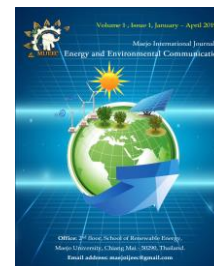




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## RESEARCH REVIEW

### Membrane fouling issues in anaerobic membrane bioreactors (AnMBRs) for biogas production

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#### ABSTRACT

Anaerobic Membrane Bioreactor (AnMBR) technology in recent years has been actively used for municipal and industrial wastewater treatment. Also, AnMBR technology has been considered as an alternative wastewater treatment application over conventional activated sludge system. AnMBRs are best possible operated with flat sheet, hollow fiber, or tubular membranes both in the microfiltration or in the ultrafiltration, but on ceramic membrane use has not been reported widely. AnMBRs are a desirable technology that needs additional research efforts and development. However, membrane fouling, which continues a major problem for all membrane bioreactors, seems much more serious under anaerobic than aerobic conditions. In this review, membrane fouling issues (including membrane fouling mechanism, classification, influent parameters, and mitigation) were discussed and summarized. Moreover, in fouling control, biogas sparging and recirculation (i.e. methane production) were addressed. Lastly, future research perspectives relating to its application and membrane fouling research are planned.

#### 1. Introduction

Because of a risen concern in sustainability within wastewater management, there has been growing attention in recent years in the study of anaerobic wastewater treatment (Robles et al., 2012). The anaerobic membrane bioreactor (AnMBR) has the potential to be a more sustainable wastewater treatment technology than conventional processes because of the low requirements for energy and nutrients, low sludge production and the potential to generate methane of

the anaerobic process (Metcalf and Eddy, 2003; Donga et al., 2016).

The anaerobic degradation (AD) of complex organic matter to methane (CH<sub>4</sub>) and carbon dioxide (CO<sub>2</sub>), which includes the interaction of four different metabolic groups of bacteria, namely hydrolytic, acidogenic, acetogenic and methanogenic bacteria (Nakaoka et al., 1992; Pantawong et al., 2015; Unpaprom et al., 2015; Unpaprom and Ramaraj, 2016) present, in general, some considerable advantages when compared to aerobic treatment. These are: less production of sludge, low nutritional needs, ability to deal with high organic

loads, low cost and finally biogas ( $\text{CH}_4$ ) production (Kappell et al., 2005; Saddoud et al., 2007; Le-Clech, 2010). However, due to their higher investment costs and their somehow complex operation, anaerobic processes are not always implemented. Furthermore, they are significantly influenced by several factors like the type and variability of wastewater, the type of organic contaminants in the influent, its pH, etc. (George et al., 2012).

Biodegradability of a substrate and its potential to produce biomethane through AD process (Ramaraj et al., 2015; 2016a,b,c,d; Wannapokin et al. 2017, 2018; Chuanchai and Ramaraj, 2018; Van Tran et al. 2019a,b). In AnMBRs, biomass can be effectively maintained inside the reactor offering optimum conditions for organic matter degradation without any carry-over of Suspended Solid (SS). By integrating membranes to anaerobic municipal wastewater treatment, enhanced effluent quality in terms of COD, SS and pathogen counts can be reached in comparison with conventional anaerobic processes, and a stable treatment performance can be obtained to meet strict discharge standards (Kocadagistan et al., 2007; An et al., 2009; Ho et al., 2010; Liao et al., 2016). AnMBRs have been reported that it can provide a possibility for the agricultural use of the treated effluent for non-potable purposes in many regions suffering from water shortage (Martinez-Sosa et al., 2011). Agricultural use of treated effluents generally demands extensive pathogen removal along with the availability of macronutrients. Since macronutrients such as ammonium and orthophosphates are not removed by anaerobic bioprocesses and pathogens can be retained by the membrane unit (Saddoud et al., 2006; Ellouze et al., 2009), permeates of AnMBRs are certainly of interest for agricultural use (Norton-Brandao et al., 2013). In addition to achieving high effluent qualities, a shorter start-up period is required for AnMBRs in comparison to UASB systems, which is one of the major advantages in the treatment of especially low-strength wastewaters (Álvarez et al., 2006; Hu et al., 2006; Lin et al., 2011).

Despite the mentioned advantages, membrane fouling is considered as a major operational challenge for AnMBR process and has been investigated comprehensively for both aerobic and anaerobic MBR. The fouling decreases permeate flux and membrane lifespan. AnMBRs are generally operated at higher biomass concentrations compared to aerobic MBRs, impacting rheology and thus, reactor hydraulics and pumping (Kayawake et al., 1991; Choo et al., 1996; Nagaoka et al., 1996; Sainbayar et al., 2001). To complement the current knowledge on AnMBR fouling, this review paper was mainly focused on membrane fouling issues regarding AnMBR technology. Finally, the main conclusions and the future perspectives were presented.

## 2. Membrane fouling issues

### 2.1 Membrane fouling mechanism

Membrane fouling is a major problem that actively effects membrane application. As shown in Figure 1, membrane pore-clogging and sludge cake deposition on membranes are known as membrane fouling in membrane applications which is usually the main fouling factor (Lee et al., 2001). Membrane fouling results in a reduction of permeate flux or an increase of transmembrane pressure (TMP) depending on the operation mode (Meng et al., 2009).

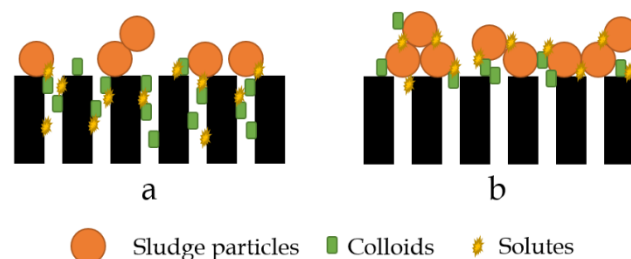


Figure 1. Membrane fouling mechanism (a) pore clogging (b) sludge cake deposition

Meng et al. (2009), had reported that membrane fouling occurs due to the following mechanisms:

- (1) adsorption of solutes or colloids within/on membranes;
  - (2) deposition of sludge flocs onto the membrane surface;
  - (3) formation of a cake layer on the membrane surface;
  - (4) detachment of foulants attributed mainly to shear forces;
  - (5) the spatial and temporal changes of the foulant composition during the long-term operation (e.g., the change of bacteria community and biopolymer components in the cake layer).
- Mainly, the unwanted deposition and growth of microorganisms, colloids, solutes, and cell debris within/on membranes are the membrane fouling. From the complex bacteria community of the activated sludge, it is shown that the fouling behavior in MBRs and AnMBRs are more complicated than the other membrane applications. Generally, as shown in Figure 2, a three-stage fouling history might be proposed (Cho and Fane, 2002; Zhang et al., 2006):

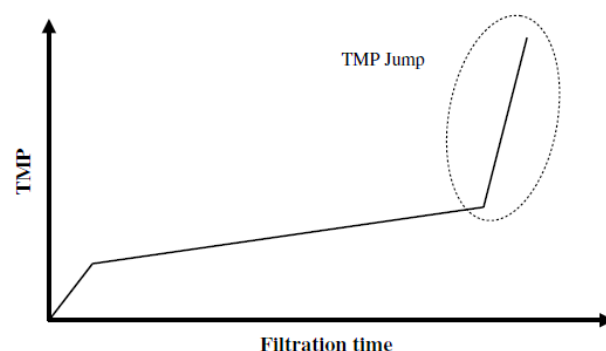


Figure 2. TMP jump illustration

- Stage 1: TMP rapid rise at initial short-term;
- Stage 2: TMP weak rise in long-term;
- Stage 3: TMP sharp jump (Cho and Fane, 2002).

TMP jump occurrence has been shown in Figure 2. Due to TMP jump which significantly decreased flux, suddenly change of the biofilm or cake layer on the membrane surface can cause by this phenomenon (Cho and Fane, 2002). Also, dead bacteria can release extracellular polymeric substance (EPS) that can coat on membrane surface. The increase of EPS concentration had been reported that it related to TMP jump and flux declining rate (Zhang et al., 2006).

## 2.2 Classification of membrane fouling

Membrane fouling can be classified into reversible and irreversible fouling generally based on cleaning method. Reversible fouling, is caused by surface attached foulant, refers to fouling which can be removed by physical cleaning such as backwashing. Irreversible fouling, is caused by pore strongly attached foulant, refers to fouling which can be removed by chemical cleaning. However, irreversible fouling cannot be removed completely, some of them will remain on the membrane pore and surface permanently (Lin et al., 2013).

Membrane fouling can be also classified into biological, organic and inorganic fouling. Biological fouling is caused by bacteria cell which they had interaction the membrane. Biomass that contain cell debris and colloidal particles, it can be appeared as biological fouling. Because of their small size, they can pass through pores of membrane and block easily. Some studied have been reported that some bacteria prefer to grow in membrane surface than in sludge bulk. These bacteria are the main microbial community that releases EPM into membrane and cause biological fouling (Gao et al., 2010; Lin et al., 2011).

Organic fouling refers to biopolymers such as protein or polysaccharide. Due to the small size, they can deposit and act like a glue on membrane surface. For inorganic fouling, struvite ( $\text{MgNH}_4\text{PO}_4 \cdot 6\text{H}_2\text{O}$ ) existed as main inorganic foulants identified earliest in AnMBR systems.  $\text{K}_2\text{NH}_4\text{PO}_4$  and  $\text{CaCO}_3$  can appear also in the system (Nagata et al., 1989). the presence of cations in the influent and sludge suspension, which is the origin of inorganic elements in cake layer, can affect significantly on precipitation of inorganic foulants. the flocs or biopolymers can catch through charge neutralization and bridging effect and then enhanced filtration resistance (Seidel and Elimelech, 2002).

## 2.3 Influent parameters of membrane fouling

TMP values and cross-flow velocities are mostly influenced membrane permeate fluxes in side-stream AnMBRs respect to MBR configuration. TMP values, gas sparging intensities, and the duration of membrane relaxation influenced in submerged anaerobic membrane bioreactors (subAnMBRs). However, gas sparging applied in external configurations with tubular membranes led to an increase in the membrane permeate fluxes; so, introduction of gas/liquid

two-phase flow inside tubular membranes can additionally be a way of controlling fouling in such applications.

Shear forces play an important role in membrane fouling for external AnMBRs because they perform differently from subAnMBRs. In external AnMBRs, magnitude of shear forces to their biomass is showed significantly greater. But to apply high shear force to the system can the size of bioflocs which increase soluble biomass that can deposit on membrane. Therefore, biogas sparging is mentioned on membrane fouling removal. Also, toxic shock feeding is another option for decreasing bioflocs in AnMBRs performance. Membrane fouling can be affected by membrane material as well.

Lastly, the relationship between membrane fouling and membrane pore size have been reported significantly in many studies. The optimal pore size is related to the specific wastewater category. The larger pore is higher membrane fouling rate due to the cake formation on membrane (George et al., 2012).

## 2.4 Membrane fouling mitigation

Cross-flow AnMBRs can be able to reduce fouling by increase crossflow velocity by reach Reynold number at 2,000. This operation can significantly decrease cake formation, but it will increase operation cost. Moreover, too high cross-flow velocity can cause high shear force which negatively reduce permeate flux (Bérubé et al., 2006).

Ultrasonic irradiation is one of the applications which can effectively reduce fouling and cake formation. But higher sludge concentration requires longer ultrasonic irradiation times and higher energy to remove fouling (Sui et al., 2008).

Biogas sparging is a membrane cleaning method with biogas recirculation back to AnMBR. This technique efficiency will depend on provided biogas sparging rate. Higher biogas sparging rate showed decrease od membrane fouling. The suggested biogas sparging rate is between 10 to 25 L/m<sup>2</sup> min. Biogas Sparging also can use in both cross-flow and submerged AnMBRs. Biogas recirculation does not harm biomass which anaerobic microbial community in AnMBRs because biogas itself is generated from anaerobic condition and contains methane and carbon dioxide mainly (Vyrides and Stuckey, 2009; Xie et al., 2010).

## 2.5 Future research perspectives

Since nowadays, people concern more about sustainability in wastewater management. Anaerobic system play an important role for renewable energy from agricultural, domestic, and industrial waste. AnMBRs also is an alternative application which can produce biogas and good fuel quality as renewable energy. However, as membrane fouling cannot be avoiding, according to this review future research should including (1) Membrane fouling mechanism which can identify and characterize foulants. To understand how cake formation, microbial community behavior; (2) Development of pretreatment influent to reduce fouling; and (3) Development

of direct monitoring for information of fouling characteristic and foulant formation.

### 3. Conclusion

In this paper provides detailed information about membrane fouling issues in AnMBR. From the viewpoint of fouling removal, reversible and irreversible were reviewed. Reversible fouling can be removed by physical cleaning. Irreversible fouling can be removed by chemical cleaning, this fouling should be paid attention for long-term operation because cannot remove completely from membrane pore and surface. From the viewpoint of fouling components, biological, organic, and inorganic were classified. Each component needs a specific pretreatment and cleaning method. For fouling mitigation, increase crossflow velocity for AnMBRs and provide higher ultrasonic irradiation can reduce fouling but still cost the system operation. In the viewpoint of optimal fouling mitigation, biogas sparging is preferable for biogas recirculation back to AnMBRs. In the past decade, AnMBRs was introduced as one of the wastewater treatment applications. They can play more advantage than traditionally conventional activated sludge system. However, membrane fouling still the major topic that must be concerned.

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