A review of fouling of membrane bioreactors in sewage treatment

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Abstract Fouling in membrane bioreactors (MBRs) represents one of the most significant barriers to their more widespread implementation for both municipal and industrial wastewater treatment. It exerts a limit on the membrane permeability, i.e. the flux through the membrane per unit transmembrane pressure, and thus the productivity of the process per unit membrane area installed. As with all membrane processes, extensive investigation of factors contributing to fouling in MBRs, and the subsequent identification of ameliorative measures that may be taken to control it, has taken place since the process was first commercialised 30 years ago. Key findings of pertinent research in this area and operational experience in full-scale plants are summarised, along with the primary facets of the MBR process itself. The most recent evidence suggests that permanent fouling, i.e. fouling not substantially removed by physical cleaning (backflushing), results mainly from certain dissolved or colloidal organic materials, and such adsoptive fouling takes place at even the lowest operational fluxes. Fouling by suspended solids, on the other hand, may be largely controlled by operation below the so-called "critical" flux, which may be increased by more vigorous aeration, and/or by periodic backflushing. It is concluded that more work is required on characterisation of species responsible for permanent fouling.
Keywords Extra-cellular polymers; fouling; membrane bioreactors; sewage

Generic types of fouling

Membrane fouling in membrane bioreactors may be physical, inorganic, organic or biological. Physical fouling refers to the plugging of membrane pores by colloidal species, such that a certain proportion of the membrane surface is effectively occluded. Inorganic and organic fouling usually respectively refer to scalants and macromolecular species.

There are few scientific studies of inorganic fouling of MBR membranes by scalants in the literature, although scaling is perhaps more prevalent than is generally realised in the treatment of certain industrial wastewaters by MBRs. Most formal studies of inorganic fouling in MBRs appear to have been based on anaerobic systems, which are susceptible to fouling by struvite (Choo and Lee, 1996; Yoon *et al*., 1999). In conventional aerobic processes calcium carbonate scaling of microfiltration membranes has been observed during the operation of both flat plate and hollow fibre MBRs, and both its control and removal present significant problems. It also takes place in municipal wastewater recovery and reuse by (abiotic) microfiltration. Simple acid dosing, most often reverted to for conventional membrane filtration, is rarely an option since pH adjustment can perturb the system microbiology and promote the generation of extracellular polymeric substances (EPS). Pre-precipitation is fraught with difficulty, since the wild fluctuations in wastewater quality (and the invariably diverse chemical heterogeneity) coupled with the generally mercurial nature of calcium carbonate scaling itself makes feedback control of the dosant extremely challenging. Indeed, there appear to be no accepted strategies for in-situ amelioration scaling. In most practical cases, solutions appear to be based around *ex-situ* acid cleaning or, most preferably, identification and elimination of the source of the problem in the upstream processing operations.

Organic fouling in MBRs, on the other hand, has been much more widely studied and characterised, as has biological fouling. It has been estimated that almost half of all fouling deposits in membrane systems comprise or involve biofilms. Indeed, because biofilms require very little in the way of nutrients to remain viable, they can exist even in ultrapure water systems. It follows that they readily form on the surface of membranes in MBRs, where membranes are in contact with biomass concentrations generally in the region of 8 to 18 g/l. On the other hand, the biofilms formed on MBR membranes, particularly in submerged processes, are thought to provide some protection to the membrane since such films are more selective (i.e. more highly-rejecting) than the membrane itself. It is widely understood that the EPS generated by micro-organisms are largely responsible for organic fouling of membranes. EPS largely comprise soluble and colloidal macromolecular species, which can then foul the membrane both at its surface and internally.

Four key factors impact upon or contribute to fouling, most of which are interrelated (Figure 1):

a) process configuration,
b) membrane material, and configuration (or geometry),
c) process operation, including system hydrodynamics, and
d) biomass concentration and composition

Overall process performance is defined both in terms of rejection, i.e. removal of key contaminants, and the energy expenditure. Fouling directly impacts on the latter through reducing the hydraulic performance of the membrane, i.e. its permeability, and rarely impacts upon rejection. The above factors will thus be discussed in turn with sole reference to membrane permeability.

Process and membrane configuration

MBRs may be configured either as sidestream or submerged membrane processes (Figure 2), and the key facets of each configuration are summarised below (Table 1). Membrane modules used in MBRs are all flat plate (FP: Kubota and the PLEIADE membrane module by Orelis), tubular (as used in most sidestream systems, such as those produced by Wehrle or Weir) or hollow fibre (HF: Zenon and Mitsubishi Rayon). Of the manufacturers listed

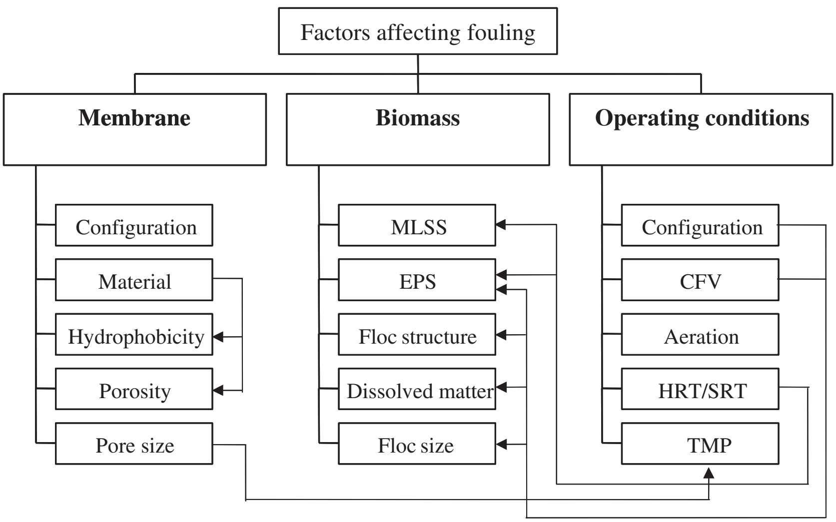

Figure 1 Factors influencing membrane fouling in MBR process (taken from Chang *et al.*, 2002)

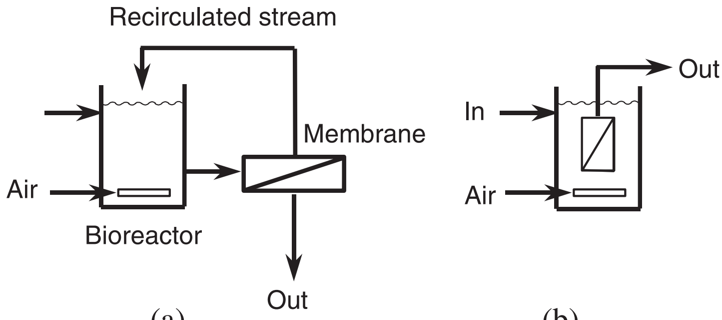

Figure 2 (a) Sidestream, and (b) submerged MBRs

the most significant in terms of recent growth and total installed area are Kubota and Zenon, both of which are submerged systems based on mechanically very robust polymeric microfiltration membranes with a quoted pore size of around 0.1 μm. As with many other membranes, the polymers are surface modified to provide the appropriate degree of hydrophilicity to minimise fouling by hydrophobic matter.

Data from existing established full-scale sewage treatment plant demonstrate differences in fouling propensity, manifested in membrane permeability, for MBRs of different membrane configuration (Table 2). For a slightly lower aeration rate the in-to-out FP module yields a permeability about double that of the HF technology, although the latter operates at a higher overall flux. This difference in fouling propensity is largely a consequence of differences in the system hydrodynamics, and results in the necessity for periodic backflushing (every 10 minutes) and more frequent chemical cleaning for the HF system.

Process operation

Flux

As with all membrane processes, fouling increases with increasing flux and, in selecting an appropriate flux for operation, a delicate balance exists between minimising the required membrane area, and so the capital cost, and minimising downtime for backwashing and cleaning, which directly impacts on operating cost. Some submerged MBRs, and in particular FP and tubular membrane geometries, operate at fluxes low enough to obviate a backwash cycle whilst still requiring relaxation (periodic zero-flux operation to achieve membrane scouring). This is the case for the plant at Porlock (Table 2). Such operation is normally regarded as being sub-critical flux, where the "critical" flux represents the limit of

Table 1 Key facets of the two MBR configurations

Sidestream	Submerged
Longest history (since early 1970s).	Most recent development (since 1990).
Membrane placed external to bioreactor.	Membrane placed in bioreactor.
Pumped system with permeation rate determined by transmembrane pressure and crossflow.	Permeate removed under hydrostatic head, with or without permeate suction, at rate partly determined by aeration.
Higher flux and hydraulic resistance; lower aeration and membrane area requirement.	Lower flux and hydraulic resistance; greater aeration and membrane area requirement.
Stabilised flux with periodic chemical cleaning.	Stabilised flux with periodic chemical cleaning for flat plate membrane configuration; short backwash cycle with periodic chemical cleaning for hollow fibre configuration.
Greater overall energy demand; greater process (hydrodynamic) control.	Lower overall energy demand; reduced process (hydrodynamic) control.

operation at a sustainable membrane permeability (i.e. a constant flux and trans-membrane pressure (TMP)). Operation above the critical flux leads to a rise in TMP, such that periodic backflushing and/or cleaning is required to remove the fouling layer. For an HF module, which is backflushable, it is more economical to operate above the critical flux in relatively short cycles. For the FP system, which is not backflushable, it is crucial to operate below the critical flux. In both cases there is thus a clear benefit in raising the critical flux, something which is generally only achievable through modifying the system hydrodynamics. Indeed, improvements made in both systems over the last few years have generally been devoted to reducing aeration requirements whilst maintaining the permeability, since aeration is the main component of the energy demand.

System hydrodynamics (crossflow velocity and aeration)

Aside from flux, the only operating parameter which can be altered to yield an immediate change in permeability is the crossflow velocity (CFV) – which then increases shear near the membrane:solution interface and so permeate mass transport through the membrane. The crossflow in a sidestream system can be increased simply by increasing the pumping rate, generally producing a roughly linear increase in flux coupled with a small decrease in permeability (Defrance and Jaffrin, 1999). In a submerged system this is not an option, and shear can only be enhanced by using aeration. A number of studies (Mercier-Bonin *et al.*, 1997; Ghosh and Cui, 1999) have demonstrated that the use of slug flow – that air lift in

Table 2 Plant operation (adapted from Judd, 2002)

Parameter	Porlock (FP)	Perthes en Gatinais (HF)
Membrane type and supplier	Flat plate MF, Kubota	Hollow fibre MF, Zenon
Flow through membrane	In-to-out	Out-to-in
MLSS, g l^{-1}	12–18	8–12
Trans membrane pressure, bar	0.1–0.15	0.2–0.5
Operational flux, $l\ m^{-2}\ hr^{-1}$	20 (ave.) – 27 (peak)	27 (ave.) – 40 (peak)
Mean aeration demand, $Nm^3\ hr^{-1}\ m^{-2}$ membrane	0.75	0.91
Mean aeration demand, Nm^{-3} air m^{-3} permeate product	32	27
Mean permeability, $l\ m^{-2}\ hr^{-1} bar^{-1}$	190	140
Backflush or relaxation cycle, min	1440 (relaxation)	10 (backflush)
Backflush or relaxation duration, min	30 (relaxation)	0.7 (backflush)
Backflush volume	–	~25% of permeate product
Clean in place, frequency	0.5% hypochlorite for 5 hrs after 9 months	1–2 times a week, hypochlorite
External chemical cleaning	–	1–4 times a year hypochlorite/ acid soak

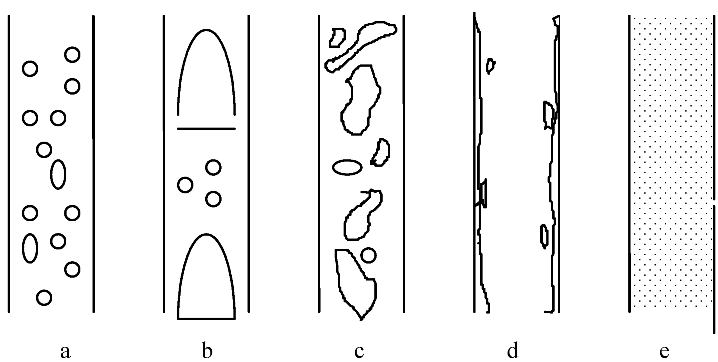

Figure 3 Flow regimes for two-phase flow; (a) bubble flow, (b) slug flow, (c) churn flow, (d) annular flow, and (e) mist flow

which the air bubbles are large enough to fill the interstices of the membrane module (Figure 3) – provides enhanced mass transfer due to the rate of change of shear produced by the rising air bubble. Bench-scale studies on MBRs have shown that slug flow producing a maximum liquid velocity of 0.1 m.s^{-1} at a transmembrane pressure of 0.08 bar is the same flux as that obtained by a pumped system operating at 0.35-0.55 m s^{-1} crossflow and 0.2-0.3 bar under otherwise near-identical conditions (Judd *et al*., 2001). A simple specific energy calculation based on these data, allowing for the energy demanded by the increased aeration, reveals the submerged system to be twice as energy efficient (2.0 kWh m^{-3} permeate product) as the sidestream (3.9 kWh m^{-3}). These data appear to corroborate other findings summarised by Chang *et al*. (2002), where estimated CFV values of 0.3-0.5 m s^{-1} for submerged aerated systems appear to produce permeabilities in the same range (60-80 l m^{-2} h^{-1} bar^{-1}) as those from sidestream systems operating at much higher CFVs (1.5-5 m s^{-1}) and TMPs (1-2.2 bar).

Hydraulic and sludge retention times (HRT and SRT)

One of the most often stated advantages of MBRs is their ability to divorce the retention of solids from floc growth, which generally relies on long hydraulic retention times. The highly selective nature of the membrane means that all suspended solids greater than ~0.5 mm are retained in the bioreactor. This makes it possible to run at longer SRTs and so high suspended solids levels (generally defined as mixed liquor suspended solids or MLSS), with a commensurate reduction in sludge waste volumes. However, the MBR sludge also appears to be less dewaterable than sludges generated in the conventional activated sludge process, something which may relate to the levels of EPS as well as the generally smaller particles in the MBR liquors (see below).

Biomass characteristics

As already stated, the biomass contains both solids and dissolved polymers, both of which contribute to fouling. However, it is only recently that attempts have been made to quantify the relative contribution of the dissolved macromolecular solutes and the suspended matter to fouling.

In the past, phenomenological studies of the impact of biomass characteristics on permeability have generally reported a linear or near-linear relationship between resistance (i.e. the inverse permeability) and MLSS (Fane *et al*., 1981; Sato and Ishii, 1991; Shumizo *et al*., 1993; Chang *et al*., 2001a). This is wholly intuitive from a simple Darcian approach. Other studies have reported impacts of operation on biomass characteristics, specifically the effect of pumping in a sidestream process on both particle size distribution (Wisniewski *et al*., 2000; Cicek *et al*., 1999a,b; Kim *et al*., 2001) and EPS levels (Chang *et al*., 2001b), the latter leading to increased membrane fouling. It has thus been postulated that the inherently lower permeabilities encountered in sidestream systems may be further reduced by fouling from EPS released from flocs as they break up under the shear stresses imposed. However, whist some empirical correlations have been proposed for permeability as a function of dissolved organic carbon (Ishiguro *et al*., 1994) and COD (Sato and Ishii, 1991), there is little or no consistency between trends reported between studies.

Recently reported correlations based specifically on EPS levels suggest a small decrease in filtrability with colloidal EPS compared with no trend with extractable EPS (Rosenburger and Kraume, 2002). It is becoming apparent that the dissolved/colloidal component of the mixed liquor contributes significantly to fouling, with reported contributions to the overall fouling resistance ranging from 5% (Defrance *et al*., 2000) to 52% (Wisnieski and Grasmick, 1998), and may be responsible for permanent, sub-critical fouling. Recent reports from the groups at New South Wales (Cho and Fane, 2002), Montpellier

(Ognier *et al.*, 2002) and Cranfield (Le Clech *et al.*, in press) have all identified a sudden increase in resistance after hundreds of hours of operation under sub-critical flux conditions. This is combined with a very slow increase in membrane resistance, attributed to fouling by EPS (Cho and Fane, 2002). It is not known whether such behaviour is common to all MBR operations or arises only under certain conditions for specific membrane materials. However, the implication is that critical flux applies only to suspended solids and has no bearing upon the sustained operation in the presence of dissolved and colloidal macromolecular species, and EPS specifically.

Conclusions

Fouling in MBRs results from the deposition of solids from the biomass suspension and solutes and colloidal material in the form of extracellular polymeric substances (EPS). The extent to which each of these components foul the membrane depends both upon their levels in the mixed liquor and the design and operation of the MBR. Recent studies suggest that the EPS can foul the membrane under conditions of sub-critical operation, such that the steady-state operational cycle time is reduced to a few hundred hours. However, actual full-scale operational plant do not appear to suffer from this limitation, generally operating for up to 6 months between chemical cleans with only relaxation. There is clearly a need to better understand solute and colloidal deposition in membranes during the operation of MBRs.

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