Membrane Fouling for Wastewater Treatment in Membrane Bioreactors

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Abstract: Membrane bioreactors (MBRs), which use membranes to separate biomass from biological effluent, offer a number of benefits over traditional treatment. Membrane fouling in MBRs, however, limits their broad usage since it lowers productivity and raises operating and maintenance expenses. To analyse, model, and regulate membrane fouling processes, there has been a lot of study and development recently. However, due to the complexity of the biomass matrix, which is very heterogeneous and contains living microorganisms, coherent and well-structured theories on membrane fouling are not yet accessible. Membrane fouling in MBR systems can occur on the membrane surface or inside the membrane pores and can be either reversible (removable by physical washing) or irreversible (removable by chemical cleaning alone). Although the inherent heterogeneity of the system makes it extremely difficult to establish a general model to describe membrane fouling in such a process, the nature and extent of fouling in MBRs is strongly influenced by three factors: biomass characteristics, operating conditions, and membrane characteristics. Low-flux operation, high-shear slug flow aeration in submerged configuration, periodic air or permeate backflushing, intermittent suction operation, or addition of powdered activated carbon are some of the fouling control approaches that have been examined (PAC). Only PAC addition is not being utilised in current massive deployments.

CE Database keywords: Membrane; Wastewater treatment; Sewage; Biomass; Biological treatment.

Introduction

Urban and industrial wastewaters are frequently treated biologically, utilising microorganisms to break down organic contaminants, such as in the activated sludge process (ASP). The ASP produces a lot of extra sludge in addition to needing big aeration and sedimentation tanks. The technique also has issues with bulking and foaming in the solid-liquid separation. The membrane bioreactor (MBR) is an alternative technique that combines the clarifying and settlement stages of the traditional ASP into a single, integrated operation. The benefits of the MBR over traditional treatment have been discussed (Stephenson et al. 2000). Benefits include a smaller environmental impact and less sludge being produced because the bioreactor is kept at a high biomass concentration. The system can handle significant variations in influent quality, and because of the efficacy of the filtration process, high-quality product water is produced, allowing for direct reuse of the effluent for nonpotable uses. Moreover, a higher nitrification rate may

achieved because an aeration tank can hold a large number of slowly growing nitrifying autotrophs (Chiemchaisri and Yamamoto 1993; Fan et al. 1996; Kishino et al. 1996; Nah et al. 2000).

In spite of these benefits, membrane fouling prevents the MBR process from being used widely. The most significant issue affecting system performance, as with other membrane separation procedures, has been demonstrated in recent reviews covering membrane applications to bioreactors (Chang et al. 2001b; Kim et al. 2001). Fouling causes a decrease in permeate flux, which necessitates more frequent membrane cleaning and replacement, raising operating expenses. Hence, the majority of MBR investigations seek to recognise, look into, manage, and model membrane fouling (Chang and Lee 1998; Tar- dieu et al. 1998; Wisniewsky and Grasmick 1998; Defrance and Jaffrin 1999a; Ognier et al. 2001). Membrane fouling is the result of interactions between the components in the activated sludge liquor and the membranous material. Included in the latter are elements of the substrate, cells, cell waste, and microbial metabolites like extracellular polymeric compounds (EPS). The composition of the suspended biomass is not fixed; for instance, in batch systems, the endogenous phase of the biomass may have more lysed cells and cell detritus than the log growth phase. Moreover, the feed water composition and the MBR operating conditions used both affect the composition of the activated sludge liquor. The wide range of operating conditions, feedwater matrices, and the limited information reported in most studies on the composition of suspended bio-mass have made it challenging to establish any generic behaviour pertaining to membrane fouling in MBRs specifically, despite the fact that there have been many investigations of membrane fouling published. In this review, all pertinent studies are appraised so as to provide an

insight into membrane fouling in the membrane bioreactor process, mainly with reference to municipal and industrial wastewaters.

Membrane Bioreactor Process

The MBR process consists of a suspended growth biological reactor combined with a membrane unit either located external to the bioreactor (sidestream) or mounted directly within it (submerged or immersed). For the sidestream configuration, a high cross-flow fluid velocity provided by a recirculation pump is designed to reduce deposition of suspended solids at the membrane surface. Although this configuration is simple and provides more direct hydrodynamic control of fouling, the energy demand is relatively high. The submerged configuration, on the other hand, relies on coarse bubble aeration to produce in-tank recirculation and suppress fouling. Although the energy demand of the submerged system can be up to two orders of magnitude lower than that of sidestream systems (Dijk and Roncken 1997; Gander et al. 2000a), submerged systems operate at a lower flux and so demand more membrane area.

Aerobic MBRs are commonly used for domestic wastewater, ''night soil,'' industrial wastewater, and municipal water treatment (Zaloum et al. 1994; Urbain et al. 1996; Zhang et al. 1997; Ragona and Hall 1998; Chang et al. 2001c). Anaerobic MBRs have been mainly applied to industrial wastewaters of high organic strength (Nagano et al. 1992; Anderson et al. 1996; Choo and Lee 1998; Fakhrulrazi and Noor 1999; Ince et al. 2000; Hu and Stuckey 2001; Lee et al. 2001). Anaerobic bacteria have slower growth rates than aerobic bacteria and so produce less residual sludge but require a relatively long retention time. Moreover, anaerobic biosolids exhibit poor settleability due to their diffusible and filamentous nature (Elmaleh and Abdelmoummi 1998). Therefore, anaerobic MBRs offer similar advantages over conventional processes as MBRs. In cases where complete removal of nitrogen is required, MBR processes adopting aerobic-anoxic cycling to obtain maximum denitrification have been used (Silva et al. 1998). This paper reviews aerobic, anaerobic, and aerobic-anoxic systems.

Membrane Fouling

Membrane fouling in MBRs is attributed to the physicochemical interactions between the biofluid and membrane. As soon as the membrane surface comes into contact with the biological suspension, deposition of biosolids onto it takes place leading to flux decline. Since this cake layer is largely readily removable from the membrane if an appropriate physical washing protocol is employed, it is often classified as *reversible* fouling. On the other hand, internal fouling caused by the adsorption of dissolved matter into the membrane pores and pore blocking is considered *irreversible* and is generally only removed by chemical cleaning. However, restricting categorization of membrane fouling to reversible or irreversible is somewhat simplistic. For instance, gel layer formation over a membrane surface is most often irreversible although it is notionally reversible since it forms a cake layer. Some kinds of fouling by pore blocking and adsorption may be partially reversible depending on the strength of adhesion and the vigor of the physical wash.

Resistance-in-Series Model

Many empirical and theoretical models have been proposed to describe the membrane fouling phenomena, the simplest being the resistance-in-series model (Field et al. 1995; Silva et al. 2000; Lee et al. 2001);

$$J = TMP/(\eta \cdot R_t) \qquad (1)$$

$$R_t = R_m + R_c + R_f \qquad (2)$$

where J=permeate flux; TMP=transmembrane pressure; η=dynamic viscosity of the permeate; R_t=total resistance; R_m=intrinsic membrane resistance; R_c=(reversible) cake resistance caused by the cake layer deposited over the membrane surface; and R_f=(irreversible) fouling resistance produced by adsorption of dissolved matter (*pore narrowing*) and/or pore blockage within the membrane (*plugging*). According to this model the flux is inversely proportional to the total resistance, the latter being the sum of individual, supposedly discrete resistances.

The resistances are conventionally measured through a series of filtration experiments comprising pure-water filtration, sludge filtration, and pure-water filtration following filter cake removal. However, such experiments are not always practical and, in any case, assume complete decoupling of all resistances. In spite of this, many authors identify components of resistance in their work. For example, the reversible fouling component has been considered as comprising a gel layer resistance coupled with a concentration polarization resistance, these being additive (Choo and Lee 1996a; Al-Malack and Anderson 1997). However, the validity of differentiating between the various resistance components on the basis of arbitrary physical tests is questionable, and some authors prefer to quote a single resistance value, R_f, including all resistances offered other than that of the clean membrane (Gander et al. 2000b; Kwon et al. 2000; Chang et al. 2001c).

Table 1 shows a summary of common values of resistances in the literature. It should be noted that each resistance value depends strongly on experimental conditions; e.g., biomass characteristics, membrane material and pore size, and operating conditions. In general, resistance values for anaerobic MBRs are much higher than those of aerobic systems largely due to the higher organic loading rates and the occurrence of high levels of fine colloidal matter and inorganic precipitation in anaerobic processes.

Empirical Models

Constant pressure filtration behavior is typified by a rapid flux decline at the start of filtration followed by a more gradual decrease until a steady state or a pseudo-steady state flux is reached. Four filtration models, originally developed for dead end filtration (Grace 1956), have been proposed to describe the initial flux decline. All models imply a dependence of flux decline on the ratio of the particle size to the pore diameter. The standard blocking and cake filtration models appear most suited to predicting initial flux decline during colloid filtration (Visvanathan and Ben Aim 1989) or protein filtration (Bowen et al. 1995). According to Bowen and co-workers, four consecutive steps had been defined: (1) blockage of the smallest pores, (2) coverage of the larger pores inner surface, (3) superimposition of particles and direct blockage of larger pore, (4) creation of the cake layer. It seems reasonable to expect all these blocking processes to prevail in a polydisperse filtration process such as MBR.

Mass Transport Models

Although the steady state models are based on highly restrictive hypotheses (spherical, inert, mono-disperse particles, low concentration solution, etc.), these equations have been used to predict flux behavior in MBRs. Models based on Brownian diffusion, as promoted by concentration polarization, are known to substan-

Table 1. Typical Resistance Values in Membrane Bioreactor System

MBR type	Resistance ($10^{11} \times m^{-1}$)	Membrane	Wastewater	Micro-organism	References
Sub–HF	$R_m = 8$, $R_c = 24$, $R_f = 1$	MF (0.2 μm), Polymeric	Domestic	Anoxic +Aerobic	Parameshwaran et al. (1999)
SS–TB	$R_m = 2.3$, $R_T = 41.1$	MF (0.1 μm), Ceramic ($TiO_2 + ZrO_2$)	Domestic +Industrial	Aerobic	Defrance et al. (2000)
SS–FP	$R_m = 11$, $R_p = 1732$, $R_{ef} = 337$, $R_{if} = 11$ (Fouling due to inorganic precipitation)	UF (20 kD), Fluoropolymer	Industrial	Anaerobic	Choo and Lee, (1996a)
SS–FP	$R_m = 13$, $R_c = 1230$, $R_f = 5$ (Fouling caused by floc breakage)	UF (20 kD), Polyacrylonitrile	Synthetic	Aerobic	Chang et al. (2001b)
Sub	$R_m = 3$, $R_c = 2890$, $R_f = 12$ (Fouling due to high organic and colloidal nature)	MF (0.5 μm), Mixed esters of cellulose	Piggery	Anaerobic	Lee et al. (2001)
SS–TB	$R_T - R_m$ (=Fouling resistance); ~100 (Depending on CFV)	MF (0.14 μm), Ceramic (TiO_2)	Synthetic	Anaerobic	Elmaleh and Abdelmoummi (1998)
Sub–FP	$R_T = 30 - 2000$ (Depending on organic loading rate)	MF (0.2 μm), Polysulfone	Synthetic	Aerobic	Nagaoka et al. (1998)
Sub–TB	$R_m = 3 - 5$, $R_c = 3 - 48$, $R_{pl} = \sim 10$ (Depending on TMP)	MF (0.5 μm), Alumina	Domestic	Aerobic	Shimizu et al. (1996b)
Sub–HF	$R_m = 22$, $R_c + R_p = 75 - 120$ (Depending on MLSS)	MF (0.01 μm), Polyacrylonitrile	Domestic	Aerobic	Shin et al. (1999)
SS–TB	$R_m = 3.6$, $R_T = 29$	MF (0.05 μm), Zircon oxide	Domestic	Aerobic	Tardieu and Grasmick (1998)
Sub–FP	$R_T = 1 - 6$ (Depending on operation time)	MF (0.4 μm), Polyethylene	Domestic	Aerobic	Ueda and Horan (2000)
Sub–HF	$R_T = 0.2 - 0.6$ (Depending on cleaning regime)	MF (0.1 μm), Polyethylene	Domestic	Aerobic	Ueda et al. (1997)

Note: Sub: submerged MBR; SS: sidestream MBR; HF: hollow fiber; FP: flat plate; TB: tubular; R_m: membrane resistance; R_p: polarization resistance; R_{ef}: external fouling resistance; R_{if}: internal fouling resistance; R_{pl}: plugging resistance; R_c: cake resistance; and R_T: total resistance.

tially underpredict the flux obtained during colloid filtration (Belfort et al. 1994). To resolve this ''flux paradox,'' back transport models such as inertial migration, shear-induced diffusion, and surface transport have been developed. The key aspect of all back transport models is the implication of the influence of particle size and recirculation rate, or cross-flow velocity (CFV), on flux. Both shear-induced and inertial lift models predict more significant fouling in the cases of small particles and/or large difference between back-transport and permeate velocities. More fouling is also predicted at lower CFVs.

For MBRs, these models have been used to determine the minimum size of compounds likely to deposit under specific flow conditions, i.e., CFV values (Tardieu et al. 1998; Choo and Lee 1998; Huisman and Tragardh 1999; Wisniewski et al. 2000). For example, for a fixed CFV of 0.5 m s^{-1}, the shear-induced diffusion model predicts that floc particles up to a size of 4 μm and up to 7 μm deposit on the membrane for flux of 12.5 and 25 L m^{-2} h^{-1}, respectively (Tardieu et al. 1998). Tardieu and his co-workers also found the inertial lift model to predict an even greater fraction of floc deposition under the same conditions (up to 15 and 20 μm, respectively). Not surprisingly, particle deposition is very limited under turbulent flow conditions (CFV ~4 m s^{-1}) where the back transport velocity is actually greater than the permeate flux. Hydrodynamic factors have a profound effect on the initial flux decline, where smaller particles (having a small back transport velocity) result in the exponential flux decline usually observed (Choo and Lee 1998). Hence, these mass transport models provide a better understanding of the role of hydrodynamics in MBR fouling (Tardieu et al. 1998), though the role of dissolved and colloidal species is neglected in such models.

Experimental Evaluation of Membrane Fouling

Although it is difficult to establish a general rule about membrane fouling in MBRs, the nature and extent of fouling are strongly influenced by three factors: biomass characteristics, operating conditions, and membrane characteristics (Fig. 1). These are considered in turn below.

Biomass Characteristics

Activated sludge is a complex and variable heterogenous suspension containing both feedwater components and metabolites produced during the biological reactions as well as the biomass itself. Many individual components of the mixed liquor, ranging from flocculant solids to dissolved polymers such as extracellular polymeric substances (EPS), can contribute to membrane fouling.

Mixed Liquor Suspended Solids and Dissolved Matter

At the early stages of MBR development, many researchers have given attention to the effects of the mixed liquor suspended solids (MLSS) concentration on the membrane fouling. Fane et al. (1981), for example, reported membrane resistance to increase linearly with MLSS, and Yamamoto et al. (1989) also reported that the flux decreased abruptly if the MLSS concentration exceeded 40,000 mg L^{-1} in a submerged system.

MLSS concentration is considered to impact directly upon cake layer resistance, as surmised from conventional cake filtration theory, the cake resistance, R_c (m^{-1}) often being expressed as (Shimizu et al. 1993; Chang et al. 2001a)

$$R_c = \alpha \cdot v \cdot C_b \tag{3}$$
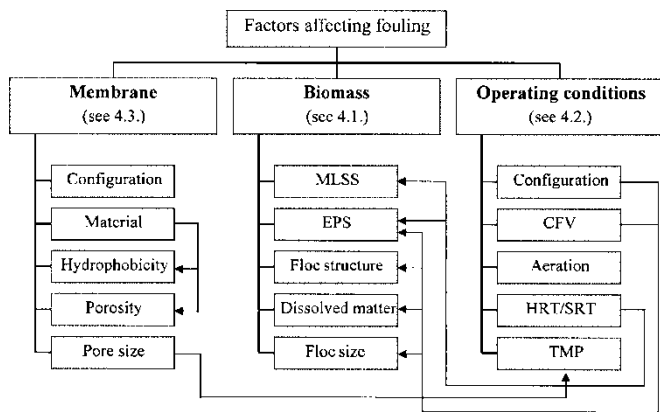
Fig. 1. Factors influencing membrane fouling in membrane bioreactor process

where a=specific cake resistance (m kg^{-1}); v=permeate volume per unit area (m^3 m^{-2}); and C_b=bulk MLSS (or mixed liquor volatile suspended solids (MLVSS) concentration (kg m^{-3}). MLSS concentrations for aerobic MBRs typically range from 3,000 to 31,000 mg L^{-1} (Brindle and Stephenson 1996). However, Lubbecke et al. (1995) showed MLSS concentrations up to 30,000 mg L^{-1} to be not directly responsible for irreversible fouling, and that viscosity and dissolved matter more significantly impact on flux. Ueda et al. (1996) observed the increase in viscosity to yield a substantial suction pressure increase, which consequently causes the MBR to fail.

Dissolved species impact both on internal and external fouling, the latter being promoted by concentration polarization. Ishiguro et al. (1994) proposed the following general correlation between flux and dissolved organic carbon (DOC)

$$J = a + b \ \log(\text{DOC}) \qquad (4)$$

where a and b=empirical constants.

Harada et al. (1994) found accumulation of soluble substances, rather than incremental increases, in the MLSS, to substantially affect flux decline in anaerobic MBRs. While aerobic MBR foulants largely originate from EPS and other metabolic products, fouling in anaerobic systems can arise from precipitation of inorganic scalants such as struvite, $MgNH_4PO_46H_2O$ (Choo and Lee 1996a; Yoon et al. 1999). Struvite can foul anaerobic systems where ammonium and phosphate ions are produced during anaerobic decomposition of organics in the wastewater.

Empirically derived flux prediction equations based on the combined effects of MLSS, dissolved matter, and viscosity have been developed. Sato and Ishii (1991) produced the following empirical relationship describing filtration resistance in terms of MLSS, chemical oxygen demand (COD), TMP, and viscosity (h) for a sidestream MBR

$$R = 842.7 \ \text{TMP} \ (\text{MLSS})^{0.926}(\text{COD})^{1368}h^{0.326} \qquad (5)$$

Krauth and Staab (1993) proposed the following equation accounting for the influence of the MLVSS for a sidestream MBR:

$$J = J_0 \cdot e^{k(\text{MLSS-MLVSS})R/\text{MLVSS}} \qquad (6)$$

where J_0=initial flux; k=empirical constant depending on TMP; and R=Reynolds number.

Some researchers have additionally included the effect of hydrodynamics on flux. Shimizu et al. (1996a) expressed the steady state flux (J_{SS}) of a submerged hollow fiber MBR as a function of MLSS concentration, superficial air velocity ($u*$), and a membrane geometric hindrance factor (ϕ) dependent on module configuration

$$J_{SS} = K \cdot u* \cdot \phi \cdot \text{MLSS}^{0.5} \qquad (7)$$

Many research groups have extensively investigated the contribution of specific mixed liquor species to membrane fouling. Defrance et al. (2000) reported that the relative contributions of suspended solids (SS), colloids, and dissolved matter to the resistance to filtration caused by fouling were 65, 30, and 5%, respectively. The same study carried out by Bouhabila et al. (2001) showed the biomass relative contribution on fouling to be 24, 50, and 26% for SS, colloids, and solutes, respectively. Through fractionation of the mixed liquors of activated sludge into floc cell, EPS and dissolved matter, Chang and Lee (1998) identified EPS as being the main component contributing to fouling resistance. Both these studies, however, showed the sum of the resistances provided by each constituent to be greater than the measured total resistance, indicating that individual fouling resistances were not additive. Wisniewski and Grasmick (1998) fractionated the activated sludge suspension into settleable particles (from particle size above 100 μm), supracolloidal-colloidal fraction (nonsettleable particle with a size ranging from 0.05 to 100 μm), and soluble (obtained after filtration with 0.05 μm membrane). They revealed 52% of the total resistance to be attributable to the soluble constituents.

Particle Size Distribution
Many researchers have sought to establish the influence of particle size on the cake layer resistance. According to the well-known Carmen–Kozeny equation applied to conventional filtration, specific resistance (a) is a function of particle diameter (d_p), porosity of cake layer (s), and particle density (p) as follows (Baker et al. 1985):

$$a = 180(1-s)/(p \cdot d_p^2 \cdot s^3) \qquad (8)$$

Eq. (9) is obtained by putting together Eqs. (3) and (8)

$$R_c = 180(1-s)/(p \cdot d_p^2 \cdot s^3) \cdot v \cdot C_b \qquad (9)$$

R_c is thus strongly dependent on cake particle size: the smaller floc size, the greater cake resistance. In general, the particle size of an activated sludge floc ranges from 1.2 to 600 μm (Jorand et al. 1995). However, the shear force arising from pumping during cross-flow filtration results in the breakup of biological flocs, generating fine colloids and cells forming which then form a denser cake layer on the membrane (Wisniewski and Grasmick 1998; Kim et al. 2001). According to Wisniewski et al. (2000), the suspension produced after the floc breakup consists mainly of particles having a size of around 2 μm corresponding to flux declines. Cicek et al. (1999a,b) revealed the average diameter of particles in a sidestream MBR system to be ∼3.5 μm, with 97% of the particles being smaller than 10 μm, whereas the ASP mixed liquor contained flocs ranging from 20 to 120 μm in size. The resulting a values, measured by vacuum filtration, of the MBR sludge were determined as 2.4×10^{15} m kg^{-1} compared to 2.1×10^{12} m kg^{-1} for that of the ASP. On the other hand, the floc size in the submerged MBR (20–40 μm) appears to be greater than that of sidestream (7–8 μm) due to the reduced shear stress (Zhang et al. 1997).

Floc breakup exposes the EPS present inside the floc structure as well as increasing the EPS level in bulk solution, leading to severe membrane fouling (Chang et al. 2001b), as explained below. It has been reported that floc breakup also leads to a loss

of biological activity (Brockmann and Seyfried 1996; Ghyoot et al. 1999a,b; Chang et al. 2001b), change in microorganism population (Rosenberg et al. 1999) and decreasing settleability (Cicek et al. 1999a,b).

Extracellular Polymeric Substances „EPS‟

An activated sludge floc can be defined as a microbial entity that is formed by different species of biomass. The constituents of the floc are embedded in a polymeric network of EPS. Since EPS provide a highly hydrated gel matrix in which microorganisms are embedded, they provide a significant barrier to permeate flow in the MBR.

Microbial EPS are high molecular-weight mucous secretions from microbial cells. They can play an important role for floc formation in activated sludge liquors (Sanin and Vesilind 2000; Liao et al. 2001). The EPS matrix is very heterogeneous, with polymeric materials arising including polysaccharides, proteins, lipids, and nucleic acids (Bura et al. 1998; Nielson and Jahn 1999).

Recently, many MBR studies have identified EPS as the most significant biological factor responsible for membrane fouling. Chang and Lee (1998) correlated the EPS levels and membrane fouling quantitatively. These authors examined the EPS levels in activated sludge in various physiological states, and found there to be a linear relationship between membrane fouling and EPS levels. Nagaoka et al. (1996, 1998, 1999) similarly linked hydraulic resistance to EPS levels in the aeration tank, including empirical parameters for EPS production and degradation, developing a phenomenological model to predict fouling and to evaluate the effects of loading rate, flux, and shear stress on bioreactor performance. Huang et al. (2001) found soluble organic substances with high molecular weights, mostly attributable to metabolic products, to accumulate in the bioreactor. These had a negative influence on membrane permeability: accumulation of 50 mgTOC (total organic carbon) L^{-1} resulted in 70% decrease in flux. In addition, EPS levels of 23 mg L^{-1} have been found to produce a six- to sevenfold increase in the internal fouling resistance (Chang et al. 2001b). The fouling propensity of specific EPS components has also been studied. Shin et al. (1999) attributed 90% of the cake resistance to EPS and found resistance to vary with the ratio of protein and carbohydrate in the EPS. Mukai et al. (2000) found the protein to sugar ratio of the EPS to influence permeate flux during ultrafiltration, the permeate flux decreasing with an increasing protein content. In a study illustrating the effects of EPS on membrane fouling carried out by Kim et al. (1998), addition of powdered activated carbon to the MBR was shown to increase permeability by reducing dissolved EPS levels from 121–196 to 90–127 mg/gVSS.

Most studies of the effect of EPS on membrane fouling rely on extraction of EPS from the sludge flocs. However, relatively large amounts of EPS can originate from unmetabolized wastewater components and bacterial products arising either from cell-lysis or cell-structural polymeric components (Dignac et al. 1998). Thus, the quantitative expression of flux as a function of EPS concentration has an inherent limitation.

Little information is currently available on EPS membrane fouling mechanisms. For the membrane filtration of marine bacteria SW8, Hodgson et al. (1993) proposed that EPS and cells were co-deposited during membrane filtration, with EPS filling the voids between the cells, forming a barrier of high hydraulic resistance. Since most resistance is attributed to the cake layer on
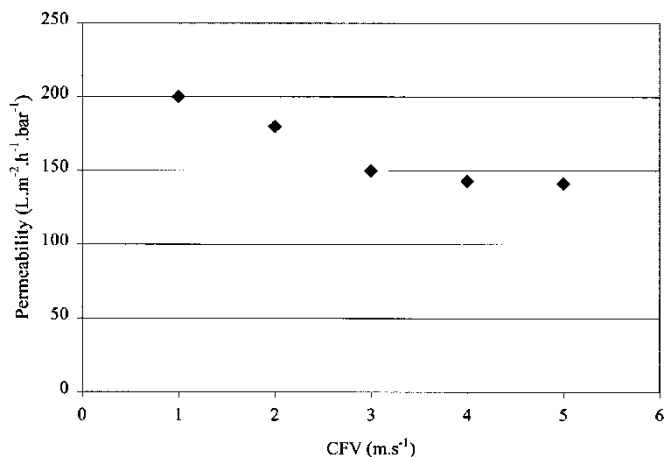

Fig. 2. Effect of cross-flow velocity on permeability (from Defrance et al. 1999a).

the membrane surface rather than internal fouling (Table 1), this hypothesis could be expanded to explain EPS fouling of the MBR.

Operating Conditions

Cross-Flow Velocity

Both experimental and empirical studies have revealed cross-flow velocity (CFV) to be a major influence on membrane fouling (Madaeni 1997; Tardieu et al. 1998; Defrance and Dalfrin 1999a). The CFV affects the mass transport of particles away from the membrane surface, and thus the resultant cake layer thickness, by increasing the shear and so shear-induced diffusion (see the section mass transport models below). A higher shear stress can be developed by moving the membrane, rather than the water adjacent to it, by using a vibrating or spinning membrane (Ohkuma 1994; Lu et al. 1999). However, while some mechanical shear-enhancement membrane separation processes exist commercially, none have been employed for MBRs.

A higher flux, 100 L m^{-2} h^{-1} compared to 25 L m^{-2} h^{-1}, sustained for a longer period (100 h compared to 6 h) was obtained when operating at CFV=4m 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 CFV. However, a small and constant increase in TMP was always noted, presumably due to the interaction between the membrane and the sludge (soluble and colloidal fractions). Flux as a function of CFV appears to follow a similar trend for MBR liquors as for other matrices (Tarleton and Wakeman 1994). The stabilized flux—the maximum flux measured, which is dependent of TMP—has been shown to increase almost linearly with CFV (Defrance et al. 1999a). However, increasing the flux by raising the CFV produces a concomitant rise in the TMP (Fig. 2), leading to a lower permeability. Literature hydraulic data show significant variation for different matrices, membranes, and configurations (Table 1), in particular, membrane material (see membrane characteristics below). While a relationship between CFV and flux is perceptible, that between CFV and permeability is less so. According to critical flux analysis by Madaeni et al. (1999), who demonstrated a similar relationship between flux and CFV, CFV has been shown to have a greater effect at high MLSS levels and smaller membrane pore sizes.

Table 2. Hydraulic Performance Versus Cross-Flow Velocity

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[γ]	2.2	8	4	Ghyoot et al. (1999a)
3[δ]	2	153	77	Ghyoot et al. (1999a)

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

Studies carried out using monodispersed particles have revealed opposing effects of CFV. With small particles a CFV increase increases flux, while for larger particles the CFV has no, or even a negative effect on permeate flux. Tarleton and Wakeman (1994) suggested that this phenomenon was due to particle classification at the membrane surface. During the incipient stages of filtration it is 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 CFV. Evidence for particle classification has been provided by results obtained with polydisperse solutions (Tarleton and Wakeman 1993). These results also suggested that a critical size arises where CFV has little or no effect on flux decline. Classification may provide an explanation of reported operational traits in MBRs. It has been shown that the sludge supernatant, containing the finer particles, causes initial fouling (Bouhabila et al. 1998; Wisniewski and Grasmick 1998).

In sidestream MBRs, increasing the CFV can be achieved by increasing the pump speed. Even for experiments carried out under biologically steady state conditions, it is possible that reported flux changes at different CFV values may be attributable to biochemical as well as hydraulic changes (Tardieu et al. 1998). As well as the effects on EPS release [see the section ''Extracellular Polymeric Substances (EPS)'' above] shear produced by pumping breaks up microbial flocs generating a larger number of fine colloidal particles which can form a denser cake layer on the membrane surface (Kim et al. 2001). This effect is exacerbated by the use of a rotary pump, rather than a centrifugal pump.

Aeration
In a submerged MBR, CFV is created by aeration which not only provides oxygen to the biomass, leading to a better biodegradability and synthesis of the cells, but also maintains the solids in suspension and scours the membrane surface and so suppresses fouling (Dufresne et al. 1997). The air-induced cross-flow can efficiently remove or at least reduce the fouling layer on the membrane surface. Equating a bioreactor equipped with baffles to an internal-loop-airlift reactor allows the CFV to be calculated theoretically (Ishida et al. 1993; Kishino et al. 1996; Liu et al. 2000). The CFV can also be estimated by the use of a flow velocity meter directly immersed in the reactor (Ueda et al. 1996, Shimizu

et al. 1996a,b). In both cases, the CFV is reported to range between 0.3 and 0.5 m s^{-1} (Table 2).

Cui and his co-workers (Cui 1997; Li et al. 1997; Ghosh and Cui 1999) have mathematically modeled wall shear variation and distribution around a large bubble in a tubular tube, producing *slug flow* to create air lift. Results were related to the mass transfer coefficient. Corroboratory permeate flux data have subsequently been provided by an experimental study of a submerged tubular MBR operated with slug flow (Judd et al. 2001; Chang and Judd 2002).

Study of the relative effect of the MLSS concentration and airflow rate on fouling (Dufresne et al. 1997) has shown aeration to be roughly half as significant as MLSS level. Moreover, the effect on permeate flux of increasing aeration was found to be independent of MLSS (Bouhabila et al. 1998), even if a higher airflow is required at higher MLSS due to the concomitant viscosity increase (Muller et al. 1995; Gunder and Krauth 1999).

A comprehensive study of the effect of aeration on fouling has been carried out by Ueda et al. (1996) on a hollow fiber submerged MBR. These authors measured air uplift velocity using an electromagnetic flowmeter, the reading giving an approximation of CFV. These authors found aeration for turbulence promotion to become more critical for intermittent operation, and that fouling suppression by aeration was mainly through agitation of the membrane fibers. An optimum value of the airflow rate was identified beyond which further increases had no effect (Table 3), an observation subsequently repeated by Bouhabila et al. (1998, 2001). Ueda et al. (1996) also reported that any reduction in the airflow had negative effect on the TMP. Although original performances could be restored following a short-term reduction in aeration rate, longer-term reduction led to a rapid accumulation of material on the membrane surface. These authors also surmised that a more densely packed module could enhance fouling amelioration due the greater effective CFV attained, a similar conclusion being reached by Liu et al. (2000) in optimizing the design of a hollow fiber membrane module MBR.

Design parameters reported by authors for different submerged systems are summarized in Table 3. In this table, the *aeration intensity* refers to the airflow rate to the membrane area ratio. The

Table 3. Airflow Rate Effect on Flux in Submerged Systems

Airflow rate ($m^3\,h^{-1}$)	Gas hold up (h^{-1})	Aeration intensity ($m\,h^{-1}$)	Flux ($L\,m^{-2}\,h^{-1}$)	Config.	Reference
0.55	0.8	0.05		HF	Bouhabila et al. (1998)
42	2	10	12	HF	Ueda et al. (1996)
3.2–4.3	0.6–0.8	1.9–2.5	1.1–2.2	HF	Dufresne et al. (1998)
0.8	0.8	8	12	FP	Dufresne et al. (1997)
100	11	2.2	31	FP	Gunder et al. (1999)
80	13	1	20	FP	Gunder et al. (1998)
78	35	0.9	18	FP	Gunder et al. (1998)
0.72	0.009	0.7	8.3	HF	Visvanathan et al. (1997)

table shows that changes in aeration intensity of more than an order of magnitude do not appear to yield a commensurate increase in flux.

Hydraulic Retention Time and Loading Rate

An indirect action of hydraulic retention time (HRT) on fouling in a submerged hollow fiber MBR has been reported by Visvanathan et al. (1997), who noted reduced fouling (i.e., no TMP increase) at higher HRT values, postulating that a rapid formation of a compact layer on the membrane surface took place at longer HRTs. Given that the MLSS concentration was reported to change from 3 g L^{-1} for an HRT of 12 h to 7 g L^{-1} for 3 h HRT, it is evident that the accompanying change in hydraulic resistance is related to the MLSS (see section on Biomass characteristics above): a shorter HRT provides more nutrients to the biomass, and leads to a greater biological growth and so a higher MLSS (Dufresne et al. 1998). MLSS is also directly influenced by organic loading rate (OLR), though Nagaoka et al. (1998) concluded, from their study of the effect of loading rate on the operation of a flat sheet MBR, that fouling was not greatly influenced by threefold change in OLR for flux and OLR values of around 4 $L\,m^{-2}\,h^{-1}$ and 2 $g\,L^{-1}\,d^{-1}$ respectively.

Solid Retention Time and Sludge Age

Solid retention time (SRT) is directly linked to the net production of excess sludge and significantly affects biological performance by altering sludge composition (Urbain et al. 1998; Bouhabila et al. 2001). The most obvious result of SRT variation is on MLSS concentration. By increasing SRT from 5 to 30 days, Xing et al. (2000) noted an apparent MLSS concentration increase from 2.5 to 15 g L^{-1}. Decreases in both EPS concentration (Chang et al. 1998) and slight increases in mean particle size (Huang 2001) have been reported at longer sludge ages, though these effects both appear to be very small. Though longer SRTs inevitably lead to both higher MLSS values, the latter increasing from 3 to 7.5 g L^{-1} on increasing the SRT from 5 to 20 days according to Fan et al. (1999), both Fan and co-workers and Bouhabila et al. (1998) have reported reduced fouling rates at the longer sludge ages. However, high viscosity liquors associated with very high MLSS concentrations can lead to excessive fouling, according to Ueda et al. (1996).

Like the HRT, the SRT cannot be varied without important changes in sludge composition. The direct effect of SRT on fouling seems once again difficult to reveal. As a general trend it was shown and now accepted that the shorter the HRT and the longer the SRT, the higher the MLSS concentration. It is then suggested that HRT and SRT cannot be considered as direct fouling causes but rather like parameters influencing factors like MLSS, particle size distribution, and EPS, which can then be directly related to fouling rates. Clearly, as with HRT, SRT only indirectly impacts on fouling.

Flux and Critical Flux

Flux selection provides the most significant factor in determining fouling rate. At high convection rates towards the membrane (i.e., at high flux), colloidal aggregation and heterogeneous deposits are observed. Rapid reversible fouling then takes place, predominantly through formation and compaction of the cake layer produced by the flocculant biomass material. Internal and/or irreversible will also take place more rapidly at higher fluxes, more or less in agreement with theoretical predictions. On the other hand, it has been reported that fouling is not observed provided the flux is maintained below some specific value. It is the evaluation of this so-called *critical flux* which forms the focus of many submerged MBR studies.

The critical flux concept was introduced by Field et al. (1995), and has since been cited in many papers concerned with the fouling limitation in MBR operation. The hypothesis for MF is that on start-up there exists a flux, the critical flux, below which a flux decline does not occur. Its value can be taken as being equal to that of either (a) the clean water flux under the same overall conditions (Fig 3) or (b) some other sustainable flux. Operation below the critical flux is called subcritical flux operation or non-fouling operation (Howell 1995), and is expected to lead to little or even no irreversible fouling.

The first definition of critical flux relates to the flux-value where the solution permeability deviates from the pure water permeability (Fig. 3). Only few authors (Kwon and Vigneswaram 1998; Huisman et al. 1999) employ this method or definition, since MBR permeability is always smaller than the equivalent clean water one. A more common practice is to incrementally increase the flux for a fixed duration for each increment, which leads to a stable TMP, at low fluxes, and the TMP starts to increase with time for more significant fluxes (Fig. 4). The TMP increase indicates a greater resistance to permeation provided by a growing polarization layer, cake formation, or fouling (Chen et al. 1997). The highest flux for which TMP remains stable is the critical flux, and is dependent on parameters such as MLSS, membrane material, and system hydrodynamics (Table 4).
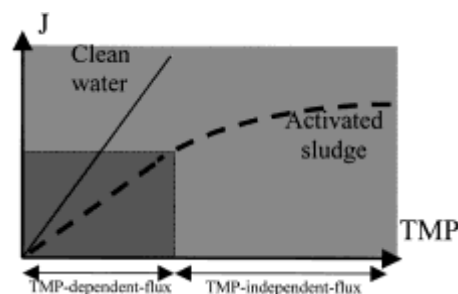
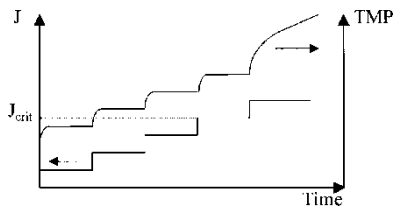**Fig. 3.** Two filtration operating zones

Fig. 4. Typical flux data using flux step method

While the critical flux concept has proved an invaluable tool in conventional membrane process design, its validity is questionable in MBR processes where fouling rates only approach zero at very low flux and, ultimately, impractical values. It has recently been observed that sustained periods of operation at very low fouling rates, very close to the ''critical flux,'' can lead to an initially slow linear increase in TMP which eventually becomes rapid and catastrophic (Wisniewski et al. 2000).

At low flux, visual observation of the membrane with a microscope confirms the absence of floc deposition. However, it has been shown that the composition of the adsorbed material on the membrane was very close to that of the supernatant of the mixed liquor (Defrance et al. 2000). The small but linear TMP increase observed at low fluxes thus appears to arise from deposition of the solute and colloidal fractions of the sludge, which are likely to interact with the membrane in the incipient stages of filtration. Operating under critical flux conditions does not appear to nullify insidious irreversible fouling by species possibly originating from lysed cells (e.g., fine colloids and macromolecules).

Washing, Cleaning, and Pretreatment
The successful operation of a MBR plant requires careful management of fouling, since its complete avoidance is not possible. Recent improvements in fouling control have led to more favorable projections of membrane life, significantly decreasing overall costs.

Membrane cleaning comprises intermittent physical cleaning (usually backwashing) and periodic chemical cleaning. Chemical cleaning is expected to completely recover membrane flux, but produces toxic or contaminated wastewater because most cleaning agents are caustic and/or contain detergents and oxidizing agents such as hypochlorite (Baker and Dudley 1998; Cicek et al. 1998b; Ragona and Hall 1998; Tardieu et al. 1998; Roberts et al. 2000). Acid cleaning is often proposed for ceramic membranes (Fan et al. 1999). On the other hand, physical methods can produce a stable flux without secondary chemical contamination but are more frequent and generally require more energy. Successful membrane cleaning procedures generally employ some combination of two techniques, with some workers experimenting with more advanced mechanical methods, such as agitator-induced flushing (Ahn and Song 2000).

For submerged aerobic MBRs, intermittent suction provides an alternative method for suppression of fouling (Yamamoto et al. 1989; Chiemchaisri et al. 1993; Liu et al. 2000). Temporary cessation of suction creates back transport of permeate which then helps to dislodge the cake layer. Intermittent filtration has also been shown to improve the hydraulic performance of sidestream MBRs, both for aerobic (Defrance and Jaffrin 1999b) and anaerobic (Choo and Lee 1996a; Wen et al. 1999a,b) systems. In one reported case, partial cleaning of the membrane was achieved through its intermittent collapsing under reduced feed pressure (Till et al. 1998).

It is common practice in submerged hollow fiber (HF) systems to periodically backflush the membrane, i.e., pump permeate back through the membrane into the feed channel, to remove the deposited cake layer (Bouhabila et al. 2001). The effectiveness of this technique depends on the nature of the fouling mechanism. If pore blocking has occurred or the cake layer is strongly adhered, it may be fairly ineffective. Inorganic materials precipitated on the membrane surface and pore walls—such as calcite and struvite—are not readily removed by backwashing (Yoon et al. 1999).

Backflushing with air through the membrane is often employed for aerobic MBRs (Chiemchaisri et al. 1992; Scott et al. 1998; Choo and Stensel 2000). Air sparging prevents the compaction of the cake layer and reducing the internal pore clogging of the membranes. By employing a 15 min filtration cycle with a 15 min air backwashing cycle at 1.5 bar air pressure, Parameshwaran et al. (1999) demonstrated a 90% improvement in flux compared with continuous suction.

Addition of powdered activated carbon (PAC) to the MBR has been shown to increase permeability by improving the hydraulic properties of the cake, principally through increasing both its bulk permeability and reducing its compressibility (Kim et al. 1998). In addition, PAC addition contributes to an increase in the biosolids back-transport velocity (Park et al. 1999), thereby reducing the cake thickness. PAC is also thought to reduce internal fouling by direct and rapid adsorption of dissolved foulants onto the carbon surface. For treatment of landfill leachate, the permeate flux was reduced almost to zero within the first hours of operation when no PAC was employed, whereas on addition of 1% PAC to the bioreactor the permeate flux improved up to 24 L m^{-2} h^{-1} (Pirbazari et al. 1996).

Membrane Characteristics

It is well known that membrane characteristics such as pore size, porosity, surface energy, charge, roughness, and hydrophilicity/hydrophobicity, etc., have a direct impact on membrane fouling. Effects of pore size on membrane fouling strongly depend on the feed solution characteristics, in particular, particle size distribution. Shimizu et al. (1990) correlated the flux with the pore size for sidestream MBR treatment of methanogenic wastes. The au-

floc size distribution nor EPS concentration are obtained or even inferred from the MLSS measurement, which in the majority of cases is the sole mixed liquor quality parameter reported. This may partly explain why widely ranging correlations for MLSS and flux have been reported, with the MLSS exponent values ranging from 0.5 to 1.

A further complication to fouling characterization the change in the physical, chemical, and physiological characteristics of the mixed liquor both with feedwater quality and with time, such that the extent to which these characteristics can be

Table 4. Critical Flux Values Determined with the Stepwise Method

MBR	Membrane (pore size in μm)	MLSS (g L^{-1})	Hydrodynamics	Step length (min)	Critical flux (L m^{-2} h^{-1})	Reference
SS–TB	Millipore GVWP (0.22)	4	CFV = 1 m s^{-1} Re = 2300	30	60	Madaeni et al. (1999)
Sub–HF	Zenon Polymer (0.1)	14.8	Airflow rate = 1500 L h^{-1}	80	30	Bouhabila et al. (1998)
SS–TB	Millipore GVHP (0.22)	2.5	Re = 2520	30	22	Cho et al. (1999)
SS–TB	Kerasep, Techsep Ceramic (0.1)	10	CFV = 4 m s^{-1}	60	95	Defrance and Jaffrin (1999b)
SS-TB	Ceramic (MWCO = 300 kDa)	8	CFV = 0.5 m s^{-1}	—	<12.5	Tardieu et al. (1998)

thors showed that 0.05–0.2 μm pore sized membranes produced the maximum flux among membranes ranging from 0.01–1.6 μm in pore size. Larger pore size does not always lead to greater flux due to internal fouling. For example, Chang et al. (1994), investigating the effect of pore size on flux from alcohol-distillery wastes, found the flux produced from 0.05 μm pore size to be higher than that from 0.4 μm membrane for otherwise comparable filtration conditions. Choo and Lee (1996b) determined the optimum pore size based on the particle size distribution of anaerobic broth. These reports all emphasize the importance of pore clogging by fine particles during membrane filtration.

Available membrane materials comprise ceramic or polymeric. Ceramic materials such as aluminum, zirconium, and titanium oxide (Al_2O_3, ZrO_2, and TiO_2, respectively), show superior hydraulic, thermal, and chemical resistance, as indicated by the permeability data referring to a TMP of around 2 bar produced by Gyhoot and co-workers (Table 2). Although application of ceramic membranes to aerobic or anaerobic MBR has been studied (Shimizu et al. 1989; Ahn et al. 1998; Scott et al. 1998; Cicek et al. 1999a,b; Defrance and Jaffrin 1999a; Tardieu et al. 1999; Wen et al. 1999a,b; Yoon et al. 1999; Defrance et al. 2000), polymeric membranes are more commonly used due to the expense of the ceramic materials, which are also largely limited in geometry to tubular monoliths.

Several studies have demonstrated the importance of hydrophobicity of membrane materials. It is known that hydrophilic membranes yield the higher fluxes because of the hydrophobic nature of the interaction between the membrane and biomass (Chang et al. 1999; Futumura et al. 1994; Madaehi et al. 1999). This demands that the naturally hydrophobic polymeric materials, such as polyethylene, polypropylene, polyvinilydene fluoride, and polysulfone, are surface modified with some hydrophilic functional group (Knoell et al. 1999; Wilkes et al. 1999; Wang et al. 2000). Unmodified and modified (with hydrophilic monomers) polyethersulfone membranes can be compared, and reveal that the modified membrane presents a 25% increase in hydrophilicity, a 49% decrease in (bovine serum albumin) biofouling, and a 4% increase in albumin retention compared with the unmodified membrane (Pieracci et al. 1999).

Conclusions

Fouling is a common problem in membrane processes but is made more difficult to predict and control in a MBR due to the highly heterogeneous nature of the bioreactor mixed liquor, and, in particular, the effect of the active microorganisms (i.e., the biomass). The latter generate products, collectively termed extracellular polymers (EPS), of high fouling propensity. Of key significance with respect to fouling, therefore, is the EPS concentration and speciation, since different chemical components of the EPS have different fouling propensities. Fouling is also affected by the floc size and size distribution, with smaller flocs being generated in sidestream systems due to the shear created by the pump. Neither

determined by adoption of appropriate operating conditions is limited. In general, an increase in SRT and/or organic loading rate increases MLSS concentration and impacts on EPS levels, though not in a predictable way as far as the latter is concerned. Opera- tion at high MLSS levels is generally desirable since this implies reduced sludge yield and a smaller footprint, but an excessive MLSS produces a prohibitively high viscosity which inhibits mass transfer. As with all membrane processes, turbulence, as promoted by aeration in submerged MBRs or pumping to produce somewhat higher effective cross-flow velocities in sidestream sys- tems, increases mass transfer and reduces fouling as manifested in the higher value of the critical flux (the flux at which no fouling is observed). However, identification of critical flux itself appears problematic due to recently noted long-term irreversible fouling behavior in submerged MBR processes.

While many physical cleaning techniques have been recom- mended complete abatement of fouling appears only possible through periodic application of chemicals, even for low-flux sub- merged systems employing high-shear slug flow aeration. It is apparent from this review that a more optimal design and opera- tion of MBRs is likely to come about as a result of an improved understanding of the interaction between operating conditions, in- cluding backflushing, and the behavior of key foulants.United Utilities, and Yorkshire Water.

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