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Recent advances in membrane fouling control in wastewater treatment processes

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Abstract

Membrane bioreactors (MBRs) are systems performing biological wastewater treatment with membranes utilized for solids separation. These systems have a wide range of applications since they offer some important advantages over conventional processes (e.g. high solids removal, low sludge production etc.). One of their main drawbacks, however, is the occurrence of membrane fouling – the occlusion of membrane pores by the various components found in the mixed liquor. Factors contributing to this phenomenon are various and stem from all the aspects of the treatment process, including membrane-, biomass- and wastewater characteristics as well as operating conditions. Efficient fouling control requires a thorough insight into reactor operation and the mechanisms leading to membrane fouling in the first place. While there are some universal remedies, proper tackling of this problem requires an individual approach tailored to the system of concern, since best results originate from the utilization of several methods together. This review outlines novel and emerging methods having a potential to contribute to sustainable and economical membrane fouling mitigation in the future.

Key words

Biological processes, wastewater treatment, membrane bioreactors, membrane fouling

1. INTRODUCTION

The first use of membrane bioreactor technology has been reported in 1969 by Smith et al. [1]. It was utilized with ultrafiltration membranes in a pilot-scale plant treating industrial wastewater. Even though the initial systems had many disadvantages, such as high capital costs and challenges in operation (mainly due to excessive membrane fouling), MBR slowly started to gain recognition and popularity. The advantages that allowed the MBR to start replacing many of the conventional systems widely in use are its low footprint (since no settling tank is necessary), high treatment efficiency, low sludge production, ease of retrofitting to existing systems etc.

Membranes utilized in MBR systems have a pore size range of 103 - 10-4 µm. Based on it, the process is called microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) or reverse osmosis (RO). Another division among MBR systems takes into account the system setup. Namely, the membrane modules may be located in the treatment tank itself (submerged system) or in a separate tank (side-stream system). For either of the mentioned configurations, the membrane itself can be in the form of a flat sheet or hollow fibers. All of these options are an important aspect of reactor design and operation and need to be carefully chosen based on the specific needs of the system in question.

2. FACTORS AFFECTING MEMBRANE FOULING

Due to the vast number of factors involved in wastewater treatment, there is no single system which suits all applications. Each system comes with its inherent advantages and disadvantages, and the membrane bioreactor systems are no exception. Apart from the aforementioned advantages, there are certain drawbacks as well when compared to conventional systems (CAS). Those include process complexity, relatively high capital and operating costs, increased foaming propensity and, most prominently, membrane fouling.

Membrane fouling can best be described as occlusion of membrane pores leading to decrease in filtration flux (in case of constant pressure operation mode) or increase in transmembrane pressure (TMP; in case of constant flux operation mode). It is not to be confused with clogging which occurs between membrane sheets or fibers inside modules/cassettes. There are many factors that affect the fouling propensity of a given system, and they can be broadly grouped into: membrane-, wastewater-, and biomass characteristics, as well as operating conditions (Figure 1).

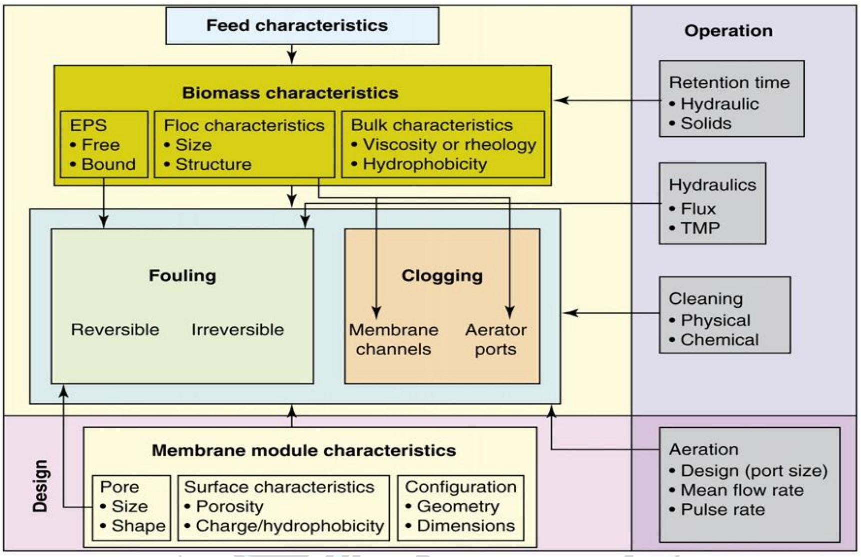

Figure 1. Interrelationship between MBR parameters and fouling [2]

2.1. Membrane characteristics

The proper choice of membranes for MBR systems is of crucial importance for the proper functioning and maintaining of the system. Membranes differ in the following aspects: material, pore size, porosity, hydrophobicity, charge, module etc. [3].

Membranes utilized in MBR systems are generally of polymeric nature, although inorganic membranes are also being used, as well as support materials (mostly textile) which are utilized for dynamic MBR (DMBR) systems. The main property they have to display is durability when exposed to various chemicals, varying pH levels, oxidants, varying temperatures as well as mechanical wear. While inorganic membranes are quite robust, their high price and limitations in their manipulation render them a relatively unpopular choice for MBRs. The most commonly used polymeric membrane material is PVDF (Polyvinylidene Difluoride), but others are also popular and include PTFE (polytetrafluoroethylene), polyolefins, PSF (polysulfonate), CA (cellulose acetate) etc. The support materials used for DMBR systems are generally of a larger pore size than commercial membranes and can be obtained at a much lower cost. They include different textile materials, woven and non-woven meshes etc.

Pore size has to be determined by considering the size of particles from the feed solution, since in the event that the two are similar, an increase in fouling probability occurs. Hereby, pore size distribution and the average pore size are the parameters considered. Porosity refers to the fraction of pores/voids in a material. Hydrophobicity is important since hydrophobic membranes are more prone to fouling, as they interact more closely with hydrophobic components of the feed solution. In order to alleviate this drawback, such membranes may be surface-modified. Charge is similarly important due to interactions on the membrane surface. Finally, module design and placement directly correlate with feed flow and particle occlusion, and have to be optimized in order to preclude fouling or clogging.

2.2. Wastewater characteristics

While domestic wastewater is mostly of similar composition everywhere, industrial wastewater is much more specific and differs largely based on the type of industry and the processes being applied. The influent wastewater affects both the biomass in the reactor as well as the membrane directly. Parameters such as turbidity and suspended solids concentration may represent the effect on the membrane, while COD, nutrient content, and potential toxicity exert their effect largely on the biomass. Other important parameters include temperature, pH, alkalinity etc.

2.3. Biomass characteristics

Biomass characteristics depend on its composition, the type of wastewater as well as the operating conditions. The main characteristics considered in MBR fouling studies include floc structure and floc size distribution, MLSS concentration, dissolved matter and EPS concentration etc. [3].

2.4. Operating conditions

The specific parameters that govern reactor operation and have a direct or indirect effect on membrane fouling are: aeration, HRT, SRT, F/M ratio, TMP/critical flux, hydrodynamics configuration, crossflow velocity etc. [3]. The reactor concentration of oxygen affects microbial growth and metabolism directly, but aeration in MBR systems also has an additional role. Namely, coarse air bubbles applied on the membrane surface in submerged systems cause shear stress which can maintain the thickness of the cake layer at a tolerable level [4]. When it comes to HRT, SRT and the F/M ratio, these parameters mostly affect the microbial biomass, i.e. its growth, flocking, EPS concentration etc. TMP and critical flux, on the other hand, are directly related to the fouling process and thus have to be closely controlled. Operating the reactor at a flux higher than the critical flux value leads to excessive fouling.

3. CLASSIFICATION OF MEMBRANE FOULING

There are various classifications of membrane fouling found in the literature. One of them is based on the possibility to remove fouling using specific cleaning processes. Namely, Park et al. [3] divide it into reversible and irreversible fouling, meaning that the former can be removed by physical or chemical cleaning (or a combination thereof), whereas the latter cannot be removed whatsoever. After irreversible fouling accumulates to a certain extent, the only way to recover initial flux/TMP values is to replace the membrane. Additionally, reversible fouling is further divided into recoverable and irrecoverable. Recoverable fouling can be removed by simple means, i.e. physical cleaning, backwashing etc., whereas irrecoverable requires the use of chemicals.

Another way fouling is classified is based on the place of its occurrence. Accordingly, it can be cake layer deposition or internal pore fouling. The former refers to the accumulation of solids on the surface of the membrane, whereby they form a so-called cake layer. That layer acts as a secondary membrane, with its own pores and permeability. This property is being exploited in DMBR systems, since the original membrane, called support material, has a relatively large pore size. On the other hand, internal pore fouling occurs when particles smaller than the pore diameter get stuck inside, thereby decreasing the amount of permeate that can pass through.

A further way of classification is according to the solids deposition pattern. Hereby, the solids that enter the pores of the membrane may either cause its narrowing or block it completely. Those that accumulate on the surface, however, form a cake layer, as previously mentioned (Figure 2).

4. ESTABLISHED MEMBRANE FOULING CONTROL METHODS

When it comes to membrane fouling control, there are two principle ways to approach the issue: membrane cleaning and fouling prevention.

Membrane cleaning can be physical in nature, including processes such as air sparging, intermittent aeration, backwashing (with air or permeate) as well as sponge scouring. It can also be chemical, which is usually the submersion of the membrane in an acid and/or basic solution. Physicochemical methods such as chemically enhanced backwashing are also in use.

Prevention of membrane fouling is performed in different ways, the most common of which is pretreatment of the influent wastewater. This involves methods such as coarse and fine screening, grit removal, primary sedimentation etc. Pretreatment is performed in conventional systems as well, but has a special importance in MBR systems due to their propensity for fouling. Another way of fouling prevention is operation of the reactor at subcritical flux level, as well as close control of sludge parameters, including MLSS, HRT, SRT, DO, F/M ratio.

One of the more advanced approaches to the problem of membrane fouling is biological control. There are several established methods, such as quorum quenching, enzymatic digestion, as well as utilization of nitric oxide (NO) and bacteriophages.

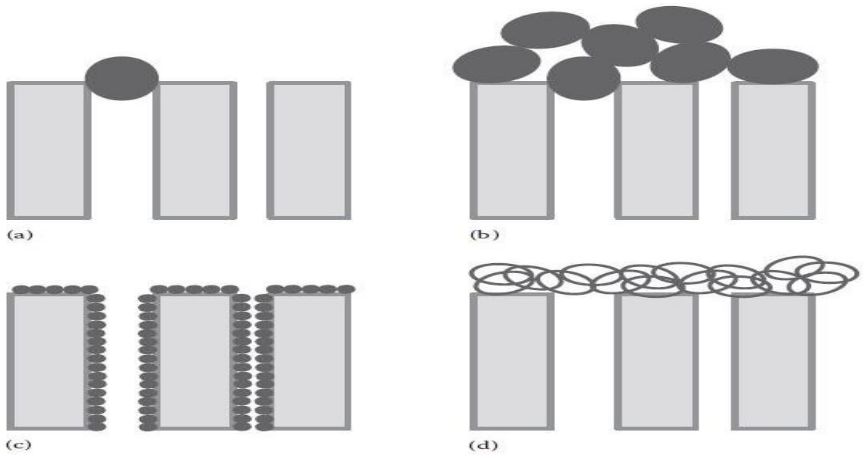

Figure 2. Mechanisms of membrane fouling according to solids deposition pattern: a) complete blocking, b) internal pore blocking, c) intermediate blocking, and d) cake formation [3].

Quorum quenching is basically the inhibition of quorum sensing (QS), a means of bacterial communication through signal molecules which enables them to produces biofilms [5]. The main approaches hereby are prevention of the production of these signal molecules, interference with the receptor of the signal or inactivation of the signals [6]. An example of the latter can be found in studies [7] and [8].

When it comes to enzymatic digestion, it can be applied on different levels, be it to biodegrade the aforementioned signal molecules or the biofilm directly. The latter works by targeting EPS, the main building material that connects bacteria into a biofilm. Several groups reported success applying this approach ([9], [10], [11]), but some concerns remain due to the short catalytic lifetime and loss of free enzymes.

Nitric oxide is a biological messenger molecule which signals bacteria to disperse their biofilm [5]. It has been shown to work for a wide range of bacterial species. The downside to this approach is the low solubility of NO in water.

Bacteriophages are a type of virus which specifically attacks bacteria, wherein it propagates eventually killing its host. In wastewater treatment applications, they have been shown to inhibit or disrupt biofilm formation on membrane surfaces ([12], [13], [14]). One of the major disadvantages of this method is the high specificity of bacteriophages against the target bacteria.

Another important approach to fouling control is through the use of electricity-based methods, the most prominent being electrocoagulation and electrophoresis.

Electrocoagulation works in-situ by creating metal cations at the anode (e.g. Fe^{3+}, Al^{3+} etc.) which then act as coagulating agents, reducing the charge difference between particles in the solution and thereby enabling them to coagulate into larger flocs. The method has been proven efficient ([15], [16]) and has a major advantage over chemical coagulation – no chemical sludge is produced. However, with this approach, care has to be taken to provide optimized conditions so as not to cause bacterial inactivation.

Electrophoresis applications, on the other hand, exploit the negative charge found on particles in the aqueous solution of the reactor. Namely, a direct current (DC) electric field applied close to the membrane drives off the particles from the membrane and towards the anode ([17], [18]). The main limiting factors in the utilization of this method are electrode corrosion and high energy consumption.

There is a variety of methods dealing with modifications of the properties of both membranes and modules. Membrane modification is most often performed by surface modification to decrease the hydrophobicity, which is performed either by coating or grafting a functional group. Another prominent method involves modification of the surface morphology to reduce microbial deposition. When it comes to module modification, it most often comes down to alterations of shape in order to create more favorable hydrodynamic conditions.

Finally, dynamic MBR systems are viewed as an improvement over conventional MBRs when it comes to cost and membrane fouling. These systems rely on support materials with a larger pore size, on top of which a secondary (cake) layer forms (dynamic membrane). This layer is composed of solids from the solution, and has a pore size comparable to that of microfiltration membranes. Due to the fact that mostly inexpensive textile materials and meshes are used as support material, these systems have a lower capital cost than conventional MBRs. Furthermore, since biomass retention is performed by the secondary layer, the support material is much less prone to fouling. Flux/TMP recovery is mostly as easy as removing the cake layer. However, the drawback of DMBRs is that initial effluent quality is low, until the secondary membrane is fully formed (unless pre-formed dynamic membrane is used).

5. ESTABLISHED MEMBRANE FOULING CONTROL METHODS

Being a major drawback of MBR systems, membrane fouling is being addressed by a considerable number of studies. New fouling control methods are being devised regularly, and the following sections describe some of the most promising among them.

5.1. Addition of adsorbents

Adsorbents are porous compounds having the ability to bind different molecules from the surrounding medium to their surface. When it comes to MBR systems, these compounds can help control membrane fouling by removing organics and other pollutants from solution, or providing a surface for attached growth of biomass. The most commonly applied adsorbents are powdered and granular activated carbon (PAC and GAC, respectively) [19]. PAC concentration has to be determined carefully, since the small size of the particles coupled with a high dosage can worsen the fouling of membranes. GAC, on the other hand, has a larger particle size and is therefore suitable as biologically activated carbon, which is covered in the next section.

5.2. Mechanically assisted membrane aeration scouring

This method encompasses the addition of abrasive particles to the reactor solution so as to enhance the membrane surface scouring. Most often used are GAC, plastic beads or other biofilm carriers. With the exception of GAC, they all perform a dual role, acting both as abrasive for the membrane and as a surface for attached growth of the biomass. GAC has the added property of being a potent adsorbent (in this context termed biologically activated carbon), and as such has been shown to be able to provide a 20-60% enhancement of flux ([20] and [21]). Additionally, the property of having both suspended and attached growth in the same reactor contributes significantly to the wastewater treatment efficiency of the system. Not all types of biofilm carrier work for all systems, though, so more research is needed in this field.

5.3. Novel membrane developments

Increase in turbulence near the membrane surface has been shown to be an efficient tool against membrane fouling. This is generally accomplished with rotating and vibrating membranes. The idea behind this is to provide a force that would be able to scour or shake off loosely bound particles on the surface of the membrane, before they achieve a stable integration into the existing cake layer/biofilm. Different studies showed the benefits of this approach, whereby a higher rotation speed has been associated with higher fouling mitigation, but only up to a certain threshold ([22], [23], and [24]).

5.4. Ultrasonic cleaning of membranes

An advanced method of physical cleaning of membranes is with the use of ultrasonic sound waves. These waves are in the frequency range of >20kHz and act by agitating the particles in the membrane, thereby loosening them and causing them to become detached. The main advantage of this method is high flux recovery and the possibility to apply it in-situ [25].

5.5. Cell immobilization

This method limits the free movement of bacterial cells by one of two ways: attachment to a support (mostly biofilm carriers) or cell entrapment (CE) with the use of porous polymer matrices [26]. The former mainly refers to operating an attached growth system, whereas the latter can also offer some level of protection of the biomass from toxic compounds. Materials applied for the matrices range from natural materials such as alginate, agar, and carrageenan, to synthetic polymers including polyacrylamide, polyvinyl alcohol, xanthan gum etc. [5]. An important advantage of CE is that in addition to preventing the biomass from reaching the membrane, it also lowers the levels of bound SMP and EPS, which are known to contribute to fouling.

5.6. Improvements in chemical cleaning

The main innovation when it comes to chemical cleaning of MBR membranes comes in the form a novel biosurfactant – rhamnolipids. These compounds offer advantages in the form of lower cost, higher solubility and less toxicity than conventional methods [5]. Their mode of action is based on biofilm reduction and detachment [27]. It was also reported that, when added during reactor operation, they increased contact between bacteria and lipid molecules, thereby enhancing their removal [28].

5.7. Novel biological control methods

D-amino acids are compounds shown to be able to trigger biofilm disassembly even in trace amounts [29]. Since they can be produced and secreted by a number of bacterial species, they offer a low-cost strategy in fouling control. However, D-amino acids have been shown to be species-specific, which significantly limits their application.

In addition, some naturally derived compounds have also shown promise in biofilm inhibition. Those include extracts from ginger [30], garlic [31], ginseng [32] and brominated alkylidene lactams [33]. Their advantage is that they have low- to no toxicity to the biomass, and may offer a cheap solution depending on their accessibility.

5.8. Addition of engineered nanomaterials (ENMs)

Various engineered nanomaterials (ENMs) can be utilized in MBR systems for efficient fouling control. These materials have specific properties which make them particularly suitable for this purpose, such as antimicrobial ability, photocatalytic activity, and hydrophilicity. Among these ENMs are silver nanoparticles (NPs), graphene, graphene oxide, fullerenes, carbon nanotubes, titanium dioxide NPs etc. [34]. They can be used as membrane surface additions or supplied to the reactor directly. Some of the disadvantages of these systems include the potentially high cost and limited accessibility, as well as limited photocatalytic activity in systems with a high turbidity.

6. CONCLUSION

Membrane fouling is the major disadvantage of MBR systems and has, accordingly, received much attention in the literature. A large number of factors contributes to this phenomenon, which makes its mitigation much harder a task. Apart from established methods, researchers all over the world are constantly devising new ways to enrich the toolbox of reactor operators in their struggle to get fouling under control. Often several methods have to be used in conjunction in order to achieve the best results, and there is no single approach which suits all systems. Therefore, a delicate balance has to be found between gains (lower fouling propensity, improved flux etc.) and losses (cost, higher sludge production etc.) in the application of fouling control methods.

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