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Literature Review on Membrane Biofouling Occurring in MBR and Its Related Technologies for Greywater or Wastewater Treatment

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ABSTRACT. This review provides an overview of membrane biofouling of the present MBR and its advanced related technologies to treat greywater and wastewater for solving the global water reuse problem. MBR is a competitive technology for wastewater treatment. HMBR, MBMBR, OMBR have shown a better membrane performance and fouling resistance than MBR. Most of the studies considered in this review were discussing factors causing membrane fouling and the fouling mechanism. In MBR and its related technologies, key microbial species in wastewater biological treatment are different from each other. There are several kinds of bacteria that could adhere to membrane surface to induce severe membrane fouling while treating domestic water such as greywater and blackwater. However, the ones causing membrane fouling are all related to filamentous species, which could cause negative effects on membrane fouling. The biomass growth in HMBR and MBMBR is both suspended and attached, while the biomass in MBR and OMBR are just suspended. Autotrophic-heterotrophic bacteria ratio and anaerobic-aerobic ratio are key parameters for microbial community and contaminants removal. Different microbial species prefer to remove different contaminants. They are affected not only by influent source but also by some environmental parameters. Nowadays, the reason for membrane fouling focuses on extracellular polymeric substances (EPS) and soluble microbial products (SMP), mostly excreting by microorganism in MBR system. EPS/SMP could help bioflocs attachment on the membrane surface in the first stage fouling and cause the accumulation on membrane surfaces and within the pore structure for cake formation and pore blocking respectively, which are the dominant fouling modes. To control EPS, it's a good way to control factors influencing biomass growth rate and biomass performance, such as sludge loading rate, HRT and organic loading rate, and control filamentous bacteria overgrowth. To control SMP, it can be achieved by adjustment of operation parameters (SRT, HRT, DO concentration, temperature, aeration) and addition of adsorbents or coagulants. Membrane fouling, which still remains a major problem for all membrane bioreactors, still needs further discussion.

Keywords: membrane fouling, MBR and its related technologies, wastewater treatment, microbial community, EPS/SMP

1. Introduction

Since wastewater is produced by a variety of sources (cooking, bathing, manufacturing, agriculture, cleaning), the contaminants in wastewater are varied and numerous. Wastewater treatment plants have a National Pollutant Discharge Elimination System permit that determines the type and amount of contaminants they can discharge into the waters. Most permits have a regulation of biochemical oxygen demand (BOD), total suspended solids (TSS), pH, coliforms, and organic nutrients. BOD is a measure of the amount of organic material in the effluent from wastewater plant. TSS are both organic and inorganic solid materials suspended in water. Coliform bacteria are found in abundance in wastewater influent, but the numbers are decreased through the disinfection process. Nutrients, especially nitrogen and phosphorus, could cause eutrophication, which shows extensive growth of algae, aquatic plants, and plankton. Contaminants, such as metals, total dissolved solids (TDS), pharmaceuticals, and endocrine disruptors, can be detrimental to water reuse. Some of their effects on the environment, humans, and water reuse are unknown.

The demand for clean water is vast, no matter for human consumption, agricultural application, or industrial use. Interest in wastewater reuse after treatments is growing, due to water scarcity (Verstraete et al., 2009; Vyrides and Stuckey, 2009), limited wastewater storage capacity as well as increasingly stringent wastewater discharge permits (Onnis-Hayden et al., 2011). Moreover, problems occurred in Canada have brought water quality after wastewater treatments to the forefront of public consciousness. Canadians desire not only water with low organic or mineral contaminants, but also without biological entities, such as bacteria, pathogens, and viruses. Therefore, wastewater treatments that are reliable, cost efficient, and effective in removing large amounts of contaminants are required (Cicek, 2003). Usually, water from recycling systems should follow four criteria: hygienic safety, aesthetics, environmental tolerance and technical and economical feasibility (Harza, 2006).

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These requirements ask for more advanced wastewater treatment technologies to meet the water quality of effluent.

Recently, more attention has been paid to the membrane bioreactor (MBR) technology for wastewater treatment due to its higher efficiency in contaminant removal, excellent effluent quality, low/zero sludge production, compact size and lower energy consumption (Liu et al., 2010). A membrane bioreactor (MBR) combines the activated sludge with a membrane separation process (Melin et al., 2006). However, the widespread application of MBR is constrained by membrane fouling, which reduces the production rate and increases the complexity of membrane filtration operations (Lim and Bai, 2003). To solve this problem, a hybrid membrane bioreactor (HMBR) is an attracting solution for using suspended carriers as supporting media for biofilm development in the aeration tank. HMBR not only improves the efficiency of biodegradation, but also enhances the nitrification process (Mnch et al., 2000). Alternatively, another wastewater treatment technology based on a combination of biofilm processes and membrane filtration technology is a moving bed membrane bioreactor (MBMBR), which are excellent for its high efficiency in the removal of organic carbon, ammonium, nitrates, and nitrites (Ivanovic and Leiknes, 2008). MBMBR could show suspended and attached growth of microorganisms together (Chu and Wang, 2011; Martín-Pascual et al., 2012; Pal et al., 2012). What's more, there has been increasing interest in a novel combination of forward osmosis (FO) and biological process for wastewater treatment, named as the osmotic membrane bioreactor (OMBR) (Achilli et al., 2009). Compared to conventional MBR, OMBR presents some unprecedented advantages: low energy consumption, higher theoretical water flux, higher rejection of the FO membrane and high quality effluent, as well as low fouling potential (Wang et al., 2010; Yap et al., 2012).

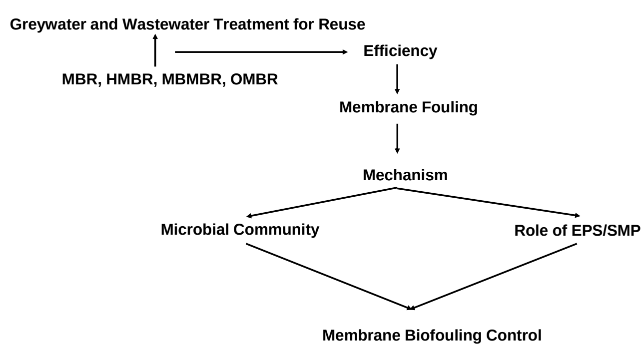

Figure 1. Flow chart.

The developments of cost-effective membrane manufacturing technology and increasingly stringent regulations for the discharge of effluents have given an impetus to the application of membrane bioreactors for wastewater and reuse (Yang et al., 2006). However, membrane fouling, which results in smaller permeate flux and higher operational costs, has been the main barrier to the widespread application of membrane bioreactor. Fouling also increases the undesired frequency of membrane cleaning and replacement, which would bring higher operational cost and shortening of membrane life (Hong et al., 2002). Even though MBR related technologies are introduced for achieving better fouling control for their more effective process design and operation procedures, membrane fouling could still have a negative effect on membrane operation. Yang et al. (2009) reported that the overgrowth of filamentous bacteria in the MB-MBR resulted in severe cake layer and induced a large quantity of EPS, which would do great harm to membrane filtration. For OMBR, membrane fouling can occur not only in the MBR it-self, but also in the downstream RO system (Vrouwenvelder and Van der Kooij, 2001). Specifically, high concentrations of dissolved organic compounds in the effluent can cause severe RO membrane fouling, leading to reduction of water flux and deterioration of treated water quality (Barger and Carnahan, 1991).

To complement the current knowledge on MBR and its related technologies fouling, this review paper mainly focuses on three issues (Figure 1). Firstly, this paper will introduce MBR and its advanced related technologies to treat greywater and wastewater for water reuse. Secondly, it will summarize the function of microbial community on membrane fouling, and key microbial species in MBR, HMBR, MBMBR and OMBR and their differences. Factors influencing their growth rate and performance and their contribution to contaminant removal will be discussed. Lastly, through collecting information on microbial community difference analysis, the role of EPS/SMP on membrane fouling and control strategies, this review attempted to understand mechanism of membrane biofouling, influencing factors causing membrane biofouling and strategies for the control of membrane biofouling.

2. The Application of Membrane Bioreactors and Related Technologies in Greywater and Wastewater Treatment

The classification of household wastewater is usually into greywater and blackwater (Haruvy, 1997). Greywater is defined as wastewater produced from domestic activities such as dish washing, laundry and bathing, whereas blackwater consists of toilet water (Eriksson et al., 2002; Santasmasas et al., 2013).

Interest in the membrane bioreactor (MBR) technology for wastewater treatment has increased because of the strict regulation on water quality, need for water reclaimation, and increase of cost efficiency with the improvement of membrane technology (Choi et al., 2002). The advantages of MBR technology for wastewater treatment include: (1) capability of conducting high volumetric organic loading rates and small reactor volume due to increased biomass concentration (Huang et al., 2001; Boehler, et al., 2007); (2) improved effluent water quality for water recycling because suspended solids and bacteria larger than the membrane pore size are retained by membrane (Rosenberger et al., 2002); and (3) complete and stable nitrification owing to the retention of slow-growing nitrifying bacteria at a prolonged solids retention time (SRT) (Yoon et al., 2004; Li et al., 2006).

However, the widespread MBR application is limited by membrane fouling, which decreases the production rate and increases the complexity of membrane filtration operations. In order to control membrane fouling, many studies have been conducted. Among them, the one attracting wide attention is a hybrid membrane bioreactor (HMBR). Some studies pointed out the ability of the attached biofilm to adsorb small biological flocs and colloidal matter, so that the membrane fouling in HMBR can be reduced (Liu et al., 2010). Liu et al. (2010) presented that a pilot-scale HMBR was developed by introducing biofilm carriers into a CMBR for municipal wastewater treatment. The results indicated that the HMBR apparently improved the organic removal. The COD removal rate increased from 90.4 to 94.2%. The HMBR also improved the nutrients removal effectively. Considering NH_4^+-N, TN and TP, the HMBR improved the removal rate by 4.2, 13.7 and 1.7%, respectively. The speed of TMP accumulation in the HMBR was apparently slowed down, indicating that the HMBR reduced membrane fouling significantly. Yang et al. (2009) compared the filterability of sludge suspension in HMBR and MBR for assessing the suspended carriers influence on the sludge suspension. The suspended carriers in HMBR had negative effects on the filterability of the sludge suspension. The increase rate of TMP for HMBR was far lower than that of MBR during long-term operation. The mean particle size of sludge suspension in HMBR decreased more sharply than that in MBR. Thus, the resistance increasing rates in HMBR were greater than that in MBR with the prolonging of operation time. Ravindran et al. (2009) presented that the HMBR with PAC and microorganisms was effective in removing nitrate, NOM, THMFP, alachlor, and MS-2, meeting the overall treatment objectives. The technology appears to be ideally suited for small-scale systems such as well-head applications. The studies showed that the initial fouling occurred due to the exposure of the membrane to the feed with fast flux decline, and the period of rapid fouling with substantial flux decline rates, while slow fouling with low flux decline rates.

An alternative technology for wastewater treatment is a hybrid system, in which an MB for biodegradation of soluble organic matter is coupled with an MBR. MBMBR has the potential to utilize the best characteristics of both biofilm processes and membrane separation (Ivanovic and Leiknes, 2008). By this technology, the biofilm system may decrease the concentration of suspended solids and increase the extent of membrane fouling. In relation to organic matter and nutrient removal, several studies show that MBMBR technology have obtained COD removal efficiency greater than 93% (Yang et al., 2009; Yang and Yang, 2011; Martín-Pascual et al., 2014). Martín-Pascual et al. (2014) investigated the MBMBR had yields of organic matter removal close to a membrane bioreactor operating with higher MLSS. This technology could reduce the energetic demands and fouling problems associated with MBR technology. The MBMBR system removed 93.44 ± 2.13% of COD, 97.73 ± 0.81% of BOD_5, respectively. In another study, a MBMBR exhibited better total nitrogen removal efficiencies (>70%) than those of a CMBR at the same operating conditions (Yang et al., 2009). Yang and Yang (2011) investigated an intermittently aerated MBMBR to achieve SND via nitrite. Results demonstrated that intermittent aeration was an effective approach to achieve nitrition removal and the COD/TN ratio was another key factor affecting TN removal. The activities of nitrite oxidizing bacteria were inhibited and could recover under subsequent continuous aeration. Yang et al. (2009) studied that a MBMBR was studied for simultaneously removing organic carbon and nitrogen from wastewater. COD removal efficiency averaged at 95.6% during 4 months. The MBMBR system presented good performance on nitrogen removal at different COD/TN ratios. When COD/TN and the total nitrogen (TN) load was 8.9 and 7.58 mg/L h respectively, the TN and ammonium nitrogen removal efficiencies were over 70.0 and 80.0%, respectively, and the removed TN load reached to 5.31 mg/L h.

Due to more stringent regulations, extensive treatment of wastewater is becoming increasingly important. An energy effective innovative osmotic membrane bioreactor (OMBR) is currently under development (Cornelissen et al., 2008; Achilli et al., 2009). In the OMBR, waste is fed into a reactor which is continuously aerated to supply oxygen for the biomass. Through osmosis, water diffuses from the bioreactor, across a semi-permeable membrane, and into a lower water chemical potential DS. The FO membrane plays as an obstacle to solute transport and provides high rejection of the contaminants. The diluted DS is sent to a reconcentration process which reconcentrates the DS and generates a high-quality product water (York et al., 1999; Cath et al., 2005). It is known that conventional wastewater treatment processes could not obtain the effective removal of trace organic contaminants (Ternes et al., 2004). However, OMBR can offer an enhanced removal efficiency for hydrophobic trace organics (De Wever et al., 2007). Alturki et al. (2012) reported a novel osmotic membrane bioreactor (OMBR) for wastewater treatment. 25 out of 27 trace organic compounds with molecular weight higher than 266 g/mol were removed more than 80%. It was controlled by the interaction between physical separation of the FO membrane and biodegradation process. The removal efficiency of the other 23 compounds with molecular weight less than 266 g/mol was very scattered. Thus, the removal efficiency of low molecular weight compounds by OMBR appears to depend mostly on biological degradation. In OMBR, the FO membrane allows high rejection of various containments and mineral salts, and results in very high quality effluent water. This high rejection of the FO membrane could facilitate the removal and recovery of phosphorus from wastewater. Qiu and Ting (2014) reported a new way to manage phosphorus recovery from wastewater by an OMBR. PO_4^{3-}, Ca^{2+}, Mg^{2+} and unconverted NH_4^+ could be rejected by the forward osmosis (FO) membrane and enlarged within the reactor. The precipitates were mostly amorphous calcium phosphate (ACP) with phosphorus content > 11.0%. Most importantly, this process can recover almost all the phosphorus, except for the portion assimilated by bacteria. High concentrations of organic matter and NH_4^+-N in the effluent can cause severe fouling of RO membranes, leading to reduction of water flux and deterioration of water quality (Barger and Carnahan, 1991). Achilli et al. (2009) presented a novel submerged OMBR system. Long-term water fluxes with activated sludge operated at a solids concentration of 5.5 g MLSS/L were only 18% lower than water fluxes using doubly deionized water feed. The removal efficiencies for

TOC and NH_4^+-N were greater than 99 and 98%, respectively, suggesting a better compatibility of the OMBR with downstream RO systems. Moreover, the OMBR system required substantially less backwashing for restoring water flux to approximately 90% of the initial water flux then CMBR.

3. Composition and Characterization of Microbial Community Used in MBR and Its Related Technologies

Many studies could provide fundamental information to relate microbial community dynamics with the factors which contribute to membrane biofouling in MBR and its related Technologies. Key microbial species in different MBR systems and in various living conditions such as suspended or attached state would cause biofouling with various velocities and extent. Some parameters for controlling microbial composition containing oxygen content and nutrient concentration would interact with microbial community. Some identification methods could provide more details on microorganism analysis, including species morphology and gene analysis.

3.1. MBR

The microbial community plays an important role in MBR system (Judd, 2008; Drews, 2010). Calderón et al. (2011) found that the main populations of bacteria, which cause membrane fouling, were *Firmicutes* at a percentage of 42.3%, and *Alpha-proteo-bacteria*, at a percentage of 30.8%, while the populations of archaea were affiliated to the *methanosarcinales* and *methanospirillaceae*. *Sphingo-monadaceae-related* bacteria and *Methanogenic archaea* were found to be components of biofouling, even after chemical cleanings. Bugge et al. (2013) reported that the morphology of sludge flocs was single cells, microcolonies, and filamentous bacteria with or without epiphytes. The floc structure is tight, dense flocculated with low levels of filamentous bacteria, leading to the low impact on sludge floc structure. Viceversa, as poorly flocculated sludge flocs, shows an open structure with high levels of filamentous bacteria. For well flocculated sludge, the dominating filamentous morphotypes belonged to Mycolata, and for poorly flocculated sludge, the predominating filamentous besreria belonged to M. parvicella accompanied Mycolata, which was the dominating filament. These two groups caused foaming problems. Other sludge contains a mixtuer of Mycolata, M. parvicella and Type 0041. The dominant bacterial functional groups, nitrifiers, denitrifiers, and polyphosphate accumulating organisms (PAOs) would have a huge ability on nitrogen and phosphorous removal. The growth mode of the bacterial populations within the flocs was single cells or weak porous cell aggregate, strong microcolonies with tightly packed cells, or filaments. Among the nitrifiers, the ammonium oxidizing bacteria (AOB) among *Betaproteobacteria* showed that primarily Nitrosomonas were most abundant with small *Nitrosospira* populations. Nitrite oxidizing bacteria (NOB) belonged to the genus *Nitrospira*. The PAOs were *Accumulibacter* and *Tetrasphaera*. The number of filamentous bacteria was high with 20 ~ 35% of the biomass. They were primarily *Microthrix*, belonging to *Chloroflexi*, and some low numbers of *H. hydrossis*, belonging to TM7 (Bugge et al., 2013).

The living mode of microorganism in MBR has a great influence on contaminants removal efficiency. There are two different sets of submerged MBRs: suspended (without media) and attached growth (with moving media) (Sombatsompop, et al., 2006). Khan et al. (2011) studied the performance of attached growth (AG) and suspended growth (SG) membrane bioreactors (MBRs) in treating domestic wastewater. The result showed that AG-MBR had a higher removal efficiency of COD, TN and TP than SG-MBR. It infers that small bioparticles have higher microbial activity and the growth of complex biomass within suspended carriers resulted in improved TN and TP removal in AG-MBR. However, some integrated MBRs with both suspended and attached could show a much higher organics removal efficiency. Ho and Sung (2010) investigated an anaerobic membrane bioreactors. The result showed biomass can be both suspended and attached with suspended biomass increasing, which played the most important role with microbial activity, and mainly contributed to organics removal.

Autotrophic-heterotrophic bacteria ratio and anaerobic-aerobic ratio are key parameters for microbial community composition. Organic nutrition would affect autotrophic-heterotrophic bacteria ratio, and anaerobic-aerobic ratio would influence contaminants removal. It is well known that high ratio of COD/TN means the richness of organic substrates in MBR system, where heterotrophic bacteria can grow quickly and utilize most of oxygen and nutrition. Thus, as a kind of autotrophic bacteria, the activity of nitrifiers is restrained, resulting in a low nitrification rate. However, in Liu et al. (2008) research, the improvement of COD/TN ratio brought the increase of the removal rates for both of NH_4^+-N and TN. In the system, the ratio of COD/TN was very low, and the concentration of organic nutrition was not the main parameter affecting nitrifiers growth, but was the key index controlling the denitrification. The enhanced denitrification with higher ratio of COD/TN could remove the adverse effects of NO_x^--N with high concentration on nitrifiers growth. However, if the system was short of nutrition with high concentration of NO_x^--N, the concentration of NO_x^--N would become higher by accumulating and restraining both of denitrification and nitrification in the aerobic and anaerobic area greatly. Tran et al. (2013) discovered that the higher heterotrophic and autotrophic activities of the MBR were attributed to the harsh alternating anoxic and aerobic conditions, which selected for populations with inherently faster growth rates. Liang et al. (2010) investigated that *Nitrosomonas* was the dominant AOB, while *Nitrospira* and *Nitrobacter* species were dominant NOB. Higher nitrifying activities should be related with more diversity of nitrifying bacterial populations in MBR. Thus, metabolic selection through alternating anoxic and aerobic processes has the potential of higher bacterial activities and could improve nutrient removal.

3.2. HMBR

A hybrid membrane bioreactor (HMBR) was developed by adding biofilm carriers into a CMBR. HMBR simultaneous contains suspended and attached biomasses in the bioreactor. Such a novel method effectively increased the total quantity of biomass in the bioreactor and improved the organic removal. Since the porous suspended carriers could supply anoxic or anaerobic condition for anaerobe such as denitrifying bacteria even in aerobic system, the difference in the microbes and morphological composition between HMBR and CMBR exist. Liu et al. (2010) investigated that a pilot-scale HMBR process employing suspended and attached biomass simultaneously in the aeration tank was run for wastewater treatment, combining Kaldnes K3 biofilm carriers with a mixed liquor suspended solids (MLSS) of 3500 mg/L. As mentioned previously, the HMBR apparently improved the organic removal. The COD removal rate increased from 90.4 to 94.2%. It also improved nutrients removal effecttively. Average NH_4^+-N, TN and TP removal was enhanced by 4.2, 13.7, and 1.7%, respectively. The SRT of the attached biomass is higher than that of the suspended biomass, and this makes it available for the growth of nitrifying microorganisms preferentially on the carriers and consequently results in a higher NH_4^+-N removal. Many studies indicate that under suspended biomass, simultaneous nitrification-denitrification (SND) was affected by both DO and the floc size. Lim et al. (2004) observed a slight difference in microbial community structure between the anoxic and the aerobic condition. It is known that larger floc size could result in much higher TN removal than smaller floc size does in the same bioreactor under similar DO conditions (Zhang and Qi, 2007). In a HMBR, the function of the biofilm attached to the carriers was similar to that of larger biological flocs, because their increased size and inner space could provide better conditions for forming anoxic zones for denitrification within the attached biomass. Miura et al. (2007) reported that the pre-coagulation and sedimentation process had a larger impact on bacterial community structures than MLSS concentration did, because the influent water quality of the HMBR (turbidity, TOC, DOC, TP and pH) were significantly different from that of the CMBR. Especially, the effluent pH was lower, which probably caused the different microbial communities in the HMBR. Moreover, most incoming microorganisms could be removed by the precoagulation and sedimentation, which also significantly influenced the bacterial community structures in the HMBR.

3.3. MBMBR

An alternative to the CMBR is to combine a biofilm reactor with membrane filtration, which could enhance the system performance and decrease the effect of suspended solids on membrane fouling (Leiknes et al., 2006; Leiknes and Ødegaard, 2007). This system was indicated as a moving bed membrane bioreactor (MBMBR). Microbial community in a MBMBR system develops in different types of aggregates, such as suspended flocs or attached biofilms. The immobilization of microorganisms has benefits in the development of slow-growing microbial species, such as nitrifying bacteria, and also can support a proper environment for aerobic and anoxic microorganisms (Liang et al., 2010). Therefore, differences in the bioactivity of the mixed liquor and the biofilm can occur. The MBMBR is a mixed reactor with the biomass carriers. They could not settle due to their lower density compared to water, even with biomass on it. The carriers are moving through the bioreactor by aeration. The surplus sludge washed off from the carriers or built in the sludge flocs in the suspension partly precipitate on the bottom. The settled sludge was released off by opening the drain valve of the reactor. Jabornig and Favero (2013) reported that a total suspended solids concentration between 100 and 500 mg/L could be maintained under a drained volume of 2 L/week. A loading rate of the biomass carriers with 0.004 kg BOD/m^2 d resulted in a biofilm area of 9.4 m^2 or roughly 30 L of biomass carriers. Thus, the adaptation of the microorganism to the environment and growth on the biomass carriers stabilized the degradation process and improved the system performance. Martı´n-Pascual et al. (2014) investigated that the MBMBR had yields of organic matter removal with a lower MLSS concentration, close to a CMBR with higher MLSS. Since then, it is possible to reduce the MLSS concentration without decreasing the removal efficiency of the system. In addition, if the MLSS concentration is lower than that of the MBR, the energetic costs of the MBMBR are lower than the MBR, because less aeration is required. Thus, this technology could reduce both fouling problems and energetic demands.

Some researchers have done a lot of work on microbial community analysis and the factors affecting their composition in MBMBR system. Yang et al. (2009) studied the respective respirometric activities of heterotrophic bacteria, ammonium oxidizers and nitrite oxidizers at the steady time of each phase. It implied when COD/TN ratio was stepwise decreased from phase III to phase V, the relative abundance of heterotrophic bacteria decreased gradually in the CMBR and hardly changed in the MBMBR, which can be deduced that the ability of shock loading endurance was better in the MBMBR than in the CMBR. The relative abundance of ammonium oxidizers and nitrite oxidizers had no regular change in both reactors. Yang et al. (2009) also compared sludge in MBMBR and CMBR. The sludge color was yellow in the MBMBR and khaki in the CMBR. Microbial species were plentiful in the MBMBR. Filamentous bacteria and protozoa organisms including *Ciliates*, *Vorticella*, and *Amoebae* were observed abundantly. Microscopic examination revealed that a certain amount of *Metazoans* including *Rotifers* and *Nematodes* also presented in the MBMBR. While in the CMBR, the dominant community of bacteria was *Epistylis* and Vorticella. A small quantity of *Amoebae* and *Rotifers* was found in CMBR. It can conclude that the microbial multiformity in the MBMBR was richer than that in the CMBR. However, this study also showed that the membrane fouling was more severe in the MBMBR than in the CMBR. It inferred that the filamentous bacteria in the MBMBR caused a thick and dense cake layer on the membrane surface.

Some environmental factors changing could lead to AOBs (those that oxidize ammonia to nitrite) and NOBs (those that oxidize nitrite to nitrate) community changing. DO concentration is a key factor affecting nutrition and sludge bulking (Sliekers et al., 2005). Under low DO concentration, AOBs have outcompeted NOBs, based on the higher oxygen affinity of AOBs than NOBs (Zhang and Qi, 2007). Winkler et al. (2012) reported that under anoxic conditions, the decay rate of AOB was zero, while the decay rate of NOB was invariable, almost equaling. Yang and Yang (2011) also verified that the activities of the NOBs were inhibited under the intermittently aerated mode, but could be covered under continuous aeration. On the contrary, AOBs did not exhibit any impact with the anoxic disturbance.

3.4. OMBR

An OMBR is an innovative membrane bioreactor for wastewater reclamation, combining activated sludge treatment and forward osmosis (FO) membrane separation with a post-treatment. The driving force is a concentration difference over the membrane surface using a draw solution at the membrane product side, drawing pure water from the feed water side towards the product side. Cornelissen et al. (2011) conducted an OMBR research to investigate its performance on wastewater reuse. It could be concluded that (1) FO performance (water flux) depended on the temperature due to viscosity effects of the feed solution; (2) FO performance was independent of activated sludge type with different compositions in COD and conductivity; and (3) Higher flux values were obtained for FO membranes facing the draw solution side due to lower internal concentration polarization effects.

OMBR has a high organic nutrient removal efficiency and its microbial community is easily affected by environmental conditions (such as temperature and salinity). Especially, some functional bacteria, such as AOB, NOB, and denitrifying bacteria are more sensitive to elevated salinity conditions (Moussa et al., 2006; Osaka et al., 2008). Hence, the elevated salinity is likely to influence microbial biological activities as well as system performance. It indicated that the organic matter removal constantly reached up to 98% during the OMBR operation, although there is significant accumulation of TOC within the reactor. The reason for TOC accumulation is because of the FO membrane having a much smaller pore size. The high rejection of the FO membrane (> 99%) caused significant DOM accumulation within the reactor. This DOM could be contributed to EPS release and the metabolic products and intermediates generation by microorganisms (Tan et al., 2015). Tan et al. (2015) also indicated salt accumulation at the beginning of the operation caused mild deterioration of nitrifying activity in the reactor, leading NH_4^+-N to accumulate to 10.0 mg/L. This is for AOB are generally slow growing and sensitive to environmental conditions changes (such as temperature and salinity), AOB activities are easily inhibited (Lay et al., 2010). This may relate to the shift in activated sludge bacterial community structures in the first 2 ~ 3weeks. However, biodegradation of TOC was not significantly affected, and remained relatively stable at the same level even after adding silver nanoparticles, as system additions, possibly due to the diverse bacterial community in the reactor. Qiu and Ting (2013) presented significant accumulation of organic matter and NH_4^+-N, indicating the bioactivity deterioration resulted from salt accumulation. With the salinity increasing, high salt-tolerant new species has taken over almost all the dominant species. AOB community presented significant succession among species of *Nitromonas*. For NOB, *Nitrospira* was not significantly influenced. However, *Nitrobacter* would be washed out within the first 10 days. It could be observed that significant succession of denitrifying bacterial community from *a*- to *c*-*Proteobacteria* members also presented in the system.

In MBR and its related technology systems, there were several kinds of bacteria which could adhere to membrane surface to generate severe membrane fouling while treating domestic water such as greywater and blackwater. Guo et al. (2008) found that the bacteria inducing membrane biofilms could probably be *Pseudomonas sp*. and *Nitrobacter sp*. when dealing with bathing wastewater. Xia et al. (2008) used SEM and FISH to confirm that there were certain bacteria on the membrane surface which caused membrane fouling. The bacteria species were likely to be *Pseudomonas sp*., *O*. *anthropi sp*. and *Enterobacter sp*., respectively. Wastewaters from the toilets, as blackwater, are extremely harmful to the aquatic environment because they have high concentrations of suspended solids, uncountable number of microorganisms (including fecal bacteria, pathogenic bacteria, and even viruses) and large quantities of ammonia and organic pollutants (Knerr et al., 2011). Abundant microorganisms especially subminiature animals were found in the mixed liquids, such as *Aspidisca sp*., *Vorticella sp*., *Suctoria sp*., and *Rotifer sp*.. The microbial communities and their activities were similar to those in the conventional activated sludge processes. Moreover, *Aeoloosma hemprichii* was also detected, which always survived in the anaerobic condition (Li et al., 2007). Thus, distinctive microorganisms are presented in different water sources. They would have no doubt leading to a deep influence on membrane performance.

In MBR and its related technology systems, the filamentous species could cause negative effects on settling properties, and they have an impact on the filterability. The effect of the common filamentous species on the filtration properties under different modes of MBRs would have an influence on fouling. In HMBR system, the decrease of the membrane resistance, especially the cake layer resistance, eventually brought about a much slow increase of TMP and a prolonged filtration cycle. The large decrease of extracellular products, could provide an explanation of membrane fouling control by the HMBR operation (Liu et al., 2012). In a short-term OMBR experiment, a gradual salinity in the reactor could cause a decrease in the water flux and sludge production, and significantly changed the biomass characteristics. The increase in the fouling of the FO membrane was much less severe than that of the MF membrane as the MLSS concentration increased from 0 to 20 g/L. Fouling of submerged FO membranes can also be effectively controlled by aeration (Luo et al., 2015). However, the rate of membrane fouling in MBMBR was about three times higher than CMBR despite the latter had suspended solids two times higher than the former. The reason is that the overgrowth of filamentous bacteria resulted in a thick and compact cake layer. Thus, it could be speculated that the overgrowth of filamentous bacteria in the MBMBR resulted in severe cake layer, which would cause membrane flux reduction, and further induce membrane fouling problem (Yang et al., 2009).

Therefore, it should also be possible to “manage” the microbial populations in MBR and its related technologies in order to optimize filtration properties to control membrane fouling by selecting effective microorganisms and appropriately regulating environmental factors (wastewater type, F/M ratio, and oxygenset-point) working on the biomass for target microbial species growth and biological activity. Such indented management is well needed for membrane fouling control and can likely be carried out also in MBR and its related technology systems.

4. Extracellular Polymeric Substances and Soluble Microbial Products

Biofouling is the result of interactions between the membrane surface and those of the biomass or sludge consisting of microbial cells, or aggregates, microbial secretion products, cell constituents derived from lysis, and viruses (Liao et al., 2004). Based on a few studies (Chang et al., 2002; Le-Clech et al., 2006), biofouling can be characterized on the basis of three fouling patterns: EPS/SMP adsorption to the membrane surface; pore clogging by cells; and cake formation arising from the deposition of cells or aggregates.

MBR biomass consists of large amounts of particulate, colloidal and dissolved fractions, all of which contain potential foulants. After initially MLSS concentration was thought to control fouling rate, the focus has quickly turned to slimy and sticky substances which could be bound to the flocs or suspended freely. These groups of compounds are mostly named extracellular polymeric substances (EPS) when they are bound to the flocs or soluble microbial products (SMP) when suspended freely in the supernatant.

Some bacterial species would exist in the membrane and excrete sticky matter such as EPS and SMP to induce severe membrane biofouling (Meng et al., 2009). It was also reported that the changes in the EPS and SMP characteristics could be influenced by changes in the microorganism communities (Laspidou and Rittmann, 2002). Ji et al. (2010) showed a linear relationship between EPS production rate and biomass growth rate. Ng and Hermanowicz (2005) showed approximately a 60% increase in EPS production for biomass growth rates increasing from 0.04 ~ 0.18 h^{-1}, but approximately a 20 % decrease in EPS for biomass growth rates increasing from 0.18 ~ 0.26 h^{-1}. Kim and Jang (2006) indicated that the shift in the bacteria communities such as filamentous bacteria was a major factor for indicating the changes of the EPS and SMP. Therefore, the production of EPS and SMP seems to depend on the kind of microorganisms involved and the system conditions.

Large amounts of EPS and SMP can originate from wastewater components and bacterial products either from cell-lysis or cell-structural polymeric components (Chang et al., 2002). EPS are of biological origin, participating in the formation of microbial aggregates and containing insoluble materials (sheaths, condensed gel, capsular polymers, attached organic material, and loosely bound polymers); whereas, SMP are considered soluble EPS (soluble macro-molecules, colloids, and slimes) (Laspidou and Rittmann, 2002). EPS and SMP consist of polysaccharides (PS), proteins, lipids, nucleic acids, etc., originating from cell lysis, microbial metabolites or wastewater components (Tsai et al., 2008). Usually, PS and proteins are supposed to be the major components that contribute to fouling. Some studies indicated that protein and carbohydrate as predominant of EPS/SMP contribute to the decrease of the perme- ate flux and further cause serious membrane fouling (Hodgson et al., 1993; Chang and Lee, 1998). Protein presented a great positive correlation to fouling resistance. However, carbohydrate, presented a moderate positive correlation possibly due to the low amounts. Then, protein was the major factor in EPS/SMP affecting membrane flux. Thus, the composition and quantity of the organic fraction of the EPS/SMP would have a correlation with the membrane fouling.

The presence of soluble and suspended EPS leads to the accumulation of this material on membrane surfaces and within the pore structure (Chang and Lee, 1998). Especially, smaller particles can cause more severe membrane fouling than the larger particles. It may be assumed that particles of sizes close to or smaller than the membrane pore sizes can contribute to membrane fouling through internal and external pore blocking (Bai and Leow, 2002). Then, SMP is much easier to lead to internal pore blocking, which is the main reason for internal membrane fouling. This may change the friction factor in the flow channels and cause a decrease in the flow area, which leads to greater TMP (Liao et al., 2004). The functions of EPS matrix are multiple and could aggregate bacterial cells in flocs and biofilms, forming a protective barrier around the bacteria, and adhering to surfaces (Laspidou and Rittmann, 2002). Based on its heterogeneous and changing nature, EPS can generate a highly hydrated gel matrix in which microbial cells could be embedded (Liu and Fang, 2002). They can be explained for the creation of an important barrier to permeate flow in membrane processes. What's more, during filtration, EPS and SMP could adsorb on the membrane surface, block membrane pores and form a gel structure on the surface of the membrane, providing a possible nutrient source for biofilm formation and a resistance to permeate flow (Rosenberger and Evenblij, 2005). EPS and SMP could help to aggregate and stabilize the matrix of biopolymer and microbes, and then promote bioflocculation. The bioflocculation by cation bridges could be linked with membrane fouling through the EPS and SMP characteristics (Kim and Jang, 2006). Therefore, investigation of the performance of the biological aspects of membrane fouling, with the characteristics of EPS and SMP, is required to reduce membrane fouling.

In the operation of MBR systems, the membrane fouling was mostly resulted from cake layer formation with an average contribution of 86% (Bani-Melhem et al., 2015). Cake formation has been found to be the main factor controlling both the applicable membrane permeate fluxes (Jeison and Van Lier, 2007) and the critical fluxes (Jeison and Van Lier, 2007). Small flocs, EPS and inorganic materials played an important role in the cake formation process, with the cake layer being found to have a highly heterogeneous structure (Lin et al., 2011). Cake sludge was found to have smaller particle size distribution, keeping much higher specific filtration resistance, 1.5 times more EPS and evidently different microbial community than the bulk sludge (Lin et al., 2011). Ye et al. (2005) found that the cake resistance was higher as the amount of EPS increased. Based on this observation, they raised a sigmoid relationship between the EPS and cake resistance. In Chang et al. (2002) research, it was reported that 90% of the cake resistance were attributed to EPS and the cake resistance changed with the ratio of protein and carbohydrate in the EPS. It can be concluded that most resistance is attributed to the cake layer on the membrane surface rather than internal fouling. In addition, Lijuan Deng proposed higher MLSS concentration and overgrowth of filamentous bacteria could cause higher cake resistance, because the sludge flocs with amounts of filamentous bacteria could produce a higher amount of SMP on membrane surface to form sticky and non-porous cake layer (Deng et al., 2014). Moreover, the SMP can be readily deposited onto the membrane surfaces by permeation drag to affect the first stage of fouling (Bae and Tak, 2005). It was obtained that the SMP could reduce the porosity seriously by filling the void spaces between the cells to form a strong cake.

Grey water (GW) treatment has received considerable attention as a valuable source for wastewater recycling and reuse during the last few years. Guo et al. (2008) studied the role of EPS in MBR operation during bathing wastewater treatment. Influent bathing wastewater had high lineal alkylbenzene sulphonates concentration, low carbon strength, and total phosphorus concentration. NH_4^+-N was the main composition of total nitrogen. The EPS could accumulate not only in the mixed liquor (EPSs), but also on the membrane surface (EPSm) (Li and Yang, 1996). In the first stage, the EPSs could change from 20 to 33 mg/g VSS during day 1 ~ 10, which means there was an impact on the inoculated sludge to produce more EPSs. In the second stage, the sludge gradually adapted to the wastewater and EPSs concentration increased steadily from day 20. EPSs increase from days 20 to 60 was smaller than that from day 60 to 90 (in the third stage). This was because the microorganisms in the mature stage were older than those in the steady stage, so that more EPSs were accumulated due to the interception of the membrane. The change of EPSm differed from that of EPSs. The EPSm concentration increased during the entire operation time. The reason was that the microorganisms attached to the membrane surface gradually. Significantly, the results of analyzing EPSs and EPSm did not show a significant difference during the operation of the first experimental stages. A small difference was observed in the last stages which might be partially due to the production of SMP as results of the increase in microbial growth (Bani-Melhem et al., 2015).

For understanding characterization of EPS and SMP, we could start from the activated sludge properties including SSs, dynamic viscosity, hydrophobicity, and zeta potential, and then investigate the correlation between these properties and membrane fouling resistance, to find the internal relationship with respect to EPS and SMP. The extraction of EPS was based on a cation ion exchange resin (Dowex-Na form) method (Frølund et al., 1996). EPS was normalized as the sum of carbohydrate and protein, which were analyzed using phenol/sulfuric-acid method and folin method (Lowry et al., 1951), respectively. The sludge floc size was determined by focused beam reflectance measurement. And the mean particle size was adopted to characterize its effect on membrane fouling. The soluble microbial products (SMP) were characterized as soluble chemical oxygen demand (COD) of activated sludge supernatant. The dynamic viscosity was determined using a rotational viscosity meter. The relative hydrophobicity was evaluated similar to Wilén et al. (Wilen et al., 2003). The flocs were first shaken in order to break them into small particles, and the supernatant was sampled for zeta potential measurement (Chang et al., 2001). For the sludge samples the membrane fouling tests and activated sludge analysis were performed within 10 h to avoid changes in sludge characteristics.

Sludge loading rate and correspondingly HRT and organic loading rate (OLR) are main operating parameters affecting the production of bound EPS since they govern biomass growth and decay. Rosenberger S et al. (2002) reported that there were high bound EPS concentrations and high sludge viscosity as F/M ratio increased. The formation of bound EPS is growth-related and is produced in direct proportion to substrate utilisation (Laspidou and Rittmann, 2002). Thus, the increase of organic loading rate or F/M ratio will induce the generation of more bound EPS. In addition, aeration intensity, dissolved oxygen and feed substrates have been proven as important parameters affecting bound EPS. Li and Yang (2007b) cultivated activated sludge with different carbon sources including glucose and sodium acetate, and different SRTs of 5, 10 and 20 d. The sludge fed on glucose had more EPS than the sludge fed on acetate. For any of the feeding substrates, the sludge had a constant tightly bound EPS value regardless of the SRT, but the loosely bound EPS content decreased with the SRT, indicating that SRT is more important than feed substrates on the control of bound EPS. A more recent investigation also showed that the protein/carbohydrate ratios of feedwater correlated strongly with bound EPS composition (Arabi and Nakhla, 2008). It was found that with increasing P/C ratio of feedwater, the P/C ratio of bound EPS also increased slightly, but both protein and carbohydrate concentrations decreased. It can be concluded from these studies that there are several factors either alone or combined with each other that play an important role in the formation of bound EPS. Thus, these factors are keys to control bound EPS.

What's more, filamentous bulking has been found to have a strong influence on MBR fouling (Meng and Yang, 2007). The overgrowth of filamentous bacteria leads to a sharp increase of bound EPS concentration and then induces the increase of sludge viscosity and sludge hydrophobicity. In addition, the filamentous bacteria can enlace and fix the foulants on the membrane surface. Some studies showed control methods of filamentous bacteria in MBR. Chudoba et al. (1973); Caravelli et al., (2003) indicated that filamentous bulking can be controlled by selectors, optimization of operating conditions, addition of coagulants and chlorine. Liu and Liu (2006) suggested to provide sufficient DO and alkalinity for the sludge, because the filamentous bulking is caused by the low DO of sludge suspension or low pH of feedwater in many cases.

Due to the membrane rejection, the SMP is more easily accumulated in MBRs, which results in the poor filterability of the sludge suspension. Iritani et al. (2007) reported that the contribution of the supernatant to the membrane fouling of an anaerobic activated sludge is almost 100%, indicating that SMP is the controlling factor in microfiltration of activated sludge. Several attempts have presented that polysaccharide-like substances in SMP contribute more than protein-like substances to membrane fouling (Rosenberger et al., 2006; Yigit et al., 2008). Some investigations presented that the occurrence of SMP in MBRs impacts on membrane fouling significantly, and SMP concentration and SMP composition would determine its fouling propensity. The control of SMP concentration in MBRs is crucial. In general, the control of SMP can be achieved by two approaches: adjustment of operation parameters (i.e., SRT, HRT, DO concentration, temperature, aeration) and addition of adsorbents or coagulants to reduce SMP concentration.

SMP Control could realize though adjustment of operation conditions. Barker and Stuckey (1999) summarized the process parameters (feed strength, HRT, OLR, SRT, substrate type, temperature, biomass concentration and reactor type) affecting the production of SMP in conventional activated sludge process. In MBRs, it is feasible to control SMP concentration in MBRs by selecting suitable operation parameters. Shin and Kang (2003) reported that SMP concentration in the MBR reactor and effluent increased to some extent and then became stable, and finally decreased at a long SRT. Liang et al. (2007) observed that accumulation of SMP in the MBR became more significant at short SRTs for the treatment of readily biodegradable synthetic wastewater. Moreover, SMP are actually eliminated to a large extent through biodegradation, adsorption or other mechanisms (Drews et al., 2006). Drews et al. (2007) observed that DO and nitrate concentrations appeared to have an impact on SMP elimination and SMP elimination could be lower at low DO concentration. The low DO concentration could also lead to poor flocculation, then, higher DO could give rise to a better filterability of sludge suspension (Kang et al., 2003; Jin et al., 2006). Sudden temperature changes led to spontaneous SMP release and increase in fouling rates (Drews et al., 2007). Morgan-Sagastume and Grant Allen (2005) found that sludge flocs deflocculation occurred under a temperature range from 30 to 45 ℃, which caused an increase in SMP concentration. To achieve low SMP concentrations, a sufficient supply of oxygen is required and sudden temperature change should be avoided (Drews et al., 2007). What's more, substrate type or feedwater composition affects the formation and elimination of SMP. McAdam et al. (2007) observed that carbon substrate had a great influence on floc stability. Acetic acid resulted in the production of high concentrations of small particles (i.e., colloids and solutes) due to the weakly formed flocs. Ethanol, on the other hand, encouraged the growth of strong flocs that were capable of withstanding shear.

SMP control could achieve by addition of adsorbents/coagulants. Addition of adsorbents or coagulants into sludge suspension can decrease the level of solutes and colloids or enhance the flocculation ability. The addition of powered activated carbon (PAC) is a simple and convenient method for fouling control. The PAC can not only incorporate into the bioflocs forming biologically activated carbon (BAC) (Ying and Ping, 2006), but also adsorb biopolymers in the sludge suspension. It suggests that PAC addition can improve membrane flux significantly; but, if the addition of PAC is beyond the optimal value, it will do harm to membrane permeation. Thus, the improved performance of the MBR requires regular replacement of aged BAC with fresh PAC (Ying and Ping, 2006). Coagulants can remove SMP by charge neutralisation and bridging (Wu et al., 2006). Addition of an optimum calcium concentration could induce lower SMP concentration, lower hydrophobicity, lower concentration of filamentous bacteria and better flocculation, which resulted in the reduction in cake layer resistance and pore blocking resistance (Kim and Jang, 2006). Attempts have been also made to use alum, ferric chloride, and chitosan as coagulants or filter aids (Ji et al., 2008; Koseoglu et al., 2008; Song et al., 2008). Zhang et al. (2008) reported that ferric chloride was found to be a preferred coagulant to reduce both SMP with MW > 10 kDa in the supernatant and sludge flocs in the range of 1 ~ 10 mm. Wu et al. (2006) showed that polymeric coagulants could provide more positive charges and longer chain molecules for filtration reinforce of sludge suspension than monomeric coagulants.

Even though most studies suggested that both EPS and SMP could play an import part on membrane fouling, some studies still have doubts on the role of SMP. SMP has close relationship with EPS, since SMP is soluble EPS, which is produced by EPS release, cell lysis, hydrolysis products (Laspidou and Rittmann, 2002; Rosenberger and Kraume, 2002). However, some studies showed that the influence of SMP on membrane fouling was mainly caused by EPS. Lee et al. also reported that the influence of SMP on membrane fouling could be ignored (Lee et al., 2003). Thus, the effect of SMP on membrane fouling varied from one study to another, because the experiment conditions were different from each other, and produced discrepant results. Therefore, the relationship between EPS and SMP, their functions on membrane fouling and the fouling mechanism still need to go further research, however, the importance role of EPS and SMP could not be ignored.

5. Conclusions and Recommendations

To remove contaminants and pathogens from various wastewater, this review provided MBR and its related technologies such as HMBR, MBMBR, OMBR to treat domestic wastewater and specific wastewater. However, membrane fouling is a major road block that prevents the applications of MBR and its related technologies for wastewater treatment and reuse. The factors causing membrane fouling, mechanism of membrane fouling, and fouling control strategies were reviewed.

In the coming few years, membrane fouling is still a hot issue in research and application of MBRs and its advanced related technologies. According to recent studies, the future research on membrane fouling should include:

(1) Optimizing operation conditions of MBR and its related technologies to make sure excellent membrane performances in contaminants removal and enhancing biodegradation process.

(2) Regulating microbial community in MBR and its related technologies to facility floc formation and reduce sludge bulking in wastewater treatment.

(3) Regulating the parameters affecting microbial species growth and bioactivity to increase contaminant removal efficiency in greywater and blackwater.

(4) Developing methods of the visualization and characterization of membrane fouling in advanced MBRs related technologies to observe the formation of membrane foulants.

(5) Discovering EPS/SMP roles on different kinds of membrane fouling, such as microfiltration, ultrafiltration, nanofiltration, reverse osmosis to understand how they work on cake formation and pore blocking on and within these membranes.

(6) Developing more effective and easy methods to control and minimize membrane fouling. Removable fouling could be realized by physical methods, but the key issue is to reduce their capital costs and enhance membrane hydrodynamic conditions to go against this kind of fouling. For irremovable fouling, the way to limit the deposition of foulants onto the membrane surface should be developed.

(7) Investigating the occurrence and fate of SMP in MBR systems, the dynamic process of SMP production and elimination, and the accumulation and detachment of SMP on or within the membranes.

(8) Developing membrane anti-fouling materials to paint on the surface or modify current membranes. Since the potential influence of adsorbents/coagulants on microbial community or biomass metabolism is unknown, these adsorbents/coagulants might have a potential environmental risk. At this point, some natural sources of anti-fouling materials are welcomed due to its benefits to the biomass in the system and in the environment in receiving waters.

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