SIXTH FRAMEWORK PROGRAMME

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EUROMBRA

Membrane bioreactor technology (MBR) with an EU perspective for advanced municipal wastewater treatment strategies for the 21st century.

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D5 - Literature Review on Aeration and main Operating Conditions in Membrane Bioreactors

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Introduction

Because of their great potentiality in terms of water disinfection and as they offer the possibility to reuse water, MBRs are showing a growing interest and a rapid development. Many MBR plants are now in operation worldwide for domestic waste waters and are replacing the conventional treatment (activated sludge + settling). Besides, numerous academic studies are also focusing on MBRs (Yang et al., 2006).

The objective of this report is to synthesise some available knowledge on the interactions between main operating conditions and filtration performances in MBRs.

Filtration performances can be influenced (see figure 1) by strong interactions between the membrane system (membrane structure and shape, module design), the biological fluid and its composition, (which are themselves influenced by the bioprocess operation), the filtration operating parameters (permeate flux, aeration for fouling control or removal, relaxation, backwashes).

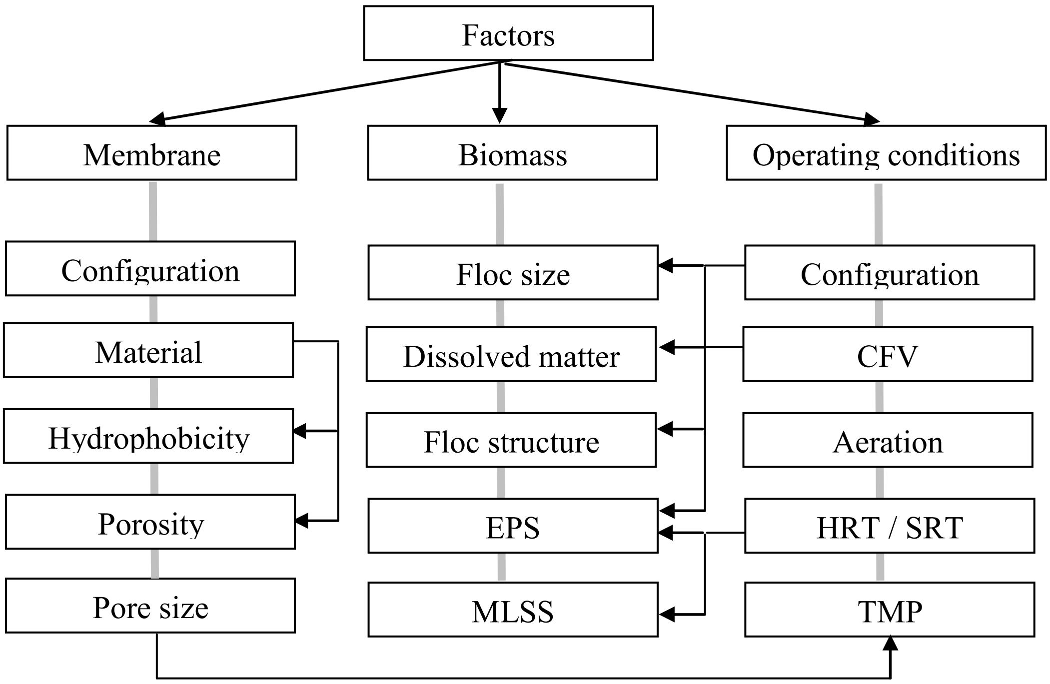

Figure 1 : Factors influencing membrane fouling in MBR process [from Chang et al. (2002)]

Indeed filtration performances can be limited by membrane fouling and the aim of most studies about MBR process is to prevent or to limit fouling in order to enhance system performances. For that reason it is necessary to consider the parameters influencing fouling that can considered as linked to three factors : membrane characteristics, operating conditions and biomass characteristics (Chang et al, 2002) .

For that reason, these different aspects will be introduced successively in this report.

A specific attention will be given to the conditions of aeration for filtration operation, which appear clearly as a key point as well for fouling control as for energy considerations.

A. Presentation of filtration systems in MBRs

On a filtration point of view, MBR systems can be defined and classified according through key points of the filtration process: membrane, filtration mode, module design and filtration process. The objective of this paragraph is mainly to present the main definitions of the filtration system that can be useful to describe MBRs.

A.1 Membrane and filtration modes

Most MBRs are based on the use of microfiltration or ultrafiltration membranes; membranes can be in the shape of hollow fibres, tubular or flat sheet In the case of hollow fibres or tubular membranes, filtration can be operated from outside to inside the membrane (which is the case for all hollow fibres) or from inside to outside the membrane (only for tubular membranes).

Inside/Out Outside/In

Figure 2: Outside/in and inside/out filtration

A.2 Module design

According to Fane (2002), the module configuration defines the membrane arrangement, the packing density, the filtration mode and the hydraulic distribution throughout the membrane network. Module configuration also determines the energy requirement and the ease of membrane regeneration. Three main types of modules are used in MBR processes for wastewater treatment, which are based on flat sheet membranes, tubular membranes or hollow fibre membranes.

The classification chosen in this project (cf Deliverable D10) distinguishes “carterised” and “non-carterised” membranes. Membranes can be inserted in a carter or in a cassette, and will the be considered as “carterised” or will only be gathered together to build a set without external rigid walls (and so called “non-carterised”). Carterised systems are confined systems in which membrane motion is limited by the carter walls and the liquid and air flows are also limited by these walls. Non-carterised systems allow more freedom for membrane motion but also do not fix any limit for the flows, which are more difficult to define and to control.

Carterised Submerged (non carterised)

Figure 3 : Definition of caracterised/non carterised system

A.2.1 Flat sheet module

A flat sheet module is an assembly of flat sheet membranes. Each membrane is housed within a rectangular box which collects the permeate. For MBRs application, the space between membrane sheets must be at least of 6 to 7 mm [Sofia *et al.,* 2004].The main examples of flat sheet membranes in MBRs are the KUBOTA membranes. In such modules hydrodynamics is easily controlled for one-phase flow. In case of gas/liquid flows description of flows is much more complicated [Ducom *et al.*, 2003]. Mostly, flat sheet configuration does not allow to perform backwashes.

A.2.2 Tubular module

Tubular modules may result from an association of numerous tubes, single membrane tube or multi-channel membrane tubes. The tubes are linear and rigid. They are mainly operated in outside/in filtration. Sludge deposit within the tubes can be minimised and membrane permeability controlled if high liquid velocities and shear stresses are used. In MBRs, the inner tube diameter must be higher than 5 mm to prevent tube clogging. In MBRs the tubes can be polymeric as in STORCK or Milleniumpore systems or mineral as in the Degrémont MBR. As examples, Mayer *et al.* (2005) used a tubular membrane module with 5.2 mm inner diameter tubes (109 tubes). Chang *et al.* (2002) worked with a module composed of 9.5 mm inner diameter polyethersulfone tubes (20 tubes).

Figure 4 : Multi-channel membrane

The following table shows some examples of tubular membrane geometries for use in MBRs.

Table 1 : Examples of tubular membrane geometries for use in MBRs

	External diameter *(mm)*	Specific area *(m^2/m^3_{module})*
Mineral membrane	8	250
	12	200
	20	120

A.2.3 Hollow fibre module

In MBR system, hollow fibre modules are composed of bundles of fibres with 1.5 to 2.5 mm inner diameter. These modules are mainly operated in outside/in filtration to avoid fibre clogging. Several bundle configurations have been developed for hollow fibres and studied for application to MBRs. For example, at lab scale the following modules can be found:

- U shaped bundle:
[Espinosa-Bouchot *et al.,* 2003; Yu *et al.,* 2003; Van Kaam *et al.,* 2004]
It contains one or several bundles of fibres disposed in 'U' (figure 5) and potted together at bottom of the module. This arrangement can only be developed with low inner diameter fibres and deformable fibres.

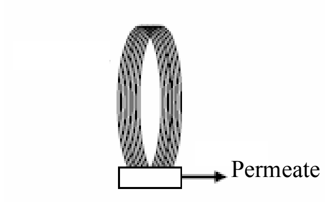

Figure 5 : U-shaped bundle

- One-side potted and freely movable
[Espinosa-Bouchot *et al.,* 2003]
Each Fibre is glued internally at its top and the bundle is potted at the bottom. Fibres are free to move independently at their top and are fixed at their bottom (figure 6).

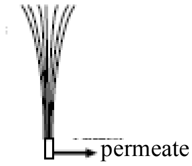

Figure 6 : one-side potted and freely movable fibres

- Right and tended fibres module
[Hong *et al,* 2002; Lim *et al,*. 2003]
Fibres are potted together on both sides. They are right and slightly tended (figure 7). This configuration does not allow fibre motion.

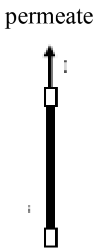

Figure 7 : Right and tended fibres

- Right and released fibres
[Hong *et al.,* 2002; Delgado *et al.,* 2004]
These modules have the same configuration than the previous one but the fibres are not tended (figure 8). Thus in this case, fibres are more free to move in between their two fixation points.

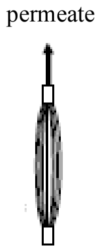

Figure 8 : Right and released fibres

- Right fibres module in horizontal position
[Ueda *et al.*, 1997]
They are composed by right and non–tended fibres positioned horizontally (figure 9). Feed water flows vertically through the bundle. As a consequence hydrodynamics and fouling phenomena might be very different than in the vertical position. However, this module configuration might favour fibre breakage.

Figure 9 : Right fibres module in horizontal position

A.3 Process operation

Different kinds of process operation modes can be distinguished, which depend on:
- the liquid velocity inside the carter (in the case of carterised systems),
- the use of aeration or not during filtration,
- the location of the membrane compared to the bioreactor,
- the recycling of concentrate in the bioreactor area.

A.3.1 Aeration or not

Additionally to air aeration for the biomass, air can be injected at the membrane bottom in order to limit fouling of the membrane surface or clogging of the bundle. Air is introduced below the membrane assembly and is supposed to be ideally distributed to optimise the air scouring action across the membrane surface. The ideal air flow and flow pattern for this purpose are not clearly known today. For providing oxygen to the biomass it is evident that small air bubbles are required to increase oxygen transfer between the gas and the liquid phase. However the ideal aeration mode for preventing fouling is not clear today even if numerous studies claim that big bubbles would be preferable.

Aeration for fouling prevention can be operated using specific air injectors, independently to the air injection for biomass. Operation with sequential air injection is also been used at industrial scale, for example in the ZENON systems. The objective of this sequencing is to decrease operating costs due to the energy consumption induced by air sparging.

A.3.2 Location of the membrane system compared to the bioreactor

According to the definition proposed in D10, two different membrane systems locations can be distinguished: inside the activated sludge tank (so called “submerged”) or external to it (so called “side-stream”).

A.3.2.1 Membranes submerged in the activated sludge tank

As proposed by Yamamoto in 1988, membranes can be completely submerged in the activated sludge tank (figure 10).

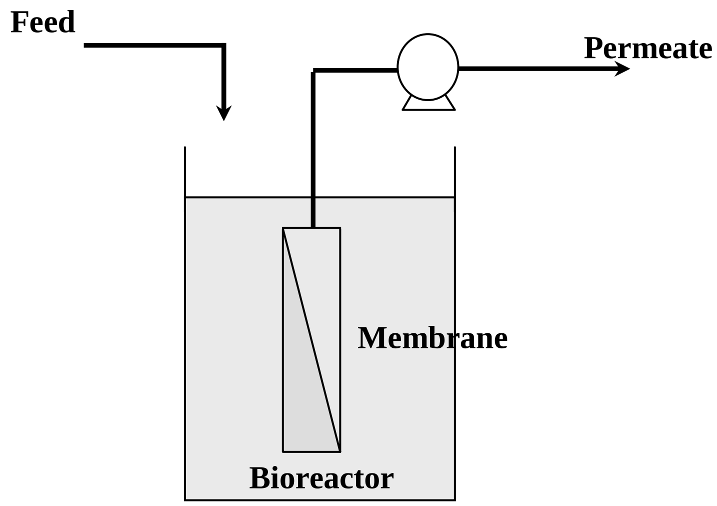

Figure 10 : submerged membranes

The permeate is sucked through the membrane wall with a pump. Air is injected at the membrane bottom in order to limit fouling. Recent systems are using two different aeration systems: one injects fine dispersed bubbles which are necessary for biomass growth and one generates coarse bubbles to control fouling. The liquid flow close to the membrane is only due to permeate flow through the membrane wall and to a liquid motion induced by the bubbles.

This process is commercialised by industrial firms for instance with flat sheet membranes (Kubota, Zenon) or tubular membranes (Milleniumpore).

A.3.2.1 External membranes system

The second possible location for the membrane systems is to be external to the activated sludge tank (figure 12).

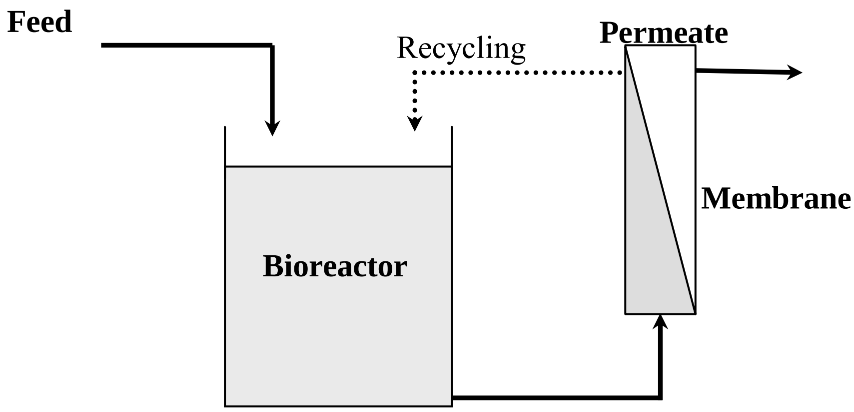

Figure 11: external membrane system or "side-stream" system

The membrane system is thus outside the bioreactor. If the concentrate is recycled to the bioreactor a liquid velocity is generated in the membrane system, the flow patterns and the recycling rate are linked. For tubular membranes, high velocities and high recycling rates are generally imposed. In hollow fibres or flat sheet membranes, the range of liquid velocities is lower. Low liquid velocities are used to decrease the particles concentration in the membrane system. High liquid velocities are used with the aim to control the formation of the deposit. If the concentrate is not recycled, the liquid velocity in the carter is negligible, but the water recovery by the filtration system is 100%, which can be called a "dead-end" system. This process is commercialised by industrial firms with hollow fibres as Polymem, Degrémont, Mitsubishi, Memcor, Zenon.

Possible advantages and drawkacks of these two MBR configurations are presented in Table 2.

Table 2 : Advantages and disadvantages of MBR configurations (Till and Malia (2001))

Submerged membranes	External membrane system with high recycling rate and high velocity
Aeration cost high (~90%) Very low pumping costs (higher if suction pump is used (~28%) Lower flux (larger footprint) Less frequent cleaning required Lower operating costs Higher capital costs	Aeration cost low (~20%) High pumping costs Higher flux (smaller footprint) More frequent cleaning required Higher operating costs Lower capital costs

There are today very little information about external membranes operated with a low recycling rate or velocity, like the one which is under development by Polymem or like so called air-lift systems. Increasing the level of information about these systems is one of the goals of the EUROMBRA project.

A.4 Comparison of membrane systems

There are very few studies showing a comparison of membrane systems. Numerous studies are describing MBRs operation and are providing data on a single system. But comparison of these data is very risky. Indeed, it is very difficult to compare systems which are very different, operated with different conditions with different waste waters, at different scales (from lab-scale to industrial scale) and characterised using different analytical procedures..

One of the aims of the EUROMBRA project is to establish, through the large network of partners, a consistent database to compare quantitatively the performances of different MBRs systems under controlled conditions.

As an example, Fane (2002) proposed a qualitative comparison of the characteristics of some membrane modules (Table 3).

Table 3: Characteristics of tubular membranes, flat sheet membranes and hollow fibre membrane, adapted from Fane (2002)

Characteristics	Tubular membranes	Flat sheet membranes	Hollow fibre membranes
Arrangement	External - recycling	External / submerged	External / Submerged
Packing density	Low	Moderate	High
Energy demand	High (turbulent flow)	Low-moderate (laminar flow)	Low
Cleaning	Efficient + physical cleaning possible	Moderate	Back washing possible
Replacement	Tubes or element	Sheet	Element

The following table (table 4) introduces typical values of fouling resistance for different MBR systems. It is clear that systems are difficult to compare as the resistance is often given for different filtration time and permeate flux, so for different values of total filtrated volumes. Moreover aeration conditions are also often different and expressed using different units: air volume/liquid, liquid volume/h or air volume/membrane area/h.

Table 4 : Example of cake resistances values in MBR systems

MBR system	Wastewater	Permeation Flux ($L.h^{-1}.m^{-2}$)	Filtration Time (d)	Aeration ($m^3.h^{-1}.m^{-2}$)	Resistance (10^{11} m^{-1})	References
Submerged HF	simulation	12.5	25	36 64.8	175 100	Li and Wang (2005)
Submerged HF	Raw water	20.8	70	nc	60 (pore fouling) 308 (stable sludge cake) 32 (dynamic sludge film)	Chu and Li (2005)
Submerged HF (Zenon)	Real wastewater	10	nc	1.2 4.8	90 30	Bouhabila (2001)
Submerged HF	Real wastewater	From 1.5 to 10	Increase of flux every 20 min	1.2	Up to 2500	Lesage (2005)
Submerged HF	Real wastewater	5	67	0.1	160	Massé (2006)
Submerged non-woven tubular	Synthetic wastewater	16.7-50 16.7-50	nc	118.8 79.2	11-33 2-5	Chang (2006)
Submerged Tubular (Milleniumpore)	Sewage	From 8.3 to 82.5 every 8	Increase of flux every 15 min	2.1	From 1.7 to 7.2	Le Clech (2003a)

B. Influence of operating parameters on filtration performances

Different operating parameters seem to have an influence on filtration performances but conclusions proposed by the authors can be different depending on the MBR system they are using. In this paragraph, we will successively introduce information about the influence of membrane material, permeate flux, and information on long term operation. Paragraph (D) will focus on aeration and more specifically on the role of operating parameters in relation with aeration conditions and the MBR system configuration.

B.1 Impact of membrane material

For the treatment of domestic waste waters as for drinking water production, organic membranes are today more often used, partly because operating conditions (pressures, temperatures …) are close to those recommended for their use.

In MBRs, membrane fouling is inevitably linked to the interactions between the membrane material and the components of the activated sludge (feed components, cells, microbial metabolites).

It is generally considered that extracellular polymeric substances (EPS) are playing an important role in fouling [Chang and Lee, 1998]. EPS compounds are mainly proteins, polysaccharides and humic substances. Proteins show high affinities with hydrophobic surfaces whereas carbohydrates are relatively much more hydrophilic than proteins [Mukai *et al.*, 1999].

One important characteristic of membranes in MBRs is thus their hydrophilic/hydrophobic comportment. Choi *et al.* (2002) tested the behaviour of three membranes in MBR conditions: a very hydrophilic (cellulose acetate), an hydrophobic (polyethersulfone) and an intermediate (sulfonated polyethersulfone). The more hydrophobic membranes were much more sensitive to fouling than the hydrophilic membrane, rejection rate of soluble EPS being the same. Even if the initial flux of the hydrophobic membrane was higher than the hydrophilic one, the final fluxes were almost equal for all three membranes [Mukai *et al.*, 1999]. Moreover, Ma *et al.* (2000) suggested that hydrophobic interactions are stronger than hydrophilic interactions, and thus supports the previous result. For some authors, the ratio proteins/polysaccharides is considered as more important than the total amount of EPS for the attachment of flocs and solutes on the membrane surface [Mukai *et al.*, 1999, Lee *et al.*, 2003]. Furthermore in the case of hydrophobic membranes, at the starting of a filtration step preferential hydrophobic adsorption leads to an irreversible attachment of sludge particles and solutes on the membrane, with a high protein/carbohydrate ratio. So the membrane is getting more and more hydrophobic with time and fouling is getting more and more significant too [Lei and Zhou, 2005]. Furthermore, Yamato *et al.* (2006) compared two hydrophobic polymeric membranes, made of polyethylene (PE) and polyvinylidene fluoride (PVDF) during filtration of the same wastewater. It was shown that PVDF is superior to PE in term of prevention of irreversible fouling in MBRs: most of the fouling observed for the PE membrane was irreversible while reversible fouling was dominant for the PVDF membrane. Moreover the origin of fouling matters was shown to be different for the two membranes: t dissolved matter causes irreversible fouling of PE membrane and carbohydrates induce reversible fouling of the PVDF membrane [Yamato *et al.*, 2006].

Membrane electric charge can also be considered as influencing membrane performances. Ma *et al.* (2000) operated filtration of an *E coli* suspension with neutral and charged membranes. It was found that the neutral membrane surfaces were most effective in reducing *E. coli* fouling. This result is not surprising because *E. coli* has both negatively and positively charged surface groups. So that, charged membrane surfaces develop stronger interactions with charged feed than neutral membrane surfaces.

Nevertheless, it seems that influences of membrane surfaces properties and thus of membrane material are important mainly in the case of short filtration duration [Ma *et al.*, 2000]. Actually, Ma *et al.* (2000) reported that after 1 hour of filtration, membranes are sufficiently fouled so that deposit controls the filtration behaviour and in this way, membrane properties have less influence over longer durations.

Finally, it is necessary to remind that micro-organisms have the ability to biodegrade cellulose acetate (CA) membranes for that reason, CA membranes are no more used in MBRs.

As a conclusion, membrane material (hydrophily/hydrophoby, surface charge) has a strong influence on fouling in MBRs and on its reversibility. It mainly influences the first filtration instants.

B.2 Impact of permeation flux

The permeation flux is a primordial parameter for the MBR process optimisation [Gui *et al.*, 2002]. In a MBR, at the time of the filtration, particles, colloidal compounds and a part of soluble compounds are retained by both membrane and filtration cake. During the filtration operation a material deposit onto the membrane takes place and causes a fouling which increases with increasing the permeation flux [Yang *et al.* (2006)].Thus, higher the permeation flux, higher the material deposit and so, higher the membrane fouling, the others operating parameters being constant.
Moreover, according to the Darcy's law, increasing the permeation flux leads to increase the TMP and hence due to the cake compressibility, to reduce the material deposit porosity and permeability reducing the filtration performance.

The concept of critical flux appeared firstly in 1995 and was introduced by Field et al. (1995), it is defined for microfiltration like flux below which a decline of permeability with times does not occur and above which fouling is observed. It is widely recommended to use a permeation flux below the critical flux in the view to reduce the membrane fouling.
However, even with sub-critical flux filtration conditions, membrane fouling occurs due to irreversible adsorption of mixed liquor soluble compounds and a decrease of TMP is observed after some critical filtration period [Pollice *et al.*, 2005].

Actually the value of the critical flux is strongly dependant of operating conditions such as suspension properties, membrane properties and hydrodynamic conditions [Pollice *et al.*, 2005, Bacchin (2004)]. So, a value of critical flux is related to process configuration, plant size and nature of influent. These parameters impact upon the shear stress and the foulant potential of mixed liquor, and thus, influence the value of critical flux.

In conclusion, in the literature data no available study makes the comparison between different permeation fluxes in long term filtration conditions. Nevertheless, in short term conditions, it is generally reported that higher the filtration flux, higher the membrane fouling. So that, filtration conditions have to be adjusted in order to minimize cleaning frequency or to maximize permeate flux. Thus the total cost will determinate the optimal strategy.

B3. Comportment during long term operation

A strategy for reducing membrane fouling is to filter the biological suspension at low fluxes, below the critical flux. Nevertheless, even if operation with low permeation flux reduces membrane fouling, a lost of process productivity with filtration time inevitably occurs.

According to numerous authors under sub-critical and long term filtration conditions, TMP time-variation shows two successive steps: a fist one where the increase in TMP remains weak and a second one in which a sudden jump in TMP occurs. As an example, these two steps are visible on the figure 12, and Table 5 shows the TMP variation for each step and the total MBR production.

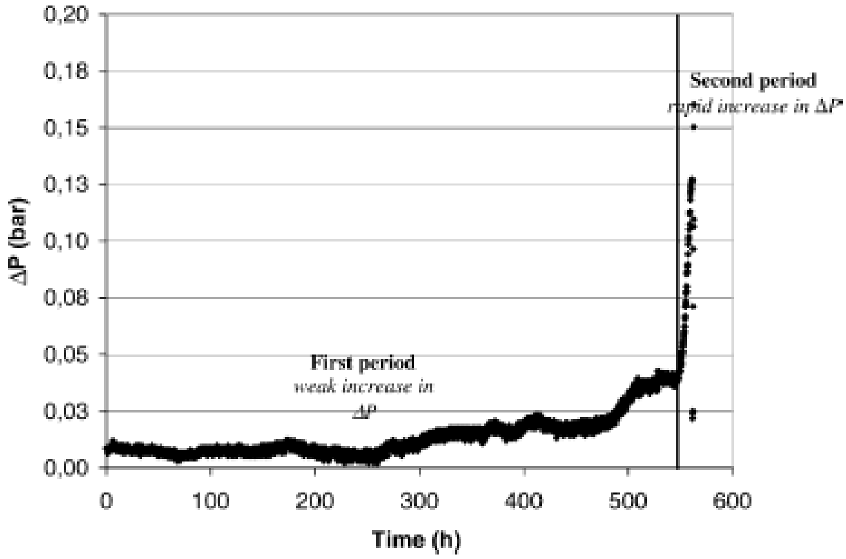

Figure 12 : Transmembrane pressure time-variation during long-term constant flux filtration in a MBR for stabilised biological conditions (Ognier et al., 2004)

Table 5: TMP variation for different MBRs operated at constant flux

Authors	Operation conditions	Total production (m^3/m^2)	First period Time (h)	First period d(PTM)/dt (kPa/h)	Second period Time (h)	Second period d(TMP)/dt (kPa/h)
Massé (2005)	Submerged hollow fibres Polysulphone Pore size 0.2 µm C_V = 0.59 – 0.54 $kg_{COD}/m^3/d$ J = 5 L/h/m²	2.88 – 12.12	576	0.04	2184	0.003
Orantes et al. (2005)	Capillary membranes Polysulphone Pore size 0.1 µm C_V = 0.4 – 0.8 $kg_{COD}/m^3/d$ J = 2.3 – 4.6 $L/h/m^2$	2.1 - 4.2	800	0.0002	108	0.27
Orantes et al. (2005)	Capillary membranes Polysulphone Pore size 0.1 µm C_V = 1 $kg_{COD}/m^3/d$ J = 2.3 – 4.6 $L/h/m^2$	20 - 28	6 000	0.00001	230	0.1
Ognier et al. (2004)	Tubular membrane Alumina Pore size = 0.05 µm C_V = 3 $kg_{COD}/m^3/d$ J = 10 $L/h/m^2$	5.8	550	0.036	30	1.08
Yu *et al.*, (2003)	Hollow fiber membrane Polyvinylidene fluoride Pore size = 0.22 µm C_V = 0.24 $kg_{COD}/m^3/d$ J = 23.4 $L/h/m^2$	7.3	300	0.015	12	2.88
Cho *et al.* (2002)	Plate membrane Polyvinylidene fluoride Pore size = 0.22 µm C_V = 6.5 (4-12) $kg_{COD}/m^3/d$ J = 30 $L/h/m^2$	12.75	360	0.05	65	0.25

In the literature, different and complementary explanations are proposed for this phenomenon. Two assumptions are presented here:

- According to Ognier *et al.* (2004), interactions between solute compounds and membrane provoke a low fouling and cause a reduction of membrane pores number. Thereby, this phenomenon alters the membrane characteristics in regard with filtration performances. So, despite a global constant flux, there is an increase of the local flux because of the filtration area decreases due to irreversible membrane fouling by EPS. The local flux rises over time leading an increase in the TMP too. After a critical filtration period time the local flux reaches the critical flux provoking a rapid deposit of suspended matters onto the membrane. The second step begins since local fluxes in few areas of membrane exceed critical flux value. So, even if the global permeation flux value is inferior to critical flux, a sudden TMP jump, about 1 Pa/s for the work of Ognier et al. (2004), is observed after a critical period. This step is characterised by a high hydraulic resistance which, according to Ognier *et al.* (2004), is due to a strong deposition of suspended matter on the membrane wall.

- Massé (2004) proposed to explain the TMP jump by the accumulation of soluble EPS within the biological suspension and by the sensitivity to pressure of cake containing proteins. Indeed, EPS and mainly proteins are retained and accumulated on the membrane surface and can modify the cake structuration or create a compressible gel layer on/inside the cake increasing the compressibility of the cake layer. Thus proteins accumulation could lead to a higher compressibility of the foulant deposit. This mechanism was also considered by Shiau *et al.* (2004). Massé (2004) suggested that fouling mechanisms for the two observed steps are different. The first TMP increase may be due to EPS adsorption, colloidal particles and flocs deposit on the membrane surface and also in the membrane pores for the colloidal particles and as seen previously the second step may be due to a higher compressibility of the foulant layer involved by proteins adsorption.

To resume, under sub-critical conditions and for long term filtration the observed membrane fouling behaviour (TMP or flux increase following two steps) is commonly the same in the literature data. Nevertheless the fouling mechanism understanding, and mainly the reason why the TMP jump is starting, is not yet elucidated.

A complementary approach to understand fouling mechanisms during these two steps is based on membrane observations using destructive methods. Two authors directly observed the membrane after use and autopsy. Cho *et al.* (2002) observed fouled membranes using a SEM (Scanning Electron Microscope) and showed that during the first step of weak TMP increase, EPS were the main compounds of the foulant layer. Li *et al.* (2003) proceeded to a direct observation of particles deposit on the membrane by DOM (Direct Observation Through the Microscope), they observed that actually a particles accumulation occurred already in sub-critical conditions. That is confirmed by Orantes *et al.* (2004), who pointed out a large biomass deposit throughout capillary fibre network without any drastic TMP evolution [Orantes *et al.*, 2004].

In a general manner, long term filtration behaviour in sub-critical conditions is quite unknown. So far, even if permeation flux is fixed under critical flux, no available models can predict the duration of this steady-state critical time period [Massé, 2005, Le Clech *et al.*, 2003] nor the time at which the drastic TMP increase will appear. Moreover, Le Clech et al. (2003) claims that, although a MBR is operated at sufficient low flux, no sustained operation is possible indefinitively.

B4. Air sparging during filtration (case of aerobic MBR)

In MBR processes, the use of bubbling to control fouling is of paramount importance. Two complementary strategies may be involved: air sparging to prevent membrane fouling during filtration runs and/or air sparging to remove fouling (sequential filtration/gas sparging steps). In case of aerobic MBRs, the air-liquid flow used against fouling is different from the one used for mixed liquor aeration and oxygen mass transfer.

Even if most of MBRs in operation at industrial scale are using air inputs for fouling control, optimisation of air sparging is not a solved problem due to the lack of understanding on mechanisms induced by bubbles (Martinelli et al., 2006). Moreover air sparging or aeration is a key problem for the process as it represents one of the main operating costs. The major reason for this misunderstanding is that each module configuration and membrane systems (which are in constant evolution) presents specific flow pattern and thus transfer phenomena.

In this context, this paragraph will successively summarise operating data for aeration of large scale MBRs, hydrodynamics characteristics for different modules and effect of aeration parameters on filtration performances.

B4.1 Operating conditions for aeration in large MBR

Some examples of operating conditions for MBRS operated with air sparging are given in Table 6.

Table 6: Aerations conditions for different full-scale MBRs
[adapted from D1, Churchouse (1997), [b] Günder et al. (1998)]

Membranes	System Capacity	Flux	Aeration Conditions
	(m3/day)	($L.m^{-2}.h^{-1}$)	($m^3.m^{-2}.h^{-1}$)
Flat Sheet			
Kubota *	1.9 - 13	20 - 33	0.56 – 1.06
Brightwater *	1.2	27	1.28
Toray *	0.53 – 1.1	21.6 – 25	0.4 - 0.54
Huber *	0.11	24	0.35
Colloide *	0.29	25	0.5
Submerged plate 0.2-0.45µm (240m²) [a]	100 (sewage)		0.92
Hollow Fibre			
Zenon *	48 - 50	18 – 25	0.29 – 0.4
M. Rayon *	0.38	10	0.65
USF Memcor *	0.61	16	0.18
Asahi-kasei *	0.9	16	0.24
KMS Puron *	0.63	25	0.25
Submerged HF (0.2 µm) [b]	24 (product)	13.3	0.94

The following observations can derive from this table:

- Whatever the MBR configuration or membrane geometry, the permeate flux in large systems is today always in the same range: between 10 to 33 $L.h^{-1}.m^{-2}$, and mostly between 20 to 27.
- The air flow rate, expressed here as a ratio to membrane area, varies between 0.24 to 1.28 $m^3.m^{-2}.h^{-1}$ (so with a factor of 4) without clear relationship between permeate flux and air flow rate.
- The aeration conditions are similar for the two membrane geometry (HF and FS).Till and Malia (2001) claimed that Kubota system are operated with a minimum air requirement of 0.75 m3.m-2.h-1, which is in the range of the values introduced in table .

Moreover, when the plant capacity is increasing, the aeration flow rate per membrane area is not increasing.

We have to point out here, that very often on industrial plants the parameter available on aeration is the ratio air flow rate/membrane area. It is easy to calculate but it is probably not the pertinent parameter to characterise the flow pattern in the membrane system and thus its efficiency to prevent fouling. Other possible parameters will be introduced in the following paragraph.

B4.2 Two phase flow pattern in MBR

The description of aeration in MBR has to raise again what is known on air-liquide flow in a tank or a column without membrane and particles. The different parameters commonly used are listed here:

- the superficial liquid velocity Ug_s defined as $U_{g_s} = \frac{Q_g}{S}$ [m.s-1]
- the superficial liquid velocity Ul_s defined as $U_{l_s} = \frac{Q_l}{S}$ [m.s-1]

where Qg and Ql are the gas and liquid flow rate respectively, and S the section area offered to the flow, (=total area – area occupied by membranes) .

Superficial velocities are calculated as if each phase was circulating discretely in the free section.

- the mixture velocity Um defined as $U_m = U_l + U_g$ [m.s-1]
- the air-injection factor ε defined as $\varepsilon = \frac{U_{g_s}}{U_{g_s} + U_{l_s}}$ [dimensionless]
- the mixture Reynolds number $\mathrm{Re}_m = \frac{D_h \cdot U_m \cdot \rho}{\mu_{(20°C)}}$ [dimensionless]

where Dh [m] is the hydraulic diameter, defined as $\mathrm{D_h} = \frac{4 \cdot \mathrm{S_W}}{\mathrm{L_W}}$, where Lw [m] is the wetted length, Sw [m²] the wetted surface, $\mu_{(20°C)}$ [Pa.s] the liquid dynamic viscosity at liquid temperature and ρ [kg.m-3] the density of the sewage.

Capillary phenomena may have an important effect on flow characteristics. It is thus important to distinguish aeration in confined volumes (in carters…) and in non-confined volumes (fibres inside big tanks…for example).

In confined volumes, different two-phases flow patterns can be observed (figure 13) and can be classified according to the value of ε :

- bubble flow, ε < 0.2 : air bubbles are dispersed in the liquid phase
- slug flow, 0.2 < ε < 0.9 : flow comprises alternate gas slugs and liquid slugs
- annular flow, ε > 0.9 : a continuous gaseous phase is flowing in the centre of tube or channel

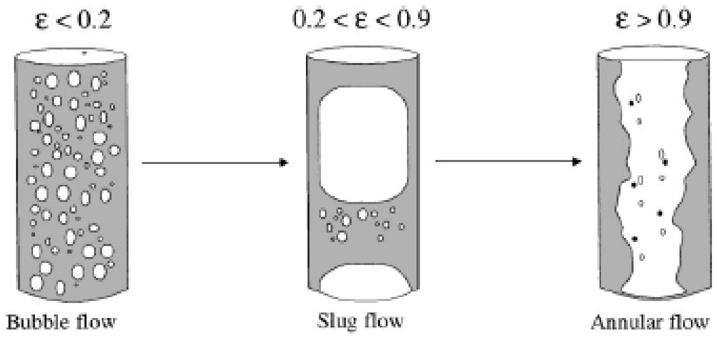

Figure 13 : Gas-liquid two-phase flow patterns in confined volumes

However, as shown is Figure 14, when the liquid volume is no more confined, spherical caps may occur. In this last configuration, there is a lack of knowledge on flow characterisation.

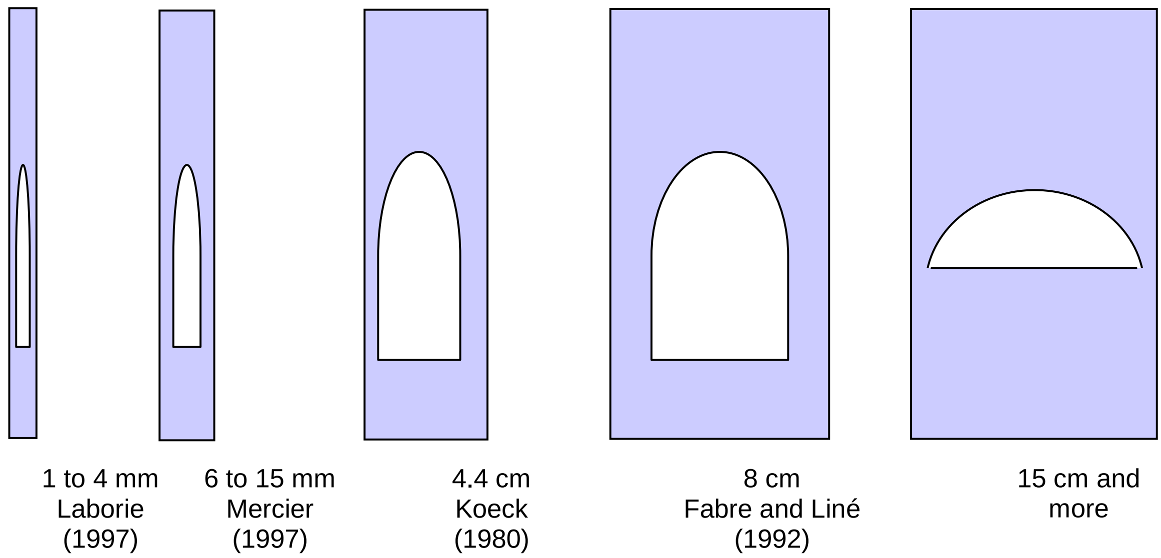

Figure 14: Gas-liquid two-phase flow patterns in confined volumes

As mentioned in D2, membrane modules may be inside the activated sludge tank (submerged) or external to it (side-stream) and membranes may be carterised or not : depending on the MBR design, all kinds of flow patterns described above, may be observed. The lack of knowledge about hydrodynamics in MBR induces a difficulty to correlate filtration performances with the two-phase flow characteristics. One question is still fundamental and was not yet solved: what is the pertinent aeration parameter to control fouling?

Hence, from a process point of view, three parameters are important: the volume of liquid around the membranes (or the degree of confinement), the membrane area and the aeration flow rate. Numerous studies are focusing on this topic but it remains difficult to compare or synthesise results. For instance, concerning the mixture Reynolds number, which was often considered as a pertinent parameter in a confined inside/out filtration [Laborie et al], Espinosa et al [2006] clearly showed that Rem is not a pertinent parameter to characterise outside/in filtration in an other confined system, which is a carterised side-stream system operated at low liquid velocities: indeed experiments performed at an identical Rem but with different liquid and gas velocities lead to different fouling resistances.

Another key point is "what is the good parameter to express the aeration flow in order to have a generic approach of its efficiency and a good capability of extrapolation from lab scale to industrial scale:
- air flow-rate per membrane area?
- air flow rate per permeate flow rate?
- air flow rate per volume of liquid (total liquid volume in the reactor?
- or volume in the bundle?)
- air flow rate per wetted surface ?
- superficial air velocity ?

One of the objectives of the EUROMBRA project in WP3 will be to provide some comparative data in order to conclude on this point.

Table 7 summarises the available range of operating conditions for different MBR con figurations.

reference	Membrane	V reactor	S memb	Ratio S/V	air flow rate related to membrane area	air flow rate related to volume reactor	effluent
		m^3	m^2	m^2/m^3	$m^3/h/m^2{}_{memb}$	$m^3/h/m^3{}_{réac}$	
Benitez et al, (1995)	HF/S/C	0.009	0.300	33.333	0.45	15	wastewater
Bouhabila et al. (1998)	HF/S/NC	0.003	0.016	6.400	9.4 to 53	60 to 340	wastewater
Bouhabila et al, (1998)	HF/S/NC	0.020	0.500	25.000	0.30 to 1.7	7.5 to 42.5	wastewater
Bouhabila (2001)	HF/S/NC	0.020	0.500	25.000	1.2 to 3.6	90	sludge
Choi et al (2003)		0.070	0.400	5.714	11	60	raw water
Delgado (2002)	HF	0.170	0.930	5.471	1.9	11	synthetic
Guibert et al (2002)	HF/S/C	21.900	46.500	2.123	2.3	6.2	synthetic (bentonite)
Günder (1998)	HF/S	9.000	80.000	8.889	60	533	wastewater
Ozaki and Yamamoto (2001)	FS/S/C	0.12	0.3	2.6	$0.010-0.060\ m.s^{-1}$		activated sludge
Hong (2002)	HF/S/NC	0.018	0.300	16.667	1.2	20	synthetic
Ji (2006)	HF/S	0.009	0.150	16.667	0.27 – 0.80	4.4 – 13	synthetic
Madec (2000)	HF/S/NC	1.200	42.000	35.000	0.095	3.3	synthetic (bentonite)
Shimizu (1996)	HF/S/NC	0.450	8.000	17.778	0.53	9.3	domestic
Ueda et al.. (1997)	HF/S/NC	21.400	4.000	0.187	0 - 18	3.4	wastewater
Massé (2004)	HF/S/NC	0.016	0.2	12.5	0.1	1.3	
Espinosa (2005)	HF/E/C	0.0027	1.2	446	0.33	0 - 150	Synthetic (bentonite)
Lee et al. (2003)	HF/S/NC	0.007	0.1	14.3	2.5	34	Synthetic wastewater
Visvanathan (1997)	HF/S	0.080	1.000	12.500	20.10^{-5}	25.10^{-4}	domestic
Ognier (2004)	T/E/NC	0.016	0.200	12.66	Anoxic	-	synthetic (ethanol)
Orantes (2004)	HF/S/NC	0.050	0.250	5	9.6-19	48-96	synthetic (acetate)
Provenzi (2004)	HF/S/NC	0.030	0.080	2.667	15	40	synthetic (ethanol)
Bodzek (1996)	T/E/C	0.025	0.050	2.000	7.8	15.6	domestic

B4.3 Effect of aeration on filtration performances

For each MBR configuration, it is important to evaluate mechanisms which are really responsible for the enhancement of membrane performances. Up to now, different phenomena are reported as a possible way to induce modification of filtration performances:

- Turbulence or secondary flows in the bulk suspension,
- Modification of stresses on membrane surface and/or local pressure field,
- Membrane motion and shaking induced by air sparging (in case of outside-in hollow fibres).

The effects of air sparging have to be discussed in relation with the different membrane geometry: tubular, hollow fibres and flat sheet membranes.

B4.3.1 Tubular membranes

Many studies reported an increase of permeation flux for tubular membrane filtration [Bouhabila *et al.*, 2001, Dufresne *et al.*, 1997, Ueda *et al.*, 1996, Li et al. (1997), Vera et al. (2000), Cui and Wright (1994)(1996), Mercier-Bonin et al. (2000)]. As instance, the critical flux was increased by 50% with a very low air velocity (0.02 m/s) for microfiltration of synthetic waste water and real sewages. With the same effluent (Real sewage at 3 g/l), the limiting flux was increased by a factor two (Le Clech et al. (2005)). In filtration of dextran and bovine protein (BSA), Cui et al. (1996) have observed an improvement of permeate flux up to 320% with a gas-liquid flow (Ul: 0.31-1.57 m/s, Ug=0-0.35 m/s, ε=0.011-0.71).

Optimisation of aeration conditions

The effect of aeration on filtration improvement is higher for higher MLSS concentration in raw water (Cui and Wright (1994, 1996)). The same result has been observed by Abdel-Ghani (2000) for filtration of surfactants. In the same way, a higher effect of aeration at higher transmembrane pressure was observed. These two results have been explained by the authors by the reduction of the polarisation layer induced by air and liquid slug: the impact of aeration is all the more evident when membrane fouling is high.

About the optimal air flow rate, results may be more hazy. Some studies reported an increase of the permeation flux with an increase of the air velocity (figure 15) and other one reported a more significant effect of air for moderate liquid flows, low transmembrane pressures and a high proportions of gas (Vera et al. (2000)). This tendency could be explained by the difference of cross flow velocities (CFV) used in these studies. The range of CFV corresponds to liquid velocities around 0.25-0.55 m/s for Le Clech et al. (2005) and 0.5 – 3 m/s for Vera et al. (2000).

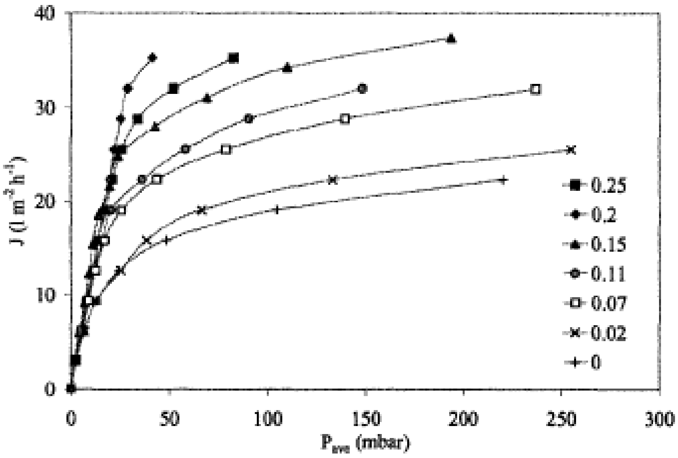

Figure 14: J vs TMP at various U_G values (m.s^{-1}) in a submerged MBR with real sewage (MLSS : 3g/L) [Le Clech et al., 2005]

About optimal aeration characteristics, Li et al. (1997) studied the influence of air bubble frequency and size (bubbles volume from 2.2 to 8.3 mL and frequency from 0.05 to 1 s-1) in 12.7 mm tubes for HAS et β IgG proteins solutions. An increase of the bubble volume induced an improvement of permeate flux but there was a limiting bubble volume size above which no more increase of permeation flux can be obtained. Same kinds of results have been obtained about the influence of the bubble frequency.

Possible explanations are based on different phenomena: (i)an increase of shear stress on the membrane surface, (ii)intermittence of air and liquid slugs which inverts the direction of the stress and (iii)pressures variations in the slipstream of slugs, with an increasing in local turbulence (Mercier-Bonin et al. 2000a).

Le Clech et al. (2005) compared sidestream and submerged configurations for a tubular membrane geometry. Experiments showed similar fouling behaviour observed for the submerged system operated at Ug= 0.07-0.11 $m.s^{-1}$ and the side-stream system operated with a liquid velocity in b the range of 0.25 to 0.44 m.s-1. Analysis of flux step experiments also showed that the effect of Ug in the submerged configuration is greater than this of CFV in the side-stream system. Moreover the effects of Ug was shown to be greater for higher permeate fluxes. Effects of air velocity are shown in figure 14.

To conclude, optimisation of aeration conditions for tubular membrane remains under way.

B4.3.2 Flat sheet membrane

The influence of air was mainly tested at lab-scale, with different suspensions (model suspensions or sludges). Most authors have observed a good efficiency of air sparging in a flat geometry.

Evidence of Flux enhancement due to aeration

For baker yeast and bentonite suspension, Mercier-Bonin et al. (2000) found a flux improvement from 70 to 200 % for ultrafiltration and from 0 to 280 % for microfiltration ceramic membranes (Tami, Ltd.) with air sparging on an external system.
Lee et al. (1993) have ultrafiltered micro-organisms (Escherichia coli) at 0.4 bar. They found a 520% flux improvement with a 4.2 L/h air flow rate of (velocity are not available in the paper) and a 11.4 L/h liquid flow rate ($\varepsilon = 0.27$) with a micro-organisms concentration of 2.1 %, in comparison with a higher liquid velocity (19.8 L/h) without aeration.

Ducom et al. (2002) found, in an external carterised module, that whatever the superficial liquid velocity, for particle suspensions air sparging allows a significant increase of the permeate flux, which confirms that the two-phase flow prevents particle deposit. Indeed, for bentonite filtration, the permeate flux increased by 70% for the lower liquid velocity and the higher gas velocity, which was 1 $m.s^{-1}$ (Ducom (2001)).

For sludge suspensions, Ozaki and Yamamoto (2001) have observed an important decrease of the sludge accumulation with high aeration velocity (0.049 m/s) for flat sheet membranes (Nitto Denko Co., Ltd.) with a channel width of 1 cm. Sludge accumulation is divided by 3 compared to an aeration velocity of 0.031 $m.s^{-1}$ (Figure 15).

Influence of module design on flux enhancement

Ozaki and Yamamoto (2001) studied, with a flat sheet module immersed directly in the bioreactor, the dependency of sludge accumulation on aeration intensity for different flow channel widths. They concluded that sludge accumulation and filtration resistance are dependent on aeration intensity, and are less dependent on flow channel width and MLSS concentration (figure 15).

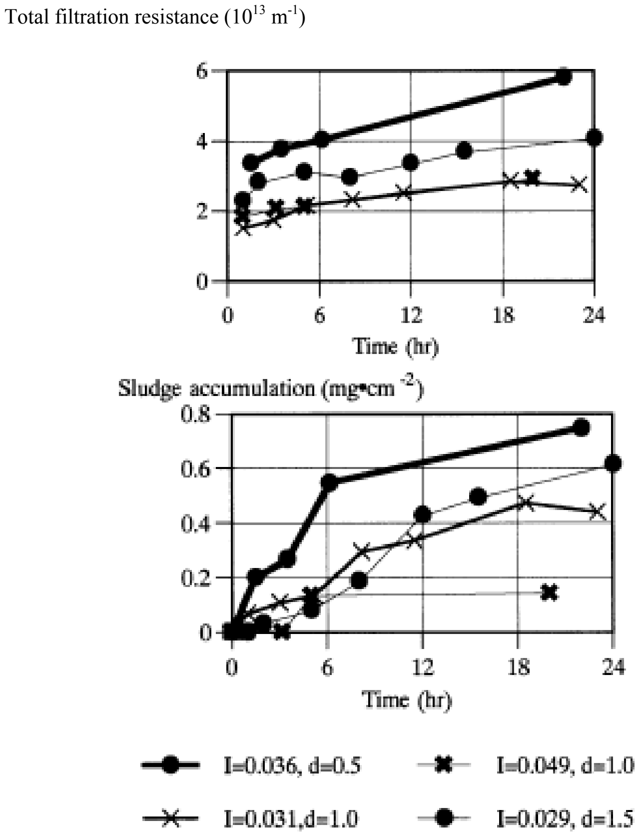

Figure 15: Variation of total filtration resistance and sludge accumulation for different aeration intensities (I in m.s^{-1}) and flow channel width (d in cm) [from Ozaki and Yamamoto, 2001]

Many studies explained the benefic effect of aeration for flat sheet membranes by the increase of the shear stress (Ozaki and Yamamoto (2001), Lee *et al.* (1993), Mercier-Bonin et al. (2000), Ducom et al. (2002)).

A comparison of two air diffuser systems was performed by Sofia et al. (2004), comparing a 200mm long x 10mm wide coarse bubble diffuser with six holes at 20mm apart and each hole had a diameter of 2mm, and a fine bubble diffuser of porous media with pores of 0.5mm approximate diameter. They measured crossflow velocity at the centre of the flat sheet membrane unit. They found higher crossflow velocity values with fine air bubbling. Nevertheless, after a certain aeration intensity (0.017 m^3.m-^{2}.s^{-1}), crossflow velocity reached a plateau level (0.69 m/s). They conclude that it is not cost-effective to operate the reactor at aeration intensity above 0.017 m^3.m-^{2}.s^{-1}.

In Kubota systems, Howell et al. (2004) have observed that cake fouling rates (from simulated settled sewage water) and residual fouling are dependant on gas flow rate but the relation ship is strongly dependant on the permeation flux.

B4.3.3 inside/out hollow-fibre membrane

This configuration exists but cannot be used in MBRs system. The principal application today is for drinking water. We will just here give some tendencies that could give some elements of comparison with the other geometries, as the flow inside hollow fibres can sometimes be very close to a flow outside hollow fibres in a carterised system.

Effects of air injection on particular fouling were identified by Laborie (1998). The characteristics of the deposit are modified in presence of gas-liquid flow: its porosity and thickness are increasing and its specific resistance is decreasing when the air flow grows. For this kind of membranes, a slug flow was observed for small values of ε ($\varepsilon \geq 0.1$) (Laborie, 1998) and enhancement of flux permeate was from 20 to 200% for clay filtration.
An experimental measurement of sheer stresses on the surface of hollow fibre tubes membrane was used to correlate the flux enhancement with the hydrodynamics parameters of the two-phase flow. This enhancement seems to be linked to mixing and turbulence generated by air bubbles in the liquid phase, rather than the intermittency of shear, even if under some conditions, it seems to play a role in the control of the deposit.

B4.3.4 outside/in hollow-fibre membrane

Evidence of flux enhancement by aeration
In these configurations, air sparging always allows an increase of filtration performances (Cui *et al.*, 2003, Chang et Fane, 2002). Benefits of bubbling are clearly shown when comparing deposit resistances with and without bubbling, as shown in figure 16 for a yeast suspension. The benefit of bubbling is clearly evident as permeate flux is increased 3-6 fold by a modest gas flow. For Espinosa et al. (2003), the operating transmembrane pressure in wastewater treatment, could be reduced up to 3 times with the use of air sparging.

Bubbling can be considered as a way to reduce the magnitude of readily removable resistance (Rr) and the 'irreversible' resistance (Rir), which represents fouling by fine colloids and debris in the feed – bakers yeast (Cui *et al.* (2003).

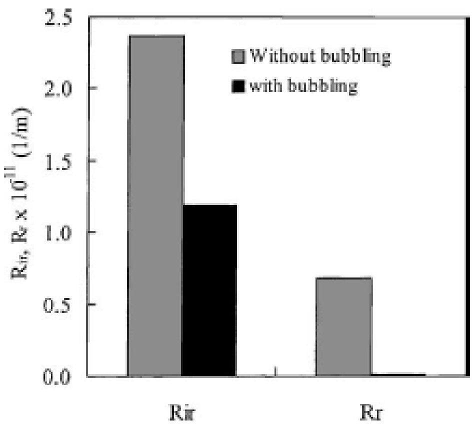

Figure 16 : Comparison of filtration resistance after 90 minutes filtration (Chang and Fane (2000).

A 5 g/L clay suspension was filtered in a Zenon module ZW500 (filtration area of 42 m²) (Madec, 2000). Critical flux was increased from 32 to 80 L/h/m² when air flow rate was increased from 8 to 42 m^3/h.

Optimisation of aeration conditions

Most studies have pointed out the positive influence of air sparging below a limit value of gas velocity and the existence of a limit aeration flow-rate beyond which a further increase has no effect on fouling removal [Ueda *et al.* (1997), Wicaksana *et al.* (2005), Espinosa et al. (2003), Chang and Fane (2001)]. For wastewater treatment Wicaksana *et al.* (2005) have determined an optimal air flow value of 5 $m^3_{air}.h^{-1}.m^3_{liquid}$. Espinosa et al observed a limit gas flow rate value of 72 $m^3_{air}.h^{-1}.m^3_{liquid}$, beyond which there is no more benefit to continue increasing the air flow rate for the filtration of clay suspensions.

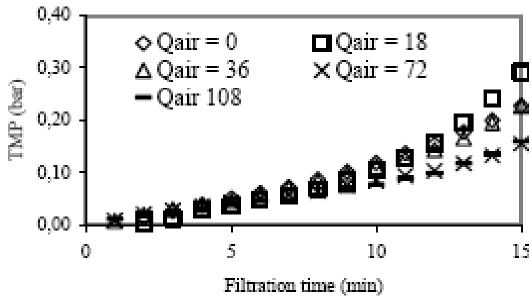

Figure 17 : TMp time variation for different air-flow rates (m^3 air $.h^{-1}.m^{-3}$ liquid) during filtration time with a permeate flux of 7 $L.h^{-1}.m^{-2}$ [from Espinosa et al, 2003]

However, Chang et al. (2006) observed an increased filtration resistance with increasing aeration intensity at constant flux for non-woven membrane tubes with a synthetic wastewater.

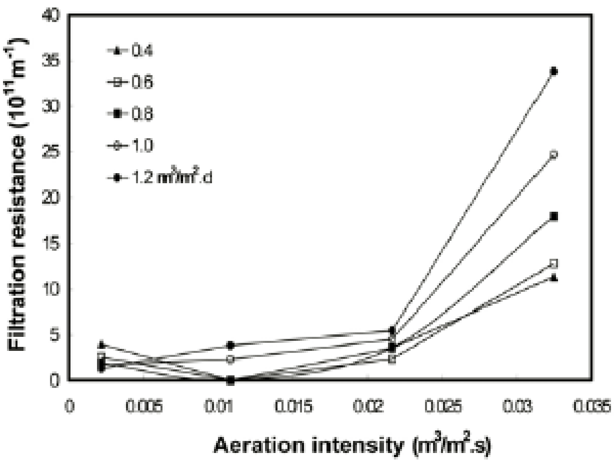

Figure 18 : filtration resistance versus aeration intensity for different initial fluxes (MLSS : 5 g/L) (Chang et al., 2006)

The main explanation for the differences found between different authors in literature lies in the fact that the authors do not operate in the same range of gas velocity and some authors only explore the lower part of the curve, that means below the limit gas velocity value. Moreover two phenomena could also contribute to the explanation, (i) a cross-flow along the membrane surface is created and induced a shear stress generating the backtransport or re-entrainment of floc particles from the membrane surface (Ueda et al., 1997), (ii) another effect of air bubbling, especially at the high aeration intensity case, is the breakage of floc particles. Shear-induced floc breakage can result in the enrichment of smaller size floc particles in the bioreactor (Wisniewski et al., 2000).

For outside/ in membrane, the influence of aeration on filtration performances has to be linked with the module configuration since both the two-phase flow and fibre motion depend on fibre diameter, tightness, length and density (Wicaksana *et al.*, 2005), Delgado *et al.*, 2004).

The last important parameter is the kind of aeration performed in the MBR. Sofia et al. (2004) have compared injection with small bubbles and coalesced bubbles at same superficial velocity: the smallest ones induced best performances. In the contrary, in this study, Madec (2000) has concluded that the size of bubble (in range 1-10 mm) had no effect on membrane performances. This problem of which kind of bubble is more interesting for membrane filtration is remaining even if first quantitative analysis is very promising (Martinelli et al. 2006).

a) Aeration and fibre diameter

Fibre diameter has an influence on fibre motion, permeation flux and distribution of suction pressure. The problem is that longitudinal pressure drop inside the fibres is increasing when fibre diameter (for a same length) is decreasing.

In Chang and Fane (2001) publication, for an air flux of 3.6 m3/h/m²memb and above a 30 L/h/m²permeate flux, suction pressure is increasing more quickly for small fibre diameters (0.65mm) than for big ones (2.7mm). Without aeration, this suction pressure variation is similar for both sizes of fibres. This may be explained by the fibre movement amplitude which is decreasing when the fibre diameter decreases (from 3 to 1cm for 0.65 to 2.7mm diameter, Wicaksana et al. (2005)). As a conclusion, Chang and Fane (2001) indicated a clear advantage for the smaller fibre diameters. Same tendency was obtained from the membranes provided by two different manufacturers (US filter and AKZO hollow fibres, both with a 0.2 µm pore diameter -cf figure 19).

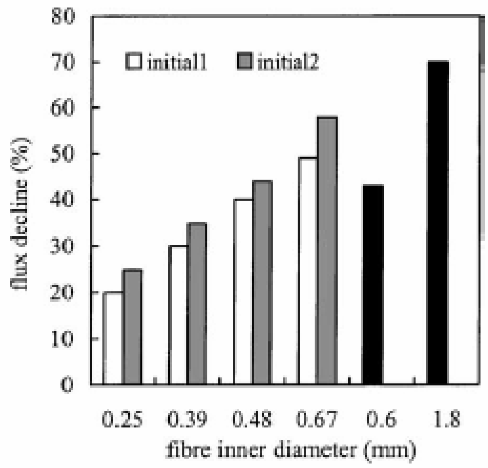

Figure 19 : Effect of fibre diameter on flux decline in filtration with submerged hollow fibres (ug = 0.2 m/s, initial flux 1, range 52-64 L/m²/h, initial flux 2, range 86-89 L/m²/h) – white and grey : US Filter, black : AKZO, (Chang and Fane (2001)).

However, it is important to add that Sridang *et al.* (2004) have compared critical flux values with bentonite (0.2-2 g/L) for different fibre diameters and the critical flux obtained with a big external fibre diameter (2mm) is twice higher than the one obtained for a smaller external fibre diameter (0.72mm).

Influence of fibre diameter is dependant on the fibre material, permeability and of the operating permeate flux.

b) Aeration and fibre Length

Many studies have reported that, when aeration is used, fouling velocity decreases more rapidly during filtration when fibres are shorter because longitudinal pressure loss increases with fibre length (Carroll and Booker (2000)).
Moreover, Fibre length has also an influence on fibre ability to move (Kim et al. (2004)). Wicaksana et al. (2005), have studied the influence of fibre length on their ability to move . They found a relation of proportionality between these two parameters.

c) Aeration and fibre tightness

Fibre tightness of fibres can influence the fibre mechanical resistance but it modifies at first the fibre motion.
The advantage of loose fibres is shown in Figure 20 (Chang and Fane (2002)), which shows suction pressure profiles for a submerged module at an average permeate flux of 30 $L.h^{-1}.m^{-2}$ and a bubbling flow rate of 7 L/min. The loose fibre (with a tightness of 95%) shows a constant suction pressure whereas the tight bundle (fibre length = fixed end spacing so with a tightness of 100%) shows a steady rise in suction pressure.

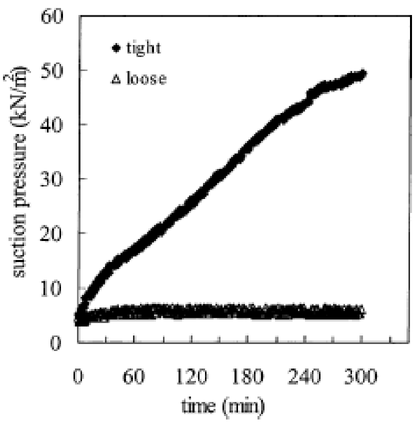

Figure 20 : Suction pressure profiles for tight and loose hollow fibre bundles (5g/L yeast suspension, Jm = 30 L.h-1.m-^{2}, Vg = 7.3 L/min) (Chang and Fane (2002)).

A relation between tightness and fibre length seems to be available. Long and relaxed fibres are stirred uniformly by air bubbles, which could create a high shear stress. For short and tense fibres, fibre motion induced by aeration is not sufficient enough for fouling removal, according to Delgado et al. (2004). Wicaksana *et al.* (2005) found an optimal fibre tightness of 96%, which is the ratio of distance between fixed ends and fibre length.

For a carterised module operated with low liquid velocities, in which membranes are fixed at one end and free at their top, Espinosa (2005) has demonstrated that a condition for a good fouling removal is fibre motion and that air flow rate has to be chosen high enough to obtain this mobility.

In conclusion, it is well known that fibre tightness plays a major role in fouling removal or prevention but associated physical phenomena are not actually well-known. Fot that reason further studies are necessary to understand phenomena involved in fibre motion and in fouling removal.

d) Aeration and fibre orientation

Two main orientations are possible to manufacture for membranes used in MBRs. They can be placed either horizontally or vertically as represented in Figure 21. For the first one orientation, bubbles are going up generating turbulence which reduces particles accumulation and for the second one, bubbles can get entrapped in gas slugs located between some groups of fibres. (Cui *et al.* 2003).

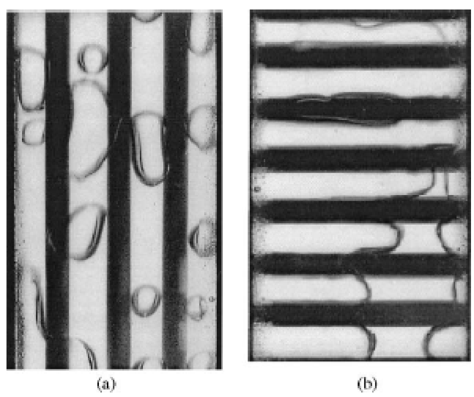

Figure 21 : Characteristics of bubbles with (a) axial and (b) transverse fibre array (Chang and Fane (2000))

Some studies have showed the interest to use vertical fibres. For instance, Chang *et al.* (2002) have shown that flux decrease is less important (from 5 to 15%) for hollow fibres in vertical position than in horizontal position for filtration of yeast with a 0.2 m/s gas velocity. In figure 22, Cui (1993) compares the permeate flux in vertical and horizontal membranes and the flux is clearly more important for the vertical tube.

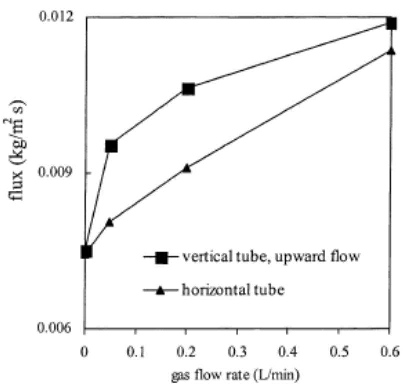

Figure 22 : Effect of tube orientation on gas sparged UF (Cui (1993)).

e) Aeration and fibre density

Only a few of studies have focused on the influence of fibre density or of packing density on aeration efficiency and further studies are necessary.

Kiat *et al.*(1992) showed that fouling becomes critical when fibre density is higher than 20 fibres/cm². Fibre density influences the air bubble size and so local hydrodynamics, which could explain these results.

Moreover the position of aeration injection is important [Yeo et al. (2006)] and more precisely the distance between bubbles and fibres: phenomena induced by aeration are different if bubbles rise across the fibre or if they are not in contact with the module (Martinelli 2006).

B5. Effect of gas sparging on rejection

Gas-liquid flow has an effect on hydrodynamics so it should have an effect on solute rejection. As an example, Cui et al. (1993, 1994) worked with a PVDF tubular membrane (nominal MWCO of 100 kDa).Using gas-liquid flow dextran rejection was increased from 89 to 95 % and from to 80 to 93% for the 162 and 87 kDa dextran respectively.
This means that a gas-liquid flow might impact the retention of other soluble compounds, and today the question is open about the possible effect of air sparging on SMP retention in MBRs.

B.6 Use of gas sparging in anaerobic systems

Some MBRs applications are operated in anaerobic conditions. Even for anaerobic systems, use of gas sparging can be used to enhance permeate flux.

Indeed, Kayawake *et al.*(1991) used biogas to enhance the permeate flux in ceramic tubular membranes in a submerged MBR. The permeate flux was increased from 4.6 to 8.3 L.h-1.m-² for a biogas velocity of 2.10-6 m/s.

For a side-stream system, Imasaka *et al.* (1989) concluded on the better performances when filtration is operated with both the liquid and the gas phase: a higher liquid flux, a lower energetic consumption and a lower membrane fouling are achieved.

B.7 Effect of Air sparging for fouling removal

In the field to drinking water production different examples show how air sparging can be used to remove fouling after a filtration step, that is formally performed without aeration.
Air can be injected periodically during cleaning periods of ultrafiltration membrane, as in the AirFlush process commercialised by Storck (Verberk et al., 2000 ; Futselaar, 2000) or in the process (Laborie, 1998) for drinking water. Aeration can also be used during backwashes to control remaining fouling in dead-end filtration (Remize, 2006).
These processes are recent and aim to minimise energetic consumption (Laborie, 1998) and water consumption (Remize, 2006) during backwashes with water.

In MBRs applications, aeration can also used for fouling removal.

The cake-removing efficiency was improved by intensifying the air flow without increasing the air flow rate (Ueda *et al.*, 1997). Therefore, it was suggested to concentrate membrane modules over a smaller floor area in order to enhance the aeration intensity.
The cake-removing efficiency of aeration does not increase proportionally with the increase of the air flow rate, and an optimal gas flow rate value of about 0.7 $m^3.min^{-1}$ is recommended, on the basis of the observation curve of suction pressure for a permeate flux of 0.37 m/d (figure 23).

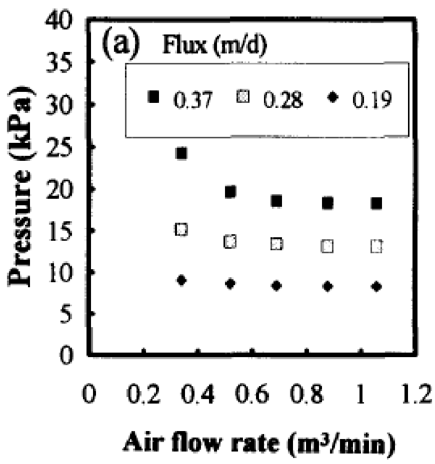

Figure 23 : Suction pressure vs. air flow rate for different permeate fluxes (Ueda et al. (1997)).

Chang and Fane (2002a), using 0.2 µm hollow fibres found that aeration is more efficient to remove a part of particles deposit for fibres with a smaller external diameter. At the end of filtration, air flow rate was directly increased from 0 to 5.4 $m^3_{air}.h^{-1}.m^2_{membrane}$ for two different diameters. As a consequence, suction pressure was decreasing immediately in small fibre diameter (0.65mm). On the contrary, suction pressure was maintained constant for big fibre diameter (2.7mm).

B8. Effect of liquid velocity

For systems with a reciculation of liquid, it is important to have some information about the influence of liquid velocity. This concerns external recycled systems at high velocity as well as those with low liquid velocity induced by recycling or airlitft. However, due to different phenomena, effects are very different for systems oprated at high liquid velocity (high energy demand) and at low liquid velocity (low energy demand).

For instance, a higher permeate flux (100 $Lm^{-2}h^{-1)}$ compared to 25 $Lm^{-2}h^{-1}$, sustained for a longer period (100h compared to 6h) was obtained when operating at cross-flow velocity equal to 4 $m.s^{-1}$, rather than 0.5 $m.s^{-1}$ [Tardieu et al., 1998]. Visual observations of the ceramic membrane used also revealed no floc deposition when the system was operated at high liquid velocity. It was postulated that the deposited cake is formed from the finer particles present in the feed while the coarser particles are preferentially removed by the scouring action of the liquid cross flow.

This shows that the choice of the liquid velocity has to take into account the energy consumption. Indeed, lab-scale studies give the same conclusions, the higher liquid velocity, the higher permeate flux. However energy consumption computed from lab-scale data is often not representative, and fullscale experimentations are necessary. The liquid velocity is reported to range between 0.3 and 0.5 $m.s^{-1}$ for submerged systems, 1 and 5 $m.s^{-1}$ for external membrane systems with high velocities (cf table 8) and 0.01 and 0.1 $m.s^{-1}$ for external membrane systems with low velocities.

Table 8: Hydraulic performances vs. liquid velocity for sidestream and submerged high velocity MBRs [from Chang et al., 2002]

Cross-flow velocity ($m\ s^{-1}$)	TMP (bar)	Flux ($L\ m^{-2}\ h^{-1}$)	Permeability ($L\ m^{-2}\ h^{-1}\ bar^{-1}$)	References
Submerged				
0.5*	0.3	21	70	Ishida et al. (1993)
0.4*	0.15	12	80	Ueda et al. (1996)
0.3–0.5*	0.3	17	57	Shimizu et al. (1996a)
Sidestream				
2.5	2.8	66	24	Krauth and Staad (1994)
1.5–3.5	1	60–80	60–80	Trouve et al. (1994)
2.2	2.2	9	4	Bailey et al. (1994)
2	0.2–0.3	23–68	115–227	Sato and Ishii (1991)
4.7^{α}	1.8	125	70	Ghyoot et al. (1997)
2.9^{β}	0.7	127	181	Ghyoot et al. (1999b)
1.5^{χ}	2.2	8	4	Ghyoot et al. (1999a)
3^{δ}	2	153	77	Ghyoot et al. (1999a)

Note: *Estimated values. $^{\alpha,\beta,\chi,\delta}$Experiments carried out by Ghyoot and co-workers under these conditions (type of membrane—MLSS concentration in $g\ L^{-1}$): $^{\alpha}$ceramic MF-4; $^{\beta}$polymer (PVDF) UF-7; $^{\chi}$polymer (polyethersulfone) UF-4; $^{\delta}$ceramic MF—from 5 to 18.

Espinosa (2006) studied the complementary influence of gas velocity and liquid velocity on fouling resistance in an external system operated with very low liquid velocities. For this system operated at a low liquid velocity, critical fluxes are increasing when the air superficial velocity increases until a limit value of about 0.08 to 0.1 m/s. However critical fluxes are decreasing when the liquid velocity is increasing which can be explained by the sensitivity of air flow patterns to the liquid velocity.
Moreover, the effect of the air is then (in certain conditions) more effective that the effect of the water at the same superficial velocity in the system. Video observations of the gas flow for both operating conditions showed that lower cake resistances were obtained when the gas velocity is higher (at a constant Rem) because the gas is in shape of big slugs that are able to make the fibre move whereas when the air velocity is smaller the bubbles are smaller. Results obtained in term of critical flux and critical transmembrane pressure for different liquid and gas velocities are introduced in figure 24.

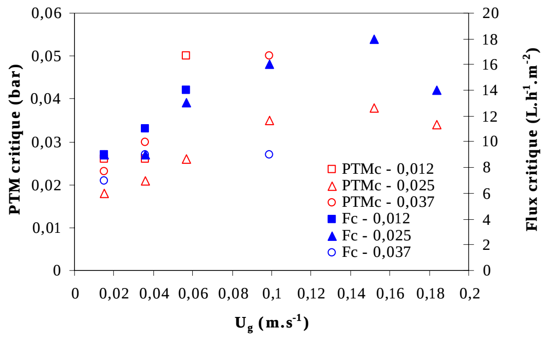

Fig 24. Critical TMP and flux as a function of air superficial velocity and liquid velocity in a side-stream low liquid velocity system [from Espinosa et al, 2006]

B9. Effect of module configuration

This part of the report concerns the effect of module parameters on filtration performances for a given aeration or velocity.

B9.1 Influence of pore size diameter

Experiments have been carried out at Anjou Recherche (Tazi-Pain et al, 2002) in order to investigate the impact of the membrane molecular weight cut-off (MWCO) on the COD removal. Results are presented in for three industrial effluents. Membrane mean pore size has not or very little impact on the COD removal in the MWCO range studied.

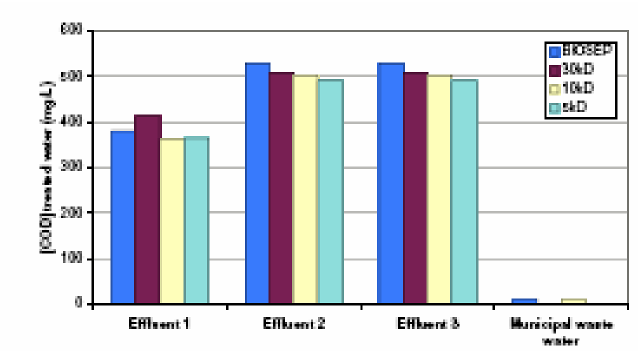

Figure 1 : Impact of the membrane MWCO on COD treated water [Tazi-Pain et al. (2002)]

B9.2 Influence of fibre length

The longer the fibre, the higher the fibre movement but the higher the fibre weakness. Carroll and Booker (2000) have studied the influence of length fibre on permeate flux. They explain that particles accumulation is more important in big flux aeras (where the membrane is more productive). There is a non uniform axial profile so a non uniform profile of fouling. When permeate flux is decreasing, fouling velocity becomes sensitive to the fibre length and profiles of flux are becoming more uniform. Flux decrease is always slower when fibre length is higher. Indeed, a total relative permeate flux decrease of 43 and 14 % respectively for fibres of 0.2 and 0.6 m length.

Kim et al. (2004) have compared initial fouling velocity for different lengths of fibres during filtration of bentonite (100 mg/L). Fouling velocity was found increasing when fibre length is increasing, with or without aeration. The only difference is that fouling velocities are considerably lower with aeration than without.

B10. Effect of Backwash sequences

Backwashes are physical methods which make it possible to control membrane fouling in MBRs. During backwashes water coming from the stored permeate or air flow backward through the membrane and results in a partial removal of deposit matters on the membrane surface. Indeed backwashes are mainly efficient for the removal of the accumulated deposit on the membrane surface which mostly constitutes the reversible fouling, whereas pore blocking resistance is not completely eliminate and particularly in the case of high forward filtration fluxes.

Following a backwashing, membrane regains a part of its permeability, and permeation flux or TMP are partially restored. Nevertheless, an irreversible loss of productivity might be observed with filtration time.

Different backwash procedures for MBRs are presented in table 9.

Table 9 : Examples of different backwash procedures

Backwash duration	Backwash interval	Backward flux or TMP	Forward flux or TMP	Membrane type	References
15 s.	15 min.	96 L/h.m²	48 L/h.m²	Hollow fibre membrane	Smith *et al.* (2005a)
5 – 10 min.	8 s.	80 – 90 KPa	18 – 72 L/h.m²	Inside/outside UF module	Jiang *et al.* (2003)
5 min.	1 h.	20 Kpa	10 kpa	Inside/outside UF module	Katsoufidou *et al.* (2005)
35 s.	10 min.	Not communicated	18 L/h.m²	Hollow fibre membrane	Rosenberger *et al.* (2002)
60 s.	60 min.	100 KPa	10 L/h.m²	Hollow fibre membrane	Hernandez Rojas *et al.* (2005)

In the literature data, the term of "back-pulsing" is often employed. It refers to a cyclic process of forward filtration followed by backward filtration which consists in an extremely rapid pulse (pulse duration is generally less than 1 second). Backpulsing occurs every few seconds throughout the process [Ma *et al.*, 2001], whereas backwash interval periods and backwash duration can last respectively few dozen of minutes and few minutes [Smith *et al.*, 2006a].

For long term filtration, both backwashing and backpulsing have a positive effect in term of TMP increasing or permeation flux conservation [Yang *et al.* 2006]. Ma *et al.* (2000) reported that the long term net flux with backpulsing is approximately twice greater than that without backpulsing. Smith *et al.* (2006a) reported that backwashing during membrane filtration removes most of the reversible foulants leading to reduced TMP increases and permeate flux decline. So that, backwashing appears to be a key for longer filtration time before intensive physical and/or chemical cleaning.

In contrast, according to Ma *et al.* (2000; 2001) the cake layer formed on the membrane surface without backwashing has a positive effect on the filtration control. Actually, it plays a role of secondary membrane and can avoid the penetration of small particles into the membrane and thus can limit internal fouling. Even if baskwashing is effective for reducing the permeability decline with filtration time, it provokes an higher internal membrane fouling and the recovered flux after long term filtration with frequently backwashing is lower than that of membrane fouled without backwashing.

In a general manner, the backwashing frequency, duration and backward flux are important parameters for successful long term operation of filtration. Nevertheless these parameters are impossible exactly to pre-determine in MBR processes, they depend upon many parameters such as the permeate flux, membrane properties and foulant concentration [Smith *et al.*, 2006a, 2006b].

<u>Optimal backwash duration</u>

Optimal backwash duration is one which terminates after the entire removal of the reversible layer. Indeed, too short backwash duration results in the failure of the complete removal of reversible components. Moreover, according to Smith *et al.* (2005) the other problem of such a short duration is that any foulants removed from the membrane surface are not sufficiently propelled from the proximity of the fibre bundle. So that, at the beginning of the following filtration cycle these foulants are very close to the membrane surface and the probability they

immediately deposit onto the membrane surface, thus resulting in a continued TMP increase, is very high.
Otherwise, a too long backwash duration removes effectively the entire reversible layer but in term of permeate production, an unnecessary additional quantity of permeate is used reducing the productivity of the system. This also leads to higher energy consumption [Smith *et al.*, 2006b].

The productivity of the system is represented by the following equation:

$$J_{net} = \frac{J_p T_p - J_B T_B}{(T_P + T_B)A}$$, where:

J_{net} (L/h.m²) is the permeate productivity of the system, J_p is the permeate flow rate, J_B is the backwash flow rate, T_p is the duration of the permeate production cycle, T_B is the duration of the backwash sequence and A is the surface area of the membrane.

Optimal backwash interval

The problem of optimising backwash interval is similar to that of the backwash duration. A too long backwash interval involves an inefficient backwashing in regard with the removal of the reversible layer and a too short backwash interval results in a productivity loss in term of permeate production.
Indeed, a long filtration period induces the development of a thick cake layer on the membrane surface. Besides it leads to the compression of this external layer which becomes increasingly an irreversible foulant layer [Chen *et al.*, 2003]. Although irreversible fouling is unable to be on line controlled, continued control of the reversible foulant layer building up impacts on the degree of irreversible fouling.

Smith *et al.* (2005) reported that for an MBR system with hollow fibre membrane and fed with synthetic wastewater the optimal backwash interval is when the TMP increase is about 3% of the maximum allowable pressure increase.

Backward flux or TMP

In the literature data, values of backward flux or TMP are unequal and do not follow the same trend. Nevertheless most backwashes are operated at higher pressure or flux than forward filtration. Indeed, Ma *et al.* (2001) used a lower TMP for backpulsing than for forward filtration in the view to minimise permeate consumption whereas Smith *et al.* (2005a) worked with a backwash flux which was higher than the permeate flux. In the same way, Katsoufidou *et al.* (2005) worked at a forward pressure of 10 kPa and at a backward pressure of 20 kPa : they found that for backwashing carried out at pressures higher than 34 kPa, membrane compaction occurred leading to a significant membrane flux reduction.

To conclude, backwash sequences improve in a general manner the filtration behaviour in an MBR system. For long term filtration they induce a better flux or TMP conservation. Key parameters for a successful backwash operation are backwash duration, backwash interval and backwash flux. However, for each MBR system these parameters are different and must be adjusted to the system. Indeed, mainly the foulant propensity of the biological solution, the permeate flux and the shear stress along the membrane determine the operational parameters of the backwash operation.

B11. Effect of Relaxation sequences

Application of relaxation sequences during filtration time in an MBR is an other physical method which mitigates membrane fouling. Relaxation sequences are sequences during which filtration is stopped. Hong *et al.* (2002) showed that MBR membrane performance was clearly improved by such an intermittent filtration. Nevertheless, after each relaxation sequence, the membrane permeability is only partially recovered, indicating that irreversible fouling occurs [Hong *et al.*, 2002] or that longer relaxation times would be necessary. Thus, pressure relaxation is only able to remove the reversible foulant layer , reversibility being considered at an acceptable time-scale.

Gui *et al.* (2002) explained that deposition on the membrane surface depends on the balance between the velocity toward membrane surface due to membrane flux and the backtransport velocity induced by shear force. This phenomenon is explained on the following figure, where Vb is the backward transport velocity, Vf is the forward transport velocity and Vs is the cross flow velocity over the membrane surface.

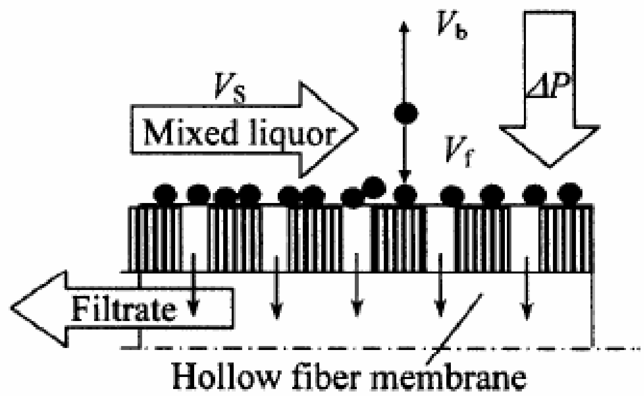

Figure 26 : Membrane filtration process, Gui et al. (2002)

The backward transport of feed particles is mainly related to the cross flow which is strongly dependent on the aeration rate near the membrane and the particle concentration [Gui *et al.*, 2002].

During a filtration operation and when permeation flux is above the critical flux, V_f is higher than V_b and suspended solids accumulate onto the membrane surface, thus constructing the cake layer. Consequently during relaxation time, Vb is the main phenomenon which makes the particles in movement and hence backtransport removes reversible foulants from the membrane to the bulk solution. The backward transport is due to two phenomena:
Concentration gradient in suspended solids between the cake layer and mixed liquor: particles not irreversibly attached to the membrane surface diffused away from the membrane surface thanks to the concentration gradient.

- Air scouring. Indeed Hong *et al.* (2002) reported that in order to maintain a certain steady membrane permeability during the filtration operation of a MBR, the cake layer need to be removed by a shear. In most of MBR systems, it is implemented by an uplifting flow of bubbling air which is supplied by air diffusers located at the bottom of the membrane, this method is called "air scouring" [Hong *et al.*, 2002]. Thereby, the backtransport of reversible foulant during relaxation sequences is strongly enhanced by the air scouring.

Gui *et al.* (2002) showed that the TMP increase rate decreases with an increase of non-suction time. But in the same way, for a backwash sequence, it exists an optimal duration after which pressure relaxation has no effect and constitutes a permeate productivity loss. This optimal time occurs after the complete removal of reversible foulant layer [Hong *et al.*, 2002].
In an other side, the longer the filtration time, the more difficult the cake layer removal. Long periods of filtration time are likely to compress the cake layer and hence to form irreversible deposit which accelerates fouling of MBR [Li *et al.*, 2003]. This phenomenon is quite the same that this which occurs during backwash intervals. Hong *et al.* (2002) found, in the case of an MBR fed with synthetic wastewater, that with a 15 min non suction time a limiting suction time of 145 min and when the suction time is prolonged beyond this value the permeate flux was not recovered.

Compared to backwash sequences, relaxation sequences do not involve permeate consumption and hence its use reduces the productivity loss in regard with permeate production. Moreover, relaxation sequences do not require much energy consumption. Nevertheless not enough available works exist which do the comparison, regarding MBR productivity, between relaxation sequence and backwashing in the same MBR system. Thus it appears important to compare productivity and total operation cost for both MBR systems functioning with either backwashing or relaxation.

C. Influence of biological fluid on filtration performances

C1. Nature and composition of influent water

In the literature data, with the aim of studying wastewater treatment, numerous types of MBR influents can be found. In many case real wastewaters are used to feed MBR. Otherwise synthetic substrates are employed so that influent composition is entirely known in order to improve the degradation mechanisms understanding. For instance, Le Clech *et al.* (2003) reconstituted a synthetic wastewater with peptone, meat extract, urea and mineral salts; Lobos *et al.* (2004) worked with a complex substrate composed of acetate and meat juice and Orantes (2005) used an easily biodegradable influent: acetate and mineral salts.

Wastewaters present relative stability characteristics. Actually numerous studies have shown that the chemical composition of wastewater does not change significantly over time, nevertheless at the same time water flows and concentrations fluctuate [Sophonsiri *et al.*, 2004]. A literature survey done by Sophonsiri *et al.* (2004) compares total COD concentration in numerous wastewater after primary sedimentation. It ranges from 300 mg/L to 900 mg/L and the organic matter was evenly distributed in all size fractions.

Both nature and composition of the MBR influent influence the filtration behaviour. Indeed, Pollice *et al.* (2005) showed that the fouling rate of a MBR system varied with the feedwater matrix.

An important parameter in MBR feedwater characteristics is their part of slowly and easily biodegradable organic fractions. Actually, in regard with EPS are the main foulants substances in the biological bulk [Le Clech *et al.*, 2003], higher the influent part of easily biodegradable organic matter, higher the EPS concentration of activated sludge and hence higher the sludge fouling propensity. Indeed, biomass growth linked to the substrate consumption leads to the synthesis of EPS [Massé et al., 2006].

Pellegrin *et al.* (2002) studied the respirometric needs of heterotrophic populations developed in a MBR and found that the oxygen needs depend directly on the nature of substrate. They showed that exogenous respiration coefficient for the wastewater assimilation is greater than that corresponding to the acetate. The fact that the wastewater contains wider sources of easily degradable carbon in comparison with the acetate induces a greater number of active cells and therefore greater energetic needs to maintain their activities. Thus comparatively to the degradation of acetate, during to the degradation of the wastewater an important part of substrate is catabolised – which corresponds to the energy production - and this phenomenon predominates on the anabolism which produced cells and reserve materials and so EPS synthesis.
These results are supported by both studies of Pollice *et al.* (2005) and Le Clech *et al.* (2003) which showed that MBR sludge fed with synthetic wastewater had a higher fouling propensity than sludge fed with real wastewater. Le Clech *et al.* (2003) found that, in their experimental conditions, the critical flux value was around 10 l m-2h-1 for the MBR fed with synthetic wastewater (peptone, meat extract, urea and mineral salts) and around 19 l m-2h-1 for the real wateswater. Further, the fouling rate dP/dt was much higher even at sub-critical flux values for the MBR fed with synthetic wastewater than for the one fed with real wastewater.

Additionally to the biodegradability of feedwater organic compounds, the influent composition plays an important role on the filtration performances. Indeed, a nutritive deficiency involves a weak microorganisms adhesion capacity onto the membrane surface [Massé, 2004]. Thus under nutritive deficiency conditions, the cake layer is smaller than under normal conditions [Flemming *et al.*, 1988]. Furthemore, Chang *et al.* (1998) showed that sludge fed with a substrate containing a poorly nitrogen rate presented a weaker specific filtration resistance than another one fed with a substrate containing an accurate nitrogen rate for biomass equilibrium, despite the particle distribution size remained the same. They explained this difference by the sludge EPS rate, sludge with a nitrogen deficiency had a lower EPS concentration.

C2. Effect of free bacteria and floc size

Particle and floc sizes of mixed liquor in a MBR may strongly affect membrane fouling behaviour. If foulants have smaller sizes than the membrane pores, they may enter the pores and pores blocking may occur. Otherwise, if foulants are much larger, they cannot enter the pores and a cake layer may be formed on the membrane surface [Jiang *et al.*, 2003].
Most of the membrane used in MBR have pores diameters ranging from 0.01 µm to 0.1 µm (MF and UF). However, microbial flocs of MBR and free bacteria are much larger than membrane pore sizes, respectively between 10 - 50 µm and 1 – 2 µm [Jiang *et al.*, 2003, Wisniewski *et al.*, 2000, Defrance *et al.*, 2000], thus they are too big to pass through the membrane wall. This is the basis of the high rejection degree of suspended solids in MBR. So,

in regard with MBR membrane fouling, free bacteria and flocs are mainly involved in cake formation on membrane surface, which has a predominant contribution in membrane fouling [Defrance *et al.*, 2000].

Effect of floc size

The activated sludge floc is an association of microorganisms, microcolonies and exopolymeric substances secreted by bacteria [Chaignon *et al.*, 2002].
As a general rule, floc size in MBR is smaller than a that of a conventional activated sludge process [Defrance *et al.*, 2000, Massé, 2004, Cicek *et al.*, 1999]. Cicek *et al.* (1999) found that the specific resistance of MBR sludge is 2.4.1015 m.kg-1 and the one of activated sludge is 2.1.1012 m.kg-1. Thus, they supposed that such a difference might partly be explained by the floc size.

Massé, 2004, also compared the curves representing fouling velocity as a function of permeate flux for sludges sampled in a MBR and in a conventional activated sludge operated at the sludge age and MLSS concentration. He showed that the fouling propensity is significantly much higher in the MBR. Moreover, whatever the sludge age, MBR sludge shows a higher sensitivity to fouling than an activated sludge. One of the explanation for this difference lies in the difference of floc size distribution, the sludge in the MBR showing two populations among which a population of very small particles which can play an important role in fouling. A complementary explanation is linked to the difference of SMP in the two reactors, the MBR being enriched in proteins and polysaccharides in solution. The author explains that soluble and colloidal proteins and polysaccharides could participate to the difference of sludge filtration between the two systems.

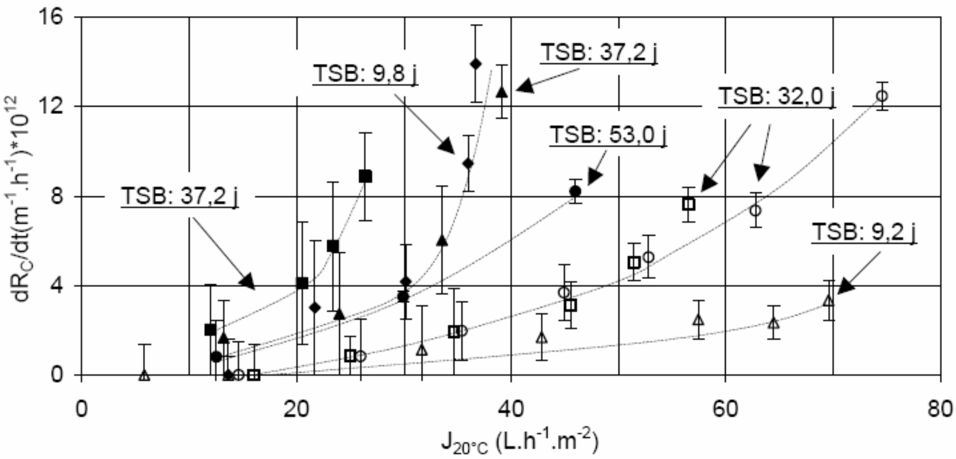

Figure 27 : Fouling velocity vs. flux for different SRT(black : MBR / white : AS) [Massé, 2004]

In MBR processes, to mitigate membrane fouling shear stresses are applied along the membrane and can be high depending on the operating conditions and of module design. Shear stress is mainly due to two phenomena: air scouring (Cf. A.7) and cross flow [Lee *et al.*, 2003, Wisniewski *et al.*, 1998]. Moreover, in the case of an external membrane module with a high liquid velocity, the sludge recirculation increases the shear stress [Kim *et al.*, 2001]. The shear stress imposed along the membrane decreases the size of bacterial flocs.

Defrance *et al.* (2000) found that after 100 days of operation in a high shear MBR, the most frequent size of flocs dropped from 300 µm to 50 µm in the aeration tank. The reduction in the floc size is directly proportional to the magnitude of shear stress and operation time. The decrease of floc size is due to a floc breakage and induces the presence in the suspension of isolated cells and microflocs [Wisniewski *et al.*, 2000]. According to Wisniewski *et al.* (1998), the floc breakage is certainly due to erosion strengths or to a rupture of the network of polysaccharides fibrils which is the support of the floc structure. In addition, consequently to floc breakage, a release of EPS from the flocs and the cells to the bulk solution occurs. Actually, inside flocs cells are usually encased in EPS matrix [Kim *et al.*, 2001]. Kim *et al.* (2001) proved this result by an UV analysis: they observed an increase of EPS concentration after 7 days of crossflow operation.

Besides, Chaignon *et al.* (2002), studied the influence of stirring on floc size. They found that the higher stirring intensity, the smaller the floc size. When they increased the stirring intensity, flocs broke up rapidly and then are more slowly reaching a steady state floc size. However in this case, they pointed out the reversibility of the phenomenon: activated sludge was able to flocculate and deflocculate under respectively lower and higher stirring speed. Steady state aggregate size is interpreted as the result of a dynamic equilibrium between floc growth and floc break up in function of shear stress intensity.

In the literature data numerous results support the idea assumed by Cicek *et al.* (1999) which was the difference between the specific resistance of conventional activated sludge and MBR sludge was due to the difference in floc size.
Lim *et al.* (2003) studied the filtration at a constant pressure of both bulking sludge and granular sludge in a MBR fed with a synthetic substrate and functioning with a hollow fibre membrane, the SRT was 10 day and MLSS concentration 3.5 g.L-1. Filtration behaviours of the both sludge are reported on figure 27.

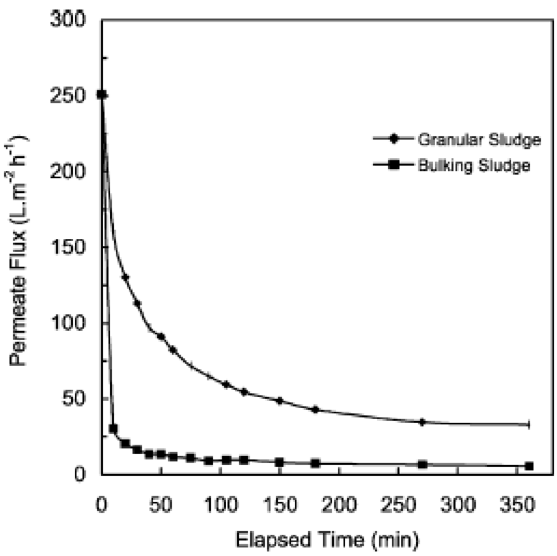

Figure 27 : Typical flux decay patterns of membrane microfiltration treating bulking and granular activated sludge wastewater, Lim et al. (2003)

The difference between these two filtration performances was attributed to the differences in the particle size distribution of the two types of activated sludge. Actually, bulking sludge had a larger particle size distribution (1-200 µm) and was centred on 30 µm whereas granular

sludge particle size ranged from 20-200 µm was centred on 100 µm. Therefore it was concluded that smaller particles caused more severe membrane fouling than larger particles.
In an other study, Ma *et al.* (2006) confirm that smaller flocs form a dense and compact cake layer on the membrane. Hence, a cake layer formed with small flocs is less permeable than a layer made with larger ones [Defrance *et al.*, 2000].

Therefore, the cake specific resistance, α (m.kg-1) is strongly dependant on cake particle size, the smaller the particles, the greater α. Even if these equations support previous ideas, Chang *et al.* (2005) reported that at low MLSS concentration α was not an accurate criterion for the estimation of cake fouling. Further, α is also dependent of the cake porosity ε which may be related to the EPS content in the cake (Orantes, 2005). Thus, particle diameter is not the only one parameter which determinate the specific cake resistance trend.

According to Kim *et al.* (2001), a small floc size, due to a high shear stress inherent in the MBR process, is not the only one factor which deteriorates the cake layer permeability. They suggested it is partly because of the reduction of floc size and partly because of the release of EPS from cells and flocs to bulk solution. They consider that EPS are relatively non-compressible and contribute to increase the cake resistance [Kim *et al.*, 2001]. They did experiments on a crossflow membrane bioreactor functioning with two types of recirculation pumps which induced high shear stresses, they found that the specific resistance of the mixed liquor increased dramatically by a factor of 50 after floc breakage. They assumed it was due to both a decrease in floc size and a release of EPS. Moreover EPS may cause adsorption and pore blocking which increases membrane resistance.

Li *et al.* (2005) studying a submerged MBR functioning with hollow fibres membrane, plotted the air scouring rate versus the critical flux. They observed a critical air scouring rate beyond which further increasing aeration intensity could not cause higher critical flux. They suggested two reasons. The first one is at the super critical air scouring rate, the effect of air scouring on the improvement of the ratio of gas hold-up and thus the intensity of turbulence was no more evident. The second one is a negative effect of large amount of smaller particles arising from strong shear stress on membrane permeability caused by the high air scouring rate. Thus, it is appeared that shear stress should be optimised because on the one hand it prevents rapid deposition of particles onto the membrane surface but on the other hand it could cause a floc breakage and hence involve a stronger membrane fouling by increasing the cake layer resistance [Jiang *et al.*, 2003].

Effect of free bacteria

Firstly, free bacteria may cause internal or external pore blocking and hence they may involve irreversible fouling [Lim *et al.*, 2003].
Then, presence of free bacteria in MBR mixed liquor may cause biofouling [Lim *et al.*, 2003, Chae *et al.*, 2006, Orantes, 2005]. Biofouling may be initiated with the deposition of individual bacteria cells on the membrane surface and then, the cells subsequently multiply and form a biofilm. Biofilm formation presents few steps. First, a reversible adsorption of free cells on the membrane surface followed by an irreversible attachment due to EPS secretion from bacteria. Then, bacteria multiply causing the biofilm growth [Orantes, 2005].
Many authors consider the biofilm as a second membrane, so-called dynamic membrane, which protects the MBR membrane absorbing soluble and colloidal foulants [Holdich *et al.*,

1990]. Moreover it was found that biofilm may have a positive effect on process performances and effluent quality [Holdich *et al.*, 1990, Jiang *et al.*, 2004]
Thus, the presence of a biofilm leads to an increase in the total hydraulic resistance of the membrane, due to an increase in the cake resistance, but protects the membrane against adsorption and pore blocking, i.e. irreversible fouling [Kim *et al.*, 2001]. Kim *et al.* (2001) found that after a floc breakage the change of irreversible fouling resistance was negligible whereas the cake resistance was increased by a factor 10.

C3. Effect of SMP and EPS

It is generally admitted that microbial products, called extracellular polymeric substances (EPS) are playing an important role in fouling [Chang and Lee, 1998]. EPS are present inside flocs (called bound EPS) or there are soluble and present in the supernatant of mixed liquor (SMP: Soluble Microbial Products). Bound EPS structure flocs and permit the attachment of biomass to a surface, in case of MBR it is to the membrane surface and form the filtration cake.

It is difficult to give a relation between bound EPS and fouling. Indeed, Reid *et al.* (2004), Rosenberg *et al.* (2002) and Massé et al found no relation between sludge filterability and concentration in bound EPS although Mikkelsen and Keiding (2002) found that filterability is better when this concentration increase.

It is the same problem for SMPs. SMP can have a relative importance too on the filterability of mixed liquor but their precise part is not clearly defined in the literature [Orantes, 2005]. Bouhabila (1999), comparing sludge and supernatant filterability, considers soluble EPS as the main responsible for fouling: an increase of polymer concentration in supernatant increases clogging. This tendancy is confirmed by Rosenberger and Kraume (2002), who found that composition of the liquid phase effect most filterability of activated sludge, a major influence being the concentration of suspended EPS : the higher the suspended EPS concentration, the lower the filtration index. Kim et al. (2001) also report a decrease of filterability with increasing suspended EPS concentration. On the contrary, Lee *et al.* (2003) did not find any parameter (soluble EPS total quantity, dissolved organic carbon, contact angle) which influence clogging due to supernatant.

Influence of SMPs on nitrogen removal changing intermittent aeration cycle was studied by Nagaoka and Nemoto (2005). They found that in the 120 minutes-cycle reactor, TMP increased more rapidly than in the 10 minutes-cycle reactor. They explained that the reason might be that SMP of more than 1000 kDa are produced more rapidly in the A20 minutes-cycle condition. They also found that three peaks at around 100, 500 and 2000 kDa are prominent in EPS for both cycles and higher molecular weight SMP are decomposed to smaller molecular weight SMP on the membrane surface.

Rosenberger with partners from different European groups (2005) compared the performances of different MBRs and each partner found a relation between fouling and polysaccharides, with a decrease of filtration index, specific flux and an increase of fouling rate, resistance increase, when polysaccharide concentration is increasing. A linear relationship between fouling rate and polysaccharide concentration was also proposed by Lesjean et al. (2004). So, on this basis, the first conclusion pointed out the role of polysaccharides in fouling.

However, a more recent publication from the same group (Drews et al. (2006)) working over longer term experiments and different water resources showed that it is not so evident to conclude about the influence of polysaccharides only. And no more correlation can be proposed between fouling and polysaccharides. It is apparent that the fouling rate tends to increase with increasing polysaccharides concentrations, but data show a larger scatter around the linear correlation reported by Lesjean et al. (2004). One main conclusion is now that the different EPS and PS fractions have to be identified in more details in order to better describe the interactions between organics and membranes.

On another side, Massé showed clearly that fouling velocities are clearly increasing when proteins or polysaccharides concentrations are increasing in the supernatant. Moreover, determination of cake specific resistance for different transmembrane pressures showed that the cake layer can be compressible and cake compressibility seems to be mainly related to the composition in proteins in the supernatant. This means that proteins are playing a major role in cake structuration and compressibility either by building bridges between particles or by forming gel layers. He also showed that the concentration of proteins and polysaccharides in the supernatant depends on sludge residence time. So SRT can be considered as an important parameter that really influences the sludge composition and thus fouling and more precisely cake specific resistance and compressibility.

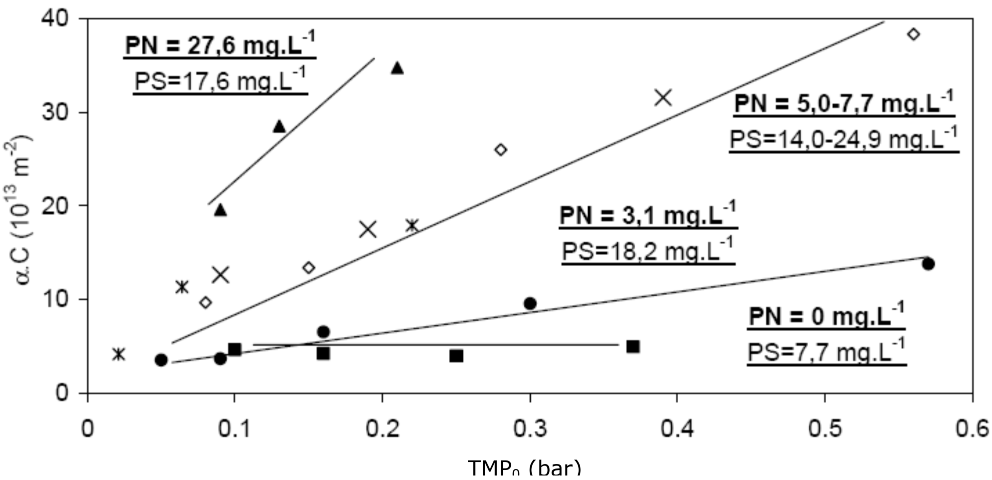

Figure 28 : αC vs. TMP for different proteins or polysaccharide concentrations (PN= proteins, PS= carbohydrates) [Massé, 2004]

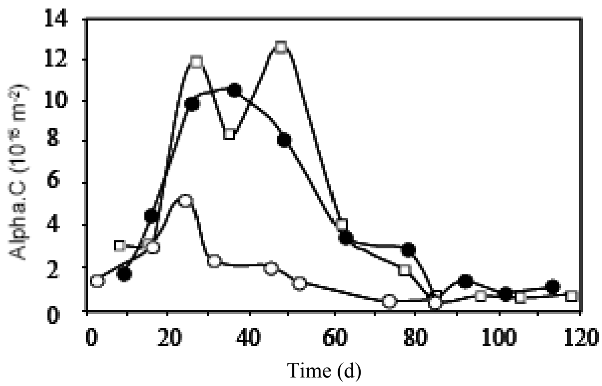

Figure 29 : Evolution of supernatant product α.C for three SRT
(● 10 days □ *20 days* ❍ 30 days) [Bouhabila, 1999]

Furthermore, Figure 29 shows that for a fixed SRT, the α.C product is increasing during the first 30 days and decreases afterwards. This variation of α.C can be linked to the variation of EPS concentration in supernatant [Massé, 2004], this underlines the fact that EPS in supernatant should be implicated in short term fouling.

Conclusion

From this literature study, it appears that even if numerous studies have focused on MBRs it is still difficult to find some generic results because of the complexity of this problem and because of the lack of concerted approaches and of the large number systems and influent waters used.

Among the different operating parameters, aeration appears to be a key point as well for process efficiency than for module design. Aeration is the most common end efficient method used to avoid fouling. However the choice of aeration conditions is often very empirical. A better knowledge is necessary on aeration effects on fouling/fouling reversibility in relation with module design and properties of the biological fluid since mechanisms which take place during filtration with air injection are not well described. However the combination between recent techniques for hydrodynamics characterisation and the development of CFD (Computational Fluid Dynamics) allows a better understanding of effects of two-phase flow hydrodynamics on filtration performances and to conclude that module design should be more thought in relation with the possible use of air.

It seems today evident that microbial products and their soluble part are playing a significant role in membrane fouling. However important differences in the literature results are observed depending on the system used and on the influent water. Some studies are claiming that soluble proteins are the main problematic species whereas others are focusing on polysaccharides. The difference in these results is probably linked to the fact that for instance organics are only characterised by global parameters and that more attention should be given to small (nanosize) particles. Further studies are necessary to elucidate the role of SMP and colloidal particles on fouling mechanisms in MBRs and the key questions that we have to face now are : what category of proteins or polysaccharides are involved in fouling ? what is the contribution of soluble compounds which are yet present in the influent ? What is the contribution of colloids and nanoparticles?

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