sustainable fluxes aren't excessively higher than those for aerobic MBRs.

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REFERENCES:

Aquino, S.F., Hu, A.Y., Akram, A. and Stuckey, D.C. (2006). Characterization of dissolved compounds in submerged anaerobic membrane bioreactors (SAMBRs). Journal of Chemical Technology and Biotechnology, vol. 81, no. 12, pp. 1894-1904.

Beaubien, A., Bâty, M., Jeannot, F., Francoeur, E. and Manem, J. (1996).Design and operation of anaerobic membrane bioreactors: Development of a filtration testing strategy. Journal of Membrane Science, vol. 109, no. 2, pp. 173-184.

Cadi, Z., Huyard, H., Manem, J. and Moletta, R.(1994). Anaerobic digestion of a synthetic wastewater containing starch by a membrane reactor. Environmental Technology, vol. 15, no. 11, pp. 1029-1039.

Chen, Y.R. and Hashimoto, A.G. (1978). Kinetics of methane fermentation. Biotechnology Bioengineering Symposium, 8, pp.269-273.

Chu, L.-., Yang, F.-. and Zhang, X.-. (2005).Anaerobic treatment of domestic wastewater in a membrane-coupled expended granular sludge bed (EGSB) reactor under moderate to low temperature. Process Biochemistry, vol. 40, no. 3-4, pp. 1063-1070.

Elmaleh, S. and Abdelmoumni, L. (1997).Cross-flow filtration of an anaerobic methanogenic suspension. Journal of Membrane Science, vol. 131, no. 1-2, pp. 261-274.

Elmitwalli, T.A., Soellner, J., De Keizer, A., Bruning, H., Zeeman, G. and Lettinga, G. (2001), Biodegradability and change of physical characteristics of particles during anaerobic digestion of domestic sewage. Water Research, vol. 35, no. 5, pp. 1311-1317.

Fawehinmi, F., Lens, P., Stephenson, T., Rogalla, F. and Jefferson, B. (2004) The influence of operating conditions on extracellular polymeric substances (EPS), soluble microbial products (SMP) and bio-fouling in anaerobic membrane bioreactors. IN: Proc. IWA Water Environment Membrane Technology [WEMT 2004], 7-10th June, Seoul, South Korea, 469-477.

Gunder, B (2001). The membrane Coupled-Activated Sludge Process IN Municipal Wastewater Treatment. Technomic Publishing Company.,Lancaster.

Harada, H., Momonoi, K., Yamazaki, S. and Takizawa, S. (1994).Application of anaerobic-UF membrane reactor for treatment of a wastewater containing high strength particulate organics. Water Science and Technology, vol. 30, no. 12, pp. 307-319.

Hu, A.Y. and Stuckey, D.C. (2006).Treatment of dilute wastewaters using a novel submerged anaerobic membrane bioreactor. Journal of Environmental Engineering, vol. 132, no. 2, pp. 190-198.

Imasaka, T., So, H., Matsushita, K., Furukawa, T. and Kanekuni, N. (1993).Application of gas-liquid two-phase cross-flow filtration to pilot-scale methane fermentation. Drying Technology, vol. 11, no. 4, pp. 769-785.

Judd, S (2006). The MBR book. Principles and Applications of Membrane Bioreactors in Water and Wastewater treatment. Elsevier,Ltd.

Kayawake, E., Narukami, Y. and Yamagata, M. (1991).Anaerobic digestion by a ceramic membrane enclosed reactor. Journal of Fermentation and Bioengineering, vol. 71, no. 2, pp. 122-125.

Lee, S.-., Jung, J.-. and Chung, Y.-. (2001).Novel method for enhancing permeate flux of submerged membrane system in two-phase anaerobic reactor. Water Research, vol. 35, no. 2, pp. 471-477.

Lesjean, B., Rosenberger, S., Laabs, C., Jekel, M., Gnirss, R. and Amy, G. (2005).Correlation between membrane fouling and soluble/colloidal organic substances in membrane bioreactors for municipal wastewater treatment. Water Science and Technology, vol. 51, no. 6-7, pp. 1-8.

Liao, B.-., Kraemer, J.T. and Bagley, D.M. (2006).Anaerobic membrane bioreactors: Applications and research directions. Critical Reviews in Environmental Science and Technology, vol. 36, no. 6, pp. 489-530.

Tchobanoglous, G., Burton, L.F, and Stensel, H.D. (2003).Wastewater Engineering: treatment and reuse.Metcalf & Eddy, Inc.. Boston : McGraw-Hill.

Wen, C., Huang, X. and Qian, Y. (1999).Domestic wastewater treatment using an anaerobic bioreactor coupled with membrane filtration. Process Biochemistry, vol. 35, no. 3-4, pp. 335-340.

POSTER
SESSION

Development of one accelerated fouling protocol until to increase the efficiency of cleaning strategies

L. Bonnet, S. Lyko, T. Wintgens, T. Melin

*Department of Chemical Engineering, RWTH Aachen University
Turmstr. 46, 52056 Aachen, Germany (E-mail: bonnet@ivt.rwth-aachen.de)

INTRODUCTION

The enhancement of MBR opened new perspectives for waste water treatment. Nevertheless fouling stayed the most inconvenient for long-term operating with notably the reduction of filtration performances. The development of maintenance cleanings strategies can permit to increase this operating time. Until to optimise these, one accelerated fouling protocol is being established until to test quickly more chemicals and to study their actions on fouling as well as membrane material.

The origin of fouling is now better defined [Le Clech, 2006]. If these last years, the research was most oriented about the impact of EPS [Rosenberger, 2006], some last conclusions are less convinced. That is to say that the sludge stays one complex matrix of components, which is specific for each case. The determination of the most of fouled components can permit to develop one model solution. But this solution could in any case reproduce the reality. Nevertheless, the comparison between fouling with real and/or model solution could permit a better understanding of fouling mechanisms.

This understanding as well as the identification of the action of chemical agents can permit to optimise the efficiency of CIP and COP cleaning. Currently, the actions of chemicals give us some generalities about their mode of actions, but we find any or only some information about the complementary of several products, with several concentrations.

EXPERIMENTAL DESIGN

Two test rigs are available to investigate the operating performance of a single hollow-fibre (PURON® and Zenon®) or a hollow-fibre bundle (PURON®, Koch Membrane Systems) (see figure 1). The test rigs are operating with an automatic control device, so that and all examined parameters are monitored online. The membrane (virgin or fouled) is submerged in the model solution. In addition to the aeration, backwashing (with or without chemical cleaning agents) can be used to remove adsorbed compounds and to simulate a filtration/backwashing process in an MBR system. The hollow-fibres membrane material is PES (PURON®) and PVDF (Zenon®)

The used solutions are supernatant of activated MBR-sludge with 0.05% NaN_3 or model solution obtained with combination of proteins (BSA), polysaccharides (Dextran), humic acids and minerals.

FOULING PROTOCOL

The main goal of this fouling protocol is to have a representative long-term fouling in a short period, that is to say a reduction of the initial permeability of 90% in about one week. In order to realise this challenge, it is necessary to have a filtration with a flux higher than the critical flux. But the most risk, in this case, is to build a layer at the surface of the membrane and not to increase the intern fouling. But some factors, like formation of gel layer or desorption, could limit this goal.

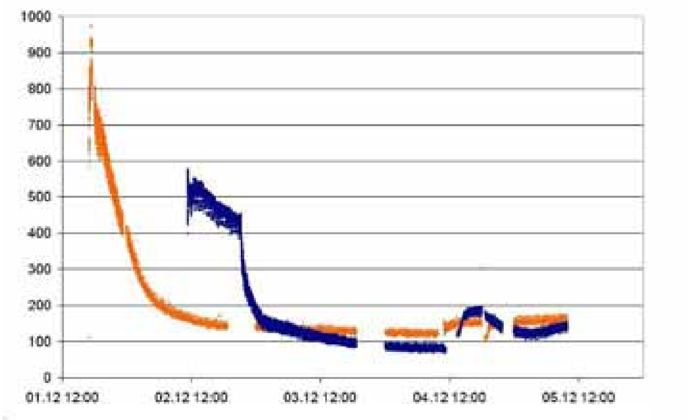

Figure 1 Permeability of two membranes during a filtration at 60 lmh with sludge supernatant

The first results permitted to observe that a filtration with high flux – bigger than 200% of the critical flux – was necessary to have a reduction of initial permeability bigger than 50%. The most problem is after to limit the formation of the gel layer. One augmentation of the ratio filtration/backwash time gives some satisfactory first results and limits the desorption phenomena. The inhomogeneity of membrane – value of initial permeability – is also one inconvenient for a good comparison between the different experiments. Nevertheless, the observed fouling is currently easy removal with chemical cleaning.

CLEANING PROTOCOL

In order to develop this second step, natural fouled membranes are used. The concentration and the contact time of chemicals are principally considered like initial conditions of cleanings. Indeed, if the influence of temperature is still demonstrated, the regulation of this for conventional plants represents one important energetic demand.

The first observations permitted to focalise notably about the origin of fouling – organic or inorganic. The measurements of permeability, MWCO as well as SEM observations could permit to give soon interesting overviews about the efficiency of cleanings as well as the destruction of the membrane material.

CONCLUSION

The establishment of one accelerated fouling protocol gives us interesting perspectives for the future, in the goal to develop efficient cleaning protocol with the combination of the both. The fouling protocol could permit also a better understanding of fouling mechanisms.

REFERENCES

P. Le-Clech, V. Chen and T. A.G. Fane: Fouling in membrane bioreactors used in wastewater treatment • REVIEW ARTICLE, Journal of Membrane Science, Volume 284, Issues 1-2, 1 November 2006, Pages 17-53

S. Rosenberger, C. Laabs, B. Lesjean, R. Gnirss, G. Amy, M. Jekel, J.-C. Schrotter: Impact of colloidal and soluble organic material on membrane performance in membrane bioreactors for municipal wastewater treatment. Water Research 40 (2006) 710-720

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Protecting an aquifer by retrofitting a municipal wastewater treatment plant with membrane filtration

Christoph Brepols, Heinrich Schäfer, Norbert Engelhardt

Erftverband, Paffendorfer Weg 42, 50126 Bergheim / Erft, Germany (Email: Christoph.brepols@erftverband.de)

Open-cast lignite mining shapes the landscape in the catchment area of the river Erft west of Cologne and inflicts the hydrology of the whole region. Groundwater levels are often low and many rivers and streams have only low flow volumes. So the effluent from the wastewater treatment plants may insert a predominant influence on water quality. Throughout the years Erftverband has equipped many WWTPs with some form of tertiary treatment. After commissioning two large scale membrane bioreactors in 1999 and 2004 the Erftverband has currently a third one under construction.

The plant in Bergheim – Glessen will have a capacity of 9,000 population equivalents with an total wastewater intake per year of 0.9 million m³ and discharges into a small stream in a groundwater protection area.

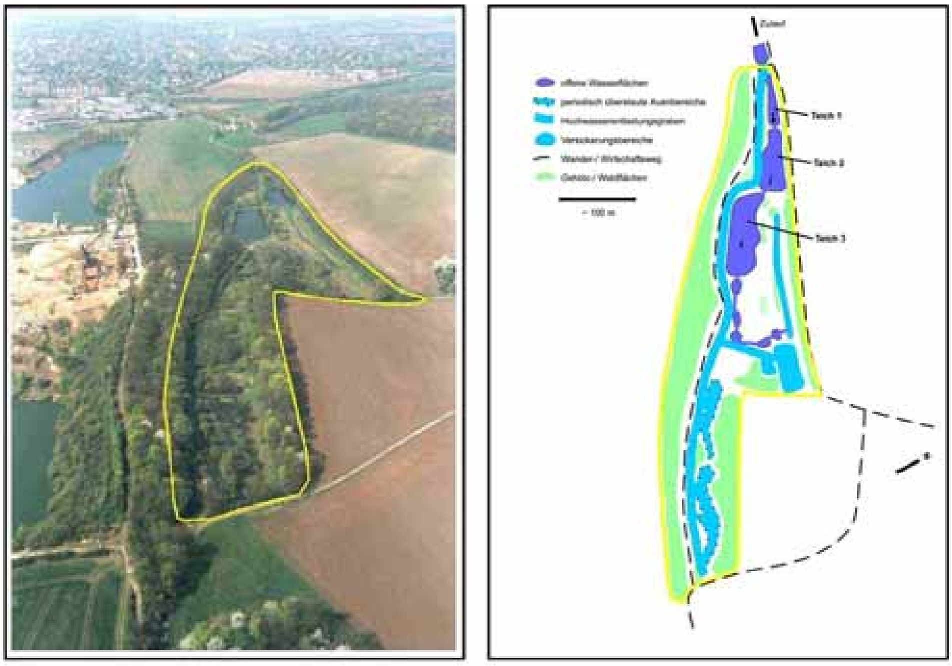

Figure 1 The wetland ‚Große Laache' west of Cologne

A few kilometers downstream the water flows into a wetland draining away in a pervious geological gravel formation. (see figure 1) As this wetland contributes to the ground water reserve of the city of Cologne, the effluent criteria are very stringent in order to sustainably protect the aquifer.

To meet these requirements the Erftverband has chosen to retrofit the existing plant by building a membrane filtration instead of the old secondary clarifier and a new mechanical pretreatment. The flow scheme is presented in figure 2.

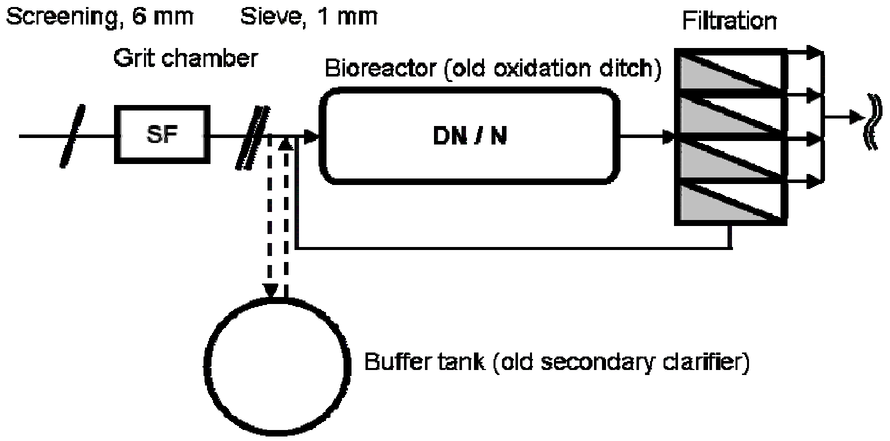

Figure 2: Flow scheme of the retrofitted WWTP in Bergheim-Glessen

The design parameters for the installation are presented in Table 1. The membrane filtration consists of submerged hollow fiber membranes installed in four separate tanks. The biomass concentration in the filtration tanks is 12 kg/m³. In contrast to this, a biomass content of only 8 kg/m³ in the bioreactor is sufficient to obtain at a sludge retention time of 25 days.
So the recycle rate is relatively low and the carryover of oxygen from the filtration can be limited, reducing the negative impact on possible denitrification.

Table 1: Design parameters for Glessen MBR

Parameter	Symbol	Unit	Value
Bioreactor volume	V	m³	1,300 m³
Sludge concentration in the bioreactor	X_{SS}	kg MLSS/m³	8
Sludge concentration in the filtration tanks	X_{SS}	kg MLSS/m³	12
Sludge retention time	SRT	d	25
Membrane surface	A_M	m²	12,096 m²
Membrane net flux	v_F	l/(m² h)	24
Membrane aeration rate	R_M	Nm³/(m²h)	0.40

The effluent values are restricted to the parameters in table 2.

Table 2: Effluent regulations

Parameter	Unit	Value
COD	mg/l	30
BOD_5	mg/l	6
NH_4-N	mg/l	1,5
P_{tot}	mg/l	0,6

The plant is scheduled to go into operation by the end of 2007. After starting up a monitoring will take place to assess the actual influence of the new plant on the aquatic system.

Characterization of Shear Forces Inside Gas-Sparged Hollow Fiber Membrane Modules

C.C.V. Chan, P.R. Bérubé, E.R. Hall

Department of Civil Engineering, University of British Columbia
6250 Applied Science Lane, Vancouver, British Columbia, Canada

Over the past decade, membrane filtration has emerged as a proven technology for water and wastewater treatment applications. Despite its popularity, the problem of membrane fouling remains the Achilles heel of membrane filtration. A common strategy to control membrane fouling is the use of gas sparging to prevent particle deposition on membrane surfaces. The optimized use of gas sparging for fouling control/prevention depends on the effective distribution of sparged gas bubbles and bubble-induced shear forces across membrane surfaces. Although there have been numerous studies on bubble-induced shear profile in confined membrane systems such as tubular membranes, there is very limited literature available that describes the shear distribution in unconfined systems such as submerged hollow fiber membrane modules. The reason for this limited knowledge is the complex and transient nature of geometry and hydrodynamics inside hollow fiber modules. Different bubble sizes and shapes delivered inside the module result in different shear forces acting on membrane surfaces. Loosely held fibers also sway laterally as a result of wakes generated by rising bubbles. The hydrodynamic conditions surrounding a hollow fiber under gas sparging are, therefore, poorly understood. This presents an obstacle in optimizing the performance of submerged hollow fiber modules with respect to energy costs associated with gas sparging.

The objective of this study is to gain a better knowledge of the hydrodynamic conditions inside the submerged hollow fiber module under gas sparging conditions. Bubble-induced shear forces acting on hollow fiber membranes in a gas-sparged module were measured using the electrochemical shear method. The advantage of the electrochemical shear method over other measurement methods (i.e particle image velocimetry) is that the electrochemical method does not have line-of-sight limitations. Additionally, since the probes were embedded flush with fiber surfaces, the electrochemical method offers the advantage of non-intrusive measurement during experimentation.

Several types of experiments were conducted to investigate:

- the mechanisms of particle back-transport (and fouling control) via bubble-induced shear forces
- the shear profiles within the hollow fiber bundle during gas sparging outside of the bundle
- the shear profiles within the hollow fiber bundle during gas sparging inside the bundle
- the effect of lateral fiber movement on shear force distribution within the hollow fiber bundle
- the effect of physical contact between fibers on shear profiles within a hollow fiber bundle

The characterization of shear profiles within the hollow fiber bundle, and the findings obtained from this study provide the insight necessary to begin to address the current knowledge gap and therefore contribute to optimizing gas sparged membrane processes with respect to energy requirement.

Keywords: Submerged hollow fiber; gas sparging; shear profile

Performance evaluation of submerged membrane anaerobic bioreactor treating different strength synthetic industrial wastewater

Marco Ferraris*[1], Carolina Innella[1], Francesca Malpei[2], Alfieri Pollice[3]

[1] ENEA, Sez. Sviluppo tecnologie e processi di recupero e riuso, S.S. 106 Jonica km419+500 Rotondella, MT (Italy).
[2] POLITECNICO DI MILANO, DIIAR Sezione Ambientale, Piazza Leonardo da Vinci 32, 20133 Milano (Italy).
[3] IRSA-CNR, Viale F. De Blasio 5, 70123 Bari (Italy).
* corresponding/presenting author: Marco Ferraris, Italy. E-mail: marco.ferraris@trisaia.enea.it,

INTRODUCTION

The use of membrane filtration for biomass retention in anaerobic bioreactors was object of study in the last twenty [1]. Membrane filtration can overcome the limits of high rate reactor technologies (e.g. UASB, anaerobic filters etc.) which are mainly related to difficulties in biomass retention under variable influent characteristics and operational conditions [2]. Previous research has mainly focused on cross-flow filtration in side-stream configurations, but severe limitations of this type of filtration systems were reported due to the loss of biomass activity caused by extreme hydrodynamic conditions [3]. The submerged membrane configuration has the potential for mitigating these problems, and the aim of the present work is to report some results of the performance of a submerged hollow fibre anaerobic MBR in terms of process effectiveness, fouling occurrence, and other operational aspects.

The use of submerged membranes in aerobic wastewater treatment field was shown to have advantages with respect to the side-stream configuration in terms of lower energy consumption and slower cake accumulation due to coarse bubble aeration below the membrane module. The latter technique is proved to be effective and appears to generate no negative effects on the biomass. Here, the use of biogas sparged instead of air is evaluated in order to extend the application of integrally submerged membrane technology

EXPERIMENTAL DESIGN

A sketch of the pilot plant is reported in Figure 1. The bioreactor has a total volume of 40 litres and it is equipped with a Zenon hollow fibre membrane (ZW1 module, 0.1 micron pore size, 0.046 m^2 surface area). Biogas is continuously recirculated and sparged under the module (15 - 20 NL min^{-1}) while excess biogas is collected in a gasometer and the total production quantified. Synthetic concentrated wastewater is prepared once a week, stored at 4°C and fed into a dilution tank where tap water is added to reach the desired concentration. Permeate is extracted by a reversible pump and used for periodic membrane backwash.

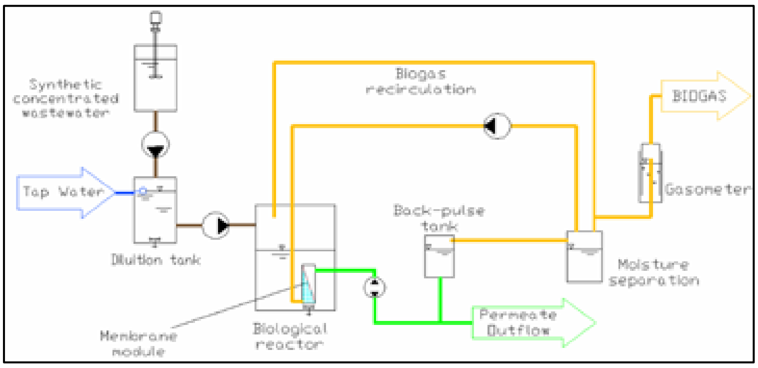

Figure 1. Scheme of the pilot scale anaerobic membrane bioreactor.

Three experimental campaigns have been conducted under mesophilic conditions (T = 37 °C) with different synthetic substrate composition and different volumetric loads (Table 1). Granular sludge from a UASB full scale plant was used as first inoculum. Membrane permeability was determined with tap water at 37°C, and then it was used to evaluate the extent of fouling and the effects of the adopted cleaning strategies.

RESULTS

The first experimental campaign was performed to test the system and showed a very fast start-up of the biological processes and a high removal efficiency of the organic substrate after few hours of operation (up to 95%). However, after 14 days of continuous operation the membrane permeability dropped due to the precipitation of salts (species not determined), possibly caused by an excessive saline load of the synthetic feed. In any case, the analysis of ammonium, phosphorus and magnesium concentrations in the feed and in the permeate lead to exclude the precipitation of struvite.

The original membrane permeability (813 $L \cdot m^{-2} \cdot h^{-1} \cdot bar^{-1}$) was restored by cleaning with a 200 ppm ipochlorite and 2 g/L citric acid solutions sequence soaking.

Table 1. Main operational parameters of the three experimental campaigns.

		First experiment	Second experiment	Third experiment
Biological process setup	**Wastewater:**	glucose + Vanderbilt mineral medium 3 gCOD/L	glucose + meet extract + yeast extract 3 g COD/L (1.4 g/L, 2.3 g/L, 0.3 g/L respectively)	glucose + meet extract + yeast extract 12 g COD/L (5.6 g/L, 9.2 g/L, 1.2 g/L respectively)
	Inoculum:	UASB granular sludge (start conc. 10 gVSS/L)	sludge from previous experiment (start at 5.5 gVSS/L)	sludge from previous experiment (start at 9.0 gVSS/L)
	Volume of reaction:	20 L	15 L	15 L
	Organic load:	1.5 g/L*day	1.5 g/L*day	6.0 g/L*day
	SRT:	No sludge wastage	100 days (second part of experiment)	100 days
	HRT:	48 hours	48 hours	48 hours
Filtration setup	**Permeation flux:**	10.4 LMH	8.4 LMH	8.4 LMH
	Permeation duration:	9 minutes	6 minutes	6 minutes
	Backpulse flux:	15.1 LMH	12.1 LMH	12.1 LMH
	Backpulse duration:	0.5 minutes	0.5 minutes	0.5 minutes

The second experiment was started with the same volumetric load of the previous one (1.5 $gCOD \cdot L^{-1} \cdot day^{-1}$) but with a higher backwash frequency (6 minutes of permeation and 0.5 minutes of backwash) and with a synthetic feed (3 $gCOD \cdot L^{-1}$) based on glucose, meat extract and yeast extract (1.4 $g \cdot L^{-1}$, 2.3 $g \cdot L^{-1}$ e 0.3 $g \cdot L^{-1}$ respectively). With this synthetic feed no problems occurred with precipitates and it was possible to operate the process for 60 days continuously without chemical cleaning and with an average COD removal of 95%. Despite a sludge growth from 5.5 $gTSS \cdot L^{-1}$ to 11.0 $gTSS \cdot L^{-1}$ observed during the test, the TMP was stable at 0.02 bar with an average flux of 8.5

$L \cdot m^{-2} \cdot h^{-1}$. This TMP value is comparable with the values reported for aerobic MBR operated for urban wastewater treatment.

In order to test the system under conditions closer to a real industrial application, the third experiment was performed by feeding the plant with a synthetic wastewater having a higher volumetric load (6 $gCOD \cdot L^{-1} \cdot day^{-1}$). In this case a stable biological process was reached after thirty days, during which the COD in the permeate was clearly influenced by high level of VFA (mainly acetic acid). The TMP during the first 20 days (during overload phase) maintained quite low values (0.02 – 0.03 bar) but a trend showing an increment of 0.62 $mbar \cdot day^{-1}$ was evident, and probably due to the observed growth of total suspended solids (from 9.6 $gTSS \cdot L^{-1}$ to 17.8 $gTSS \cdot L^{-1}$). Subsequently, precipitation of salts started to be observed in the permeate suction pipe and probably occurred also in the bioreactor, since a more rapid increment in TMP was observed. As a result after fifteen days it was not possible operate filtration any more. Analyses performed on the permeate showed struvite ($MgNH_4PO_4 \cdot H_2O$) super saturation. In this case, it was not possible to restore the filtration process even after replacing the membrane module with a new one, showing that the sludge had become not filterable at all. An important aspect is that TMP exceeding 0.25 bar caused severe cavitation in the permeate pipe, probably due to CO_2 degassing, thus the operational pressure limit for the system was much lower than those normally considered for aerobic systems.

CONCLUSIONS

The preliminary results obtained show that submerged membrane filtration in anaerobic bioreactors is possible, provided that a number of factors are considered. In the case of lower volumetric loads and when no salt precipitation occurred, it was possible to operate the system for sixty days without problems and obtaining high COD removal performance. In the case of higher load, struvite precipitation was observed, and was probably responsible for the worsening of sludge filterability.

The use of Zenon submerged membranes with biogas sparging was shown to be feasible, but cavitation can occur when the negative pressure in the suction pipe drops below 0.750 bar absolute. This can limit the operational range, so that weakly pressurized reactors may be required. The present study shows that further studies are needed on wastewater composition and load, in order to define the correct utilization area for anaerobic membrane bioreactors.

REFERENCE

[1] Stephenson, T., Judd, S., Jefferson, B., and Brindle, K. Membrane Bioreactors for wastewater Treatment, IWA Publications, London, UK, 2000

[2] A.E. Hernández, L.C. Belalcazar, M.S. Rodríguez and E. Giraldo (2002); Water Science & Technology 45 (10), 169–174

[3] Brockmann M., and Seyfried C. F. (1996); Water Science & Technology. 34 (9), 205-213

Utilisation of white water in hotel and catering industry

Katrin Gethke, Heinrich Herbst, Christopher Keysers

Institute of Environmental Engineering, RWTH Aachen University, Germany – 52056 Aachen (Email gethke@isa.rwth-aachen.deGermany)

INTRODUCTION

The water consumption in the hotel and catering industry is far higher than that in private households, which is a significant expense factor in this sector. To lower the costs and - closely connected - to protect the water resources, hotels can use decentralised waste water treatment systems to sanitise a part of their waste water for subsequent reuse. In a project of the Institute of Environmental Engineering at RWTH Aachen University, a loop system for water in a hotel is to be tested. For this, grey water shall be purified until it becomes white water with a high quality for reuse.

DESCRIPTION OF THE PROCESS AND FIRST RESULTS

In the hotel and catering industry person-related water consumption is significantly higher than in private households. Daily drinking water consumption can be measured up to 500 litres per guest. In pubs and restaurants 60 litres per guest are used, although the guests usually do not stay longer than three hours.

In the project a loop system for water in a small hotel is to be tested. The hotel comprises 50 guest rooms with a total of 90 beds, a restaurant with 100 seats and convention rooms. The most important part of the loop system is a waste water treatment plant, which transforms grey water into high-quality white water (figure 1).

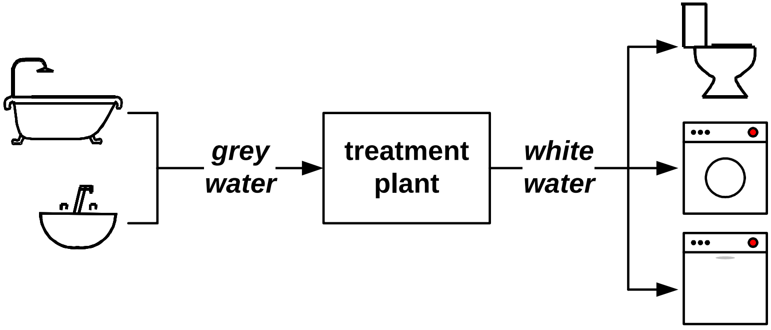

Figure 1: Flow chart of the planned loop system

The sources of the grey water are showers, bathtubs and washbasins of the hotel. This grey water has only a minor load of harmful substances. Only the load of COD and some special forms of XOC (xenobiotic organic compounds), e.g. surface active substances and scents, are high. In the project the produced white water shall be used for toilet flushing as well as for the first stages of the washing programme of the washing machine and the dishwasher. For the last two mentioned applications the white water must have a high quality, especially it must be free of bacteria and viruses.

To achieve a high-quality white water the treatment plant consists of four different stages (Figure 2). The grey water is collected in an equalising tank, whose function is to feed the next steps of the plant with a continuous flow. The next two steps comprise the treatment process consisting of an aeration tank and a membrane filtration tank. In the aeration tank the COD and, in parts, the XOC load is degraded. Membrane filtration ensures that the white water is free of bacteria and viruses. After the treatment process the produced white water is stored. To prevent a recontamination with bacteria the stored water must be disinfected. For disinfection, ozonisation or UV irradiation can be used.

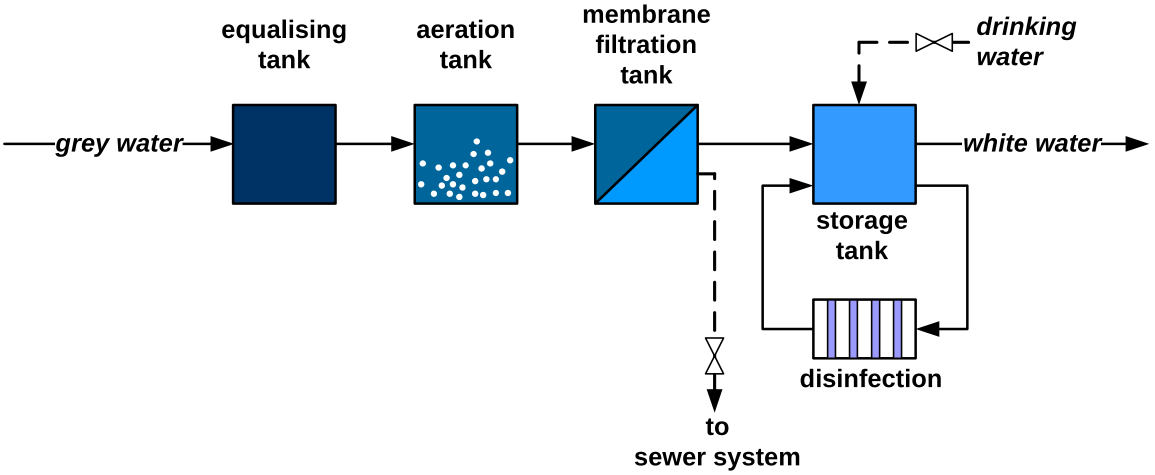

Figure 2: Flow chart of the grey water treatment plant

In preliminary tests grey water was not treated by the entire system, but only by membrane filtration using a nanofiltration unit. General hydrochemical parameters of the grey water and of the permeate, such as pH value and electrical conductivity, were monitored and are shown in Table 1. Standard wastewater parameters such as BOD and COD and the presence of microorganisms were also analysed.

Table 1: Quality of grey water and permeate of nanofiltration in preliminary tests

	COD	BOD_5	pH	turbidity	electrical conductivity	total coliforms	Escheria coli	intestinal entero-coci
	[mg/L]	[mg/L]	[-]	[NTU]	[µS/cm]	[cfu/100mL]	[cfu/100mL]	[cfu/100mL]
grey water (median)	94.0	37.0	7.48	44.0	305.0	10^7	10^5	10^2
permeate NF (rate of yield: 50%)	< 10.0	< 1.0	7.69	< 1.0	174.0	-	-	-
permeate NF (rate of yield: 90%)	14.0	< 1.0	7.59	< 1.0	264.0	0	0	0

As seen in the results of the preliminary tests the grey water could be transformed into high-quality white water by membrane filtration only.

CONCLUSION

The implementation of a water loop system in hotels lowers the fees for drinking water and waste water. In the same way the water resources can be protected. With the planned treatment plant for the described project grey water will be transformed into white water. The excellent results of preliminary tests raise hope for producing a high-quality white water, which can be used for the first stages of the washing programme of washing machines and dishwashers.

Utilization of microfiltration for regeneration of degreasing solutions

Horčičková J., Mikulášek P.

University of Pardubice, Faculty of Chemical Technology, Department of Chemical Engineering,
nám. Čs. Legií 565, 532 10 Pardubice, Czech Republic.
E-mail: Horcickova.J@seznam.cz

This paper reports the results of the study the possibility of microfiltration utilization for regeneration of degreasing solutions. The study was designed to evaluate the system performance and its ability to extend the service life and to improve the quality of degreasing solutions from surface refining processes.

The regeneration of six types of commercially available degreasing solutions was investigated and for series of crossflow microfiltration experiments were used α-Al_2O_3 microfiltration tubular membrane (0,1µm).

In other experiments the influence of pressure difference and feed velocity on steady state value of permeate flux were studied. (Fig. 1, 2)

The results of the experiments show that the regeneration of the degreasing solution by microfiltration, eventually ultrafiltration, on a suitable membrane is successful and can significantly stretch out the life span of the degreasing solutions. There are several factors, such as pressure difference or feed velocity, which influence the course of microfiltration of degreasing solutions. Nevertheless, the results obtained from the experiments cannot be generalized because the composition and type of regenerated degreasing solution – which can be changed for different types of solution – play the main role.

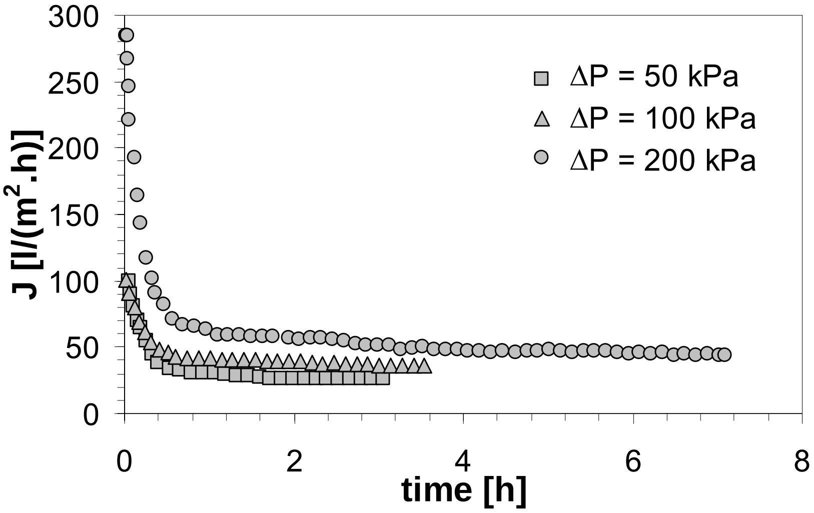

Figure 1 Influence of transmembrane pressure on permeate flux for PRAGOLOD 54 UF degreasing solution. 0,1µm α-Al_2O_3 membrane, T = 20°C, u = 1,17 m/s.

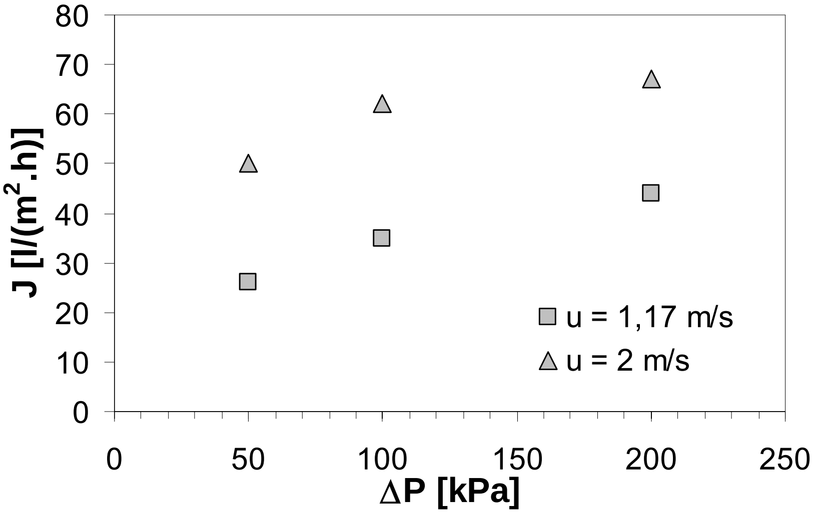

Figure 2 Effects of crossflow velocity on permeate flux for PRAGOLOD 54 UF degreasing solution. 0,1μm α-Al_2O_3 membrane, T = 20°C.

Keywords: microfiltration, degreasing solution, regeneration, surfactant, oil.

Influence of nano particles on the membrane fouling in the biofilm membrane bioreactor (BF-MBR)

Igor Ivanovic ,Markus Fitz ,TorOve Leiknes

NTNU - Norwegian University of Science and Technology, Department of Hydraulic and Environmental Engineering, S.P. Andersensvei 5, N-7491 Trondheim, Norway (E-mail: igor.ivanovic@ntnu.no

Biofilm membrane bioreactor (BF-MBR) is an alternative, hybrid system to the conventional MBR (AS-MBR) for municipal wastewater treatment. In BF-MBR the moving-bed-biofilm reactor (MBBR) for biodegradation of soluble organic matter is coupled with a submerged membrane reactor (sMBR) for particle separation 0 (Figure. 1)0. The moving-bed-biofilm reactor (MBBR) utilizes the whole tank volume for biomass growth, as well as the activated sludge (AS) reactor, but contrary to AS does not need any sludge recycle. This is achieved by having the biomass growth on carriers that move freely in the water volume of the reactor 0. This characteristics present considerable advantage over the activated sludge process, since no sludge recirculation takes place, only the surplus biomass has to be separated. Like in all membrane processes, fouling in BF-MBR is a main obstacle that needs to be understood in order to develop sustainable process.

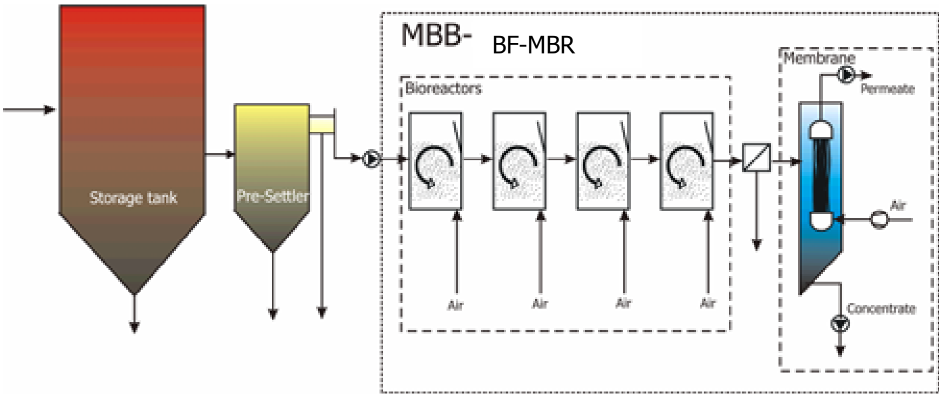

Figure 1 Schematic of the BF-MBR pilot plant

The previous studies showed importance of particle distribution in the range of 0,04 to 2000 µm, on the membrane performance in BF-MBR00. The aim of this study is to investigate distribution of nano particles in BF-MBR system and to estimate the effect on performance of UF membrane filtration unit, with a nominal pore size 40 nm (0.04 µm).

The performance of the membrane filtration unit in BF-MBR, has been investigated during the nine days operational cycle (figure 3). The constant production flux (52 LMH) was applied and the TMP development was continuously measured over the time. Organic loading rate of soluble organic matter, represented as FCOD in MBBR, was 3,2 g $O_2/(m^2day)$, while total COD removal was higher then 92%. Recovery was ~ 96 %.

Distribution of nano particles in the system was measured by Beckman Coulter N5, particle size analyser. Three sampling points were chosen; inlet-raw wastewater, effluent from MBBR and membrane reactor (middle zone). Target range for particle analysing was from 3 nm to 1200 nm.

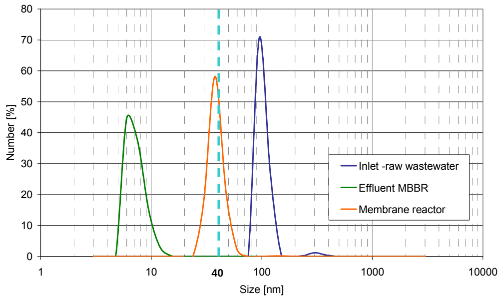

Figure 2. PSD in number % - for the chosen sampling places

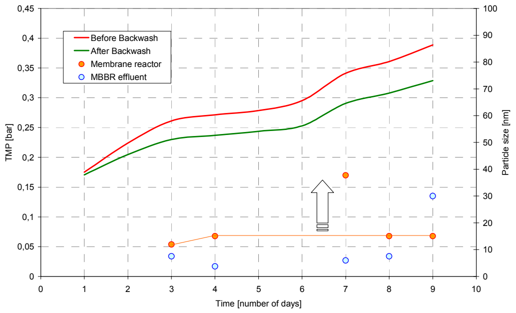

Figure 3 TMP vs.Size of the most present particle fraction in a membrane reactor

Results presented in figure 2 showing the distribution of nano particles on the seventh day. Normally the most present size of the particles during this experiment in the membrane reactor was in range 11,93 – 15,03 nm. On the seventh day the highest concentration of particles in the membrane reactor was measured at 37,76 nm. The same day was measured a higher fouling rate i.e. dTMP/dt (Figure 3). This finding guides to the conclusion, that high concentration of submicron-nano particles around the membrane pore size, could be a reason for poorer membrane performance.

Presented experiment is attempted to find more evidences for better understanding of membrane fouling, by the particles, especially nano particles. Importance of more detailed measurements of particle size in the nano range is obvious. More experiments will be performed in order to approve these results.

REFERENCES

Leiknes, T., and Ødegaard, H., The Development of a Biofilm Membrane Bioreactor, Proceedings, IWA Specialty Conference on “Wastewater Reclamation and Reuse for Sustainability (WRRS2005), 7 - 11 November 2005, Jeju, Korea. Accepted for publication in Desalination.

Ødegaard, H., Rusten, B., Westrum, T. A new moving bed biofilm reactor - Applications and results. Wat. Sci.Tech., 29(10-11), p. 157-165. .(1994)

Åhl R.M., Leiknes T., Ødegaard H., Tracking particle size distributions in a moving bed biofilm membrane reactor for treatment of municipal wastewater, Proceedings, IWA International Conference on Particle Separation, 1-3 June 2005, Seoul, Korea. Accepted in Water Science & Technology (2005).

Ivanovic I. Leiknes T., Ødegaard H., Influence of loading rates on production and characteristics of retentate from a biofilm membrane bioreactor (BF-MBR) Desalination 199 490–492 (2006)

Systematic investigation of the effect of flux enhancers on MBR sludge filterability

Vera Iversen*, Juliane Mohaupt* , Anja Drews*, Boris Lesjean** and Matthias Kraume*

* TU Berlin, Department of Chemical Engineering, Germany
** Berlin Centre of Competence for Water, Germany
E-mail :vera.e.iversen@tu-berlin.de

1. INTRODUCTION

The widespread application of MBRs is still restricted by membrane fouling which reduces the filtration performance and thus increases investment and operation costs. Soluble and colloidal materials like soluble microbial products (SMP) or extracellular polymeric substances (EPS) are considered to be major foulants in membrane bioreactors (MBRs) [1]. The removal of these substances causing irreversible fouling is thus thought to reduce the fouling of the membrane.

In addition to traditional strategies for fouling prevention, mostly trying to remedy the effects of fouling by air scour, backflush or relaxation, the new and promising method of adding chemicals [2, 3] is being investigated extensively at TU Berlin. From literature it can be seen that different additives like polymers, metal salts and others come into question. Most research groups focus on the exploration of one or few additives. Results of a comprehensive, systematic and impartial screening have not been published so far.

Previous tests with a total of 30 additives [4] have shown that some of these substances are able to reduce SMP concentration. Some already showed an enhanced flux performance in a small scale filtration unit. Besides binding SMP, the additive might also flocculate the sludge and therefore cause different filter cake properties. On the other hand they might themselves interact with the membrane in some way or affect the biomass activity [5].

Small scale filtration tests were therefore conducted in order to quantify the effect of the flux enhancers. These tests were conducted with the previously determined optimal additive concentration in sludge and with untreated sludge for comparison. The influence of the additive itself on the membrane material was also investigated with different membrane materials.

The parameters influencing filterability and their effects might be quite different for larger scale applications, also different effects might occur for long term dosing. So this work is part of a preliminary study for the use of flux enhancing additives in a pilot scale MBR (appx. 1.5m^3 reactor, 22m^2 membrane area) operated in the frame of the European project “AMEDEUS”. First pilot scale results in this plant will also be presented at the conference.

2. EXPERIMENTAL METHODS

Small scale filterability tests were conducted in a cross-flow filtration test cell designed at the department [6]. The impact of the residual (5% of optimal dosing in water) on three different membranes (two 0.2µm PVDF membranes from different manufacturers; one PES membrane, MWCO 150kD) was evaluated in 2h filtration tests with a flux of 60 L/(m^{2}h). Aerated filtration tests with MBR-sludge and optimal additive dosing were carried out under constant flux conditions. A protocol was developed to enable evaluation of different resistances (see Fig. 1).

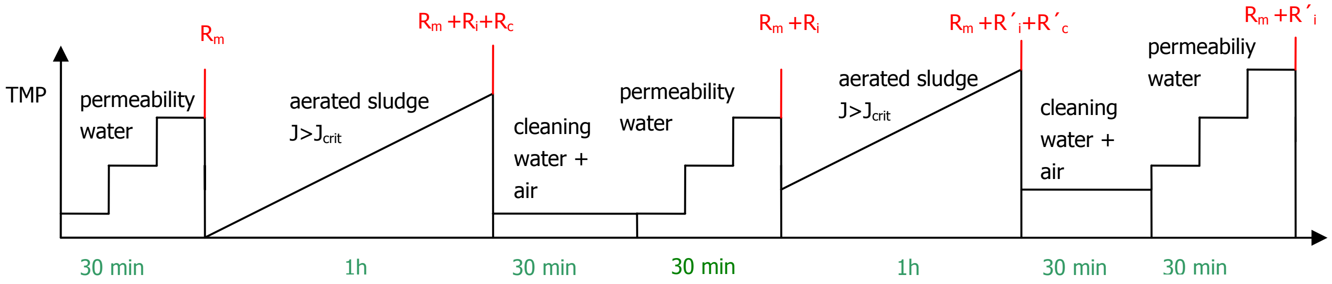

Figure 1: Filterability tests for treated and untreated sludge

The test cell was equipped with two circuits: one with the sludge and the second with tab water. First, the membrane permeability was determined with tab water. Then the second circuit was operated under aerated conditions [7] with sludge and a flux of appx. 25L/(m²h) > Jcrit for 1h. Afterwards the membrane was cleaned with water and air scouring in order to remove the cake and the water permeability was determined a second time. After 1h sludge filtration and subsequent cleaning the water permeability was determined a third time. With this information the membrane resistance (Rm), the resistance due to cake layer formation (Rc) and irreversible fouling (Ri) for the new and the used membrane (R'c, R'i) were determined in accordance to [8]. The sludge for these tests was taken in the morning from an MBR pilot and was stored under aerated conditions until the trial (max. 5 hours). Two sets of tests were conducted within one day: one with and one without additive dosing to ensure the same sludge conditions for comparison.

Large-scale tests will be done with two parallel pilot MBRs. Each plant consists of two 1m³ tanks with a working volume of approx. 0.8m³. The pilot units are located in a 20' sea container near a pumping station of the Berliner Wasserbetriebe thus drawing combined municipal wastewater from Berlin city center as influent. After the settler used as sand trap for the removal of larger particles, the wastewater flows into a stirred anoxic chamber. The following tank is aerated and equipped with a 22m² membrane module supplied by A3 water solution, Gelsenkirchen. The used membrane is made of PVDF with a pore size of 0.2 µm. TMP, flux, DO, T and pH in the membrane chamber are registered on-line.

3. RESULTS AND DISCUSSION

Fig. 2 shows results of the filtration tests with sludge. At first sight it seems that the addition of substance C-2 and the polymers 3 ,7 and 10 strongly enhances the filtration performance while an addition of substance B-5 worsens the performance in comparison to the reference sludge. At second sight it must be said that the trials on the 15/9 with addition were done first while the sludge test was done afterwards. For all other tested substances the reference sludge was tested first. In comparison to the reference sludges on the other days the resistance one the 15/9 is very small. It is therefore possible that fouling causing substances like SMP are eliminated within the 4-5h between the start of the first and the second experiment. These results further stress the importance to conduct parallel tests like the ones planned with the two pilot MBRs and not to store the sludge.

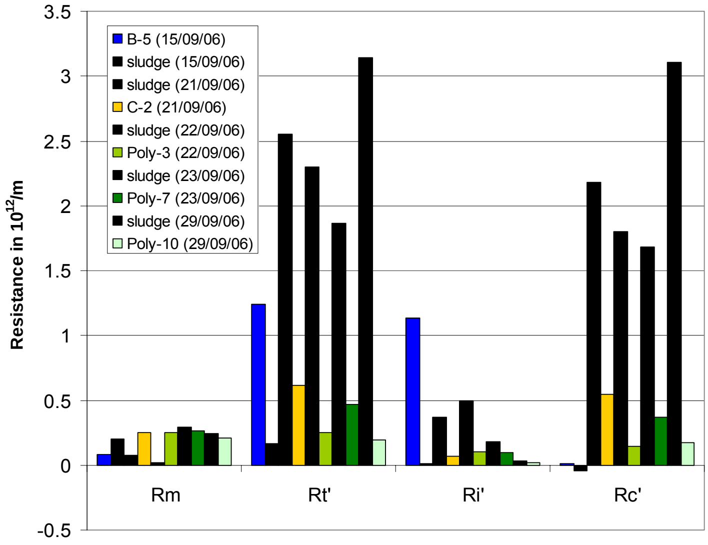

Figure 2: Effect of additive on filtration (black columns: reference sludge)

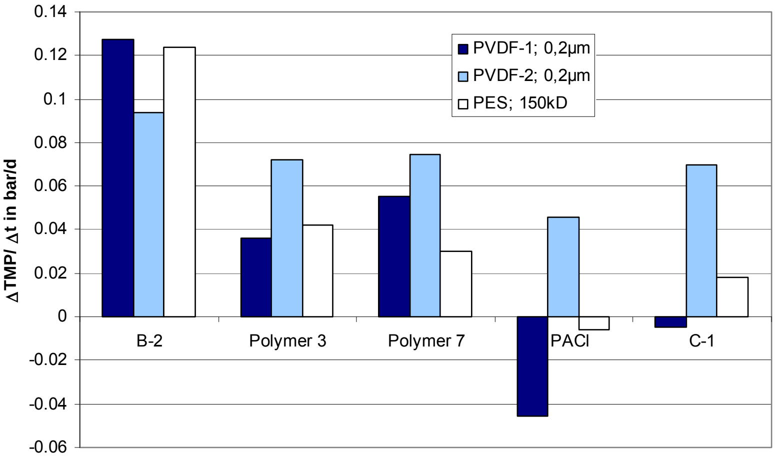

Figure 3: Impact of residual on membrane materials

The results of the residual trials are shown in Fig. 3. As can be seen, the influence of the five tested substances differs strongly for the three different types of membranes, even for the two PVDF microfiltration membranes. Therefore, it seems advisable to prescreen several substances to find the best for the respective membrane material. The fouling propensity especially of additive B-2 seems to be quite high. In general, the TMP increase is rather high due to the high flux employed

ACKNOWLEDGMENT

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REFERENCES

1. S. Rosenberger, C. Laabs, B. Lesjean, R. Gnirss, G. Amy, M. Jekel, and J.-C. Schrotter, *Wat. Res.,* vol. 40: pp. 710-720, 2006.
2. S.-H. Yoon and J.H. Collins, *Desalination,* vol. 191: pp. 52-61, 2006.
3. H.H.P. Fang, X. Shi, and T. Zhang, *Desalination,* vol. 189: pp. 193-199, 2006.
4. V. Iversen, L. Bonnet, A. Drews, B. Lesjean, and M. Kraume. *Can we control the fouling with flux enhancing chemicals?* in *EUROMBRA Workshop*. 2006. Trondheim.
5. V. Iversen, M. Mohaupt, A. Drews, B. Lesjean, and M. Kraume. *Influence of flux enhancing chemicals on membrane*

material and OUR. in *4th IWA International Membrane Conference*. 2007. Harrogate (accepted).
6. S. Rosenberger, A. Drews, and M. Kraume, *Chem. Ing. Tech.,* vol. 73: pp. 597, 2001.
7. J. Schaller, A. Drews, and M. Kraume, *Desalination,* vol. 199: pp. 507-508, 2006.
8. T.-H. Bae and T.-M. Tak, *J. of Mem. Sci.,* vol. 264: pp. 151-160, 2005.

Water Reuse Experience with ZeeWeed Ultrafiltration Membranes

Katharina Jähnel

ZENON GmbH, Nikolaus-Otto-Str. 4, D-40721 Hilden (Germany)
e-Mail Eckart.Doepkens@ge.com

Improvements in membrane technology over the last decades have lead to different approaches for water reuse. ZeeWeed Membrane Bioreactors (MBR) or ZeeWeed Tertiary Filtration Systems can be used and are a proven state of the art technology with more than 15 years of experience. The ZeeWeed technology is based upon immersed ultrafiltration membranes with an Out-In flow direction. Requirements like the EU Bathing Water Directive can be easily achieved. Table 1 shows the effluent quality from the ZW1000 plant Hailfingen (Germany).

Table 1: Quality of feed and effluent of a ZW1000 tertiary filtration at WWTP Hailfingen (Germany)

Parameter	**Unit**	**effluent secondary clarifier = UF feed**	**UF effluent**
TSS	mg/L	5 – 30	< 1
Turbidity	NTU	5 – 50	0,1 -0,2
Faecal Coliforms	MPN/100 ml	-	< 30
Enterococci	1/100 mL	-	0

If desalination of the ultrafiltration permeate is additionally required, the effluent of a ZeeWeed filtration system is well suited to feed a reverse osmosis (RO) system. SDI values of the ultrafiltration effluent are typically below 3, which is ideal for an effective RO operation.

Main focus of recent research and pilot projects are the improvement of life cycle costs, in order to allow for cost-effective installations on a global scale, especially in regions where water shortage is an issue. Several improvements regarding operation modes and membranes have been achieved during the last couple of years. Energy consumption, for example, has been reduced to 100 – 250 W/m³, depending on the operation mode.

The paper describes results obtained with the ZeeWeed® 500 and ZeeWeed® 1000 membrane series, with special focus on SDI and microbiology. The data presented include results both from existing plants of more than 2 years of operation and from pilot studies using new membrane series.

European and Worldwide Water-Reuse Applications with Kubota Submerged Membrane Bioreactor

M. Kanai*, S. Lluch** and K. Izumi***

* Kubota Membrane Europe Ltd, 8 Hanover Street, London W1S 1YE, UK
(E-mail: kanai@kubotalon.co.uk)
** Hera Amasa, S.A., Dep. Aguas, C/Paletes, 6 - 08290 Cerdanyola, Parque Tecnológico del Vallés, SPAIN
(E-mail: sergi.lluch@heraholding.com)
*** Kubota Corporation, 1-1-1 Hama, Amagasaki, Hyogo 661-8567, JAPAN
(E-mail: ki-izumi@nk.kubotalon.co.jp)

Abstract This paper reviews Water Reclamation and Reuse strategies in different countries and details Water Reuse applications employing Kubota Submerged Membrane Bioreactor Unit® (Kubota SMU) in Japan, Australia and Spain. The technologies for water reuse are achieving its importance and MBR technology to produce better effluent quality, which can be reused, draws attention for sustainable environment.

Keywords Filtration, Membrane bioreactor, Kubota submerged membrane system, Wastewater, Water reclamation

INTRODUCTION

Reclaimed water is of fundamental importance to our environment and economy. Planned, more direct use of reclaimed water contributes to an efficient and more reliable use of the water resources. It enables to reduce the impact of water shortage due to population growth or drought. At the same time it decreases the impact of human activities on the fresh water resources. Water reuse is no longer restricted to water scarce areas. It is becoming an essential component to increase the water supply reliability, both in quantity and quality, and to comply with increasing environmental standards. It is regarded that NOW is the time to consolidate and improve methodologies and technologies in order to better recognize the benefits of reuse and to enhance the implementation of water reuse schemes. Amongst, this paper focused on Water Reuse applications relied on MBR technology.

The membrane (MBR) technology –technology of membrane separation of activated sludge– is the combination of activated sludge together with micro- or ultra- filtration membranes with pore size of typically between 10 nm to 0.5 μm to produce suspended solid free effluent. The membrane filtration process replaces final clarifier step in conventional activated sludge treatment achieved by gravity only. The membrane also provides disinfection by eliminating bacteria and virus. Additionally, it enables operation at higher sludge concentration typically up to 18-20 g/l as compared to 5-6 g/l at conventional activated sludge systems, which reduces required footprint.

History of MBR technology

After initial development started in the late 60s, the MBR technology for wastewater treatment experienced rapid development from the early 1990's onwards. The first systems commercialised in the 70's and 80's were based on what have come to be known as sidestream configurations, i.e. the membrane separation step was employed in an external sludge recirculation loop, mainly with in-to-out flow through organic or ceramic tubular membranes. Due to the high energy demand, these technologies targeted only small and niche market applications such as treatment of ship-board sewage, landfill leachate or industrial effluents.

In the early 90s, the Japanese Government launched an ambitious 6-year R&D project which led to a major technological and industrial breakthrough of the MBR process: the conception of submerged membrane modules, working with low negative pressure (out-to-in permeate suction) and membrane aeration to reduce fouling. This paved the way towards a significant reduction of capital and operation costs, due to the reduction and simplification of equipment and the abandonment of the energy demanding sludge recirculation loop.

The commercialization of the submerged MBR system precipitated rapid and extensive market penetration. The first pilot-scale European submerged MBR plant for municipal wastewater was built in 1996 (in Kingston Seymour, UK), soon followed by the construction of the full-scale Porlock WWTP (UK, commissioned in 1998, 3,800 p.e.), Büchel and Rödingen WWTPs (Germany, 1999, resp. 1,000 and 3,000 p.e.), and Perthes-en-Gâtinais WWTP (France, 1999, 4,500 p.e.). A few years later only, in 2004, the largest MBR plant worldwide was commissioned to serve a population of 80,000 p.e. (in Kaarst, Germany). The size of installations has thus grown from few thousand to hundreds of thousands population equivalent in only a few years.

Kubota Submerged Membrane Unit® (SMU)

Kubota started its Membrane Business by MBR using tubular type Ultra-Filtration for a solid-liquid separation in 1988. Consequently, in 1991, Kubota developed Kubota Submerged Membrane Unit® (Kubota SMU) with Micro-filtration employing the flat-sheet membrane panels. In 1998, the first European (and the first oversea Kubota SMU) MBR application in municipal sewage treatment plant was accomplised in Porlock, Dorset, UK. Since then, there are more than 3,000 applications treating both municipal and industrial wastewater in operation employing Kubota SMU worldwide.

Accalimed Quality: Certifications awarded to Kubota SMU. Kubota SMU system meet the filtration performance and disinfection requirements for compliance with the California Water Recycle Criteria (Title 22) in USA. Both type 510 and type 515 membrane panels were given departmental certification after having demonstrated the ability to meet the governmental definition of Desinfected Terciary Recycled Water: log-5 virus reduction and daily average turbidity of less than 2 NTU. In addition, Kubota SMU system complies with the World Health Organization Criteria, the International Maritime Organization Criteria, the EU Bathing Water Directive.

RECLAIMED WATER APPLICATIONS WITH KUBOTA SMU

The public perception of reclaimed water varies between country and culture. The more direct use of reclaimed water, *i.e.* in its height, *drinking* (potable use) of reclaimed water in particular, raises more controversial opinions. In Singapore, NEWater has been successfully introduced and received majority of acceptance with a background that Singapore may face serious shortage of potable water –the contract for water supply between Malaysia expires in 2011 and 2061; whereas reclaimed water 'direct' applications received rejections from the public in Australia. Nevertheless, water recycling is regarded as one of the sensitive issues in politics because of the investment cost in new technologies –greywater recycling is still cost effective as compared to desalination though. In this section, governmental policies and accepted existing Water Reuse references are described by country.

Japan

In Japan, the driving force for water reuse was the high demand of both domestic and industrial water in cities due to the high concentration of population and industries. To solve this, the local government has encouraged the operators of large-scale buildings to install water reuse systems and use the treated intermediate water –Chuusui (direct translation is Middle Water, which ranks between Jousui (Upper Water = potable water) and Gesui (Lower Water = wastewater))– the term used to describe recycled water in Japan. Kubota SMUs are in operation under the guidelines of Tokyo Metropolitan area in particular and effluent is often reused as toilet flushing water and irrigation. Figure 1 shows a toilet flushing water reuse application at an office building in Tokyo Metropolitan area. Typical effluent quality obtained by Kubota SMU in this application is shown in

Figure 2. There are currently 48 applications employing Kubota SMU effluent for toilet flushing and irrigation water reuse.

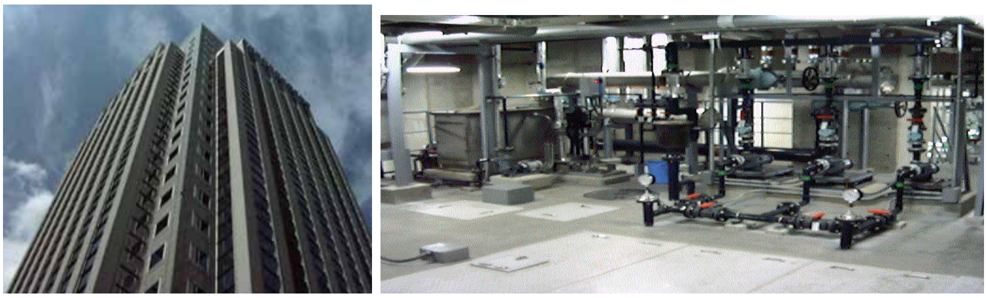


Figure 1. An office building equipped with Chuusui system with membrane surface area of 640 m^2 (design flow of 189m^3/d), operated since May 1997.

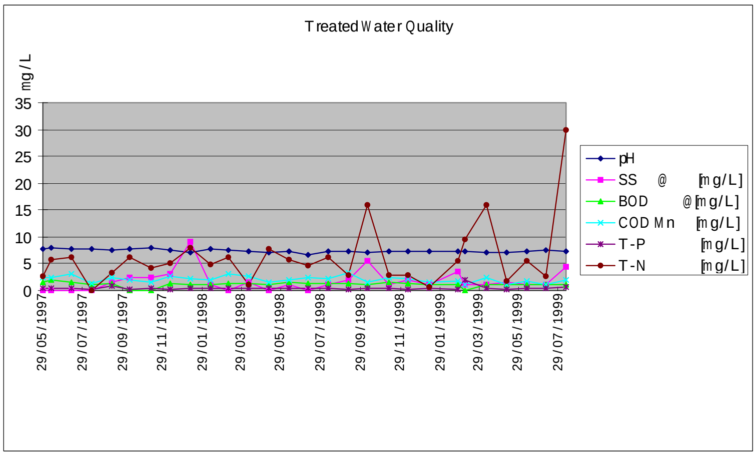
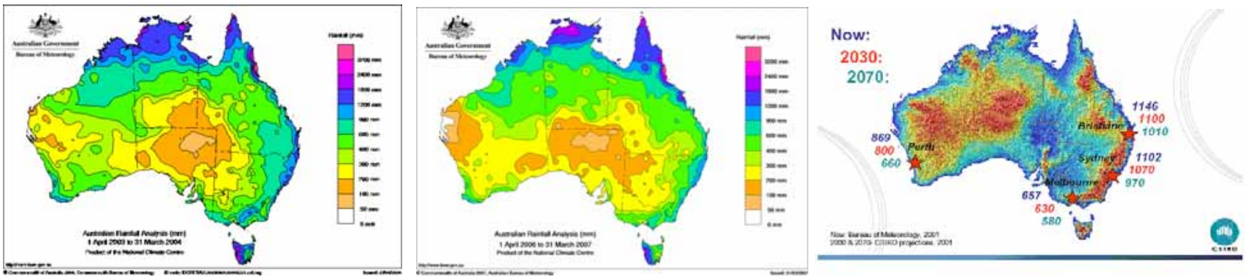


Figure 2. Typical effluent quality data by Kubota SMUs.

Australia

Severe rainfall deficiency in southeastern part in recent years (shown in Figure 3) was national driving force of water reclaiming scheme. Strategies, the guidelines/definitions of recycling water, levels of allowed reuse purposes as well as public perceptions of water reuse vary regionally. Queensland issued water recycling strategy –state government initiative to encourage water recycling that is safe, environmentally sustainable and cost-effective.



3-a 3-b 3-c

Figure 3. Australia's 12-monthly rainfall maps (3-a. Apr2003-Mar2004, 3-b. Apr2006-Mar2007, 3-c. projections of rainfall of 2006, 2030 and 2070) showing southeastern part suffers from recent short rainfall (Bureau of Meteorology, 2007).

The agriculture (irrigation) sector occupies more than 75% in the consumptive water use in Queensland (Queensland, 2000). The most common recycled water use is non-potable which includes irrigation, industrial reuse (cooling, laundries, car washing), toilet flushing, ground water recharge. In metropolitan areas in NSW, it will become compulsory to install rainwater tank to new homes to be built. Sewer mining, dual-pipeline, direct reuse of greywater and rainwater are the key projects stated in Sydney Water's Environmental plan in 2006-2011 (Sydney, 2006). Although potential reclaimed water use is high in Australia, recycled water is used only 2% in irrigation sector in Queensland as compared to 5% in California, USA. Despite the national pressure, latest public rejections towards potable use of recycled water hit the news headline and thus, one of the milestones for Water Reuse is receiving good public perception in the country. Figure 4 shows a greywater recycling system flow chart at Inkerman Oasis, Victoria.

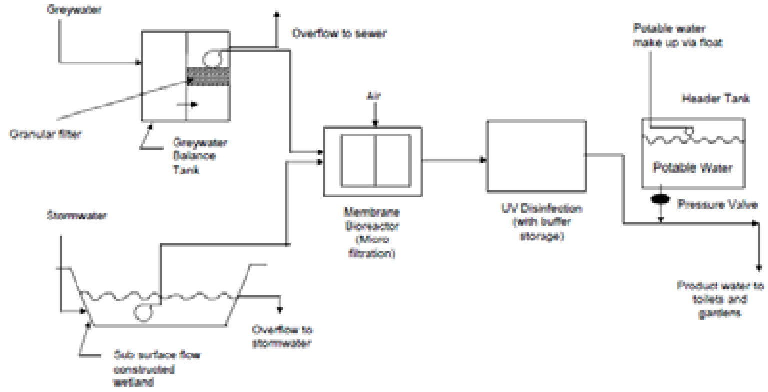


Figure 4. Greywater recycling system at Inkerman Oasis.

Spain

Spain is one of European countries where water resource has been historically scarce. Local governments encourage water reuse and offers subsidies for the cost of water reuse applications (Catalana, 2006). Industrial applications such as winery and golf course irrigation have been successfully implemented (Figure 5). Sewage plants aiming at water reuse is acquiring public perceptions and many water recycling projects are underway.

4-a **4-b**

Figure 5. A Winery (4-a) and golf resort (4-b) application where Kubota SMU effluent is reused for irrigation purpose.

CONCLUSIONS

Reclaimed water has been obtaining its importance as sustainable water due to global warming and overpopulation in metropolitan areas. MBR technology offers safe and reliable effluent to be reused for many applications. Continuous efforts to improve energy efficiency as well as to reduce capital/operational cost to appeal 'affordability' towards public will be required to gain perceptions for wider water reuse applications.

REFERENCES

Bureau of Meteorology, Australian Government (2005). Drought Statement issued 2nd June 2005.

Department of Primary Industries (2000). Agricultural Water Recycling Background Study: Prepared for Queensland Water Recycling Strategy.

Sydney Water (2006). Sydney Environment Plan 2006-2011.

Agència Catalana de l’Aigua (2006). Subsidies for water-saving and non-drinking reuse initiative.

Impact of wetting agents on filtration properties of membranes applied in municipal MBRs

J. Kochan, S. Buetehorn, C. N. Koh, T. Wintgens, T. Melin
D. Volmering, K. Vossenkaul

Institut fuer Verfahrenstechnik /Department of Chemical Engineering , RWTH Aachen University, Turmstr. 46, 52056 Aachen (Germany), E-mail: kochan@ivt.rwth-aachen.de, Phone: +49241 80 96233

Membrane bioreactors are widely used in municipal water treatment for decades of year. During the last years, the whole process has been optimized in order to achieve high permeabilities and permeate quality. Besides efforts which have been made to optimize fouling prevention, process parameters or influent quality, there is a need to investigate membrane properties and their impact on filtration performance. Membrane characterization can provide common information about pore size distribution, pore density, membrane hydrophilicity, wettability and surface charge.

This work is focusing on investigating the impact of wetting agents on membrane filtration performance. It was found out that during pure water filtration with hollow fibre PES membranes the permeability was changing when filtration tests were carried out while permeate flux was set to 20 $l.m^{-2}.h^{-1}$, 15 $l.m^{-2}.h^{-1}$, 25 $l.m^{-2}.h^{-1}$ and once again to 20 $l.m^{-2}.h^{-1}$. For each permeate flux value the trial lasted 30 minutes, i.e. permeability measurement for the same permeate flux value was repeated after 90 minutes. The measurements were conducted on the fibre test rig which is shown in the figure 1.

The results for four types of membranes are shown in the figure 2.

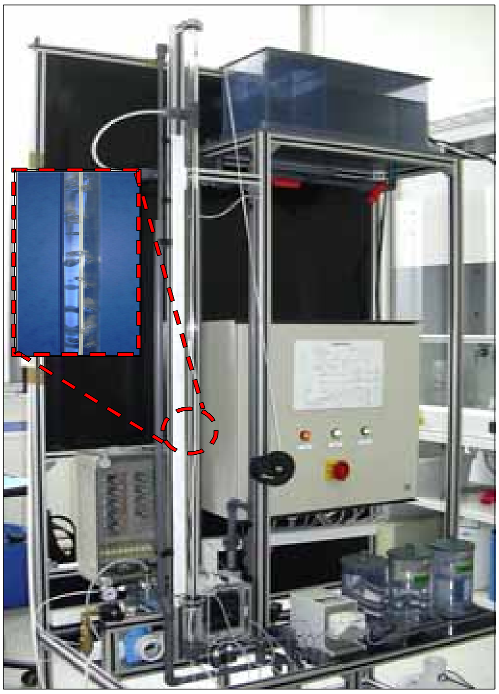


Figure 1: Single fibre test rig

This phenomenon is explained by membrane wettability. When starting filtration with a virgin membrane, not all pores are wetted. Such smaller pores are not taking part in filtration and membrane does not work efficiently. After 90 minutes of filtration the smaller pores are wetted as well and can contribute to membrane filtration performance. The increasing TMP play also an important role as the membrane pore wetting goes.

The use of wetting agents seemed to be a possibility to standardize the procedure of membrane characterization. This work will focus on different wetting agents which have a preferably low surface tension. Further step, the concentration of wetting agents, temperature at which the membrane is to be wetted and membrane exposure period will be varied. After the trial with hollow fibre membranes, flat sheet membranes will be tested. The efficiency of wetting agents applied on different polymeric materials will be investigated for PVDF, PES and CA membrane materials. The impact of TMP on wetted membrane performance will be observed on single fibre test rig as well.

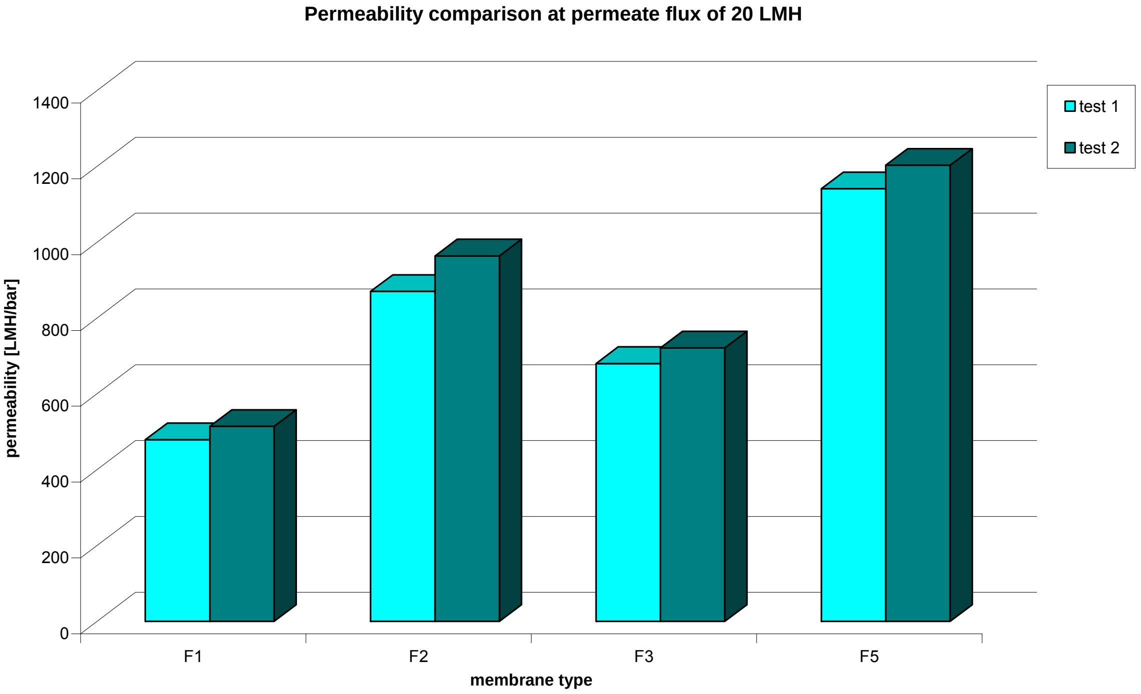


Figure 2: Permeability change of four different PES hollow fibre membranes at the beginning and the end of the filtration trial for the same permeate flux value

The overall goal of this research work is to create a standardized protocol for membrane characterization on the basis of pure water filtration.

REFERENCE

Batsch A., Melin T, Thomas H., Klee D., Hoecker H., Schaefer S., Vossenkaul K. (2005): Modifikation von Polymer-Membranen zur nachhaltigen Leistungssteigerung beim Einsatz in der Abwasseraufbereitung. Project Report, March 2005

J.S. Kang, K.Y.Kim, Y.M. Lee, Preparation of PVP immobilized microporous chlorinated polyvinyl chloride membranes on fabric and their hydraulic permeation behaviour, J. Membr. Sci. 214 (2003) 311-321

W.Y.Chuang, T.H.Young,W.Y.Chiu,C.Y.Lin, The effect of polymeric additivies on the structure and permeability of poly(vinyl) alcohol asymmetric membrane, Polymer 41 (2000) 5633

T.P. Hou, S.H.Dong, L.Y.Zheng, The study of mechanism of organic additivies action
in the poly(sulfone) membrane casting solution, Desalination 83 (1991) 343

Sample pre-treatment device for continuous EPS analysis in membrane bioreactor systems

Renata Mehrez *, Martin Jekel * and Mathias Ernst **

* Technische Universität Berlin, Department of Water Quality Control, Straße des 17. Juni 135,
Sekr. KF 4, D-10623 Berlin, Germany
(E-mail: *renata.mehrez@tu-berlin.de; martin.jekel@tu-berlin.de)*
** Technische Universität Berlin, Centre for Water in Urban Areas, Straße des 17. Juni 135,
Sekr. KF 4, D-10623 Berlin, Germany
(E-mail: *mathias.ernst@tu-berlin.de)*

Abstract
Polysaccharides and proteins are the main components of extracellular polymeric substances (EPS), which are considered to give a major contribution to the overall fouling potential in membrane bioreactor (MBR) systems. The approach of the present work is to develop an on-line EPS sensor, which will measure continuously the main components of EPS directly in activate sludge. The present study focuses on the development of a sample pre-treatment device for the subsequent analysis of polysaccharides and proteins. The sample pre-treatment device consists of a stainless steel filter with 1 µm pore diameter that filters activated sludge continuously. The specific aeration of the filter and the relaxation intervals prevent the clogging process and prolong the filter run-time. The filtration tests were conducted to determine the most appropriate filtration protocol with respect to the longest possible filter run-time. A further objective of the study was to verify if the quality of the filtrate changes under different filtration protocols and over the time with regard to measured parameters like polysaccharides, proteins and other water constituents. The filtration experiments were conducted in activated sludge from MBR pilot plant. Manually paper filtrated sample was taken as a reference sample. The results show that manual and automated filtration modes have comparable separation properties, enabling the implementation of the newly developed pre-filtration device for continuous application.

Keywords
Activated sludge; extracellular polymeric substances; membrane fouling; polysaccharides; proteins; sample pre-treatment

INTRODUCTION AND OBJECTIVES

Membrane bioreactor (MBR) technology is becoming an important innovation in the biological treatment of wastewater. Despite many advantages over conventional treatment (e.g. better effluent quality) membrane fouling still restricts the widespread application of MBR and increases both, the investment and operational costs. MBR fouling is considerably influenced by the content of the extracellular polymeric substances (EPS) in the activated sludge of the MBR system, also called soluble microbial products (SMP) (Le-Clech et al., 2006; Rosenberger et al., 2006). The continuous determination of their concentrations is an important step for understanding and controlling the fouling phenomena. Up to now the main components of EPS – the polysaccharides and proteins – are measured by means of photometric measurements, their sum is expressed as EPS content (Flemming&Wingender, 2001; Le-Clech et al., 2006). However, the sampling and analysis is performed infrequently and no consistent data about the variation of these parameters in activated sludge during a long period of time are available. Moreover the determination methods are not standardised, what leads to hardly comparable results of different research studies.

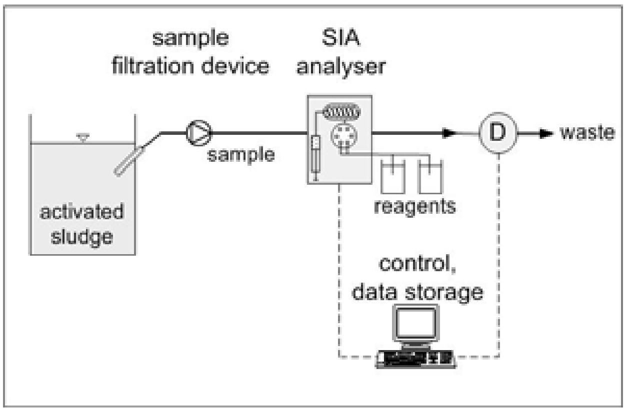


Figure 1. Components of on-line EPS sensor.

The approach of the present work was to develop an on-line EPS sensor, which will measure continuously the main components of EPS directly in activated sludge. This would allow an on-line monitoring of polysaccharides and proteins and subsequently a better understanding of fouling phenomena in terms of these parameters. The figure 1 shows the set-up of the EPS online sensor, which consists of two main components (i) the sample pre-treatment device and (ii) the SIA analyser (SIA: Sequential Injection Analysis) for measuring of polysaccharides and proteins by photometric methods (Mehrez et al., 2007). The present work focuses on the development of a sample pre-treatment device for the subsequent photometric analysis of polysaccharide and protein concentration. The sample pre-treatment device should run continuously and automatically and it should separate the particles from the activated sludge sample. However it is important that the colloidal substances, which may be partly the polysaccharides and proteins, remain in the sample. As a pre-treatment step the filtration was chosen. It corresponds to the manual sample pre-treatment usually done by paper filtration. The study's objective was to set-up the sample pre-filtration device and to verify if the different filtration protocols (flux and filtration interval was varied) change the quality of the filtrate with regard to the measured parameters.

MATERIAL AND METHODS

Pre-filtration device

The sample pre-treatment device consists of cylindrical stainless steal filter (MicraMesh, cmc Instruments GmbH, Germany) with the surface of about 50 cm² and a pore diameter of 1µm. The filter is submerged in the reactor with activated sludge. The filtration mode is out/in. At the bottom of the filter an aeration tube is fixed. The ascending air bubbles inhibit the fast formation of the cake layer and prolong the filtration time until the filter is blocked. The filtration is intermitted by the relaxation, the peristaltic pump stops and the valve opens allowing the air to come into the tube system. The filtrate contained in the tube flows backwards through the filter into the activated sludge reactor and releases the filter cake from the filter. The pump and the valve are controlled by a clock-pulse generator.

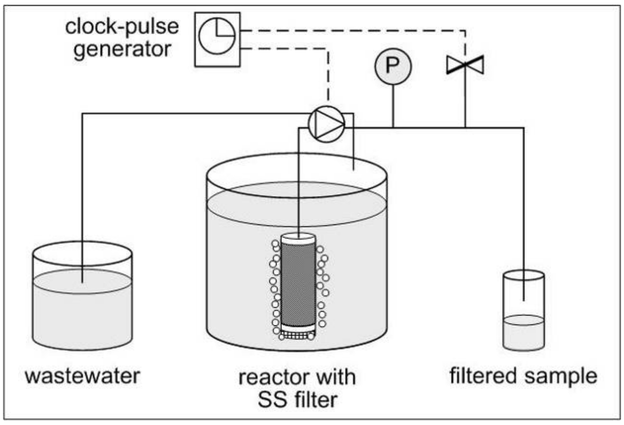


Figure 2. Scheme of the filtration experiment.

Filtration experiments

Filtration experiments were conducted with the sample pre-treatment device to determine the most appropriate filtration protocol with regard to the longest filter run-time and to the highest flux. A further objective was to verify if the quality of the filtrate changes under different filtration protocols with regard to the measured parameters. The filtration experiments were carried out in a small reactor filled with 1.9 L or 0.8 L of activated sludge from MBR pilot plant for municipal wastewater in Berlin (figure 2). The activated sludge was filtrated continuously with different flux values. Moreover the filtration interval was varied in different experiments. The table 1 summarises the most important parameters of the experiments. Municipal wastewater was continuously added to the reactor with the same flow rate as the activated sludge was filtrated. During the experiments the vacuum pressure was measured (SEN-3, 0-160 mbar, Kobold Messring GmbH, Germany). The filtration experiments were stopped after ten (experiment I) or six days (experiment III) or when the filter was clogged and the vacuum pressure was higher than 200 mbar (experiment II). After each experiment the filter was cleaned with 1 % sodium hypochlorite (Roth, Germany) for at least two hours followed by the treatment with 10 % concentrated sulphuric acid for at least two hours.

Table 1. Conditions during the filtration experiments.

PARAMETERS	Filtration I	Filtration II	Filtration III
Flux [L/(h*m²)]	10	17.5	17
Filtration / relaxation interval [min/min]	20 / 2	20 / 2	10 / 2
Experiment duration [d]	10	3	6
Hydraulic retention time [h]	32	10	12
pH- value	7.8 - 6.2	8.6 - 6.5	8.5 - 6.9
O_2 concentration [mg/L]	-	~8.6	~9.0
Temperature [°C]	~18	17.5 - 18.3	17.9 - 18.9

Samples

The filtrate and wastewater samples were taken each day, every other day respectively and analysed for different parameters. Additionally 50 mL of activated sludge was filtered manually through the paper filter (Schleicher and Schuell filter - 589/1 black ribbon, 125mm), which was pre-rinsed with 200 mL of pure water (Millipore, France). This sample was used as reference for the automated continuous filtration.

Analysis

During the filtration experiment the mixed liquor suspended solid concentration (MLSS), pH-value (WTW Micrprocessor pH 537, Germany), the oxygen concentration (WTW oxi 325, Germany) and the temperature of activated sludge was measured. Parameters like turbidity (2100N Hach, Germany), polysaccharide and protein concentrations were determined in the filtrate from manual and automated filtration and in the wastewater. Polysaccharide concentrations were analysed according to the method of Dubois et al. (1956). D-Glucose-Monohydrate (Sigma, USA) was used for calibration (2–100 mg/L). Samples were analysed at 490 nm as two or three replicates and the results are given as glucose equivalents. Protein concentrations were analysed according to Frolund et al. (1996), which is a modified method of Lowry et al. (1951). Bovine serum albumin (Roth, Germany) was used for calibration (5–200 mg/L). Samples were analysed at 750 nm as two or three replicates and the results are given as BSA equivalents.

Both filtrates and the wastewater were filtered through 0.45 cellulose nitrate filter (Sartorius, Germany), the dissolved organic carbon (high TOC Elementar Analysensysteme GmbH, Germany; thermo-catalytic oxidation) and the biopolymer concentrations (SEC, LC-OCD system by DOC-Labor Dr. Huber, Germany) was determined. The size exclusion chromatography (SEC) can be used for the characterization of organic carbon in a sample. 1 mL sample was injected into an HPLC column (Toyopearl HW-55S by Tosoh Bioscience, Japan). A phosphate buffer (Na_2HPO_4, KH_2PO_4) was used as eluent transporting the sample through the column where its components are separated according to their size.

RESULTS AND DISCUSSION

Run-time of the pre-filtration device

One of the objectives was to test the pre-filtration device under different filtration conditions and to determine the most appropriate filtration protocol in regard to the longest filter run-time and to the highest flux. Thereby (i) the filtration interval was change from 20 to 10 min and (ii) the flux from 10 to 17 L/(h*m²) (see table 1).

Influence of the filtration interval. Figure 3 shows the development of the vacuum pressure versus filtration time of experiment II and III. These two experiments have the same parameters but the length of filtration interval. The filter in the experiment III with a filtration interval set to 10 min clogs much slower. The vacuum pressure increases at the end of the experiment in the sixth day to the value of -37 mbar. The experiment II (filtration interval: 20 min) runs only three days. After this time the vacuum pressure was higher than -200 mbar and the filtration was stopped, because air bubbles penetrated into the tube system. The MLSS value, which can influence the process of filter clogging was in both experiments comparable (figure 4). The mean value of the MLSS was 6.9 g/L and 7.3 g/L for the experiment II and III respectively. In the experiment II the MLSS became smaller from the second day of the test, because of initial lost of activated sludge due to a foaming event.

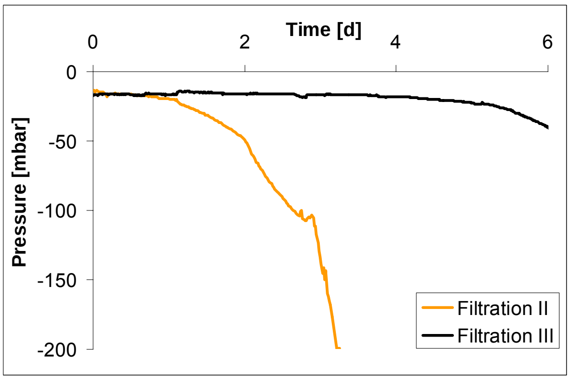


Figure 3. Development of vacuum pressure during experiment II and III.

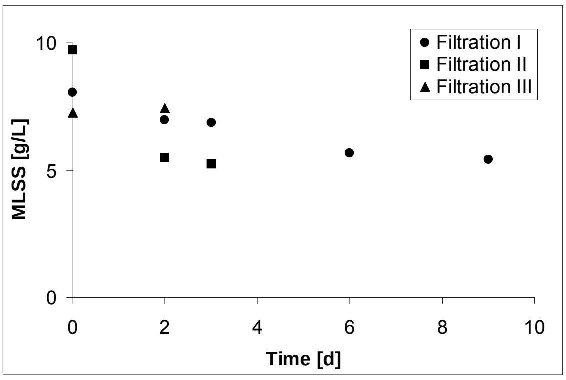


Figure 4. MLSS of activated sludge for different experiments.

Influence of the filtration flux. The higher the flux is, the more filtrate is produced, what is necessary for representative sampling of the activated sludge. The pre-filtration device should filtrate sufficient volume not only for the continuous analysis of polysaccharides and proteins, but also for the exchanging of filtrate dead volumes in the tubes between the filter and the analyser.

In the experiment I the flux was set to 10 L/(h*m²) and in the experiment II to 17.5 L/(h*m²), the filtration interval was fixed to 20 minutes. The duration of the experiments show indirectly that the increasing of the flux accelerates the filter clogging to a high extend. In the experiment II the filter have had to be cleaned already after three days. On the other hand the experiment I was stopped after ten days without any indices that the filter was clogged (no air bubbles could be seen in the tube system). Direct comparison of the pressure development can not be done, as the vacuum pressure data are not available for experiment I. The MLSS value was in both experiments comparable (figure 4). The mean value of the MLSS was 7.3 mg/L for the experiment I (up to the fourth day) and 6.9 mg/L for the experiment II.

Filtrate quality

Another objective of the study was to characterise the separation properties of the stainless steel filter with regard to the parameters like polysaccharide and protein concentrations and to describe their change over the run-time. This change can be expected because of the permanent particle deposition onto and in the filter during the continuous filtration. An activated sludge sample filtered manually with paper filter was taken as reference for this experiment.

Figure 5 shows the developing of all measured parameters (turbidity, DOC, polysaccharide and protein concentration, biopolymer concentration – (a-f)) during all filtration experiments (I-III) for manual (paper filter) and automated (stainless steel (SS) - filter) filtered sample. In most of the samples the content of the measured parameters was slightly higher in the manual filtered samples. This difference was the highest at the beginning of the filtration experiments; however it decreased with ongoing operation. After the third or the fourth day the difference got very small or even the tendency changed and the concentration in SS filtrate became a little bit higher than in the paper filtrated sample.

Turbidity. The turbidity is an integrative parameter, summarizing the optical properties of particles present in the sample. Both filtration modes (SS filtration and manual) let some particles pass, because the pore diameter of paper filter is between 10-25µm and of SS filter about 1 µm, so higher than the upper size limit of dissolved substances. The mean value of the turbidity of paper filtrate was for all filtration experiments 1.5, 2 and 0.7 NTU respectively. The turbidity in the SS filtrate was higher in the first three days, in mean about 0.8 and 1.1 NTU for the experiments I and III. Afterwards the difference decreased to a value smaller than 0.4 NTU. In the experiment II the tendency was reversed, the turbidity of paper filtered sample was higher than in SS-filtrate (2 and 1 NTU; only one measurement done).

Dissolved organic matter. The DOC a sum-parameter, which expresses the amount of dissolved organic components in a sample. With regard to this parameter the paper filter showed also higher DOC retention. In the first three days the DOC in the paper filtrate was about 1.5 and 1.8 mg/L higher than in the automated filtrated sample in the experiments I and III. This value decreased with the ongoing operation to 0.8 and 1.2 mg/L, what corresponds to a difference smaller than 5 % and 9 %. In the experiment II the DOC in both samples had almost the same value.

Polysaccharides and proteins. The polysaccharides and the proteins are the substances, for which analysis the sample pre-treatment device was developed. The comparison of the manual and automated filtration shows in general that the SS filtrate contains a little higher concentration of both substances. The difference of the polysaccharide concentration was in average smaller than 1.1 mg/L and of protein concentration smaller than 2.1 mg/L. From the second day of the experiment II the SS filtrate contained a smaller amount of polysaccharides and of proteins (mean difference was 0.7 mg/L and 1.2 mg/L respectively). The ratio of polysaccharide and protein concentrations in paper and SS filtrate varied during the experiments and no clear trend could be observed with ongoing filtration as for the turbidity and DOC.

Biopolymers. With the size exclusion chromatography the separation of the organic compounds can be done according to their molecular weight. In the figure 5-f the LCOCD chromatograms are shown for manual and automated filtration for the third day of the experiment. The first small peak of the chromatogram represents the molecules with large molecular weights like polysaccharides, proteins and other biopolymers. This fraction was identified as possible fouling active substances (Rosenberger et al. 2006, Laabs et al, 2006). The later the organic substances eluate the smaller they are. The second large peak represents smaller humic substances, humic hydrolysates and acids.

The comparison of LCOCD chromatograms shows that in accordance to the DOC measurement the paper filter retains more organic bio-polymers than the SS filter. Also humic substances and low molecular acids (figure 5-f) are better removed by the paper filter. Relating to the whole chromatogram the paper filtrate contains in average 2, 15, and 11 % less DOC than the automatically filtrated sample for experiments I, II and III respectively.

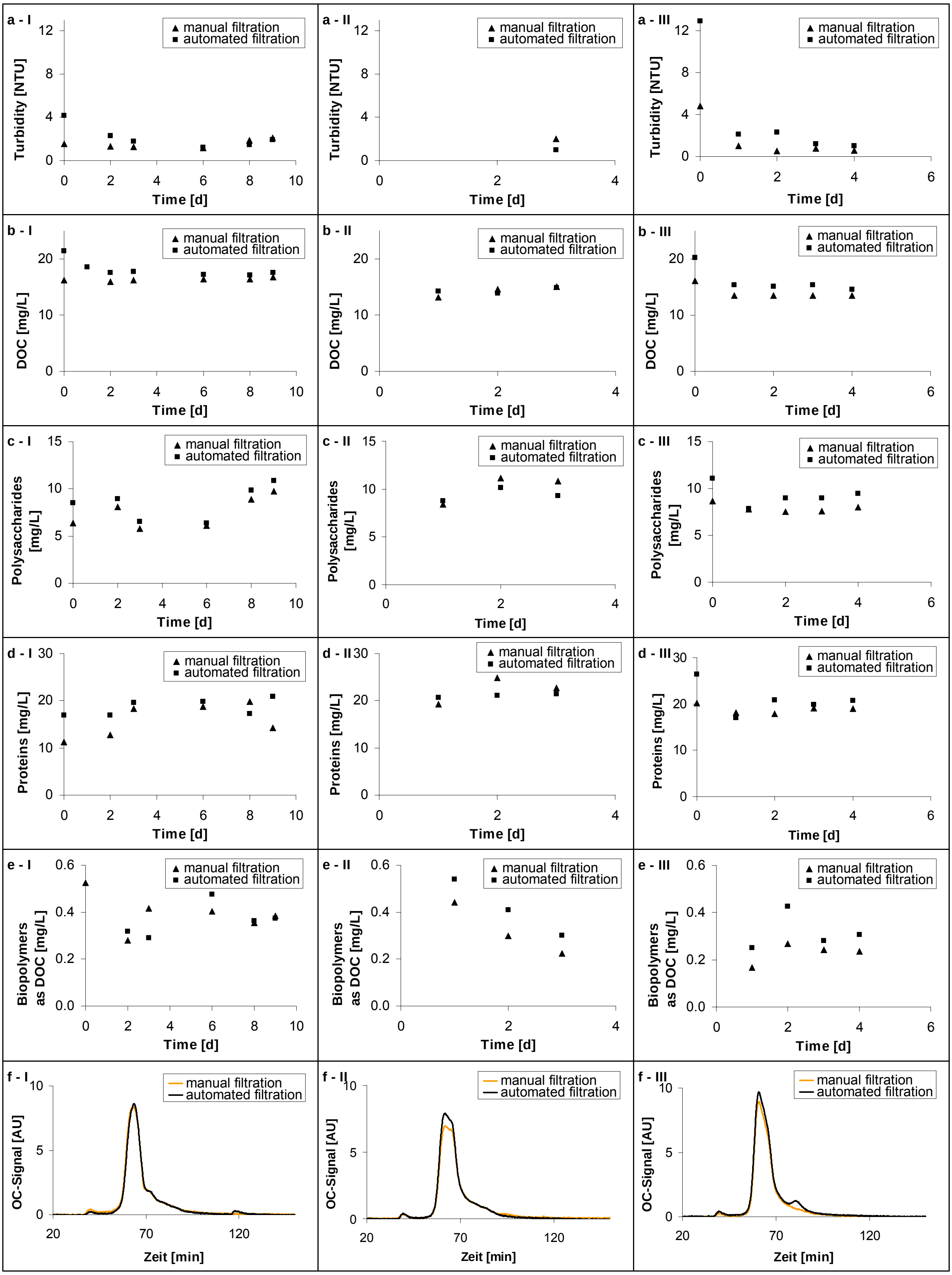


Figure 5. Comparison of measured parameters (a-f) between manual filtration (paper filter) and automated filtration (stainless steel filter) during three filtration experiments; (I) experiment I, (II) experiment II, (III) experiment III.

The generated results show that the different filtration protocols have no significant influence on the stainless steel filter separation properties. However the separation characteristics change with ongoing operation and filter blockage. It can be observed that in experiment II, the SS filter was clogged rapidly (the vacuum pressure increased after the first filtration day to the value of -36 mbar and after the second day to -50 mbar) and consequently the SS filter retains the measured parameters to a slightly larger extent than the paper filter.

CONCLUSIONS

The set-up of the pre-treatment device for the continuous filtration of activated sludge was successfully realised. The tests with activated sludge showed that a low filtration rate, relaxation and specific aeration of the filter are effective means to remove the cake layer and to prevent filter clogging. The used agents for the filter cleaning - sodium chlorite and sulphuric acid was sufficient to remove the deposited particles on the stainless steel filter surface and in the pores.

Three filtration experiments were conducted to define the boundary condition for the use of the pre-filtration device for continuous MBR operation. It could be observed that the shortening of filtration interval and the decreasing of the flux prevent the SS filter from fast blockage, what prolongs the time until the filter cleaning. The results of these short-time experiments show that the experiment I (filtration interval 20 min, flux 10 L/(h*m²)) and experiment III (filtration interval 10 min, flux 17 L/(h*m²)) are comparable with respect to the filter run-time and the filtrate quality. However, higher flux is important to guarantee the representative sampling for the subsequent polysaccharides and proteins analysis. The higher filtrate volume can ensure that the dead volume of the filtrate, which is contained in the tubes between the filter and the analyser, is exchanged completely. In one hour approximately 45 mL and 66 mL of activated sludge can be filtered with the filtration protocol of experiment I and III respectively. The filtrate dead volume can be estimated to 45 mL and the needed volume for the analysis is 1 mL. To guarantee the complete exchange of filtrate in the tubing system the filtration protocol with filtration interval of 10 min and flux 17 L/(h*m²) (exp. III) is concluded to be the most appropriate for future application in the MBR pilot plant.

In general the SS filter shows comparable separation properties as paper filter (manual filtration). However during the automated filtration more substances are passed through the filter than during the manual filtration, what can be explained by different filtration mechanisms. The paper filtration is based on the cake filtration and although the pore diameter is quite large (10-25 µm) substances with a smaller size are retained in the cake and hindered to pass the filter (see figure 5-f). During the automated filtration the filter cake is not formed on the filter surface due to the ascending air bubbles, so only the filter material presents a barrier for the particles. With ongoing filter operation both filtration modes align to each other in regard to the turbidity and DOC. The difference of the polysaccharide concentrations in both filtrates was up the second filtration day in average smaller than 0.7 mg/L and of protein concentrations smaller than 1.2 mg/L. The difference between paper and SS filter is less significant, when the measurement error is taken into account (1 mg/L for polysaccharides and 0.5 mg/L for proteins).

In experiment II the vacuum pressure increased after the second filtration day to -50 mbar. Under these conditions the measured parameters (except biopolymer concentration) were higher in the manual filtered sample. This effect should be examined in further filtration experiments, when the automated filtration runs for a longer period than considered in this study.

The results obtained in this study show that the pre-treatment device is suitable to be applied in a continuous EPS testing sensor in the MBR pilot plant. In the future work the pre-filtration device will be tested with a filtration interval of 10 min and with the flux 17 L/(h*m²) or higher for a longer time until the SS filter is completely clogged. Afterwards it will be coupled with SIA analyser and tested on the MBR pilot plant.

ACKNOWLEDGMENTS

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REFERENCES

Dubois M., Gilles K.A., Hamilton J.K., Rebers P.A., Smith F.S. (1956). Colorimetric method for determination of sugars and related substances. Anal. Chem., 28(3), 350-356.

Flemming H.-C., Wingender J. (2001). Relevance of microbial extracellular polymeric substances (EPSs) – Part I: Structural and ecological aspects. Wat. Sci. Tech., 43 (6), 1–8.

Frolund B., Palmgren R., Keiding K., Nielsen P.H. (1996). Extraction of extracellular polymers from activated sludge using a cation exchange resin. Wat. Res., 30(8), 1749-1758.

Le-Clech P., Chen V., Fane T.A.G. (2006). Fouling in membrane bioreactors used in wastewater treatment. J. Membr.Sci., 284 (1-2), 17-53.

Laabs C. N. Amy G. L., Jekel M.(2006). Understanding the size and character of fouling-causing substances from effluent organic matter (EfOM) in low-pressure membrane filtration. Environ. Sci. Technol. 40(14), 4495-4499.

Lowry O.H., Rosebrough N.J., Farr A.L., Randall R.J. (1951). Protein measurement with the folin phenol reagent. J. Biol. Chem., 198, 265-275.

Mehrez R., Ernst M., Jekel M. (2007). Development of a continuous protein and polysaccharide measurement method by Sequential Injection Analysis for the application in membrane bioreactor systems. Proceedings IWA International Membranes Conference for Water and Waste Water Treatment, Harrogate, UK.

Rosenberger S., Laabs C., Lesjean B., Gnirss R., Amy G., Jekel M., Schrotter J.-C. (2006). Impact of colloidal and soluble organic material on membrane performance in membrane bioreactors for municipal wastewater treatment. Wat. Res., 40, 710-720.

Influence of operating conditions on chemically reversible fouling in submerged MBRs.

T. Miyoshi* T. Naruse* N. Yamato* K. Kimura* and Y. Watanabe*

* Department of Urban and Environmental Engineering, Graduate School of Engineering, Hokkaido University, Kita-13, Nishi-8, Kita-ku, Sapporo 060-8628, Japan.
(E-mail: jms-mbr@eng.hokudai.ac.jp)

Abstract In this study, continuous operation of pilot-scale MBR was conducted in order to investigate the influence of operating conditions on chemically reversible fouling. Analyses of organic matter contained in mixed liquor of MBRs and the foulant desorbed from fouled membrane were carried out to elucidate the nature and behavior of organic matter which contribute to the evolution of chemically reversible fouling. Chemically reversible fouling was enhanced when MBRs were operated with higher F/M ratio. No clear relationship was observed between comprehensive indexes such as dissolved organic carbon concentration and rate of chemically reversible fouling. Relative abundance of polysaccharide-like substance in the foulant increased with the rate of chemically reversible fouling observed in continuous operation. Regardless of F/M ratio change, there were significant similarities in the monosaccharide compositions of membrane foulants desorbed from fouled membranes. In addition, the monosaccharide compositions of foulants were considerably different from those of dissolved carbohydrate in the mixed liquor of MBRs. It is most likely that some specific carbohydrate cause chemically reversible fouling preferentially.

Keywords: chemically reversible fouling, membrane bioreactor (MBR), operating condition

INTRODUCTION

Membrane bioreactors (MBRs) have several advantages over the conventional activated sludge systems and therefore are considered as a promising technology for future wastewater treatment process. For wider application of MBRs, however, problems associated with membrane fouling which results in deterioration of membrane permeability and consequently higher energy consumption should be solved. Membrane fouling can be divided into two types i.e. physically reversible fouling which can be restored by physical membrane cleaning such as backwashing and chemically reversible fouling which can not be restored by physical membrane cleaning and consequently can only be restored by chemical membrane cleaning. While physically reversible fouling can be mitigated by the implementation of regular physical membrane cleaning, filtration resistances derived from chemically reversible fouling gradually increase even though regular physical membrane cleaning is carried out. Therefore, the suppression of chemically reversible fouling is particularly of importance.

Various factors affecting membrane fouling, including membrane permeate flux, aeration intensity, sludge concentration in the reactor, solid retention time and food-to-microorganism ratio, were pointed out in previous studies. However, most of previous study did not focus on the evolution of chemically reversible fouling. Current understanding of the influence of operating conditions on the evolution of chemically reversible fouling is still insufficient.

The objective of this study was therefore to investigate the influence of operating conditions on chemically reversible fouling in MBRs treating real municipal wastewater. A pilot-scale MBR operation at an existing wastewater treatment facility was carried out. Organic matter which was related to chemically reversible fouling was desorbed from fouled membrane at the termination of the operation and was analyzed in order to investigate the futures of the components responsible for chemically reversible fouling.

MATERIALS AND METHODS

Continuous operation of pilot-scale MBRs was carried out at the Souseigawa Municipal Wastewater Treatment Facility in Sapporo city, Japan. Three MBRs were operated in parallel with the same wastewater. The wastewater was delivered from the primary sedimentation basin of the facility. Each MBR was equipped a hollow-fiber microfiltration (MF) membrane module made from polyvinylidene fluoride with a total surface area of 1.3 m^2 and a nominal pore size of 0.4 µm (Mitsubishi Rayon, Tokyo, Japan). In each reactor, aeration was continuously carried out at the flow rate of 3.5 m^3/h. Filtration was carried out with the constant flow rate of operation. Operating conditions for each MBR are summarized in **Table 1**. When membrane fouling became significant, membrane modules were taken out from the reactor and were cleaned physically and/or chemically. Physical membrane cleaning was carried out by spraying pressurized water on the membrane surface. Chemical membrane cleaning was carried out by submerging the membrane module in a solution of hydrochloric acid (pH 2) and sodium hypochlorite (500 ppm). The degree of membrane fouling was evaluated by membrane filtration resistance calculated by the following equation;

$$J = \frac{\Delta P}{\mu R_t}$$

where J is the membrane permeate flux ($m^3/m^2/s$), ΔP is the transmembrane pressure difference (Pa), μ is the water viscosity (Pa s) and R_t is the total membrane filtration resistance (m^{-1}). The evolution of chemically reversible fouling was evaluated with the filtration resistance recorded just after the implementation of physical cleaning.

To investigate features of constituents that were responsible for chemically reversible fouling, organic matters were desorbed from the fouled membrane at the termination of the continuous operation. Membrane modules in each MBRs were taken out from the reactors and were disassembled. Desorption of organic matter from the fouled membranes was carried out by soaking the membranes in alkaline solution (sodium hydroxide) at the 25 °C for 24 h.

A series of batch filtration experiments were conducted to investigate which fraction in mixed liquor (i.e., suspended solid (SS), colloidal matter and dissolved matter) was responsible for each type of membrane fouling (i.e., physically and chemically reversible fouling). Membrane filtration using mini-module was carried out by applying 40 kPa of pressure difference with a suction pump for 60 min. Mini-modules used in this batch filtration experiment were prepared with the same membrane as used in continuous operation. To investigate the filtration resistances caused by each fraction, three different solutions were prepared from an original mixed liquor occasionally collected from the pilot scale MBR and were subsequently used in the batch experiments. First, the whole mixed liquor without any treatment was examined in a batch test. Second, the supernatant of the mixed liquor after centrifugation (4800 rpm, 5 min), which should contain no SS, was examined. The difference between the filtration resistances observed in the first and second tests could be considered as the resistance caused by SS in the mixed liquor. The third type of solution was prepared by filtration of the supernatant using a membrane filter paper with a pore size of 0.45 µm. The filtration resistance attributed to colloidal matter could be estimated by subtraction of the resistance determined in the third test (caused by dissolved matter) from the resistance in the second test (caused by colloidal and dissolved matters). After filtration test, membrane surface of the mini-modules were physically cleaned by spraying pressurized water on the membrane surface. Then pure water permeability of the cleaned membrane was measured in order to evaluate the degree of the fouling which can not be restored by physical cleaning (i.e., the degree of chemically reversible fouling).

RESULTS AND DISCUSSION

Fig. 1 shows changes in filtration resistances observed in the pilot run. Membrane fouling was significant in MBR1 which was operated with the highest F/M ratio. Physical membrane cleaning was carried out for MBR1 several times but the efficiency of the cleaning was low. This indicates that chemically reversible fouling was significant in MBR1. In contrast, the filtration resistances in MBR2 and MBR3 were very small until 100 days of operation. After that, increases in the filtration resistances were observed with both MBRs and the rate of increase in filtration resistance in MBR2 was much higher than that of MBR3. Physical membrane cleaning conducted at the end of continuous operation revealed that the occurrence of chemically reversible fouling was also significant in MBR2 compared to MBR3. From these results, it can be concluded that the both physically and chemically reversible fouling were enhanced when MBRs were operated with high F/M ratios.

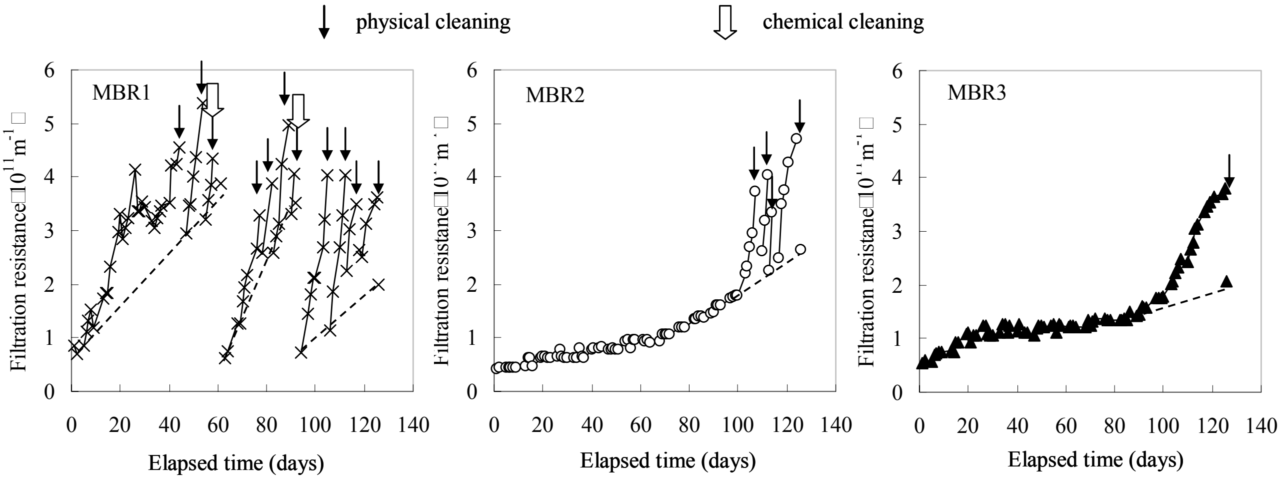

Fig. 1 Changes in the total filtration resistances in the pilot run.

Fig. 2 shows filtration resistances derived from chemically reversible fouling caused by SS, colloidal matter and dissolved matter. Seventeen batch experiments were conducted with mixed liquor collected on different occasions, and the averaged data are shown in **Fig. 2**. It can be seen from **Fig. 2** that significant chemically reversible fouling was observed with dissolved matter. It seemed that chemically reversible fouling was mainly caused by the dissolved fraction. It is therefore thought that indexes such as dissolved organic carbon (DOC), dissolved carbohydrate and dissolved protein might be related to the rate of chemically reversible fouling. In this study, however, no clear relationship between those indexes and the rate of chemically reversible fouling was established.

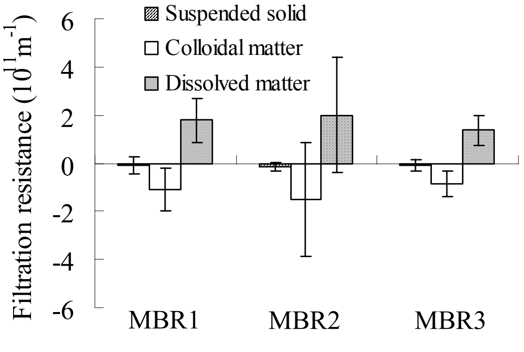

Fig. 2 Filtration resistances derived from chemically reversible fouling caused by SS, colloidal matter and dissolved matter.

Fig. 3 shows ^{13}C NMR spectra of membrane foulant desorbed from the fouled membranes at the termination of continuous operation. In the spectrum obtained for MBR1, a peak near 75 ppm, indicating the presence of polysaccharide-like substances, was significant. Polysaccharide-like substances accounted for 35%, 28% and 25% of the organic carbon desorbed from MBR1, MBR2 and MBR3 respectively. The amount of polysaccharide-like substances corresponds to the order of the F/M ratio set for each MBR. As mentioned above, severer chemically reversible fouling was observed with MBR operated with higher F/M ratio. It is therefore thought that polysaccharide-like substances are likely to cause severe chemically reversible fouling. **Fig. 4** and **Fig. 5** show monosaccharide composition of dissolved organic carbohydrate in the mixed liquor of each MBR and membrane foulant desorbed from fouled membrane used in each MBR respectively. There were significant similarities in the monosaccharide composition of all membrane foulants regardless the F/M ratio set for each MBR. In addition, the monosaccharide compositions of the membrane foulants were considerably different from those of dissolved carbohydrate in mixed liquor. These results indicate that some specific carbohydrate in the mixed liquor of MBRs might have higher affinity with the membrane and consequently cause greater chemically reversible fouling.

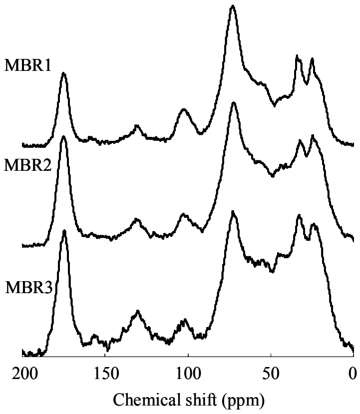

Fig. 3 ^{13}C NMR spectra of foulant desorbed from fouled membrane.

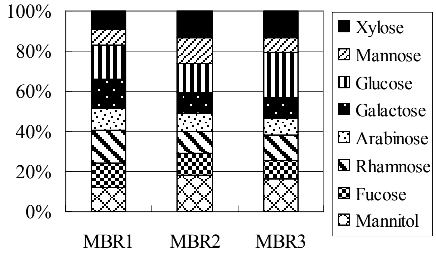

Fig. 4 Monosaccharide composition of dissolved carbohydrate in mixed liquor.

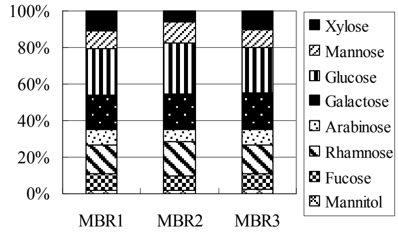

Fig. 5 Monosaccharide composition of foulant desorbed from fouled membrane.

CONCLUSIONS

In this study, the influence of operating conditions on chemically reversible fouling in MBRs was investigated. Chemically reversible fouling became significant when MBRs were operated with high F/M ratio. No clear relationship between comprehensive indexes such as DOC, dissolved carbohydrate and dissolved protein concentration in the mixed liquor and rate of chemically reversible fouling was observed. Some specific carbohydrate might cause chemically reversible fouling preferentially.

Modelling of enhanced biological phosphorus removal (EBPR) combined with post-denitrification in MBR system for domestic wastewater treatment

Johanna Mouchard

KompetenzZentrum Wasser Berlin, Cicerostr. 24 10709 Berlin, Germany
Email: johanna.mouchard@kompetenz-wasser.de

ABSTRACT

In the wastewater treatment sector, the use of conventional processes is often limited by local constraints, like quality effluent norms, land requirement, sludge disposal, etc. The Membrane Bioreactor (MBR) process, which combines biological treatment with membrane separation, is very promising technology to handle such situations. One can utilize this promising technology to the utmost efficiency by use of computer based modelling and simulation. Once the model is calibrated and validated, different hypothetical scenarios could be investigated in order to predict the effluent quality, optimize design and operating conditions and improve yield and functioning costs.

Within the framework of EU-Life "ENREM" demonstration project (Enhanced Nutrients Removal in Membrane reactor), the Berlin Center of Competence for Water together with the Berliner WasserBetriebe (BWB), has operated an MBR plant designed for advanced biological nitrogen and phosphorus removal (250 p.e.). They have investigated a post-denitrification process, without the addition of carbon source, combined with an Enhanced Biological Phosphorus Removal (EBPR). This configuration delivers high denitrification rates, and higher than the endogenous rates, as the anaerobic zone enables a group of denitryfing bacteria to store the carbon. The bacteria use this internally stored source of carbon in the anoxic zone at the end of the process. Compared with pre-denitrification operation (*figure 1*), this configuration does not need the aerobic / anoxic sludge recirculation loop and equipment and energy requirement are less important, while providing equivalent or greater removal of nutrients. The targeted effluent quality is 0.05mgP/L and 10mgN/L.

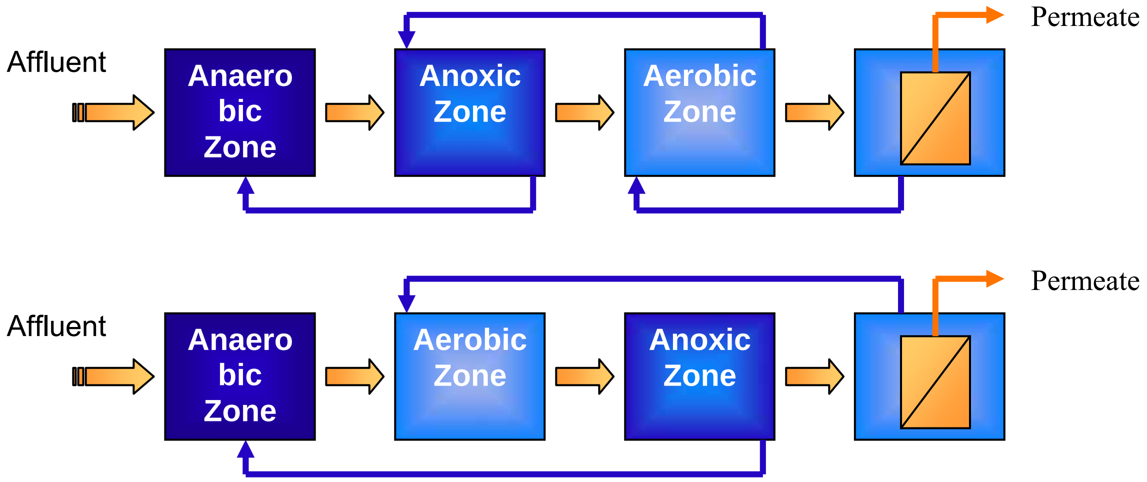

Figure 1 MBR configuration with pre-denitrification (top) and MBR configuration with post-denitrification (bottom)

Within the framework of the MBR-Train project, which provide young researchers an Early Stage Research Training on process optimisation and fouling control in MBR for water treatment, a study for six months was realized to calibrate a biological model for this system and to simulate the static (constant input conditions) and dynamic (variable input conditions) operations. The aim of this study was to optimize operation and to facilitate the design of others implementations. This study was done with Anjou Recherche (French research centre of Veolia Water, located nearby Paris), for modelling and simulation.

Delft’s model, Activated Sludge Model 2d (ASM2d) and ASM3 are among models which can be used for the modelling of EBPR wastewater treatment process. ASM3, incorporating EBPR mechanisms by introduction of the EAWAG Bio-P module, was chosen for this study. Measurement campaigns of three weeks each were performed on the ENREM demonstration plant to calibrate the model. This allowed to pay a special attention to nitrogen and phosphorus removal, and also to the effect of temperature on the biological process.

The paper will present the methodology and results of this modelling work undertaken on this unusual MBR configuration.

Module Design of Aerated Hollow Fibres modules for membrane bioreactors

Samuel Pollet Christelle Guigui, Corinne Cabassud

Institut National des Sciences Appliquées de Toulouse, Laboratoire d'Ingénierie des Procédés de l'Environnement 135 avenue de Rangueil, 31077 Toulouse Cedex 4 – France Email: samuel.pollet@insa-toulouse.fr, christelle.guigui@insa-toulouse.fr, corinne.cabassud@insa-toulouse.fr

Membranes bioreactors (MBR) are now widely used for wastewater treatment. One of the key points in process optimisation and in energy costs is related to the use of aeration for fouling removal in outside/in systems. It is still of paramount importance to understand how air/liquid flow efficiency is linked with module properties and is influencing fouling and its reversibility. Many studies have yet focused on the characterisation of two-phase flow and related filtration performances on isolated flat sheet membranes or fibres, and allow to understand phenomena at the scale of isolated fibres [1,2], or flat sheet membranes [3,4].

This paper aims to study and to characterize mechanisms at the scale of the membrane module, for a given module system. Optimal hydrodynamic parameters (air and liquid flow rates) will be determined and compared for different module properties, using specific methods to characterise fouling and its reversibility.

This paper will concern a very specific process. Outside/in hollow fibres are put in a carter located outside the activated sludge tank. A pump is recycling effluent at a low flow rate and thus generates a very low liquid velocity (up to 0.04 m.s^{-1}) in the module outside the fibres. The objective of this recycling is to decrease the concentration of suspended solids inside the module, and create mixing which is enhanced by the air flow. Influence of aeration can be studied for a large range of gas velocities (between 0 and 0.2 m.s^{-1}). Superficial velocity ranges were between 0 and 0.04 m.s^{-1} for the liquid. .

Three different modules were designed to obtain information on different parameters: fibre diameter, bundle diameter. All modules are containing one bundle of free PES fibres and aeration is the same in every module, provided by three aerators (diameter of 5mm) at the module bottom. The three modules (manufactured by Polymem) characteristics are presented in Table 1.

Table 1: Characteristics of the three modules tested.

Characteristics \ Module	**SB1M**	**LB1M**	**LB1L**
External fibre diameter (mm)	*1.45*	***2.3***	*1.45*
Bundle Diameter (mm)	*27*	*53*	*53*

All modules have the same ratio $S_{\text{fibres}}/S_{\text{bundle}}$ (66%) of fibres.

The following set of experiments was performed on each module, in order to provide elements for comparison:

Determination of the initial permeability with deionised water;

Measurement of pressure drops and gas hold up for identical liquid velocities and a large range of gas velocities (example shown in Figure 1);

Observation and determination the flow pattern (bubble or slug flows) for the different hydrodynamics conditions tested;

Characterisation of filtration ability with synthetic waters (clay suspension at 1 g/L) using the flux step method (step duration: 10 minutes and step height: 5 $L.h^{-1}.m^{-2}{}_{membrane}$) and the modified flux step method [5]- Fouling velocities and resistances were calculated for different permeate fluxes (example shown in Figure 2).

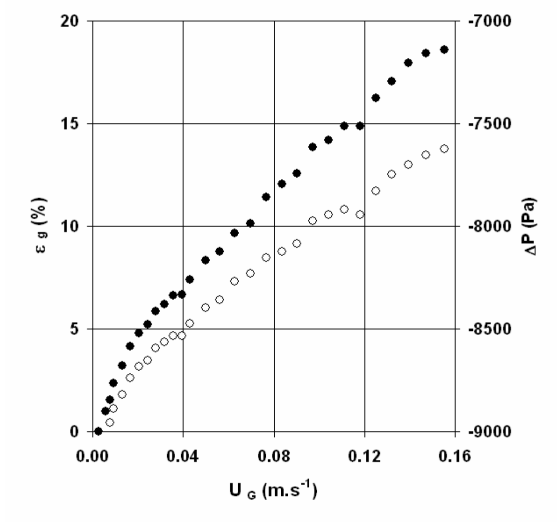

Figure 1 Gas hold up (●) and pressure drop (○) along the module vs gas velocity for a liquid velocity 0.0061 $m.s^{-1}$ – module LB1L and clay suspension 1 $g.L^{-1}$.

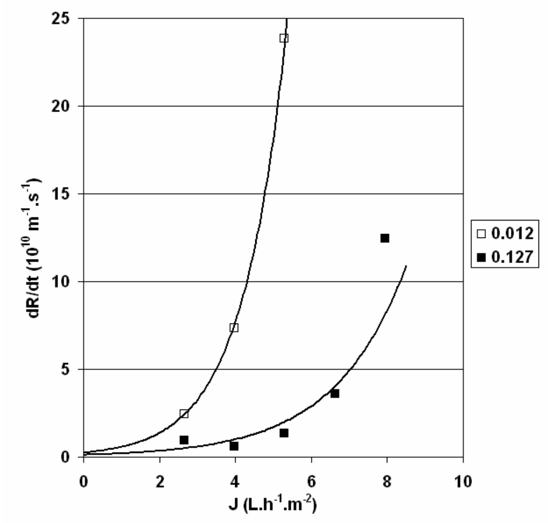

Figure 2 dR/dt vs J for two gas velocities (□ 0.012 and ■ 0.127 $m.s^{-1}$) – module LB1L and clay suspension 10 $g.L^{-1}$ – Ul = 0.018 $m.s^{-1}$

As shown in Figure 2, for a given module and a given liquid velocity, increasing of gas velocity is decreasing fouling velocity. In Figure 1, gas hold up is increasing and pressure drop is decreasing when gas velocity is increasing and these results are consistent with two-phase flow hydrodynamics. The same kinds of results have been obtained for the three modules, which allows to compare fouling abilities of the three modules in relation with operating conditions and to discuss on the relative influence of air sparging.

The aeration and filtration costs have been calculated for each module and for reference conditions to conclude on a best configuration. This work will provide series of comparable and consistent results in terms of fouling linked with hydrodynamics (liquid and air flows) with an economical aspect to conclude about the best module geometry for this specific process.

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[1] Martinelli, L., Guigui, C., Liné, A., Mechanisms induced by spherical cap bubble injection for ultrafiltration performance enhancement, ICOM, Séoul-Korea (2005).

[2] Kim, J., DiGiano, F. A., Defining critical flux in submerged membranes: influence of length-distributed flux, Journal of Membrane Science 280, 752-761 (2006).

[3] Ducom, G., Puech, F. P., Cabassud, C., Air sparging with flat sheet nanofiltration: a link between wall shear stress and flux enhancement, Desalination 145, 97-102 (2002).

[4] Ozaki, N., Yamamoto, K., Hydraulic effects on sludge accumulation on membrane surface in crossflow filtration, Water research 35, 3137-3146 (2001).

[5] Espinosa-Bouchot, M., Contribution à l'étude d'un Bioréacteur à Membranes Immergées: Impact de la configuration du module et des conditions d'aération sur le colmatage particulaire et modélisation de l'activité biologique, PhD Thesis, Insa Toulouse-France (2005).

Waste water treatment and recycling with membrane technologies

Fabian Reichwald
Krüger WABAG GmbH / Veolia Water Solutions & Technologies, Germany
Krüger WABAG GmbH Weiherstrasse 19 D-95448 Bayreuth
e-mail: Fabian.Reichwald@veoliawater.com

Krüger WABAG, part of Veolia Water Solutions & Technologies, has installed in 2003 the largest MBR plant with a membrane area of 84000 m². The advantages of the MBR plant instead of conventional process technology are the following

- The compact footprint of a membrane activated sludge plant
- Consistently high quality effluent
- Lower investment costs in comparison with conventional wastewater treatment plants using the most advanced treatment and process technology
- Funding of investment costs by the Federal State for the construction of a membrane activated sludge plant
- Increased operating experience with MBR plants and ultrafiltration technology in municipal wastewater treatment.

In the last 10 years Krüger WABAG has applied the MBR technology in various fields of activity, especially ultrafiltration in combination with reverse osmosis for wastewater recycling. One example is the wastewater recycling plant of company DuPont, which was installed by Krüger WABAG in 2000 and has been successfully working for 5 ½ years. The plant scheme is illustrated in figure 1. The plant has a maximum capacity of 110 m³/h and treats the wastewater from the own sewage treatment plant to process water, mainly consisting of the two treatment steps pressure-ultrafiltration and reverse osmosis. As a result, Du Pont is saving approx. 800.000, - € per year fresh water and wastewater costs.

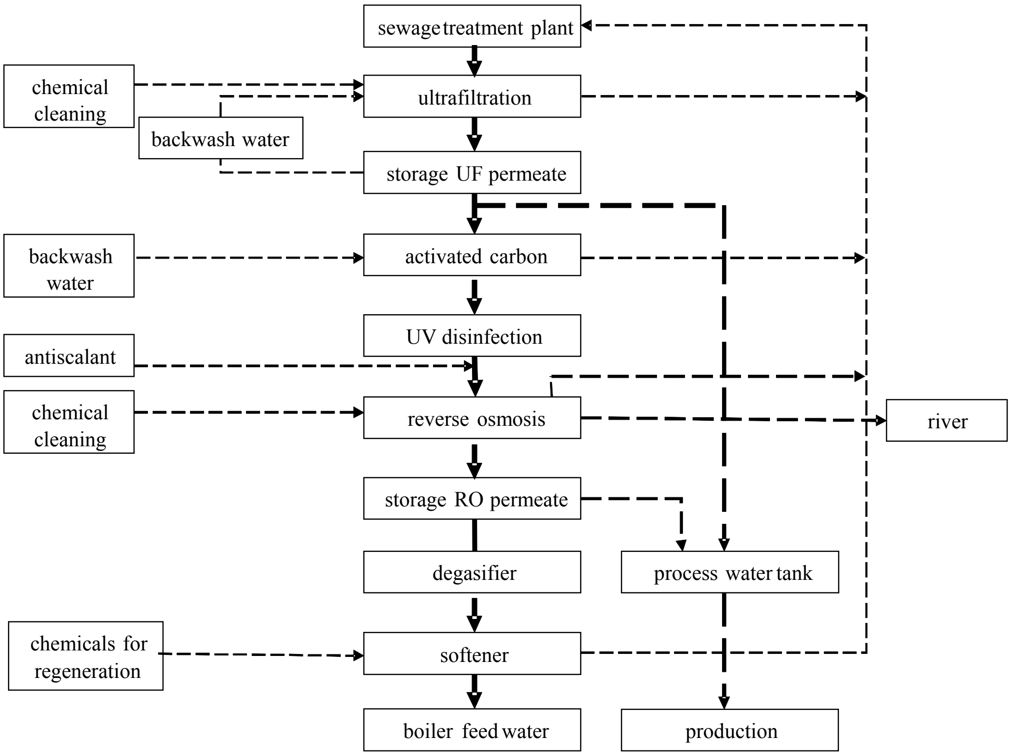


Fig. 1 Scheme Recycling plant DuPont (110 m³/h)

Krüger WABAG has installed further plants for the treatment of filter backwash water in numerous drinking waterworks, e.g. Roetgen (600 m³/h - which is one of the largest plants of the world), Egau, Dreieich, Erlangen etc. Above all the main interest is to protect and to save the natural resources groundwater / surface water.

On the swimming pool sector, however, economical aspects are the decisive facts for the installation of UF/RO plants. Krüger WABAG has installed more than 20 plants in swimming pools in the last 10 years. Here the filter backwash water is being treated to refill water by UF / RO plants. Since May 2006 the rules and regulations of DIN 19645 are applicable. Normally the amortization period of these plants is 2 - 4 years. Based on numerous treatment plants established within the last 10 years, Krüger WABAG has successfully proven that it is possible to treat wastewater economically to fresh water.

Together with ELGA Berkefeld, ELGA LabWater and RWO, Krüger WABAG form under the umbrella of Veolia Water Solutions & Technologies one of the leading suppliers in the water treatment sector. The company, with a total of more than 400 employees in Germany, Austria and Switzerland, achieved combined sales in 2005 of about € 110 million.

Link between flocculation and fouling in a MBR

Maxime Remy*, Hardy Temmink*

*Wetsus, Centre for Sustainable Water Technology, Postbus 1113, 8900 CC Leeuwarden, The Netherlands (Email: maxime.remy@wetsus.nl)

ABSTRACT

Flocculation is an important mechanism in conventional activated sludge processes because it determines the settleability of sludge. Similarly, bioflocculation determines the filterability of sludge in MBR systems. In this study, parameters related to flocculation and membrane fouling were investigated with sludge samples from a full-scale MBR treating domestic wastewater.

The results verify the importance of (bio-)flocculation on filterability. This means that future research activities on improving the performance of MBR systems should focus not only on the membranes and their operation, but also on improving the bioflocculation process.

INTRODUCTION

So far, the solution to membrane fouling is treatment of the symptoms by membrane cleaning, applying shear at the membrane surface and modifications of membrane operation and characteristics (Hwang and Lin 2002; Le Clech, Jefferson et al. 2003; Judd 2005). In this study parameters presented in literature (Table 1) as having an interest on (bio-) flocculation have been investigated and related to sludge filterability, as we believe that operation of the biological reactor and its effect on this (bio-)flocculation is equally important for fouling as the membrane and membrane operation.

Table 4 – Parameters from the sludge with effects on flocculation mechanisms.

Parameter	***Effect on flocculation***	***Reference***
Percentage inorganic material	Positive	(Urbain, Block et al. 1993)
Hydrophobicity	Positive	(Urbain, Block et al. 1993; Jorand, Boué-Bigne et al. 1998; Palmgren, Jorand et al. 1998; Wilen, Jin et al. 2003)
Floc structure; size	More compact flocs when “good” flocculation	(Wilen, Jin et al. 2003) (Urbain, Block et al. 1993)
Attached EPS	Positive Especially proteins	(Mikkelsen and Nielsen 2001); (Urbain, Block et al. 1993; Wilen, Jin et al. 2003)
Multivalent cations (Ca^{2+}, Fe^{3+})	Positive Negative effect of monovalent (Na^+, K^+…)	(Urbain, Block et al. 1993);(Wilen, Jin et al. 2003); (Sobeck and Higgins 2002); (Song and Singh 2005); (Biggs, Ford et al. 2001); (Park, Muller et al. 2006)

MATERIAL AND METHODS

Full-scale MBR

Varsseveld (51°57’15”N, 6°27’35”E) has been the first Dutch municipality to use a MBR for treating its domestic wastewater. The MBR was designed for 23.150 population equivalents with a capacity up to 755m³/h with an average of $5000m^3/d$. This MBR is composed of an anoxic predenitrification tank followed by a carousel where nitrification and denitrification occur through alternating aeration and a membrane tank in which membranes are submerged. Membranes are Zenon ZW500d with a nominal pore size of 0.035µm.

The MBR started up in January 2005 and the samples presented hereafter were taken from the carousel and the membrane tank for a period of 9 months.

Filtration characterization

Fouling was investigated by a small-scale Filtration Characterization Installation (FCI) (Evenblij, Geilvoet et al. 2005). When compared with permeability tests from the full scale, values obtained with FCI gave similar trends. The FCI was used because permeability data from full scale operation also include fouling history when compared with FCI assessment starting with clear membranes. Filtration characterization focused on the biology rather than on the membrane operating conditions. the increase in resistance after 10L/m² of permeate was extracted at a flux of 60 L/m^2*h (R_{add_10}) was used as an indication of the filterability of the samples.

Analyses

Suspended solids were removed from the supernatant by centrifugation (11min at 3000rpm), followed by paper filtration (black ribbon). The soluble fraction was obtained after filtration of the supernatant through ME 25/21 STL filters from Schleicher & Schuell (0.45µm). Non-settable compounds larger than 0.45µm are referred as “colloids”. Total and volatile suspended solids (TSS/VSS) were assessed according to Standard Methods (Clesceri, Greenberg et al. 1998).

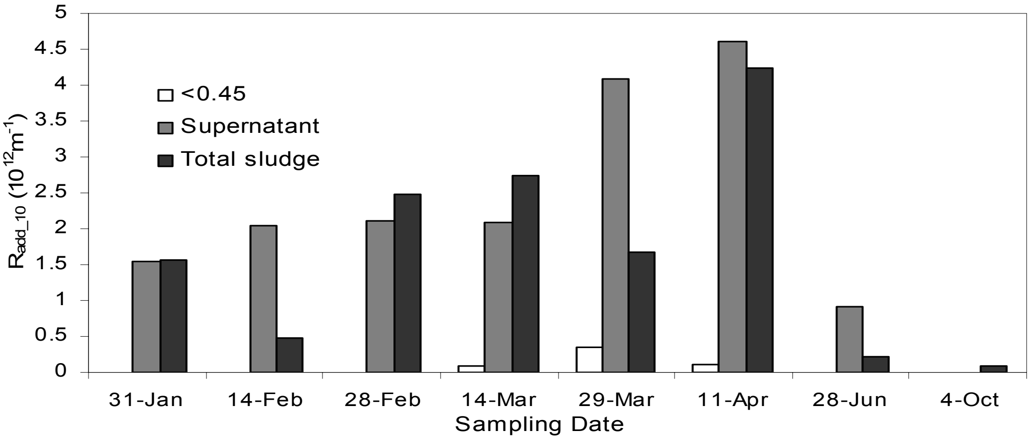
“Fixed” exocellular polymers were extracted from sludge samples by 5 hours mixing at 4°C with cation exchange resin(Frølund, Palmgren et al. 1996). Particle size distribution of sludge samples and influent were assessed with a Coulter Laser particle size analyzer. Microscopic analysis with an optic microscope was realized to assess the overall morphology and composition of the flocs

The chemical oxygen demand (COD) was measured with Dr Lange cuvette test. The sulfuric acid and phenol method was used to measure polysaccharides in sludge and waste water (DuBois, Gilles et al. 1956). Proteins were measured according to Bio-Rad Protein Assay (Bradford 1976). Intra-floc hydrophobicity, an indicator for hydrophobicity at the surface of single cells composing flocs of the sludge was determined according to ‘microbial adhesion to hydrocarbon’ (MATH) method (Rosenberg, Gutnick et al. 1980; Guellil, Block et al. 1998).

RESULTS

Filterability

To determine the contribution of different fractions of sludge taken from the membrane tank to the overall fouling, R_{add_10} was assessed (Graph 1) for the sludge after 0.45µm filtration (<0.45µm), after settling of suspended solids through centrifugation (Supernatant), and for the raw sampled sludge (total sludge).

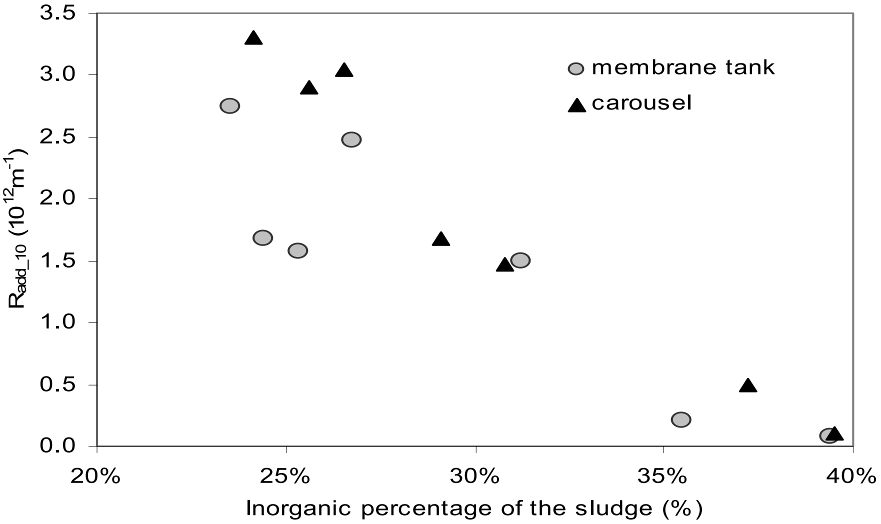


Graph 1– Evolution of R_{add_10} during filtration of different fractions of sludge

The R_{add_10} of the supernatant fraction always was around or above the R_{add_10} of the sludge whereas Radd_10 was relatively low for the soluble fracion. This indicates the important role of the non-settleable fraction in membrane fouling. From a higher R_{add_10} for supernatant compared to the sludge-water mixture a protective role of a cake layer decreasing the influence of foulants from supernatant was suspected (Graph 1).

Relation between sludge characteristics and filterability

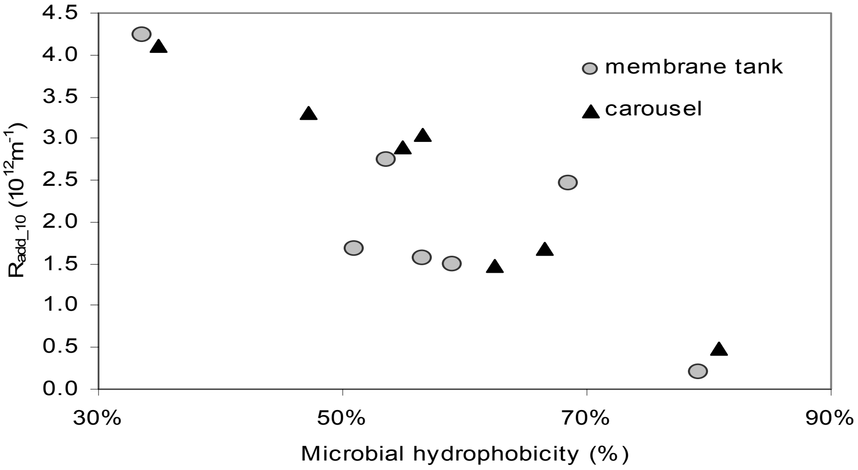
The suspended solids concentration varied between the different tanks during the study from 8 to 14g.l^{-1} with no apparent relation to R_{add_10}. This lack of relation between filterability and sludge concentration was also reported from their results by Rosenberger and Kraume 2002; Cicek, Suidan et al. 2003; Lesjean, Rosenberger et al. 2005. It however is in contradiction with results reported by others(Le Clech, Jefferson et al. 2003; Cho, Ahn et al. 2004; Itonaga, Kimura et al. 2004; Meng, Zhang et al. 2005).



Graph 2 - Relation between R_{add_10} and the percentage of ashes within the sludge.

The inorganic fraction of sludge showed a distinctive relation with sludge filterability (Graph 2). During the study, the inorganic percentage of sludge varied from 22 to 40%. Graph 2 shows a negative correlation between the inorganic fraction of the sludge and R_{add_10}..

The (intra-floc) hydrophobicity of the sludge varied between 35 and 80% during the operation of the MBR and also showed a clear relation with sludge filterability (Graph 3) with a higher filterability (lower R_{add_10}) for higher hydrophobicity.



Graph 3 - Relation between R_{add_10} and intra-floc microbial hydrophobicity.

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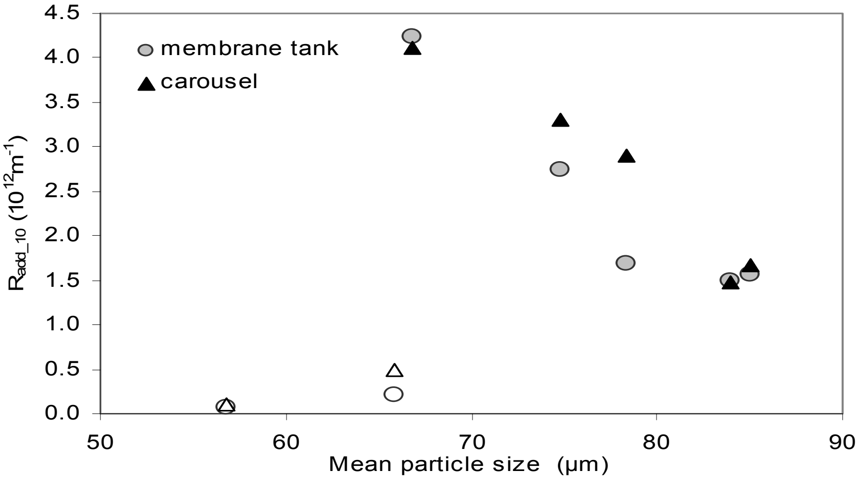
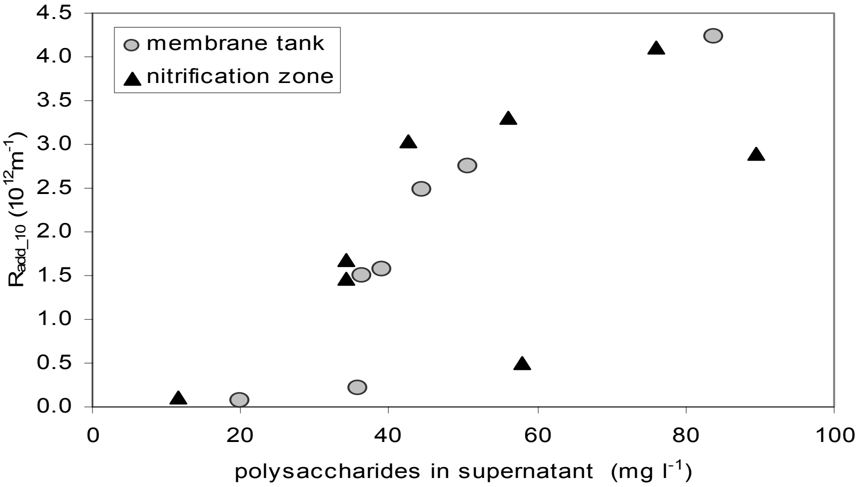
Graph 4 shows the relation between the mean particle size of sludge flocs and R_{add_10}. Mean particle size of sludge flocs varied from 55 till 85µm during the studied period.



Graph 4 - Relation between R_{add_10} and the mean particle size of the flocs (non colored points represent compact flocs).

Two different patterns were found. When the microscopic analysis of flocs showed fluffy, cotton-like structure, bigger flocs result in lower R_{add_10}. When flocs appear more compact, smaller flocs result in lower R_{add_10}. Good flocculation, resulting in tighter flocs, had a positive role on sludge filterability.



Graph 5- Relation between R_{add_10} and the polysaccharides present in the supernatant.

As already shown in Graph 1, the colloidal fraction of the sludge had the strongest negative impact on filterability. A relation between filterability and polysaccharide, protein and COD contents furthermore showed a positive relation between increasing concentration and fouling. The clearer relation was found for polysaccharides (Graph 5). This agrees with results already presented in the literature (Laabs, Amy et al. 2004; Lesjean, Rosenberger et al. 2005; Fan, Zhou et al. 2006).

DISCUSSION

A summary of different parameters found to affect sludge filterability is presented in Table 5.

Table 5 - Parameters found to have an influence on filterability of the sludge

Studied Parameter		***Effect on filterability***
Sludge concentration		None
Percentage inorganic material		Strong and positive
Hydrophobicity		Strong and positive
Particle Size distribution		Depending on flocs compactness
COD	Extracted	Negative
	Colloidal	Strong and negative
	<0.45µm	None
Polysaccharides	Extracted	Negative
	Colloidal	Strong and negative
	<0.45µm	None
Proteins	Extracted	Negative
	Colloidal	Negative
	<0.45µm	None
Ca^{2+} or Fe^{3+} in sludge flocs		Positive

When comparing parameters of interest for flocculation (**Table 4**) with those found for filterability of sludge (Table 5), it is almost a perfect match. This indicates that good flocculation is a decisive parameter to obtain suitable sludge for a MBR. Compounds said to have an effect on fouling in the literature seem to be symptoms of poor flocculation. Getting foulants better attached within the flocs should be efficient against fouling. This can be realized e.g. by decreasing shear in the reactor to avoid floc breaking of flocs, or by addition of multivalent cations or polymers to the sludge to increase binding to the sludge. Increasing dissolved oxygen could also be an option as it facilitates flocculation.

So far, research was really aimed at membranes or what was happening close to the membrane surface, coming up with solutions concerning membrane operation while what is happening with the biology, including flocs structure and strength should be more considered.

CONCLUSIONS

This research showed interesting relations between parameters of interest for sludge flocculation and filtration resistance.

Better operation of the biology that takes flocculation into account should be aimed at, besides conventional approaches to prevent membrane fouling.

Fouling is indeed a symptom of poor sludge quality with regards to flocculation. Curing the symptoms with different membranes operation and shear at the surface are still interesting, but this should be combined with a more detailed understanding of the biology and influence of operational parameters.

Parameters found to have a positive effect on flocculation such as high dissolved oxygen, longer SRT, multivalent cations loading, together with a lower shear should be investigated.

Further research on the effect of flocculation on fouling by sludge is being carried out in order to validate these results.

REFERENCE

Biggs, C. A., A. M. Ford, et al. (2001). "Activated sludge flocculation: direct determination of the effect of calcium ions." Water Science And Technology **43**(11): 75-82.

Bradford, M. M. (1976). "A rapid and sensitive method for the quantitation of microgram quantities of protein utilizing the principle of protein-dye binding." Analytical Biochemistry **72**(1-2): 248.

Cho, J. W., K. H. Ahn, et al. (2004). "Investigation of biological and fouling characteristics of submerged membrane bioreactor process for wastewater treatment by model sensitivity analysis." Water Science and Technology **49**(2): 245-254.

Cicek, N., M. T. Suidan, et al. (2003). "Impact of soluble organic compounds on permeate flux in an aerobic membrane bioreactor." Environmental Technology **24**(2): 249-256.

Clesceri, L., A. Greenberg, et al. (1998). Standard methods for the examination of water and wastewater. Washington, DC, American Public Health Association, American Water Works Association, Water Environment Federation.

DuBois, M., K. A. Gilles, et al. (1956). "Colorimetric method for determination of sugars and related substances." Analytical Chemistry **28**(3): 350-356.

Evenblij, H., S. Geilvoet, et al. (2005). "Filtration characterisation for assessing MBR performance: three cases compared." Desalination **178**(1-3): 115.

Fan, F., H. Zhou, et al. (2006). "Identification of wastewater sludge characteristics to predict critical flux for membrane bioreactor processes." Water Research **40**(2): 205-212.

Frølund, B., R. Palmgren, et al. (1996). "Extraction of extracellular polymers from activated sludge using a cation exchange resin." Water Research **30**(8): 1749-1758.

Guellil, A., J.-C. Block, et al. (1998). "Adaptation of the microbial adhesion to hydrocarbon test (MATH) for measuring activated sludge hydrophobicity." Water Science and Technology **37**(4-5): 359-362.

Hwang, K.-J. and T.-T. Lin (2002). "Effect of morphology of polymeric membrane on the performance of cross-flow microfiltration." Journal of Membrane Science **199**(1-2): 41.

Itonaga, T., K. Kimura, et al. (2004). "Influence of suspension viscosity and colloidal particles on permeability of membrane used in membrane bioreactor (MBR)." Water Science & Technology **50**(12): 301-309.

Jorand, F., F. Boué-Bigne, et al. (1998). "Hydrophobic/hydrophilic properties of activated sludge exopolymeric substances." Water Science & Technology **37**(4-5): 307-315.

Judd, S. (2005). "Fouling control in submerged membrane bioreactors." Water Science And Technology **51**(6-7): 27-34.

Laabs, C., G. Amy, et al. (2004). "Organic colloids and their influence on low-pressure membrane filtration." Water Science & Technology **50**(12): 311-316.

Le Clech, P., B. Jefferson, et al. (2003). "Impact of aeration, solids concentration and membrane characteristics on the hydraulic performance of a membrane bioreactor." Journal of Membrane Science **218**(1-2): 117.

Lesjean, B., S. Rosenberger, et al. (2005). "Correlation between membrane fouling and soluble/colloidal organic substances in membrane bioreactors for municipal wastewater treatment." Water Science & Technology **51**(6-7): 1-8.

Meng, F., H. Zhang, et al. (2005). "Application of fractal permeation model to investigate membrane fouling in membrane bioreactor." Journal of Membrane Science **262**(1-2): 107.

Mikkelsen, L. H. and P. H. Nielsen (2001). "Quantification of the bond energy of bacteria attached to activated sludge floc surfaces." Water Science And Technology **43**(6): 67-75.

Palmgren, R., F. Jorand, et al. (1998). "Influence of oxygen limitation on the cell surface properties of bacteria from activated sludge." Water Science and Technology **37**(4-5): 349-352.

Park, C., C. D. Muller, et al. (2006). " The Effect of Wastewater Cations on Activated Sludge Characteristics: Effects of Aluminum and Iron in Floc." Water Environment Research **78**(1): 31-40.

Rosenberg, M., D. Gutnick, et al. (1980). "Adherence of bacteria to hydrocarbons: A simple method for measuring cell-surface hydrophobicity." FEMS Microbiology Letters **9**(1): 29.

Rosenberger, S. and M. Kraume (2002). "Filterability of activated sludge in membrane bioreactors." Desalination **146**: 373-379.

Sobeck, D. C. and M. J. Higgins (2002). "Examination of three theories for mechanisms of cation-induced bioflocculation." Water Research **36**(3): 527.

Song, L. and G. Singh (2005). "Influence of various monovalent cations and calcium ion on the colloidal fouling potential." Journal of Colloid and Interface Science **289**(2): 479.

Urbain, V., J. C. Block, et al. (1993). "Bioflocculation In Activated-Sludge - An Analytic Approach." Water Research **27**(5): 829-838.

Wilen, B.-M., B. Jin, et al. (2003). "The influence of key chemical constituents in activated sludge on surface and flocculating properties." Water Research **37**(9): 2127-2139.

Wilen, B. M., B. Jin, et al. (2003). "Relationship between flocculation of activated sludge and composition of extracellular polymeric substances." Water Science And Technology **47**(12): 95-103.

Hydrogenotrophic Denitrification and Perchlorate Reduction of Ion Exchange Contaminated Brines Using Hollow Fiber Membrane Bioreactors

Ashish K. Sahu and Sarina J. Ergas.

Department of Civil and Environmental Engineering, University of Massachusetts, 18 Marston Hall, Amherst, MA, 01003, USA (E-mail : ergas@ecs.umass.edu)

Liquid manure, use of extensive fertilizers, inefficient septic tanks and agricultural run off are major contributors of nitrate (NO_3^--N) to groundwater. The World Health Organization (WHO) has set a contaminant limit of 11 mg/L NO_3^--N. Perchlorate (ClO_4^-) contamination in groundwater has been recorded in 38 US states and has primarily occurred in association with manufacturing of missiles, fireworks, and other industrial processes (Urbansky, 2000). No national standards for perchlorate have been established in the United States, but an advisory level of 24 μg/L ClO_4^- is presently being debated.

Both perchlorate and nitrate are highly soluble and stable in water and hence cannot be removed by conventional drinking water treatment processes such as filtration or air stripping (Urbansky, 2000). Physical and chemical treatment process such as Ion Exchange (IX), are commonly used to remove nitrate and perchlorate from groundwater. IX resins have shown to reduce nitrate and perchlorate concentrations in groundwater to <5ppm NO_3^--N and to < 4 μg/L ClO_4^-. On exhaustion of IX column, certain IX column can be regenerated using brine solution. Regeneration of IX column with brine produces high nitrate and perchlorate concentrations in brines which present a disposal problem. Studies have shown that biological treatment of IX regenerant brines can reduce nitrate to nitrogen and perchlorate to chloride using heterotrophic bacteria thereby allowing recycling of regenerant brine for further reuse (Min *et al*, 2004).

Biological treatment process for nitrate and perchlorate make use of either heterotrophic or autotrophic microorganisms. Autotrophic bacteria use inorganic carbon as their carbon source and grow slower then heterotrophs (organic carbon as a carbon source) producing less biomass and releasing fewer organic cell products (Haugen *et al*, 2002). This results in a decreased risk of biological re-growth in distribution systems and decreased disinfection byproduct formation. In this study synthetic regenerant brines containing high nitrate (50-800 mg/L NO_3^--N) and perchlorate (20 mg/L) concentrations in 0-12.5 g/L NaCl was treated using hydrogenotrophic bacteria in hollow fiber membrane bioreactors.

Hydrogenotrophic denitrifiers are autotrophs that use hydrogen (H_2) gas as their electron donor. H_2 is inexpensive, non-polluting, non-toxic and has relatively low solubility in water and hence can easily be air stripped after treatment. This research study looked at two HFMBs:

In Reactor I, 50 mg/L NO_3^--N contaminated groundwater was treated directly using hydrogenotrophic denitrifiers in a HFMB.

In Reactor II, synthetic IX regenerant brines containing 800 mg/L NO_3^--N and 12.5 g/L salt concentrations were treated using halophilic (salt tolerant) hydrogenotrophic denitrifiers in a HFMB. Reactor II was later shifted to 20 mg/L ClO_4^- in the presence of 12.5 g/L NaCl.

The objectives of this study were to: Enrich a denitrifying culture to treat 0-800 mg/L NO_3^--N in 0-12.5 g/L NaCl using hydrogen as an electron donor, Engineer bench scale HFMBs to treat synthetic IX brines containing high nitrate and perchlorate concentrations, Reduce hydraulic retention time (HRT) for economical purpose and Investigate the performance of the system in continuous and batch mode operation and in the presence of dissolved oxygen.

Hydrogenotrophic denitrifying bacteria were enriched using batch culture systems containing hydrogen, nitrate and nutrients. The batch cultures were inoculated with denitrifying bacteria obtained from wastewater plant. The cultures were enriched in a step-wise acclimation starting from 0-600 mg/L NO_3^--N and 0-9 g/L NaCl. The culture were able to reduce nitrate from 600 mg/L to 180 mg/L at NaCl concentration of 9 g/L. Lower effluent NO_3^- concentration may be attainable in the HFMB due to greater mass transfer of H_2, which is limited at high salt concentration.

HFMB modules were provided by Compact Membrane Systems Inc (Wilmington, DE, USA). Synthetic nitrified influent was supplied through the lumen of hydrophobic membranes (500 fibers, 280µm outer diameter, 0.25m length with a surface area of 0.28 m^2) and H_2 was supplied on the shell side of the reactor. Reactor I was operated in a continuous mode, while Reactor II was first operated in a continuous mode and later shifted to batch mode operation. A trans-membrane pressure of 2 psi was maintained for effective mass transfer of $H_{2.}$ A part of the effluent was recirculated to the influent line to increase mass transfer of hydrogen and decrease fouling in the fibers.

Two HFMB setups were inoculated with enriched batch cultures from wastewater treatment plant, one containing 50 mg/L NO_3-N and no salt (Reactor I) and 600 mg/L NO_3-N in presence of 9 g/L NaCl (Reactor II). The HRT for HFMB Reactor I was reduced from 15 hours to 5 hours and showed complete denitrification. An average removal efficiency of nitrate 94% was observed at 5 hr HRT.

The HFMB for Reactor II showed partial denitrification at 53 hour HRT and was shifted to a batch operation with a HRT of 53 hours. An average nitrate removal efficiency of 30% was observed in a batch mode over a period of 110 days. Reactor II was later shifted to 20 mg/L ClO_4^-. Since the thermodynamic energy yield gained by bacteria using nitrate and perchlorate (122.1 kJ/mol) as an electron acceptor is so close an attempt was made to treat perchlorate using the same innocula. An average perchlorate removal efficiency of 45 % was observed over a period of 30 days in Reactor II. Reduction of perchlorate using hydrogenotrophic bacteria at high salt concentration has not been previously reported in the literature.

The treatment of IX-regenerant brines containing high nitrate concentrations using hydrogen as an electron donor could serve as a promising technology as a hybrid system between IX and a HFMB.

REFERENCES

Haugen, K.S., Semmens, M.J. and Novak, P.J. (2002). A novel in situ technology for treatment of nitrate contaminated groundwater. Water Research, 36:3497-3506.

Min, B., Evans, P.J., Chu, A.K. and Logan, B.E. 2004 Perchlorate removal in sand and plastic media bioreactors. Water Research, 38:p. 47–60

Urbansky, E.T. 2000. Perchlorate in the Environment, Kluwer Acad/Plenum Publishers, NewYork, NY.

Electro-assisted Treatment of Sludge from Membrane Bioreactor Treating Municipal Wastewater

Devendra P. Saroj*, Giuseppe Guglielmi, Daniele Chiarani, Elisa Ferrarese, Gianni Andreottola

Department of Civil and Environmental Engineering, University of Trento, Via Mesiano-77, TRENTO-38050, Italy (E-mail: devendra.saroj@ing.unitn.it)

The increasing stress on clean water demand and stringent effluent discharge limits have led to the development of more reliable, controlled and compact wastewater treatment technologies. Membrane bioreactors (MBRs) have been proven to be well suited for satisfying most of the technical challenges now-a-days faced in wastewater treatment. By consisting intelligent coupling of filtration and biological processes, MBRs promise high quality of treated effluent, bioprocess operation at high sludge retention time (SRT), compact plant size with reduced foot-prints, and presumably, negligible sludge production. As the low sludge production is considered as one of the prominent feature of any wastewater treatment technology, several early experimental studies on MBR with zero sludge production (Houten and Eikelboom, 1997; Muller *et al.,* 1995) have been reported, however, inevitability of sludge production (concentrated though) with inert material accumulation, lower biomass activity and viability has been observed evident. MBR researches available after the first decade of submerged MBR technology have shown sludge production rates varying between 0 to 0.34 kgMLSS $\text{kgCOD}_{\text{removed}}^{-1}$ (Stephenson *et al.,* 2000). Evidently, the biological process associated with MBRs is distinct in nature and the sludge properties therefore, in terms of microbial dynamics and sludge floc/granule constituents, are different as compared to those from convention wastewater treatment. The development of associate distinct-sludge treatment technology for complementing the MBR potentialities in wastewater treatment has an obvious urge. Thereby, the current research aims at the treatment/handling of MBR-specific sludge.

The hypothesis here is based on the logic that MBR sludge is rich is inorganic contents (presence of metals etc. entrapped in sludge floc matrix) because of diverse biological process. Since these inorganics can help in electric conduction, the application of voltage across the sludge volume can lead to the electro-oxidation/lyses. Moreover, if assisted by an oxidizing agent, the performance of such a method can be improved, and that the partially treated sludge can be re-circulated to the MBR biological process leading to very high reduction in the net sludge production.

University of Trento is conducting pilot study (Approx. 200 PE) on the application of MBR for municipal wastewater treatment at Lavis WWTP (Trento, Italy). The MBR pilot plant consist separate biological tank/reactor and membrane tank where ultra-filtration hollow fiber membrane module (Zenon, ZW500d, PVDF nominal pore size-0.04µm) is immerged. The sludge treatment experiments are being carried out with the sludge from mentioned MBR.

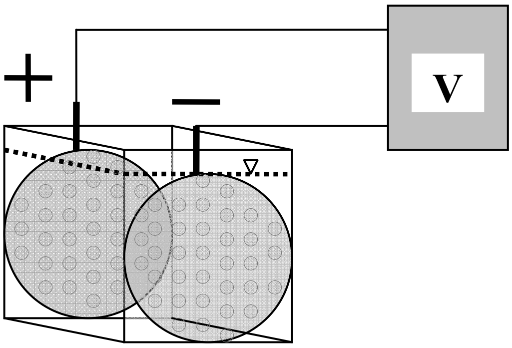


Figure 1 Schematic representation of the mini-reactor used in experiments

The electric effect on MBR sludge is being carried out in a mini-reactor (7.5 cm x 7 cm and 10 cm height, as show in Fig. 1). The dc-voltage of 15.6 V across the plates was maintained throughout the experiment (Stabilized power supply- Model: AR50; STAB, Italy). During Phase-1 experiment, one liter the MBR sludge has been concentrated by filtering (Filter paper 593 ½; Schleicher & Schuell) and reducing the volume by 50%; effect of the application of voltage, across stainless steel perforated circular plates (Circular disks- 7.5 cm dia; holes Φ 3 mm@ 1 cm; 2 mm thick) immerged fully into mini-reactor filled with concentrated sludge, have been aimed to be studied. The experiment lasted 4 hours during which the constant voltage has been maintained. During Phase-2 experiment, without sludge any concentration/volume reduction, the H_2O_2 assisted electro-treatment of the sludge has been studied along with only- H_2O_2 (without electro treatment) treatment. The Phase-2 has consisted two experiments: one with 20 ml of H_2O_2 (3 %) and 2 hours of electro-treatment, and other with 20 ml of H_2O_2 without any other assistance. The total batch volumes of 450 ml have been used for both Phase-1 and Phase-2 experiments.

Table 1 Results of Phase-1 experiment

Parameters measured*†	Filtrate of the raw sludge	Filtrate of the sludge after electro-treatment (4h)
COD, mg/L	95	1254
TOC, mg/L	33	473
Org-N, mg/L	15.3	194
Amm-N, mg/L	7.3	10.6
Nitrates, mg/L	7.2	7.4
Nitrites, mg/L	1.2	0.18

*†All the notations correspond to usual wastewater engineering terminology

Table 2 Results of Phase-2 experiment

Parameters measured	Sludge after H_2O_2 assisted oxidation		Sludge after H_2O_2 assisted electro-treatment (2h)	
	Filtrate	Total	Filtrate	Total
COD, mg/L	76	13450	57	8911
TOC, mg/L	34	1598	25	1565
Org-N, mg/L	61	613	70	621
Amm-N, mg/L	4.6	18.2	6	16
Nitrates, mg/L	<0.1	1.2	1.2	1.6
Nitrites, mg/L	0.05	0.04	0.09	0.03

The aim of the Phase-1 experiment has primarily been to observe the electric effect on the sludge matrix. From the experimental results (Table 1), it's clear that the electro-treatment plays an effective role in destruction and segregation of microbial flocs and thereby makes available a high fraction of COD in free form (High filtrate COD, TOC, Org-Nitrogen after electro-treatment). Subsequently, the Phase-2 experiment has been carried out in order to compare the relative effect of electro-treatment and the effect of H_2O_2 coupling. Results (Table 2) show that, as far as the carbon removal is concerned (TOC reduction), there is no significant advantage of coupled effect of H_2O_2 and electro-treatment; however, the electro-treatment assisted by H_2O_2 seems very promising for COD removal. Since, the Org-Nitrogen and TOC in both cases are not very different, the low COD value (8911 with respect to 13450) indicates the formation of more oxidized matter. It can then be inferred that the electro-treatment of the sludge significantly segregates/destroys the sludge flocs and while assisted by an oxidant (H_2O_2 here), improves greatly the process of organic sludge mineralization. It seems that the oxidation of organic nitrogen is less effective than that for lysed/free organics. As well, the experiment is required to be continued and it aims at the Phase-2 experiment based study on the sludge from conventional WWTP (same site) in order to verify completely the hypothesis and the relative applicability of the technology for MBRs.

Additionally, it can be commented the sludge treatment/reduction by means of advance oxidation is rather well known and several researches have been reported. However, very importantly, considering the prevalence of membrane fouling issue over other issues related with MBRs, the recirculation of treated sludge to MBR process is subject to skepticism as the possibility of potential membrane foulants in the treated sludge can not be ignored. Hence, the treatment of sludge (considered to be recirculated) has to be carried out to an extent such that the colloids/macromolecules from cell lyses have already been destroyed. The current research urges as well on the study of fouling potential of treated sludge.

Greywater Recycling with MBR Supply of hygienic service water for an office building

Dr.-Ing. Stefania Paris; Celine Schlapp
Hans Huber AG, Industriepark Erasbach A1, 92334 Berching, Germany

Keywords: Grey Water, Treatment with MBR, Treatment and Disinfection Efficiency, Quality Requirements

Abstract

The average amount of greywater (effluent from bath tubs, showers, wash hand basins, washing machines, dish washers) produced per day by a German household (new building) is 70 l per person, which is more than 50% of the total wastewater production (Mehlhart, 2001). Compared to the total domestic wastewater, greywater contains less organic pollutants and less nutrients. Greywater from bath tubs, showers or wash hand basins contains for example a by approx. two decimal powers reduced number of total and faecal coliform bacteria (E.coli) (Nolde, 1995 and Bullermann et al., 2001).

Greywater can be treated with a relatively low effort and after hygienisation reused in the household. The fresh water demand can be reduced significantly by substituting fresh water with service water. If service water is used for toilet flushing, irrigation, cleaning and car washing, more than 41 % of the domestic water consumption in Germany can be saved.

A suitable method to provide service water from greywater is for example the membrane bioreactor process. The combination of aeration and membrane filtration is able to provide a bacteria-free and virtually germ-free effluent on a small footprint. Testing over four months with the HUBER GreyUse® treatment plant, which is used to treat the greywater from sinks, washing machines and wash hand basins in the HUBER office building in Berching, has proved that there is no limitation of the biological treatment due to a lack of nutrients. The system shows a very high C-elimination. Due to its low BOD_7 concentration the permeate can be stored without problems. Besides, the effluent is solids-free and virtually germ-free. The effluent quality meets the standards, specified in sheet H201 of the German guidelines from the professional association for service and rainwater utillisation (fbr, 2005), for the reuse of treated water for toilet flushing, laundry washing and irrigation (table 1). At present, the clarified greywater is still reused for irrigation of the company's own garden. When the office building expansion will be realised, a second line will be laid in the new part of the building in order to provide for a combined use of treated greywater and rain water as toilet flush water. This will sustainably reduce the fresh water demand.

Table 1: Quality requirements on service water

Parameter	**Standard** H201$^{1)}$ (76/160/EWG)$^{2)}$	**HUBER** Service water
BOD_7	< 5 mg/l (-)	< 2.4 mg/l
Oxygen saturation	> 50% (80-120%)	> 50%
Total coliform bacteria	< 100/ml (500)	< 1/ml
Faecal coliform bacteria	< 10/ml (100)	< 1/ml
Pseudomonas aeruginosa	< 1/ml (-)	-

$^{1)}$ fbr sheet H201 $^{2)}$ EU standards for bathing waters 76/160/EWG

Contribution of nanoparticles from a biological supernatant in MF fouling

Benoît Teychene*, Christelle Guigui* , Corinne Cabassud*

*INSA, Laboratoire de génie des procédés de l'environnement, 135 Av de Rangueil, 31077 Toulouse, France (Email : Benoit.Teychene@insa-toulouse.fr; Christelle.Guigui@insa-toulouse.fr; Corinne.Cabassud@insa-toulouse.fr)

ABSTRACT

The aim of this study was to investigate the role of nanosized particles on fouling in a membrane bioreactor. Optimization of membrane bioreactors requires a better knowledge of the typology of compounds (colloidal or soluble) involved in membrane fouling and on their impacts in cake layer building and on membrane surface charge.

This knowledge is necessary to better adapt operating parameters in the MBR process or to target more precisely membrane cleaning. However, as shown by previous studies, it is difficult to conclude since different effects are observed depending on the fluid nature (synthetic, real wastewater, domestic or industrial) and operating conditions (module design, hydrodynamics and membrane material). Researchers have been working diligently at developing 'taxonomy' of NOM-membrane interactions and fluxes decline/fouling during filtration processes. For instance some authors have demonstrated the important role (positive or negative) of the solid fraction [1;2] and pointed out the presence of a low size particle population in MBRs which could explain the higher fouling ability in comparison with conventional activated sludges in which this population is absent [3]. Other studies pointed out the role of the non easily settable part, in particular proteins and polysaccharides, as the main actor of fouling [2;3;4]. However this non-easily settable part, depending on its definition, can contain both soluble and non soluble (colloids or nanoparticles) organics. Up to now, the role of nanoparticles in fouling is not clear…

This study aims to investigate the role of nanoparticles in membrane fouling in presence of solutes from a biological supernatant. Our strategy was to study the role of synthetic nanoparticules (latex and melamine) with the same mean diameter (500nm), in different environments. Different solutions/suspensions were tested, (i) supernatants (obtained after centrifugation at 10000g and 4000g) sampled from a MBR owned by Polymem (Labège, Toulouse-France), (ii) supernatants + nanoparticles.

Fouling ability of theses suspensions was characterised during filtration batch tests using a dead-end stirred cell (AMICON) using polysulfone microfiltration membranes (0.1 µm).

A preliminary characterisation by HPLC-SEC measurement of foulants present in a MBR supernatant showed that the main foulant exhibit a molecular weight distribution from several thousands to ten thousands Daltons, as reported by S.Ognier et al. and Lee et al. [5,6].

As an example Figure 1 reports HPLC-SEC measurement from a filtration test performed at 0.5 bars on a microfiltration membrane (Figure 1-a: backwashed water (500mL at 1 bar); 1-b permeate water).

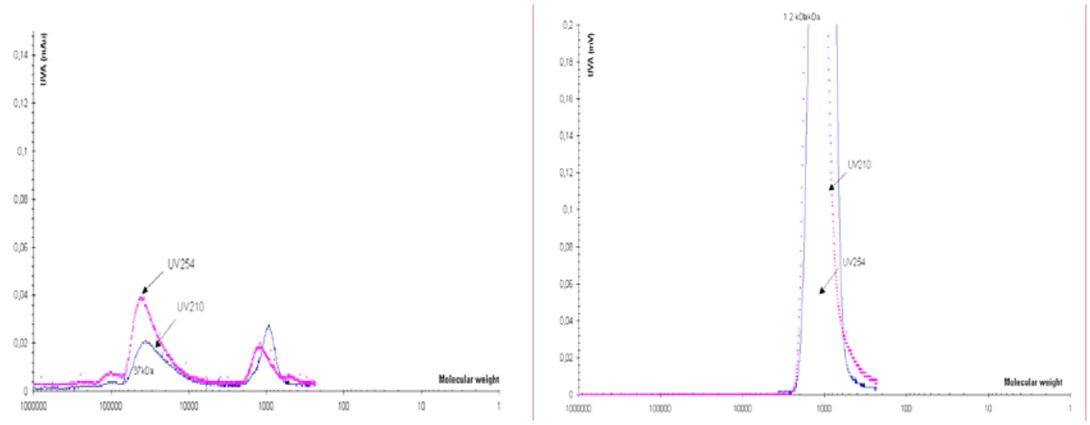


Figure 1 HPLC-SEC measurement (a) on backwashed water (b) permeate water.

The fouling ability of each solution was characterised using the pressure step method and membrane rinsing (with RO water) were performed to determine membrane fouling reversibility. A specific procedure allowed to obtain information on osmotic pressures for the different solutions. This procedure allows identifying which is the role of nanoparticles in term of membrane fouling and of non reversibility in terms of colloidal gel formation.

Particle size distribution of each suspension was measured. Membrane surfaces after filtration and rinsing were analysed by SEM and EDX. Moreover estimation of fouling effects through modification of membrane surface charge was investigated by zeta potential measurement using streaming potential method (before and after fouling).

Fouling ability for a given pressure was also characterised by determination of the cake property α.C, were α is the cake specific resistance.

Results will be presented, that will allow to compare the fouling ability and reversibility of biological supernatants depending on the way they have been prepared (centrifugation at 4000 or 10000g) and in relation with their content in nanoparticles. As an example two supernatants have the same DOC value (measurements performed after a 0.45µm prefiltration), but show a different cake specific resistance. Further experiments are actually done to confirm these results. Results with nanoparticles will also be shown.

Table 1 Filterability of two supernatant obtained during a microfiltration test at 0.5 bars (Polysulfone membrane 0.1 µm pore size; without stirring).

Supernatants	DOC (mg/L)	Pressure (bar)	α.C (10^{13})
4000 G	10~11	0,5	2,25
10000 G			1,67

First results show different filtration performances in terms of fouling ability and reversibility depending on the way the supernatant solution is prepared and on its content in added nanoparticles and it suggest a modification of membrane surface charge in term of streaming potential values.

Optimisation of phosphorous removal and denitrification processes of full-scale in-building MBR systems for urban reuse in the US

B. Verrecht[1], A. Higgins[2], R. Birks[1]

[1] Thames Water Utilities Ltd., R&D, Spencer House, Manor Farm Road, RG2 0JN Reading, Berkshire, UK, (E-mail: bart.verrecht@thameswater.co.uk)
[2] Applied Water Management, 2 Clerico Lane, Hillsborough, NJ 08844, USA

The USA plays a leading role in the development of decentralised MBR systems for urban reuse. Incentives and drivers differ depending on the region. In areas with water scarceness, e.g. California and Florida, the main driver is water conservation for reuse purposes such as golf course irrigation. However, on the east coast of the US, water scarceness is less of an issue. In metropolitan cities on the east coast, such as New York City (NYC), the main driver is the "green" image linked with the reuse of blackwater. Constructions conforming the "LEED" standards (Leadership in Energy and Environmental Design) attract more clients and potential tenants are willing to pay more rent. In-building reuse MBRs in these urban areas focus mainly on water recycling for toilet flushing, make-up water for cooling towers and irrigation of roof gardens and parks. In NYC, an extra economic incentive was introduced in 2004: buildings that can reduce their water demand by at least 25% (compared to a defined average basic usage of 261 l/person/d) receive a 25% reduction in water and wastewater charges. However, these benefits do not compensate totally for construction and production costs of the reuse systems, so at present these systems are still not economically viable.

In this paper, an overview of MBR reuse systems located in NYC and operated by Applied Water Management will be presented. Since the Battery Park City Authority requires new buildings to confirm to the LEED standards, most reuse MBRs are found in this area. The Solaire, the first environmentally responsible building in the US, was built in 2003 and has a capacity of 95 m^3/d. Other buildings, with comparable systems, include Tribeca Green (53 m^3/d), the Millennium (95 m^3/d), and several buildings currently under construction. Current research focuses on optimisation of the phosphorous removal and denitrification processes at the Solaire plant, and will be presented in this paper.

The consent for the effluent PO_4^- concentration is 2 mg/l at the Solaire, driven by cooling tower operation. Higher PO_4^- levels lead to precipitation in the cooling tower, which decreases its performance and expected lifetime. Currently, aluminum chlorohydrate is dosed at a constant rate, successfully reducing the effluent PO_4^- levels to about 1 mg/l. However, preliminary measurements indicate that influent PO_4^- levels vary largely during the day, from 18 mg/l in the morning decreasing to 7 mg/l in the evening, assumed to be from the tenants diurnal variations in behaviour (e.g. varying detergent usage during laundry, showering in the morning etc). By adjusting the dosing of aluminum chlorohydrate to varying influent flows and PO_4^- levels, a reduction in chemical usage can be achieved. A comparison with an anion-exchange technique will also be made.

The denitrification process at the Solaire is also being optimised. Issues related to the denitrification process include the presence of significant levels of dissolved oxygen (DO) in the anoxic zone from air mixing, low anoxic zone pH and associated high consumption of sodium hydroxide (NaOH) and also high MBR effluent NO_3^- levels (approximately 50 mg/l). To optimise this process, DO levels in the anoxic tank need to be reduced by both reducing the recycle flow from the membrane tank to the anoxic tank and reducing the coarse bubble mixing of the anoxic zone. The ratio of soluble COD:TKN will also be determined, to judge the possibilities for optimisation without additional carbon dosing.

A technical overview of the NYC MBR plants and the results of the proposed optimisation techniques will be presented in this paper.

Treatment of synthetic wastewater containing (RS)-MCPP using Anaerobic Membrane Bioreactor (AMBr)

Ali Yuzir* & P.J.Sallis

*Environmental Engineering Group, School of Civil Engineering and Geosciences, University of Newcastle upon Tyne, Newcastle upon Tyne NE1 7RU, UK (Email:m.a.yuzir@ncl.ac.uk)

This research investigated the feasibility of the biological treatment of wastewater containing (*RS*)-MCPP in an anaerobic membrane bioreactor. The reactor was fed with a synthetic wastewater containing brewery wastewater supplemented with (*RS*)-MCPP within range of 5 to 200mg/L and operated under methanogenic conditions with an average OLR of 1.5 kg COD $m^{-3}d^{-1}$ **(Phase I)**. The process performance of the reactor was characterised in terms of its COD degradation and (*RS*)-MCPP degradation. Throughout Phase I, the reactor achieved a COD reduction of more than 95% at an HRT of 3.3 day, suggesting the robustness and stability of AMBr system towards (*RS*)-MCPP however, an average of only 15% of (*RS*)-MCPP degradation occurred in Phase 1. During **Phase 2** the reactor achieved an average COD reduction of 94% when HRT was increased in the range of 3.3, 6.8 to 16.9 days with up to 75% (*RS*)-MCPP degradation achieved at the longest HRT.

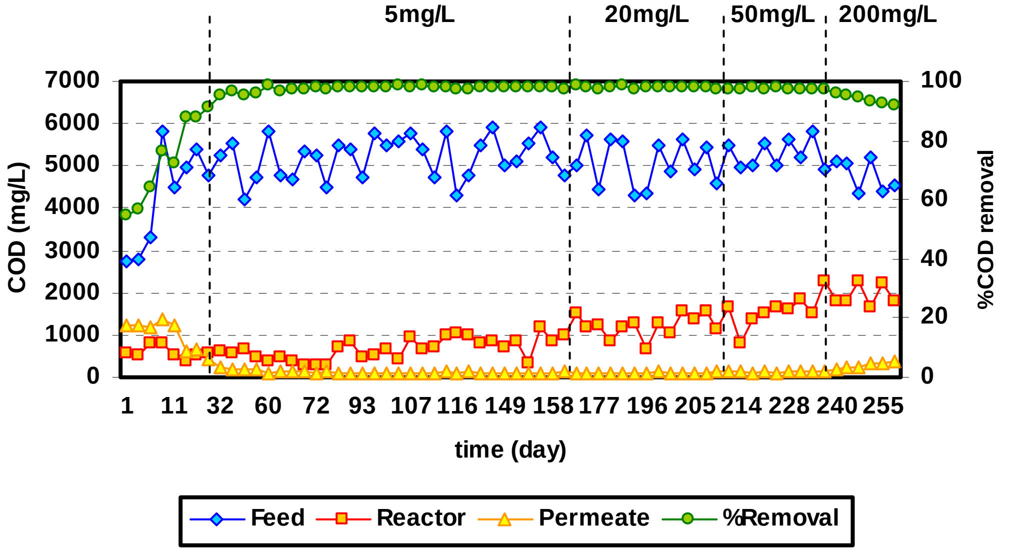


Figure 1.1 COD removal efficiency profile in AMBr at different (RS)-MCPP concentration during Phase 1

Persistent Organic Pollutants (POPs) removal in conventional WWTP and in a MBR pilot plant for agricultural reuse.

Gianluigi Buttiglieri **, Helene Bouju , Francesca Malpei *,
Politecnico di Milano, Milan, Italy
Ettore Zuccato, Istituto Mario Negri, Milan, Italy

*Contact author: francesca.malpei@polimi.it, via Golgi 39 Milano (Italy), tel. +390223996434, fax +390223996499
** Presenting author,

Several POPs have been identified in raw wastewater. They are partially removed by conventional activated sludge system (CAS) and are persistent in treated wastewater and surface waters. These substances, even at low concentrations, have high activity and can exert toxicity to living organisms (EC Directive 850/2004). Nevertheless the knowledge on this issue is still scarce and scattered. Some of these pollutants (e.g. pharmaceuticals) have not been yet regulated and standards for water quality related to these substances are not available.

Since most of these substances are of human origin the urban wastewater is considered as the main source of contamination of the aquatic environment.

Aim of the project is to study the removal of pharmaceuticals and other micropollutants (like nonylphenol and metals) from municipal wastewater, with conventional activated sludge process and MBRs. A large scale WWTP (Milano Nosedo, P.E. 1,250,000) and a pilot scale MBRs operating in parallel are investigated.

Treated wastewater from the Nosedo treatment plant enter the canal Vettabbia with low dilution ratio and is then largely reused for agriculture. This is one of the largest case of reuse of wastewater resources in Europe and therefore the monitoring of water contamination, by pharmaceuticals and drugs, is of particular interest for several reasons including reliable assessment of risks for environment (treated wastewater not reused flows into the River Lambro) and, through the food chain, for humans.

In previous works (Castiglioni et al., 2006, Zuccato et al., 2005) the mean concentrations of the pharmaceuticals were evaluated both in some Italian WWTP as well as in rivers (e.g. river Po and Lambro in the nearby of Milano-Nosedo). The total pharmaceuticals load in the effluent from the WWTP was in the range 1.0÷3.0 g/day/1,000 P.E., with total removal mostly lower than 40%.

The loads of selected pharmaceuticals in the influent to Milano Nosedo WWTP will be evaluated by HPLC-MS-MS and normalized to the population equivalent and macro-pollutants loads (COD, TSS, etc.).

The pilot MBR, fed in parallel with the Nosedo CAS, allows a comparison between POPs removal in both systems. The anoxic-aerobic MBR pilot plant (90 L anoxic, ~200 L aerobic with biomass concentration around 6 gTSS/l in aerobic tank, permeate flow of 0.5 m^3 d^{-1}) is running with hollow fibres micro-filtration membranes (Zenon® Zee-Weed-10, surface area 0.93 m^2, characteristic pore size of 0.2 µm) in same operating conditions of the full scale plant in terms of sludge age and anoxic/aerobic volume ratio (Fig. 1). Different process operating conditions (sludge age, biomass concentration, volumes, recycle ratios, etc.) will be later investigated.

Figure 1 - MBR pilot plant installed in Milano Nosedo WWTP.

The adaptation of the biomass to the chosen pollutants will be studied through the analyses of the responses to both POPs pulses and continuous flow ones. Adsorption is being estimated by lab-scale isotherms and by studying micro-calorimetry results. On the other side biodegradation analyses, using micro-calorimetry eventually validated with respirometric tests, will be carried out and the presence of potential intermediate metabolites will be investigated.

Previous results obtained, on a synthetic water spiked with Atrazine concentration of 10 g/l, show that the removal was variable (from 10 to 40%) and, anyway, slightly higher than the expected removal by only physical adsorption on the basis of lab-scale isotherms ($q=0.0253*C^{1.5328}$ with q as the adsorbed amount in µg/gSS and C the equilibrium concentration in µg/l). Biodegradation measurements were done using a micro-calorimeter BIO-RC1 (Mettler Toledo, Switzerland), but not significant biodegradation was detected. Atrazine concentration and adsorption on sludge were analysed with standard methods.

Author Index

The Berlin Centre of Competence for Water organised, together with the International Water Association, the 2nd National Young Water Professionals Conference in Germany. The conference topic was "Membrane Technologies for Wastewater Treatment and Reuse".
More than 150 scientists and professionals working in the membrane sector of the wastewater industry, coming in great extent from Germany, but also from all over Europe and oversees, attended this 3-day event. Leading-edge research projects and experiences were presented in about 50 presentations and 30 technical posters.
The present conference proceedings include all full articles submitted to the conference organisers for the platform presentations, and the extended abstracts of the posters. The best full papers will be considered for further publication in Water Science and Technology.
This conference is an initiative of "MBR-Network", the European coalition on membrane bioreactor technology, gathering about 50 European and international companies and institutions (www.MBR-Network.eu).

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