03-166

Environment Canada
Water Science and Technology Directorate

Direction générale des sciences et de la technologie, eau
Environnement Canada

Biofouling and Its Control in Membrane Separation Bioreactors: A review

By:
B. Liao, D. Bagley, H. Kraemer, G. Leppard, S. Liss

NWRI Contribution # 03-166

03-166

Biofouling and Its Control in Membrane Separation Bioreactors: A Review

B.Q. Liao, D.M. Bagley, H.E. Kraemer, G.G. Leppard and S. N. Liss

Abstract

Membrane separation technology is increasingly becoming an important innovation in biological wastewater treatment. Biofouling of the membrane is a major factor impacting the efficient and economic operation of membrane separation bioreactors (MBRs). This review summarizes the state-of-the-art progress in understanding the mechanisms and factors affecting membrane biofouling and the strategies for biofouling control. Biofouling mechanisms include the adsorption of soluble/suspended extracellular polymers on membrane surfaces and in membrane pores, the clogging of membrane pore structure by fine colloidal particles and cell debris, and the adhesion and deposition of sludge cake on membrane surfaces. Design and operating conditions of membrane modules, membrane materials, hydrodynamic conditions in MBR, process and environmental conditions of activated sludge systems and the physicochemical properties of the wastewater are the dominant factors determining membrane biofouling. Current strategies to control biofouling include periodic relaxation, backwashing, chemical cleaning and possible manipulation of hydrodynamic conditions and sludge properties. Achieving full integration of MBRs in wastewater treatment technology requires further research and development. Fundamental information on the bacteria/colloid/membrane interaction, developed through multi-method and multi-scale approaches, is particularly needed.

NWRI RESEARCH SUMMARY

Plain language title
What is the Nature of Biofouling in Water Treatment Apparatus and How Can We Minimize the Problem for the Case of Membrane Filters: A Review

What is the problem and what do sicentists already know about it?
Biofouling of water treatment apparatus poses a serious economic problem. Progress in addressing the problem is impeded by a lack of fundamental information on the biofoulants, which are mainly bacteria and their colloidal secretions.

Why did NWRI do this study?
This sub-study evolved, as one of many sub-studies, from a joint NWRI/NSERC/environmental industry/university arrangement instituted in 1997. The university partners were McMaster, Toronto and Ryerson. The major funding initially was 326 K$ coming from NSERC, supplemented by "in kind" contributions totalling 904 K$ from the other partners. This sub-study was part of a major effort to understand (1)

what flocs and biofilms do in water treatment tanks, and (2) how to manipulate their activities for maximum cost-effectiveness.

What were the results?

1. We identified a specific scientific problem of considerable importance and addressed it with new technology developed by us. Fundamental aspects of the bacteria/colloid/membrane interaction, for the case of membrane separation bioreactors, are now amenable to analysis by multi-method multi-scale approaches placed in the literature by NWRI expertise. We prepared our review to show how our work fits into a general scheme of biofouling research.

How will these results be used?

1. These results will be used to improve ongoing research programs in many organizations.

Who were our main partners in the study?

1. Natural Sciences and Engineering Research Council of Canada, National Water Research Institute, McMaster University, Hamilton, Dept. of Biology, University of Toronto, Dept. of Chemical Engineering and Applied Chemistry, Ryerson University, Toronto, Dept. of Applied Chemical and Biological Sciences, Eight Canadian industries

L'encrassement biologique dans les bioréacteurs à séparation membranaire et les moyens de le limiter : une revue

B.Q. Liao, D.M. Bagley, H.E. Kraemer, G.G. Leppard et S. N. Liss

Résumé

La séparation membranaire est une innovation qui prend de plus en plus de place dans le traitement biologique des eaux usées. L'encrassement biologique des membranes est l'un des principaux facteurs qui se répercutent sur l'efficacité et les coûts d'exploitation des bioréacteurs à séparation membranaire (BSM). La présente revue résume les plus récents progrès dans la compréhension des mécanismes et des facteurs à l'origine de l'encrassement des membranes, ainsi que les stratégies permettant de le surveiller. Les mécanismes d'encrassement comprennent l'absorption de polymères extracellulaires en solution ou en suspension par les surfaces et les pores des membranes, l'obturation des pores par des particules colloïdales ou cellulaires fines, et l'adhésion et le dépôt de gâteaux de boue sur la surface des membranes. La construction et les conditions d'utilisation des membranes et de leurs matériaux constituants, les conditions hydrodynamiques dans les BSM, les conditions de traitement et les conditions ambiantes des systèmes de traitement par boues activées, et les propriétés physicochimiques des eaux usées sont les principaux facteurs d'encrassement des membranes. Les stratégies actuelles de lutte contre l'encrassement comprennent les relaxations périodiques, le lavage, le nettoyage chimique et, peut-être, des modifications des conditions hydrodynamiques et des propriétés des boues. Une intégration complète des BSM à la technologie de traitement des eaux usées exigerait plus de recherche et de développement. On a surtout besoin d'une information de base sur l'interaction bactéries-colloïdes-membrane, information qu'il faudrait obtenir par des approches multidisciplinaires et multi-échelles.

Sommaire des recherches de l'INRE

Titre en langage clair
La nature de l'encrassement dans les équipements de traitement des eaux usées et comment on peut atténuer le problème dans les filtres à membrane : une revue.

Quel est le problème et que savent les chercheurs à ce sujet?
L'encrassement des équipements de traitement des eaux usées pose un grave problème économique. Les progrès visant à résoudre ce problème sont ralentis par un manque d'information de base sur les agents d'encrassement, qui sont principalement les bactéries et leurs sécrétions colloïdales.

Pourquoi l'INRE a-t-il effectué cette étude?
À l'instar de nombreuses autres, la sous-étude actuelle est le résultat d'une entente entre l'INRE, le CRSNG, l'industrie de l'environnement et le monde universitaire, qui remonte

à 1997. Les partenaires universitaires étaient McMaster, Toronto et Ryerson. Initialement, le principal financement est venu du CRSNG (326 k$), les autres partenaires ayant fait des contributions « en nature » totalisant 904 k$. La sous-étude faisait partie d'un effort important pour comprendre 1) l'effet produit par les flocs et les films biologiques dans les réservoirs de traitement des eaux usées et 2) comment moduler leurs activités pour maximiser la rentabilité.

Quels sont les résultats?

Nous avons identifié un problème scientifique particulier d'une grande importance et l'avons résolu grâce à une nouvelle technologie que nous avons mise au point. Certains aspects fondamentaux de l'interaction bactéries-colloïdes-membrane dans les bioréacteurs à séparation membranaire peuvent maintenant être analysés au moyen d'approches multidisciplinaires et multi-échelles élaborées et publiées par l'INRE. Nous avons préparé notre revue dans le but de montrer comment nos travaux s'intègrent à la recherche sur l'encrassement.

Comment ces résultats seront-ils utilisés?

Ces résultats seront utilisés pour améliorer les programmes de recherche courants de nombreux organismes.

Quels étaient nos principaux partenaires dans cette étude?

Conseil de recherches en sciences naturelles et en génie (CRSNG); Institut national de recherche sur les eaux (INRE); université McMaster, Hamilton, Dept. Of Biology; université de Toronto, Dept. of Chemical Engineering and Applied Chemistry; université Ryerson, Toronto, Dept. of Applied Chemical and Biological Sciences; huit industries canadiennes.

Introduction

The integration of membrane separation technology into conventional activated sludge processes for biomass separation represents an important innovation in the evolution of biological wastewater treatment technology. Membrane separation bioreactors (MBRs) operate without secondary clarification. MBRs offer several advantages over the conventional activated sludge process, including a high biomass concentration, a compact footprint, better uncoupling of the hydraulic retention time (HRT) and sludge retention time (SRT), low sludge production, resistance to shock loading, and, most importantly, superior permeate quality (Brindle and Stephenson, 1996; Manem and Sanderson, 1996; Yamamoto *et al.*, 1997; Van Dijk and Roncken, 1997; Gander *et al.*, 2000; Stephenson *et al.*, 2001).

The primary role of the membrane is to retain solids. The membrane pore size, consequently, is in the range of ultrafiltration and microfiltration, typically 0.04-0.45um. The performance of a membrane is determined by either its flux at a given trans-membrane pressure (TMP), where a greater flux is desirable, or by the TMP at a specific flux, where a lower TMP is better. MBRs can be generally classified into two categories: submerged MBRs and external crossflow MBRs (Heiner and Bonner, 1999; Gander *et al.*, 2000). In submerged MBRs the membrane modules are directly placed in the aeration tank and the permeate exits the aeration tank either by vacuum pump or by gravity (Stephenson *et al.*, 2001). For crossflow MBRs, a recirculating pump is used to transfer the activated sludge from the aeration tank to the membrane module where the biomass is separated. Water permeates from inside to outside of the module and the concentrated biomass returns to the aeration tank. At present, there are more than 500 MBR plants in the world treating municipal and industrial wastewaters (Stephenson *et al.*, 2001). Submerged MBRs are the most common systems employed for the treatment of municipal wastewaters (Manem and Sanderson, 1996; Adham *et al.*, 1999; Gander *et al.*, 2000; Stephenson *et al.*, 2001). Crossflow MBRs are mainly used for small-scale wastewater treatment applications (e.g. commercial buildings). Owing to the high-energy consumption rate of these systems larger scale applications are unlikely.

Membrane fouling, particularly biofouling, represents a significant limitation to the wide spread application of MBRs. Biofouling results in reduced performance, severe flux decline, high energy consumption, and frequent membrane cleaning or replacement (Manem and Sanderson, 1996; Nagaoka *et al.*, 1996; Nagaoka *et al.*, 1998; Tardieu *et al.*, 1996, 1998 and 1999; Wisniewski and Grasmick, 1997 and 1998; Gander *et al.*, 2000; Stephenson *et al.*, 2001; Wisniewski, 2001). The following review examines the current understanding of the mechanisms and factors associated with membrane biofouling. Strategies for controlling membrane fouling are explored, and the requirements to better control biofouling in MBRs are proposed and discussed.

Mechanisms of Biofouling in a Membrane Separation Bioreactor

Fouling is a generic term that is closely associated with deterioration in membrane performance. In practice, where flux is held constant, fouling results in an increase in TMP. Fouling can be attributed to an inorganic foulant (e.g. iron) that relates to the constituents present in the wastewater entering the MBR. Because the MBR membrane is designed to allow soluble constituents to permeate, inorganic fouling due to precipitation and concentration gradients, as is common in reverse osmosis membrane, may not be a dominant mechanism. Biofouling is specifically related to the interaction of biosolids with the membrane. This could consist of formation of biofilms or the accumulation of bioorganic material, including extracellular polymeric substances (EPS), on the membrane surface. Due to the nature of wastewater and the contents of mixed liquor suspended solids, composite fouling (i.e. a combination of biofouling and inorganic fouling) (Sheikholeslami, 1999) often occurs in MBRs. Biofouling, though, is a significant phenomenon due to the high biomass concentration in the bioreactor.

Biofouling is the result of interactions between the membrane surface and those components of the biomass or sludge consisting of bacterial cells, or aggregates, and EPS. During permeation, the mixed liquor and soluble components in water are transported to the membrane surfaces where a concentration polarization occurs due to the dewatering process near the boundary layer of the membrane surface. Based on a few studies (Choo and Lee, 1996; Tardieu *et al.*, 1996 and 1998; Roorda *et al.*, 2000; Wisniewski, 2001), biofouling can be characterized on the basis of three fouling patterns (Figure 1): adsorption of EPS to the membrane surface; pore clogging by cells; and sludge cake, or film, formation arising from deposition of cells or aggregates. All three mechanisms are likely to be involved. However, the relative importance of each mechanism is site specific and may depend on a number of factors.

The presence of soluble/suspended EPS leads to the accumulation of this material on membrane surfaces and within the pore structure (Nagaoka *et al.*, 1996; Nagaoka *et al.*, 1998 and 2000; Chang and Lee, 1998; Mukai *et al.*, 1999). This may change the friction factor in the flow channels and a decrease in the flow area, which leads to greater TMP. Both physical and chemical adsorption of EPS may occur during the permeation process. Physical adsorption involves weak interactions, which may be just a simple deposition, between EPS and membrane surfaces, and thus EPS can be removed by the shear stress of fluid mechanics, like air scouring and liquid flow on membrane surfaces. On the other hand, chemical adsorption, involving greater adhesion strength and energy between EPS and membrane surfaces, is more resistant to shear stress. Consequently, chemical agents, like acid, base and oxidants, must be used to remove chemically adsorbed EPS. Irreversible biofouling by EPS may also occur depending on the nature of interactions.

Pore clogging may occur due to cell debris and colloidal particles (Karr and Keinath, 1978; Urbain *et al.*, 1993) found in mixed liquor, whose sizes are similar to the membrane pore. During permeation these fine particles can become entrapped in the pores and thus reduce the surface area for filtration. The pore clogging decreases the flow area of the permeate resulting in a greater TMP to achieve the same flux. A number of studies have found that pore clogging may be the dominant biofouling mechanism. Backwashing and chemical cleaning are primarily the ways to reduce membrane pore clogging (Bouhabila *et al.*, 1998 and 2001; Tardieu *et al.*, 1996, 1998 and 1999; Wisniewski and Grasmick, 1997 and 1998; Gan, 1999).

Adhesion or deposition of sludge cake on membrane surfaces may also contribute to membrane biofouling (Kiat *et al.*, 1992; Ozaki and Yamamoto, 1996 and 1997; Pound *et al.*, 1997; Bouhabila *et al.*, 1998 and 2001; Tardieu *et al.*, 1996, 1998 and 1999; Wisniewski and Grasmick, 1997 and 1998; Wisniewski, 2001). In the region close to the membrane surface, a concentration gradient is expected as a result of dewatering effects. This leads to a higher MLSS at the surface of the membrane than in the bulk phase. If the mass flow of mixed liquor to the membrane surface is larger than the back transport of mixed liquor from the surface, sludge cake formation or deposition is anticipated. The transport mechanisms describing the formation of sludge cake on membrane surfaces includes shear-induced diffusion, inertial lift and surface transport (Tardieu *et al.*, 1998). Backwashing plays a major role in controlling the fouling process (Tardieu *et al.*, 1998). Air scouring in submerged MBRs is usually used to generate an adequate shear stress for removing sludge cakes from membrane surfaces, while the shear stress caused by the high flow velocity of mixed liquor in crossflow MBRs reduces sludge deposition at surfaces.

Factors Which Affect Membrane Biofouling

A number of factors may affect membrane biofouling in MBRs, as shown in Figure 2. These factors include the hydrodynamic conditions in MBRs, membrane module design (which in turn affects the hydrodynamic conditions in MBRs), the operating conditions of the biological process and membranes, membrane materials, and the physicochemical properties of the sludge. The physicochemical properties of the sludge will be directly influenced by the operating conditions of the treatment process (e.g. hydrodynamic conditions and solids retention time) resulting in subsequent impacts on membrane fouling (Liao et al., 2001).

Hydrodynamic Conditions

Hydrodynamic conditions can impact on the performance of MBRs through shear stress and air scouring. The flow velocity of mixed liquor in crossflow MBRs and the aeration intensity and time in submerged MBRs are critical parameters that can influence these phenomena and the performance of MBRs (Heiner and Bonner, 1999).

Ozaki and Yamamoto (1996 and 1997) investigated the impact of hydraulic conditions on sludge accumulation in submerged MBRs and identified that the shear rate is the main hydraulic factor that affects sludge accumulation. Shear stress caused by the cross flow velocity and the recirculating pump is crucial in controlling biofouling in the crossflow MBRs, as the shear stress induced by the recirculating pump affects not only the formation of sludge cake but also floc structure (Tardieu *et al.*, 1996, 1998 and 1999; Wisniewski and Grasmick, 1997 and 1998). At a low flow velocity of 0.5-2.0 m/sec, sludge cake layer can quickly form on membrane surfaces. A high flow velocity greater than 4 m/sec appears to prevent sludge deposition on membrane surfaces (Tardieu *et al.*, 1998). However, shear stress from the recirculating pump breaks up the flocs, which negatively impacts membrane flux (Wisniewski and Grasmick, 1998). This is further supported by a recent study from Korea (Kim *et al.*, 2001) that reported shear stress caused by the recirculating pumps affects membrane biofouling. The vane-type rotary pump imposed a much stronger shear than the turbine-type centrifugal pump and thus resulted in a much smaller floc size. The net result of shearing sludge flocs is an increase in fouling potential. Consequently, a low shear pump is recommended for the crossflow MBR process.

There have been extensive studies in Japan, Europe and North America on the impact of aeration intensity on the performance of submerged MBRs. It is generally believed that shear stress caused by the up flow of an air-water mixture on the membrane surfaces can be optimized to reduce membrane biofouling in submerged MBRs. Pound *et al.* (1997) found that the turbulence induced by the air bubbles from the aerators causes the membranes to collide and produce a self-cleaning effect. Furthermore, Ueda *et al.* (1997) reported that the air flow rate determines the sludge cake thickness in submerged MBRs. There appeared to be an optimum value above which no further decrease in sludge cake thickness was observed. When the air flow rate decreased from 700 to 350 L/min, the TMP increased from 0.2 to 0.5 bar in 20 hrs and an increased sludge cake accumulation on membrane surfaces was observed. Cake removal efficiency was improved either by increasing the air flow rate or by augmenting aeration intensity by concentrating membrane modules over a smaller floor area. More recently, in France, Bauhabila *et al.* (1998 and 2001) investigated the impact of air flow rate on the performance of hollow fiber membranes. An increase in the air flow rate from 1.2 to 3.6 $m^3/m^2/hr$ resulted in a decrease in total filtration resistance and thus increased the membrane flux by a ratio of 3. These results indicate that air scouring on the membrane surface is important in preventing sludge accumulation. A recent survey on the application of MBRs for municipal wastewater treatment (Adham *et al.*, 1999) indicates that the specific design of aerators, the location of the aerators and the air flow patterns are crucial for reducing membrane biofouling due to sludge cake formation, as these factors affect the hydrodynamic conditions in MBRs.

Module Configuration

Module design plays a major role in determining the performance of MBRs by affecting the hydrodynamic conditions in the MBR, which in turn influences the fouling rate. The packing density of hollow fibres and flat sheets, the choice of aerator and the specific location of aerator under the membranes are critical design parameters for MBRs (Adham *et al.*, 1999). Kiat *et al.* (1992) found that solids did not accumulate in the space between fibres when fibre packing density was lower than a critical value. Maximum permeate flux and productivity were obtained below and at the critical packing density, respectively. Significant solids accumulation and flux decline were observed above the critical packing density. More recently, Ozaki and Yamamoto (1996 and 1997) found that the module length and distance between membranes affect sludge accumulation on membrane surfaces. Shorter distances between membranes resulted in sludge accumulating in a shorter time. This was explained by the increase in the frequencies of collision of flocs over a shorter distance between membranes.

Innovations in the modification and optimization of module design include the reduction in leaf length, spacing adjustments, packing density, the development of new aerators, and even the location for the permeate output and aerator. An important improvement in module design is the utilization of gravity force to replace the vacuum pump to suck permeate (Kubota, Japan; Millenniumpore Ltd, UK). This design significantly reduces energy consumption.

Membrane Flux and Operating Cycle Length

Membrane permeating flux determines the mass flow of mixed liquor to membrane surfaces and thus affects sludge cake formation. It has been generally assumed that fouling should be greatly reduced when the membranes are operated at a permeate flux below a critical level determined by the magnitude of the sum of back-transport mechanisms (Field *et al.*, 1995; Howell, 1995; Tardieu *et al.*, 1998; Fan *et al.*, 2000; Fane *et al.*, 2000; Gander *et al*, 2000). However, the critical value of flux is very much system specific. The typical membrane operating flux in MBR plants for municipal wastewater treatment is about 15 gfd (Adham *et al.*, 1999).

Madaeni *et al.* (1999) found that the critical flux depends on sludge concentration and crossflow velocity. A higher critical flux is related to a higher crossflow velocity and lower sludge concentration. This is not surprising, as both a higher flow velocity and a lower sludge concentration reduce membrane biofouling. A higher critical flux is also related to a more hydrophilic membrane surface. More recently, Fan *et al.* (2000) found that membrane fouling is a function of the permeate flux and a higher permeate flux resulted in a shorter membrane cleaning interval.

The operating cycle length is important in determining the rate of membrane biofouling. Defrance and Jaffrin (1999) tested a hydrodynamic method for fouling control using intermittent filtration and backwashing. The cyclic operation consisted of alternating short periods of filtration (1-4 s) and short periods of backwashing (1-2 s) at low TMP and high velocity. The quick switch between filtration and backwashing yielded a 20% increase in permeate flux and a 10% energy reduction, compared to the conventional operating manner of MBRs with continuous filtration.

Membrane Materials and Properties

Membrane materials and properties have long been considered important factors that can affect membrane fouling. The properties include the hydrophobicity, surface charge, pore size and roughness. Presently, both tubular ceramic membrane and polymeric membranes have been used for MBRs. However, there is no direct comparative study between those two membrane materials. There is a particular lack of information regarding the role of polymeric membrane materials and properties on membrane fouling in MBRs.

Chang and Lee (1999) reported for batch filtration tests a lower fouling rate that related to a more hydrophobic membrane. It is possible to increase the hydrophilicity of a membrane via surface modification. Sainbayar *et al.* (2001) were able to increase the hydrophilicity of a membrane modified by UV grafting and this resulted in a decrease in the biofouling potential. Gan (1999) reported that there was a significant difference in the start-up transient flux with respect to pore size. A larger flux decline rate was related to a smaller pore size. However, the difference in flux diminished progressively with permeation time and smaller pore size membranes reached a stable condition earlier. This was probably due to the fact that more severe in-pore fouling might occur in larger pores. Madaeni *et al.* (1999) found that the critical flux of activated sludge is similar for different membrane pore sizes but is dependent on surface hydrophobicity.

There is only limited information on the importance of membrane surface charge with respect to biofouling. Shimizu *et al.* (1989) investigated the impact of aluminum membrane surface charge on the filtration characteristics of methanogenic waste using three types of charged aluminum membranes. It was found that the negatively charged membrane exhibited a higher filtration flux than that of the non- and positively charged membranes. The impact of membrane charge was mainly reflected by its importance in controlling electrostatic adsorption and pore clogging. No impact of membrane surface charge on sludge cake formation was observed.

Mixed Liquor Suspended Solids

The impact of mixed liquor suspended solids (MLSS) on sludge dewatering is well known for conventional sludge dewatering processes. Analogous to this is the dewatering of sludge that occurs on the surface of a membrane in MBRs. The influence of MLSS concentrations on the performance or fouling of MBRs has been studied in both laboratory and full-scale plants (Magara and Itoh, 1991; Sato and Ishii, 1991; Nagaoka *et al.*, 1996; Adham *et al.*, 1999). A higher MLSS correlates to a higher TMP or a lower permeate flux due to severe sludge cake formation on the membrane surfaces. This leads to the reduction of membrane permeability. Typical MLSS concentrations in MBRs fall in the range of 10-30g/L with a median value of 10g/L for submerged MBRs (Adham *et al.*, 1999).

Magara and Itoh (1991) found that a higher MLSS concentration is related to a lower permeate flux. A 10% variation in MLSS resulted in a 5% variation in permeate flux. Sato and Ishii (1991) developed an empirical model to correlate the membrane filtration resistance to the MLSS, soluble COD, viscosity and TMP. MLSS was found to have the second largest effect on total filtration resistance after the soluble COD concentration in the supernatant. More recently, using a pilot-scale submerged MBR, Nagaoka *et al.* (1996) found that a decrease in MLSS was related to a decrease in the filtration resistance (a decrease in TMP). This is because the increase in MLSS caused an increase in viscosity.

It has been reported that MLSS ranging from 3.5 to 7.0 g/L had little impact on membrane fouling (Fan *et al.*, 2000), implying that a higher MLSS does not cause more serious membrane fouling. It is more likely, however, that this range was below the critical MLSS levels that may lead to sludge cake formation. At these MLSS levels back transport mass of mixed liquor is always larger than the mass flow of mixed liquor to the membrane surfaces. Above the critical concentrations the net accumulation of sludge cake on membrane surfaces is proportional to the MLSS concentration.

Process and Environmental Conditions of Activated sludge (SRT, F/M, Nutrient Conditions)

Process and environmental conditions of the activated sludge system are known to play an important role in controlling sludge properties (Chao and Keinath, 1979; Andreadakis, 1993; Liao *et al*., 2001). These conditions include solids retention time (SRT), hydraulic retention time (HRT), F(food)/M(microorganism) ratio and nutrient conditions. Consequently, an influence of process and environmental conditions on the performance of MBRs is anticipated, as they affect the nature of the foulants.

Recently, a number of studies have been conducted to elucidate the role of SRT and F/M in controlling membrane biofouling (Chang and Lee, 1998; Fan *et al*., 2000; Nagaoka *et al*., 2000; Bouhabila *et al*., 2001). Using membranes with different hydrophobicity and pore sizes, Chang and Lee (1998) found that an increase in SRT from 3 and 8 days to 33 days resulted in a significant increase in the sustainable membrane flux. This could be attributed to a lower level of EPS content at higher SRTs. A similar conclusion was reached in the study of Fan *et al*. (2000) at three different SRTs (20, 10 and 5 days). It was reported that there was no need for membrane cleaning at an SRT of 20 days for more than 70 days of operation; on the other hand, a cleaning interval of 3-5 days was required for an SRT of 5 days (Fan *et al*., 2000). Nagaoka *et al*. (2000) found that the membrane fouling rate at a high F/M rate (1.5 g/L/day) was much higher than that at a low F/M rate (0.3g/L/day), due to the higher extracellular polymeric substances (EPS) content at the higher F/M loading.

There are also some studies concerning the impact of environmental and biological conditions on membrane fouling. Chang and Lee (1998) found that a foaming sludge had a much higher filtration resistance than a non-foaming sludge, mainly due to the cake resistance caused by foaming sludge. Nitrogen-deficient sludge produced less EPS and thus had a sustainable membrane flux 30-40% higher than the control sludge produced under non-nutrient-limiting conditions.

Because the foulant or biofouling originates from diverse components of activated sludge broth, it is not surprising to see the importance of sludge properties in controlling biofouling. The content and rate of membrane biofouling depend on the type and strength of interactions between membrane surfaces and each component of activated sludge broth. Until now, studies have been conducted on the floc size, EPS and viscosity, but there is little information about the role of sludge surface charge and hydrophobicity in controlling biofouling.

Particles and Colloids

Floc size has long been considered as a dominant factor in controlling sludge dewatering (Karr and Keinath, 1978). The same is true for MBRs. It is generally believed that floc size in MBRs is smaller than that in the conventional activated sludge (Zhang *et al.*, 1996; Heiner and Bonner, 1999). Recently, the influence of different fractions of the sludge on the membrane fouling has been studied (Chang and Lee, 1998; Tardieu *et al.*, 1998 and 1999; Wisniewski and Grasmick, 1998). All studies concluded that the soluble COD fraction of activated sludge broth plays a significant role in determining membrane fouling, followed by the influence of colloidal components. The larger particles were less important in determining the filtration resistance of the membrane. This is probably not surprising, as the back-transport velocity of smaller components, like colloids and solutes, is lower than that for the larger particles. More recently, Bouhabila *et al.* (2001) investigated the impact of the three fractions (suspended solids, colloids and solutes) of sludge on membrane fouling at different SRTs (10, 20 and 30 days). At all SRTs, it was clear that colloids and solutes were the dominant fractions controlling filtration resistance.

Extracellular Polymeric Substances

The importance of EPS in controlling membrane fouling has been extensively studied. A larger EPS content is related to a higher fouling rate. Sato and Ishii (1991) found that soluble/suspended EPS had the largest effect on the filtration resistance and was even more important than the MLSS. The presence of a large quantity of extracellular matrix on bacterial surfaces increased the cake resistance of bacteria (Hodgson *et al.*, 1993). These results are supported by two more recent studies (Nagaoka *et al.*, 1996; Chang and Lee, 1998; Mukai *et al.*, 1999). The accumulation of EPS in the aeration tank and on membrane surfaces resulted in an increase in viscosity of the mixed liquor and an increase in the filtration resistance of the membrane (Nagaoka *et al.*, 1996). In studying the impact of physiological and nutritional conditions on membrane biofouling, Chang and Lee (1998) found a strong correlation between filtration resistance and EPS content. Furthermore, Mukai *et al.* (1999) found that the retained ratio of proteins to sugar polymers during filtration may be one of the factors affecting the magnitude of time-dependent relative permeate flux decline. This suggests that, for permeate flux decline, there are different degrees of interactive effects of proteins and sugar polymers, whose quantity and quality in turn depend on microorganism species and their growth phases.

Nagaoka *et al.* (1998 and 2000) have developed a mathematical model to simulate temporal changes in suction pressure, flux and filtration resistance, based on the mechanisms of accumulation, detachment and consolidation of EPS on membrane surfaces. The measured patterns of flux, TMP and filtration resistance were well predicted by the model. The impact of process conditions, like organic loading, flux and shear stress, on the performance can be evaluated through the model.

Viscosity

Viscosity of the mixed liquor has a significant impact on the performance of MBRs (Magara and Itoh, 1991; Sato and Ishii, 1991; Nagaoka *et al.*, 1998). Both Magara and Itoh (1991) and Sato and Ishii (1991) found that a higher viscosity was related to a lower permeate flux. Viscosity is the third most important factor, after soluble COD and MLSS, in determining the permeate flux (Sato *et al.*, 1991). More recently, Nagaoka *et al.* (1996) found that there was a strong correlation between the filtration resistance of sludge and the viscosity. A higher TMP is related to a more viscous sludge. This is probably not surprising, as the more viscous sludge has a larger potential to cause membrane fouling.

Characterization of Foulants in MBRs

Knowledge of foulants is essential in selecting the most economic and effective cleaning method. The type and concentration of cleaning agents depend on the type of membrane and the nature of the foulants. The most effective approach to foulant identification is by extensive analysis of the foulants through a destructive autopsy (Fane *et al.*, 2000). At present, an increasing number of techniques is available for autopsy to characterize the nature and location of microbial structures (Liss, 2002). These techniques include multi-scale microscopic observation, by conventional light/optical microscopy (COM), scanning confocal laser microscopy (SCLM), two photon laser scanning microscopy (2P-LSM), atomic force microscopy (AFM) and Raman confocal microspectroscopy, conventional scanning electron microscopy (SEM), environmental scanning electron microscopy (ESEM) and transmission electron microscopy (TEM). They also include element analyses by a microprobe (energy-dispersive spectroscopy, or EDS) coupled to a scanning/transmission electron microscope (STEM-EDS), ion chromatographic (IC) analyses and GC/MS/Pyrolysis of foulants.

Most of the studies were conducted by the membrane-manufacturing industry to develop better cleaning methods for specific MBR plants. Only limited information is available in the published literature.

Correlative microscopy (CM) is a strategy of using multiple microscopic techniques (Leppard, 1992) such as COM, SCLM, TEM, and ESEM (Liss et al., 1996; Liss, 2002) which facilitates the detection, assessment and minimization of artifacts that might arise from using one technique only. Collectively, these techniques enable the microscopist to potentially visualize microbial floc or films and identify physicochemical parameters, three-dimensional arrangements of constituents, topography, chemical composition and forces governing interfacial phenomena. The use of only one microscopic technique can bias or limit the information acquired because of the artifacts that arise in specific sample preparations and the resolution constraint associated with a particular technique.

CM has been successfully used by Liss *et al.* (1996) with a minimal perturbation approach in studying natural and engineered flocs. A recent minimal perturbation approach (Droppo *et al.*, 1996a, b) involves the use of sample stabilization in low melting point agarose and a four fold multi-preparatory technique. This was found to maintain the structural integrity of the samples through the stabilization, staining and washing procedures. Nanoplast resin is particularly effective as a stabilization medium because it is a hydrophilic embedding resin that holds the fibrillar EPS (Liss et al., 1996).

SCLM is one of the most recent microscopic techniques used to study activated sludge flocs (Wagner *et al.*, 1994), and has been shown to be a useful technique in bridging the resolution gap between COM and TEM (Liss *et al.*, 1996). 2P-LSM permits examination of floc and films approaching 1 mm in thickness while minimizing photobleaching and phototoxicity (Cowan and Holloway, 1996; Holloway and Cowen, 1997; Decho and Kawaguchi, 1999).

A binocular microscope has been used to observe the nature of deposits on ceramic surfaces (Tardieu *et al.*, 1998). The thickness of a deposit was from 0-100 μm, which coincides with the observation of Shimizu *et al.* (1996). GC/MS/Pyrolysis analyses showed that the composition of the deposited foulant varied according to the biological conditions and was similar to that of the supernatant of the biological suspension after centrifugation. The deposited foulants contained a substantial quantity of mineral elements (Tardieu, 1995). In another study, Yoon *et al.* (1999) used a SEM and IC to characterize the nature of a crystalline precipitate on ceramic membranes for anaerobic treatment. A significant amount of inorganic ions, including Mg^{2+}, Fe^{3+} and PO_4^{3-}, were detected in the fouling layer, implying inorganic fouling was important under anaerobic conditions. More recently, Bouhabila *et al.* (2001) observed the existence of a deposit layer of foulant using microscopes. The deposit was a thin layer of biofilm equivalent to one or two layers of filamentous bacteria. These results suggest that both organic fouling and inorganic fouling could be important, depending on the wastewater characteristics and the nature of activated sludge.

TEM, applied to ultrathin sections of embedded samples, has been used rarely but effectively since the 1980s to examine nanoscale associations between microbes, organic colloidal materials, colloidal minerals and membrane filters (Buffle *et al.*, 1992). Recently, there has been an attempt to correlate results from multi-scale multi-method observations employing COM, ESEM, TEM and STEM-EDS (Leppard *et al.*, 1999) applied to fouled membranes. In this latter study, the ESEM (facilitated by large volume/low resolution observations from COM) was employed to bridge the scalar and resolution gaps between moderate resolution techniques (near micrometer range) and the high resolution of TEM (near nanometer range). The ESEM made two major contributions to the analysis: (1) it permitted structural descriptions of hydrated fouled membranes and their foulants in an essentially unperturbed state; (2) it oriented spatially the subsequent TEM and STEM-EDS examinations well enough to overcome the cost-prohibitive barrier of having to use TEM-based techniques alone, which would mean examining serial ultrathin sections (0.07 μm thick) taken across millimeter sized samples.

Progress with this approach has been slow because engineers and materials scientists are apprehensive about using biological preparatory techniques to prepare complex samples for ultrathin section analysis. Such techniques are both time consuming and expensive, while requiring consummate skill on the part of technicians and a fine tuning of the techniques to adapt to the specific characteristics of a given sample. Despite the apprehension, however, a multi-method TEM preparatory protocol has come into general use (Liss *et al.*, 1996); for fouled membranes, it can provide structural information which relates well to the unperturbed state, allows description of individual colloids and colloid associations with surfaces, and provides element analysis on a "per colloid" basis. Additionally, the descriptions of colloids can often be converted to valuable characterizations using literature standards based on shape, size, native electron-opacity, electron-staining characteristics, internal morphology and microprobe element analysis. A guide to the scattered literature on such characterizations can be found in Leppard (1992), in a document endorsed by the International Union of Pure and Applied Chemistry.

Figure 3a shows a view by TEM (0.003 μm practical resolution) of an ultrathin section (0.07 μm thickness) taken through a biofilm on the upper surface of a fouled membrane filter (bottom of view). The bacterial cells and their fibrillar EPS are clearly identifiable, and several kinds of colloidal debris are evident. Figure 3b shows a glancing section taken through the uppermost layer of a fouled membrane filter. This view reveals the intimate association between individual nanoscale fibrils (0.003-0.01 μm diameter) and inner membrane surfaces. Figure 4 presents a microprobe spectrum (STEM-EDS) taken from an individual selected colloid positioned at the biofilm/filter interface.

SCLM is one of the most useful tools to investigate biofilms in situ (Lawrence and Neu, 1999). A particularly useful feature of SCLM and 2P-LSM is that these can be used in combination with a variety of fluorescent molecular probes to study the spatial distribution of extracellular polysaccharides, cell viability, pH gradient, proteins, RNA, lipids, and other components of floc nondestructively. Measurements of the dimensions of colloidal matrix material and their three-dimensional disposition are realistic. Figure 5 illustrates a SCLM image showing the association of a microcolony with the surface of a membrane in a pilot scale submerged system fed municipal wastewater. The image was derived from observations of thin optical sections of a sample subjected to lectin-binding analysis (soybean agglutinin, wheat germ agglutinin and concanavalin A). The combination of the three lectins reveals the distribution glycoconjugates on the cell surface and within the film/aggregate matrix including the deposition of EPS directly on the membrane surface.

While examination of fouled membranes by microscopes is highly informative, individual autopsy techniques generally require a perturbing preparation of samples and invasion of membranes. Therefore, simple and non-invasive characterization methods are desirable. For this purpose, Fane and co-workers (Li *et al.*, 1998; Chang and Fane, 2000) developed two non-invasive methods in recent years. One is a direct observation through the membrane (DOTM) and the other involves the direct observation of the surface of the membrane (DOSM). Both methods confirmed the existence of a critical flux and directly observed the deposition of particles on membrane surfaces. These two methods provide an excellent way to monitor the sludge cake formation on membrane surfaces but not for element analyses nor for important nanoscale events.

Strategies for Controlling Membrane Biofouling and Membrane Cleaning

The strategies for biofouling control and cleaning involve both physical and chemical methods. Based on the purpose of cleaning, these methods can be classified into two categories: 1) methods for preventing biofouling; and 2) methods for membrane cleaning once fouling occurs.

Optimal Hydrodynamic Conditions and Membrane Module Design

Optimization of the hydrodynamic conditions is essential for preventing membrane biofouling or at least reducing the rate at which biofouling occurs. In a MBR plant, optimization of the hydrodynamic conditions can be achieved by controlling aeration intensity and time in submerged MBRs and flow velocity of mixed liquor in crossflow MBRs, and also by appropriate design of membrane modules. Pilot testing is required to find the optimal hydraulic conditions.

Because the design of membrane modules and spacers affects the hydrodynamics of flow and contact time, the optimal packing density of fibres and flat sheets, the geometry and location of aerators, and the orientation of fibres, have been active research topics in both industry and academia. It is believed that an optimal performance (minimized rate of biofouling) can be reached by design modification.

Backwashing

Once the membrane surface is fouled by pore clogging and sludge cake formation, backwashing is an effective way to recover, at least partially, the membrane performance (Srijaroonrat et al., 1999; Laitinen et al., 2001). In MBRs, backwashing with permeate will push the colloidal particles and cell debris from the pore structure into the mixed liquor and partially remove sludge cake from membrane surfaces. Laboratory studies indicate that backwashing dramatically improves the performance of membrane modules (Visvanathan et al., 1997; Defrance and Jaffrin, 1999; Gan, 1999; Chang and Fane, 2000; Bouhabila et al., 2001). The frequency and flux of backwashing are related to the operating and environmental conditions in the MBRs.

While both Gan (1999) and Bouhabila *et al.* (2001) found that periodic backwashing decreased internal fouling (pore clogging), the total dissolved solids (TDS) in the permeate increased with the application of a backwashing technique (Gan, 1999). In another study, Chang and Fane (2000) found that injecting air into hollow fibres and tubular MBRs is effective in controlling flux decline caused by concentration polarization and particle deposition. This was attributed to the strong turbulence caused by the two-phase flow inside the fibres and MBRs.

Chemical Cleaning

Chemical agents have been extensively used for both maintenance and recovery cleaning of membranes in MBRs. Maintenance cleaning helps to maintain flux and reduce the frequency of recovery cleaning. The inherent nature of membrane biofouling, however, makes recovery cleaning to destroy the foulants that have accumulated on membrane surfaces and within the pore structure unavoidable. There are no generic chemicals that are effective for recovering the membrane flux in every situation, as the nature of foulants changes at different locations. The chemical agents for membrane cleaning can generally be classified as acids, bases, oxidants and surfactants. A chlorine solution has been widely used as an oxidant for membrane cleaning (Gander et al., 2000). Immersion of the modules in an acid bath for membrane cleaning was also reported (Pound et al., 1997).

A knowledge of the nature of foulants will assist in the selection of chemical agents and the sequence for using more than one chemical agent. A destructive autopsy can be used to get extensive information regarding the nature and locations of foulants (Fane *et al.*, 2000).

Optimal Design of Activated Sludge Process

Because the use of MBRs in the wastewater treatment industry is relatively recent, there are opportunities to optimize performance with respect to biofouling through better process design. Pretreatment, already widely used for conventional activated sludge systems, remains important for MBRs. The presence of hair, rags and sharp materials entering the biological process will significantly reduce membrane performance. A fine screen or a high degree of primary treatment may be required for MBRs (Heiner and Bonner, 1999). Membrane fouling from these materials is not biofouling, but must nevertheless be prevented.

Currently, polymers are widely used in conventional activated sludge facilities for sludge thickening and dewatering. The MBR may remove the requirement for downstream thickening, but solids dewatering will still be required. The polymers used for dewatering should be examined not only for their solids capture efficiency in the dewatering process but also for compatibility with the MBR membranes. The presence of dewatering polymers in the liquid return from dewatering may accentuate biofouling in the MBR.

The key MBR design factor is the membrane flux. The operating permeate flux should be lower than the critical flux, which is site specific and is a function of sludge concentration and hydrodynamic conditions (Madaeni *et al*, 1999). A number of biological process design factors, such as MLSS and SRT, impact biofouling and thus flux. The sludge physico-chemical properties that affect membrane biofouling (e.g. floc size, EPS, surface charge and hydrophobicity) may be manipulated by process design (Higgins and Novak, 1997; Liao et al., 2001). For example, a relatively higher SRT and HRT may reduce the soluble COD in mixed liquor, thus reducing biofouling due to the presence of organic constituents (adsorption and pore clogging). The addition of inorganic salts may enhance flocculation, thereby reducing supernatant turbidity (Higgins and Novak, 1997) and thus reducing sludge cake formation. Other opportunities to decrease the rate of biofouling by changing the biological design parameters should also be examined.

Summary and Recommendations

Membrane biofouling is a major factor impacting the efficient and economic operation of MBRs. Three mechanisms: adsorption of EPS and other bioorganics, pore clogging, and sludge cake formation on membrane surfaces, are involved in membrane biofouling. Module design, hydrodynamic conditions, operating conditions of membrane modules, membrane materials, environmental and process conditions of a given activated sludge system and the nature of the activated sludge mixed liquor are the dominant factors affecting membrane biofouling. Several key physical issues for which optimal designs must be determined are:

1. The packing density of fibres and the space distance of the membrane modules.
2. The aeration intensity in submerged MBRs.
3. The mixed liquor recirculating flow velocity in crossflow MBRs, and
4. The shear imposed by the recirculating pump in crossflow MBRs.

Mixed liquor properties affect membrane biofouling. Four key properties are floc size, EPS quantity and quality, and the viscosity of mixed liquor. A larger floc size, a lesser EPS content and a lower viscosity are related to less membrane biofouling. Perhaps surprisingly, the colloidal particles and solutes (including soluble/suspended EPS) in the mixed liquor are more important than the suspended solids in controlling membrane biofouling.

Additional research is required to further understand and ultimately better control membrane biofouling in MBRs. The specific phenomena causing membrane biofouling must be further examined. An important starting place is to develop more knowledge about the chemical and structural nature of the foulants. Characterization of foulants using destructive autopsy must continue. Methodology used in understanding the nature of the foulants in membrane water treatment (Wiesner and Chellam, 1999) may also applicable to the MBR processes. The TEM protocols of Liss *et al.* (1996) coupled to the TEM characterization approach of Leppard (1992) should also contribute to a future understanding of the nature of foulants. Better characterization of foulants will allow the development of better cleaning strategies (for recovering flux after biofouling occurs), better membrane material selection (to prevent or slow biofouling) and better opportunities to control the biological process to minimize foulant productin (to prevent or slow biofouling).

Further information about the role of surface charge and hydrophobicity of both the mixed liquor and the membrane materials should be developed. Additionally, the relationships between the mixed liquor properties known to impact biofouling, e.g. EPS content, and the underlying operating and design variables such as SRT must be further examined. By combining this information with increased knowledge of the foulants and the fouling mechanisms, improved design and operating strategies can be implemented.

Mathematical models which predict and simulate membrane biofouling should be developed. Although models have limitations, biofouling models will allow examination and prediction of the relationships between key biofouling factors and design, operating and membrane parameters. Model calibration and verification must be accomplished by conducting comprehensive studies on the impact of specific factors on membrane biofouling in MBRs to determine the relative importance of each factor for given conditions.

MBRs are an increasingly important technology for biological wastewater treatment. Further understanding of the mechanisms of biofouling and further development of strategies to prevent biofouling as well as recover from biofouling are required to allow full integration of this technology.

Acknowledgements

Credits. Special thanks to Marcia West at McMaster University for technical support in electron microscopy.

Authors. B.Q. Liao (post-doctoral fellow at the time of this work), H.E. Kraemer (master's candidate at the time of this work) and S. N. Liss (professor and associate dean) are with the Department of Chemistry, Biology and Chemical Engineering, Ryerson University, Toronto, Ontario Canada. D.M. Bagley (associate professor) is with the Department of Civil Engineering, University of Toronto, Toronto, Ontario, Canada and G.G. Leppard (professor) is with the Department of Biology, McMaster University, Hamilton, Ontario, Canada and the National Water Research Institute of Environment Canada, Burlington, Ontario, Canada. Correspondence should be addressed to S. N. Liss, Department of Chemistry, Biology and Chemical Engineering, Ryerson University, 350 Victoria Street, Toronto, Ontario, Canada M5B 2K3; Tel: (416) 978-5000 ext. 6357, Fax: (416) 979-5044, E-mail: sliss@ryerson.ca

References

Adham S., Gagliardo P., Boulos L., Oppenheimer J., Whitman E.-J. and Trussell R. (1999) In search of pure waters. *Wat. Environ. Tech.*, Nov., 22-28.

Andreadakis A. D. (1993) Physical and chemical properties of activated sludge flocs. *Wat. Res.*, **27**(12), 1707-1714.

Arai, K. and Nagaoka, H. (1995) Effect of membrane module configuration and intermittent operation cycle in direct submerging membrane separation activated sludge process. *J. of Japan Society on Wat. Environ.* 18(4), 74-81 (In Japanese)

Beaubien A., Baty M., Jeannot F., Francoeur E. and Manem J. (1996) Design and operation of anaerobic membrane bioreactors: development of a filtration testing strategy, *J. of Membr. Sci.*, **109**, 173-184.

Bouhabila E. H., Aïm B. R. and Buisson H. (1998), Microfiltration of activated sludge using submerged membrane with air bubbling (application to wastewater treatment). *Desalination*, **118** (1-3), 315-322

Bouhabila E. H., Aim R. B. and Buisson H. (2001) Fouling characterization in membrane bioreactor. *Sep. Sci. & Tech.*, **22-23**, 123-134

Brindle K. and Stephenson T. (1996) The application of membrane biological reactors for the treatment of wastewaters. *Biotechnol. Bioeng*. **49**, 601-610.

Buffle, J., Perret, D. and Newman, M. (1992) The use of filtration and ultrafiltration for size fractionation of aquatic particles, colloids, and macromolecules, in: *Environmental Particles, Vol. 1*, (J. Buffle and H. P. van Leeuwen, eds.), Lewis Publ., Chelsea, MI. p. 171-230.

Chang I.-S. and Lee C.-H.(1998) Membrane filtration characteristics in membrane-coupled activated sludge system-Effect of physiological states of activated sludge on membrane fouling. *Desalination*, **120**, 221-233

Chang I.-S., Lee C.-H. and Ahn C. –H. (1999) Membrane filtration characteristics in membrane coupled activated sludge system: the effect of floc structure on membrane fouling. *Sep. Sci. and Tech.*, **34**(9), 1743-1758

Chang S. and Fane A. G. (2000) Characteristics of microfiltration of suspensions with inter-filbre two-phase flow. *J. of Chem. Tech. and Biotech.*, **75**, 533-540.

Chaize S. and Huyard A. (1991) Membrane bioreactor on domestic wastewater treatment sludge production and modeling approach. *Wat. Sci. Tech.*, **23**, 1591-1600

Chao A. C. and Keinath T. M. (1979) Influence of process loading intensity on sludge clarification and thickening characteristics. *Wat. Res.*, **13**(12), 1213-1220.

Chiemchaisri C., Wong Y. K., Urase T. and Yamamoto K. (1992) organic stabilization and nitrogen removal in membrane separation bioreactor for domestic wastewater treatment. *Wat. Sci. Tech.*, **25**(10), 231-240

Choo K.-H. and Lee C.-H. (1996) Membrane fouling mechanisms in the membrane-coupled anaerobic bioreactor. *Wat. Res.*, **30**(8), 1771-1780

Choo K. H., Kang I.-J., Yoon S.-H., Park H., Kim J.-H., Adiya S. and Lee C.-H. (2000) Approaches to membrane fouling control in anaerobic membrane separation bioreactor. *Wat. Sci. Tech.*, **41**(10-11), 363-371

Defrance L. and Jaffrin M. Y. (1999) Reversibility of fouling formed in activated sludge filtration, *J. of Membr. Sci.*, **157**(1), 73-84

Fan X. –J., Urbain V., Qian Y. and Manem J. (2000) Ultrafiltration of activated sludge with ceramic membranes in across-flow membrane bioreactor process. *Wat. Sci. Tech.*, **41**(10-11), 243-250

Fane A. G., Beatson P. and Li H. (2000) Membrane fouling and its control in environmental applications. *Wat. Sci. Tech.*, **41**(10-11), 303-308

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

Gan Q. (1999) Evaluation of solids reduction and backflush technique in crossflow microfiltration of a primary sewage effluent. *Resources, Conservation and Recycling*, **27**, 9-14

Gander M., Jefferson B. and Judd S. (2000) Aerobic MBRs for domestic wastewater treatment: a review with cost considerations, *Separation and Purification Technology* **18**(2), 119-130

Heiner G. and Bonner F. (1999) Is the MBR process suited to your treatment plant? *Pollution Engineering*, Dec., 64-67

Higgins M. J. and Novak J. T. (1997) The effect of cations on the settling and dewatering of activated sludge. *Wat. Environ. Res.*, **69**, 225-232

Hodgson P. H., Fane A. G. (1992) Crossflow microfiltration of biomass with inorganic membrane: the influence of membrane surface and fluid dynamics. *Key. Engg. Mater.* **61-62**, 167-174.

Hodgson P. H., Leslie G. L., Schneider R. P., Fane A. G., Fell C. J. D. and Marshall K. C. (1993) Cake resistance and solute rejection in bacterial microfiltration: the role of extracellular matrix. *J. of Membr. Sci.*, **79**, 35-53.

Howell J. A. (1995) subcritical-flux operation in microfiltration. *J. of Membr. Sci.*, **107**, 165-171

Ishida H., Yamada Y., Matsumura S. (1993) Submerged membrane activated sludge process (KSMASD)-Its application into activated sludge process with high concentration of MLSS. *2nd International Conference on Advances in Water and Effluent Treatment*, BHR Group, p321-330.

Ishiguro K., Imai K. and Sawada S. (1994) Effects of biological treatment conditions on permeate flux of ultrafiltration membranes in membrane /activated sludge treatment system. *Desalination*, **98**, 119-126.

Karr P. R. and Keinath T. M. (1978) Influence of particle size on sludge dewatering. *J. Water Pollut. Control. Fed.*, **50**, 1191

Kiat W. Y., Yamamoto K. and Ohgaki S. (1992) Optimal fiber spacing in externally pressurized hollow fiber module for solid liquid separation. *Wat. Sci. Tech.*, **26**(5-6), 1245-1254

Kim J.-S., Lee C.-H. and Chang I.-S. (2001) Effect of pump shear on the performance of a crossflow membrane bioreactor. *Wat. Res.*, **35**(9), 2137-2144.

Laitinen N., Luonsi A., Levanen E. and Nystrom M. (2001) Effect of backwashing conditions on ultrafiltration of board industry wastewaters with ceramic membranes. *Sep. Sci. Tech.*, **25**, 323-331.

Lawrence, J.R. and Neu, T.R. (1999) Confocal laser scanning microscopy for analysis of microbial biofilms. Meth. Enzymol. 310: 131-144.

Leppard, G. G. (1992) Evaluation of electron microscope techniques for the description of aquatic colloids, in: Environmental Particles, Vol.1, (J. Buffle and H. P. van Leeuwen, eds.), Lewis Publ., Chelsea, MI. p. 231-289.

Leppard, G. G., West, M. M., Husain, H. and Rabie, H. R. (1999) Microchemical and ultrastructural analysis of foulant on microfiltration membranes. Abstr. "EURO-Membrane 99", Leuven, Belgium. p. 118-119.

Li H., Fane A. G., Coster H. G. L. and Vigneswaran S. (1998) Direct observation of particle deposition on the membrane surface during crossflow microfiltration. *J. of Membr. Sci.*, **139**(1), 83-97

Liao B. Q., Allen D. G., Droppo I. G., Leppard G. G. and Liss S. N. (2001) Surface properties of activated sludge and their role in bioflocculation and settleability. *Wat. Res.*, **35**(2), 339-350

Liss, S. N., Droppo, I. G., Flannigan, D. T. and Leppard, G. G. (1996) Floc architecture in wastewater and natural riverine systems. *Environ. Sci. Technol.* **30**(2), 680-686.

Liss, S.N. 2002. Microbial Flocs Suspended Biofilms in *The Encyclopaedia of Environmental Microbiology*. G. Bitton (Editor). Volume 4 John Wiley and Sons, New York, p. 2000-2012.

Madaeni S. S., Fane A. G. and Wiley D. E. (1999), Factors influencing critical flux in membrane filtration of activated sludge, *J. of Chemical Technology & Biotechnology*, **74**(6), 539-543

Magara Y. and Itoh M. (1991) The effect of operational factors on solid/liquid separation by ultra-membrane filtration in a biological denitrification system for collected human excreta treatment plants. *Wat. Sci. Tech.*, **23**, 1583-1590

Manem J. and Sanderson R. (1996) Membrane separation bioreactor, in: Water Treatment Membrane Process, edited by J. Mallevialle, A. Argentinas, P. E. Odenaal and M. R. Wiser, McGraw-Hill, NY

Mukai T., Takimoto K., Kohno T. and Okada M. (1999) Ultrafiltration behaviour of extracellular and metabolic products in activated sludge system with UF separation process. *Wat. Res.*, **34**(3), 902-908.

Nagaoka H., Ueda S. and Miya A. (1996) Influence of extracellular polymers on the membrane separation activated sludge process. *Wat. Sci. Tech.*, **34**(9), 165-172

Nagaoka H., Yamanishi S. and Miya A. (1998) Modeling of biofouling by extracellular polymers in membrane separation activated sludge system. *Wat. Sci. Tech.*, **38**(4-5), 497-504

Nagaoka H., Kono S., Yamanishi S. and Miya A. (2000) Influence of organic loading on membrane fouling in membrane separation activated sludge process. *Wat. Sci. Tech.*, **41**(10-11), 355-362

Ozaki N. and Yamamoto (1996) An investigation of sludge accumulation mechanism on membrane threads in externally pressured crossflow filtration process using the hollow fiber module. *J. of Hydraulic, Coastl and Environ. Engg.*, **551**, 123-134

Ozaki N. and Yamamoto K. (1997) Effect pf aeration on suppression of sludge accumulation onto the membrane surface in filtration of activated sludge suspension using flat sheet membranes. Proc. 31st Annul Conf. JSWE, Sapporo 42 (In Japanese)

Pound C., Sharp, R. W. and Harvey R. C. (1997) Immersed membrane bioreactor provides reclamation quality effluent. *WEFTEC"97 Proceedings*, Vol.1, p701-712

Roorda J. H., ver dar Graaf J. H. J. M. (2000) Understanding membrane fouling in ultrafiltration of WWTP-effluent. *Wat. Sci. Tech.*, **41**(10-11), 345-353

Sainbayar A., Kim J. S., Jung W. J., Lee Y. S. and Lee C. H. (2001) Application of surface modified polypropylene membranes to an anaerobic membrane bioreactor. *Environ. Technol.*, **22**, 1035-1042.

Sato T. and Ishii Y. (1991) Effects of activated sludge properties on water flux of ultrafiltration membrane used for human excrement treatment. *Wat. Sci. Tech.*, **23**, 1601-1608

Sheikholeslami R. (1999) Composite fouling-inorganic and biological: A review. *Environ. Progress*, **18**(2), 113-123

Shimizu Y., Rokudai M., Tohya S., Kayawake E., Yazawa T., Tanaka H. and Eguchi K. (1989) Filtration characteristics of charged alumina membranes for methanogenic waste. *J. of Chem. Engg. of Japan*, **22**(6), 635-641

Shimizu, Y., Okuno, Y. I. Uryu, K., Ohtsubo, S.and Wtanabe, A. (1996) Filtration characteristics of hollow fiber microfiltration membrane used in membrane bioreactor for domestic wastewater treatment. *Wat. Res.*, **30**, 2385

Srijaroonrat P., Julien E., Aurelle Y. (1999) Unstable secondary oil-water emulsion treatment using ultrafiltration: fouling control by backwashing. *J. Membr. Sci.*, **159**, 11-20.

Stephenson T., Judd S., Jefferson B. and Brindle K. (2001) Membrane separation bioreactor for Wastewater Treatment. IWA Publications, UK

Tardieu E. (1995) Wastewater treatment by ceramic membrane bioreactor: Study of fouling mechanisms. *ECCE1*, Florence, Italy, 4-7 May, pp1237-1240.

Tardieu E., Grasmick A., Geaugey V. and Manem J. (1996) Fouling mechanisms in membrane separation bioreactor applied to wastewater treatment. Proceedings 7th world filtration congress, Budapest (May 1996)

Tardieu E., Grasmick A., Geaugey V. and Manem J. (1998) Hydrodynamic control of bioparticle deposition in a MBR applied to wastewater treatment. *J. of Membr. Sci.*, **147**, 1-12

Tardieu E., Grasmick A., Geaugey V. and Manem J. (1999) Influence of hydrodynamics on fouling velocity in a recirculated MBR for wastewater treatment. *J. of Membr. Sci.*, **156**,131-140

Thomas M. H., Judd S. J. and Murrer J. (2000) Fouling characteristics of membrane filtration in membrane separation bioreactor, in: *Membrane Technology in water and Wastewater Treatment*, edited by P. Hillis, Royal Society of Chemistry, UK, p159-165.

Ueda T., Hata K., Kikuoka Y. and Seino O. (1997) Effects of aeration on suction pressure in a submerged membrane bioreactor. *Wat. Res.*, **31**(3), 489-494

Urbain V., Block J. C. and Manem J. (1993) Bioflocculation in activated sludge: an analytic approach. *Wat. Res.*, **27**(5), 829-838

van Dijk L. and Roncken G. C. G. (1997) Membrane separation bioreactor for wastewater treatment: the state of the art and new developments, *Wat. Sci. Tech.*, **35** (10), 35–41

Visvanathan C., Yang B., Muttamara S. and Maythanukhraw R. (1997) Application of air backflushing technique in membrane bioreactor. *Wat. Sci. Tech.*, **36**(12), 259-266

Wiesner M. R. and Chellam S. (1999) The promise of membrane technology. *Environ. Sci. Tech.*, Sept., 360A-366A.

Wisniewski C. and Grasmick A. (1997) Influence of shear stresses on biokinetics and suspension filterability in a membrane bioreactor (MBR), in: *Bioreactor Bioprocess Fluid Dynamics*, BHR Group, p317-324

Wisniewski C. and Grasmick. A. (1998) Floc size distribution in a membrane bioreactor and consequences for membrane fouling. *Colloidal and Surfaces A: physicochemical and Engineering Aspects*, **138**, 403-411

Wisniewski C. (2001) Biofouling in membrane separation bioreactor: phenomena analysis and modeling, in: *Proceedings of International Conference on MBR (3)*, Cranfield University, UK.

Yamamoto K., Hiasa M., Mahmood T. and Matsuo T. (1997) Direct solids-liquid separation using hollow fiber membrane in an activated sludge aeration tank. *Wat. Sci. Tech.*, **21**, 43-54

Zhang B., Yamamoto K. and Ohgaki S., Kamiko N. (1996) Floc size distribution and bacterial activities in hollow fiber microfiltration membrane used in membrane bioreactor for domestic wastewater treatment. *Wat. Res.*, **30**, 2385.

PRINTED IN CANADA
IMPRIMÉ AU CANADA
ON RECYCLED PAPER
SUR DU PAPIER RECYCLÉ

National Water Research Institute
Environment Canada
Canada Centre for Inland Waters
P.O. Box 5050
867 Lakeshore Road
Burlington, Ontario
L7R 4A6 Canada

National Hydrology Research Centre
11 Innovation Boulevard
Saskatoon, Saskatchewan
S7N 3H5 Canada

NATIONAL WATER RESEARCH INSTITUTE
INSTITUT NATIONAL DE RECHERCHE SUR LES EAUX

Institut national de recherche sur les eaux
Environnement Canada
Centre canadien des eaux intérieures
Case postale 5050
867, chemin Lakeshore
Burlington, Ontario
L7R 4A6 Canada

Centre national de recherche en hydrologie
11, boul. Innovation
Saskatoon, Saskatchewan
S7N 3H5 Canada

Environment Canada Environnement Canada

Canada