ENVIRONMENTAL ENGINEERING SCIENCE
Volume 29, Number 3, 2012

DOI: 10.1089/ees.2010.0420

REVIEW ARTICLE

Recent Advances in Membrane Bioreactors: Configuration Development, Pollutant Elimination, and Sludge Reduction

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Received: November 17, 2010 *Accepted in revised form: April 9, 2011*

Abstract

Membrane bioreactors (MBRs) are composed of activated sludge processes and membrane filtration. Because of their unique advantages such as good effluent quality and compact structure, MBRs have been widely used for municipal and industrial wastewater treatment. Recent advances in MBR research are reviewed by focusing on development of bioreactor configurations, enhanced degradation of pollutants, and sludge reduction. Efforts of a number of novel MBR processes such as hybrid biofilm MBR, submerged rotating MBR, MBR with reverse osmosis, osmotic MBR, membrane distillation bioreactor, air-sparging MBR, and jet loop MBR for the treatment of nitrogen, phosphorous, emerging contaminants, heavy metals, and sludge reduction are summarized. Process principles, benefits, and limitations of these MBRs are discussed. According to a detailed analysis of research publications, MBR research has undergone an extensive growth in the areas of development of novel MBR configurations and application of MBRs with new purposes.

Key words: membrane bioreactor (MBR); wastewater treatment; water purification; wastewater reuse; sludge reduction

Introduction

Since the early twentieth century, activated sludge processes have been widely used in wastewater treatment (Arden and Lockett, 1914). However, the activated sludge process is usually limited by the difficulties in solid/liquid separation. To address the limitations of activated sludge process, membrane bioreactor (MBR) with combination of side-stream membranes and biological wastewater treatment was proposed in 1968 (Smith *et al*., 1969). However, wide applications of the side-stream MBRs were hindered by high energy consumption. A turning point of the development of MBRs was achieved when a submerged MBR was developed by Yamamoto *et al.* (1989). The submerged MBRs usually have lower energy consumption because of the lower suction pressure and the absence of recirculation pump.

Because of the unique feature of MBRs and particularly the significant decrease in membrane price, MBRs have been increasingly and widely used for wastewater treatments in the last decade (Judd, 2006, 2008; Yang *et al.*, 2006; Lesjean *et al.*, 2008; Wang *et al.*, 2008d; Huang *et al.*, 2010). According to the BCC research report (www.bccresearch.com, Membrane Bioreactors: Global Markets, 2008/06), >2500 MBRs have been in operation worldwide with an annual growth rate of 10.5% from 2008 to 2013. Table 1 lists the installations of MBR by some major membrane companies such as Kubota, Asahi Kasei Chemicals, Mitsubishi Rayon, GE/Zenon, Norit, and Koch with a treatment capacity larger than 1000 m^3/day. As shown in Table 1, municipal wastewater treatment was the earliest application of MBRs and is still the largest application, accounting for about 80% of all systems based on treatment capacity. In the coming years, sewage treatment will continue to be the primary use for MBR systems. However, small-scale MBR plants for tourist resorts, smaller communities, hotels, schools, aboard seagoing vessels, etc., also account for a great portion. As shown in Fig. 1, the full-scale MBR plants (especially for those smaller than 5000 m^3/day) are prevalent in North America, Europe, and recently, Asia. However, full-scale MBR references in Africa, Central/Eastern Europe, and South America are very limited. The applications of MBRs in different regions of a given country are also not the same, for example, in south-east China, where surface water is rich, the MBRs are mainly used for high-strength industrial

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Table 1. Key Information About Installations of Conventional Membrane Bioreactors Larger Than 1000 m^3/day from 2005 to 2009

Product name (company) Location of installation	*ADF (m^3/day)*	*Commission (year)*	*Waste-water*
Submerged MBRs			
Kubota MBR (Kubota)			
Ireland	1200	2005	M
Spain	1400	2005	M
United Kingdom	2892	2005	M
Spain	1200	2005	M
Germany	1860	2005	M
Israel	3000	2005	I
Germany	2910	2005	M
Spain	6800	2005	M
France	2860	2005	M
Italy	1270	2005	M
United States	7570	2005	M
United States	6813	2005	M
United States	1136	2005	M
United States	2271	2005	M
United States	3785	2005	M
United States	3785	2005	M
United States	2271	2005	M
Ireland	3000	2006	I
Spain	1575	2006	M
United Kingdom	4300	2006	M
Italy	1400	2006	M
United States	3407	2006	M
United States	2271	2006	M
United States	22,710	2006	M
United States	1938	2006	M
United States	1631	2006	M
United States	3875	2006	M
United States	1008	2006	M
France	2640	2007	M
France	1600	2007	M
France	9000	2007	M
United Kingdom	2132	2007	M
Spain	2700	2007	M
Italy	12,000	2007	M
Spain	35,000	2007	M
Italy	2200	2007	M
Turkey	2000	2007	I
Israel	3500	2007	M
United States	2271	2007	I
United States	1893	2007	M
United States	11,355	2007	M
United States	2271	2007	I
United States	2271	2007	I
United States	5148	2007	M
United States	3785	2007	M
United States	1249	2007	M
United States	1420	2007	I
United States	18,925	2007	I
Turkey	1500	2008	M
Spain	9000	2008	M
United States	3179	2008	I
United States	1136	2008	M
United States	1136	2008	M
United States	1703	2008	M
United States	1893	2008	M
Submerged MBRs (continued)			
Microza MBR (Asahi Kasei Chemicals Co.)			
China	10,800	2006	I
China	1500	2006	M
China	25,000	2007	I
China	6000	2007	I
Korea	1000	2007	M
China	100,000	2007	M
China	35,000	2007	M
Japan	1375	2008	M
China	40,000	2008	M
Korea	4200	2008	I
China	10,200	2009	I
China	8000	2009	I
Singapore	2880	2009	I
Mitsubishi Rayon MBR (Mitsubishi Rayon)			
Beijing Miyun/China	45,000	2006	M
Luoyang/China	4800	2006	I
Tianjin/China	4000	2006	I
South Korea	30,000	2008	M
Zeeweed® MBR (GE Water and Process Technologies)			
Porto Marghera/Italy	47,520	2005	I
Georgia/United States	56,800	2005	M
Jinqiao/China	30,965	2006	I
Inner Mongolia/China	31,000	2006	M
Mar Menor/Spain	20,000	2007	M
Genova/Italy	28,769	2007	M
Australia	29,000	2007	M
Dubai/United Arab Emirates	25,000	2008	M
Ashburn, VA/United States	37,584	2008	M
Peoria, IL/United States	37,584	2008	M
Daewoo MBR (Daewoo Institute of Construction and Technology)			
Jecheon-si/South Korea	1100	2009	M
Gumi-si/South Korea	8000	2009	M
Dangjin-gun/South Korea	2000	2009	M
Dangjin-gun/South Korea	1500	2009	M
Anseong-si/South Korea	2200	2009	M
Anseong-si/South Korea	3000	2009	M
PURON™ MBR (Koch Membrane System)			
Perth/Australia	1700	2006	I
Tamil/India	5000	2006	I
External MBRs			
AirLift™ MBR (Norit)			
Ootmarsum/The Netherlands	3600	2005	M
Xuzhou/China	2000	2006	I
Millsborough, DE/United States	4000	2006	M
Valyampet/India	4000	2007	I
Thutipet/India	2400	2007	I
Dubai/United Arab Emirates	17,000	2007	M
Caracas/Venezuela	2400	2008	I
DynaLift™ (Parkson)			
Delaware/United States	4353	2009	M

The data listed in this table are based on information supplied by the company, and literature review reflects the operating and design conditions at the time of the interview.

M, municipal wastewater; I, industrial wastewater; ADF, average daily flow; MBR, membrane bioreactor.

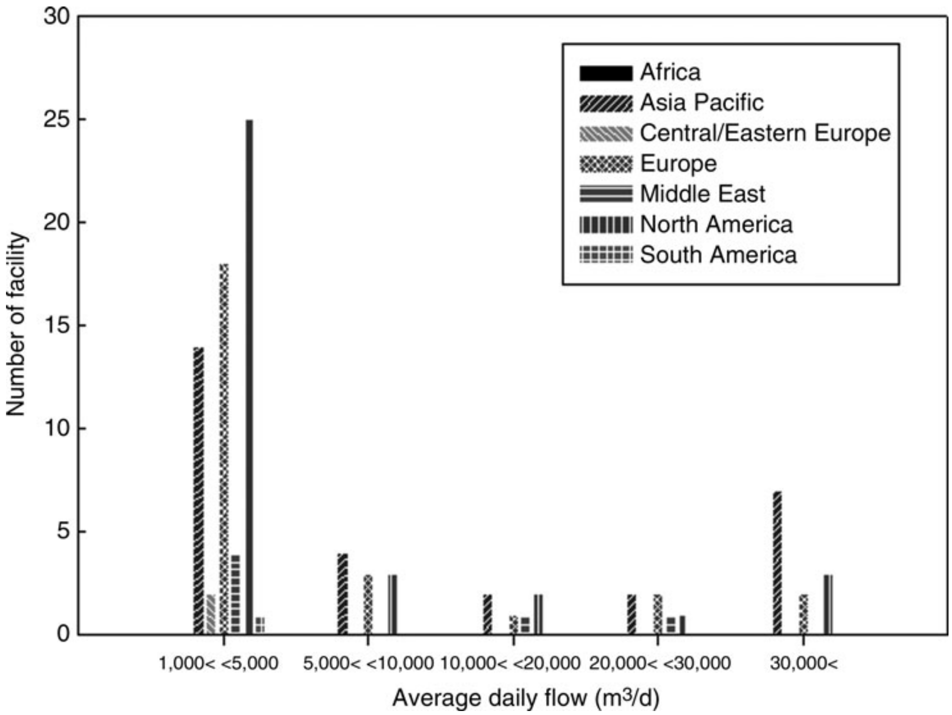

FIG. 1. Geological distribution of global MBR installation based on Table 1.

wastewater treatment, whereas in the water-poor regions such as north China they are used for municipal wastewater treatment, with a main purpose of wastewater reuse (Zheng *et al.*, 2010).

Along with the wide applications of MBRs, a number of scientific issues in relation to MBRs arose and have drawn much attention. It led to active research in MBRs, which can be revealed by the number of publications. In addition, the large-scale use of MBRs in wastewater treatment will require either new technology developments or a further decrease in the price of membranes. Other related wastewater treatment processes such as biofilm, aerobic granules, coagulation, adsorption, Anammox, and membrane distillation (MD) are integrated into MBRs either to achieve better process performance or to expand the use purpose of MBRs. Additionally, the treatment of refractory compounds such as emerging contaminants and heavy metals also boosts a wider application of MBRs. Typically, research of membrane fouling and developments of antifouling membranes, which were previously reviewed (Meng *et al.*, 2009a), have also attracted much attention. According to the literature, MBRs are also used for sludge reduction or sludge digestion.

It can be noted that considerable attempts have been made and significant findings have been also achieved. However, the operating principles, advantages, and current limitations of these novel MBR configurations, particularly for the real application of these MBRs, have not been paid enough attention. In addition, the mechanisms underlying the removal of emerging contaminants and heavy metals by the MBRs were also not fully clarified, although they were mentioned in some publications with different depth. In light of these issues, detailed analysis of past research efforts is expected to provide a clearer picture of the reported findings. Further, the current and future trends in MBRs are also of great interest to both researchers and engineers working on MBRs. This article intends to review recent developments in MBRs by focusing on novel MBR processes, pollutant elimination, and sludge reduction.

Developments of Novel MBRs

Recently, research and developments of novel MBRs have attracted more attention. As shown in Fig. 2, hybrid biofilm MBR (HFMBR), vertical submerged MBR (VSMBR), submerged rotating MBR (SRMBR), MBR with reverse osmosis (RO), osmotic MBR (OMBR), membrane distillation bioreactor (MDBR), air-sparging MBR (AsMBR), jet loop MBR (JLMBR), membrane coagulation/adsorption bioreactor fungi MBR, and anaerobic ammonium oxidation (Anammox) MBR have been developed for wastewater reuse, flux enhancement, high-strength wastewater treatment, and biological nutrient removal (i.e., nitrogen and phosphorous elimination).

New MBR configurations

HFMBR. The HFMBR was developed for either membrane fouling control or simultaneous organic and nitrogen removal. This process incorporates biofilm technology [e.g., moving-bed-biofilm reactor (Ivanovic *et al.*, 2006)] and membrane filtration into one single reactor. One feature of this process is that it can be used for high-strength wastewater treatment because of the high biomass concentration in the reactor (Artiga *et al.*, 2005). Additionally, the coexistence of aerobic zone and anaerobic zone in one biofilm can realize simultaneous nitrification–denitrification (SND) in a single reactor (Yang *et al.*, 2009). Therefore, it could be an alternative for industrial wastewater treatment, especially when space is limited.

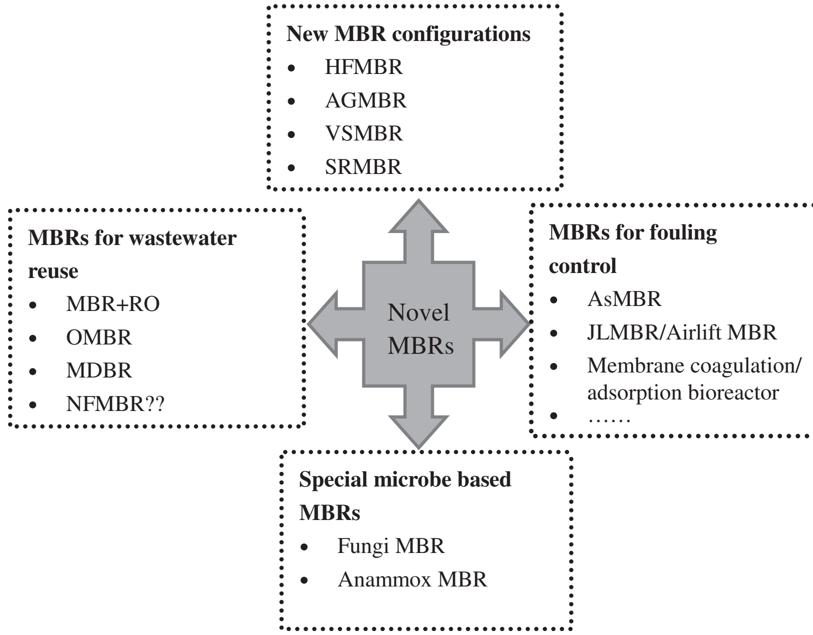

FIG. 2. Summarized illustration showing novel MBRs developed in recent years. AGMBR, aerobic granular sludge MBR; AsMBR, air-sparging MBR; HFMBR, hybrid biofilm MBR; JLMBR, jet loop MBR; MDBR, membrance distillation bioreactor; NFMBR, nanofiltration MBR; OMBR, osmotic MBR; RO, reverse osmosis; SRMBR, submerged rotating MBR; VSMBR, vertical submerged MBR.

It should be noted that the reported findings with regard to the influence of biofilm on membrane fouling are inconsistent or even contradictory. Some reported that the HFMBR not only allowed high biomass concentration, but also reduced membrane fouling evidently (Leiknes *et al.*, 2006). The reduced membrane fouling was likely due to either the fact that shear force generated by circulating media on membranes prevents the formation of cake layer (Lee *et al.*, 2006) or the fact that the biofilm carriers could entrap fouling-causing substances such as soluble microbial products (SMP) (Liu *et al.*, 2010b). Oppositely, severe fouling was found in some HFMBRs. For example, a previous study showed that the fouling rate of an HFMBR was about seven times higher than that of a conventional MBR (Lee *et al.*, 2001). One possibility for the discrepancies of the previous findings is the use of different biofilm carriers and feed wastewater, which causes differences in the production/degradation of SMP and in the biofilm characteristics.

Aerobic granule sludge, which can be considered as a special form of biofilm (Liu and Tay, 2004; Adav *et al.*, 2008; Liu *et al.*, 2009), was also integrated with MBR process (Li *et al.*, 2005; Tay *et al.*, 2007; Rui and Jin, 2008). As expected, the aerobic granular sludge MBR (AGMBR) could obtain better membrane permeability owing to the low compressibility of granular sludge. Therefore, the AGMBR not only has an excellent performance on the removal of chemical oxygen demand (COD) and total nitrogen (TN) as a result of the special structure of the granules, but also could enhance membrane permeation greatly (Yu *et al.*, 2009). Even so, the cultivation and instability of aerobic granular sludge are the major problem limiting the wider application of AGMBR (Wang *et al.*, 2008a). Additionally, the fouling mechanism of AGMBR is different from that of conventional MBR (Li *et al.*, 2005; Juang *et al.*, 2008). For example, Zhou *et al.* (2007) reported that the irreversible fouling as a result of the deposition of colloids and solutes on membranes was the major fouling mechanism of AGMBR.

The growth and metabolism of microorganisms in biofilm are different from those in sludge flocs. Thus, these two systems should have different bacterial community structures. With respect to the HFMBR, the formation of biofilm is accompanied by the attachment and detachment of microorganisms. Thus, some kind of microorganisms that have high affinity with carriers can be enriched. In the same way, these enriched microorganisms can deposit onto the membranes more readily because of the higher adhesion potential. In fact, the generation and degradation behaviors of SMP in HFMBR remain unclear so far and it could be a potential subject for future work.

VSMBR. A novel VSMBR composed of anoxic and oxic zones in one reactor was developed in an attempt to address the problems concerning effective removal of organic compounds and nutrients as well as mitigation of membrane fouling (Fig. 3a) (Chae *et al.*, 2006b). The optimal volume ratio between anoxic zone and oxic zone was found to be 0.6. The desirable internal recycle rate and hydraulic retention time (HRT) for effective nutrient removal were 400% and 8 h, respectively. Under these conditions, a pilot-scale VSMBR was fabricated and operated to remove organics and nutrients from municipal wastewater (Chae and Shin, 2007b). As a result, total suspended solid (TSS) and COD were removed by almost 100% and higher than 98%, respectively. Moreover, the average removal efficiencies of TN and total phosphorus (TP) were found to be 74% and 78% at HRT and sludge retention time (SRT) of 8 h and 60 days, respectively. The specific removal rates of TN and TP were found to be 0.093 kg N/m^3/day and 0.008 kg P/m^3/day, and the daily production of excess sludge was 0.058 kg TSS/day. A schematic illustration of the commercially used VSMBR is shown in Fig. 3b. This MBR configuration is expected to challenge current limits to the effective removal of nutrients from wastewater as well as for the reduction of excess sludge and the reduction of membrane fouling (Chae *et al.*, 2006a, 2007a; Chae and Shin, 2007b).

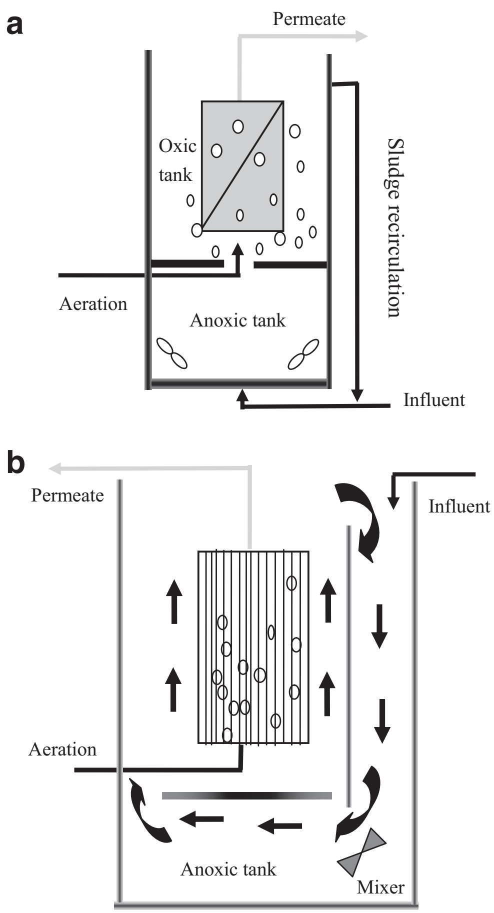

FIG. 3. Schematic diagram of **(a)** VSMBR (drawn after Chae *et al.*, 2006b) and **(b)** Daewoo MBR (drawn after www.dwconst.re.kr/tech/tech2_2.asp?boardkey = 19).

SRMBR. SRMBR is featured as a directly immersed, rotatable, rounded membrane module in a bioreactor. Rotation or oscillation of the membrane modules themselves is an alternative method to generate turbulence (Rector *et al.*, 2006; Prip Beier *et al.*, 2009). Therefore, using rotational membrane module or oscillatory membrane module in MBRs can improve mass transfer and control membrane fouling (Wu *et al.*, 2008; Bai *et al.*, 2009). As reported by Zuo *et al.* (2010), the permeate flux increased from 42.5 to 47.5 $L/m^2/h$ as the rotation rate increased from 15 to 25 rpm. A novel rotational membrane aerated biofilm reactor (MABR) was also proposed to convert ammonia nitrogen in concentrated wastewater to nitrates with high rates (Rector *et al.*, 2006; Tansel *et al.*, 2006). With respect to anaerobic MBRs particularly, the rotation or oscillation of membrane module can not only eliminate concentration polarization and fouling layer, but also agitate sludge suspension saving the energy consumption of the conventional mechanical stirring. For example, for a rotating drum mesh filter bioreactor developed for anaerobic digestion of biodegradable municipal solid waste, no fouling was experienced during the experiment at a flux of 3.5 $L/m^2/h$ (Walker *et al.*, 2009). Likewise, a vibration MBR with a spring-mass system in conjunction with a vertically suspended hollow-fiber membrane bundle could also be an interesting option for fouling control (Low *et al.*, 2009; Altaee *et al.*, 2010). The vibration of membrane module could be a more effective approach for fouling control than aeration and cross-flow velocity (Low *et al.*, 2009).

In addition to conventional MBRs, ion-exchange MBR (IEMBR), extractive MBR (EMBR), and MABR have also been applied to remove pollutants from wastewater. In the MABR, the biofilm naturally grow and attach on gas permeable membranes (Syron and Casey, 2008). The air or oxygen diffuses through the membranes and then into the biofilm. The pollutants diffuse to the biofilm from the other side of the membranes. This process can not only achieve nonbubble aeration (i.e., high oxygen transfer coefficient), but also create aerobic and anaerobic zone in the biofilm (e.g., achieving nitrification and denitrification simultaneously). In the IEMBR, the targeted pollutants such as metal ions are transported from the wastewater stream through a nonporous ion-exchange membrane into a biological compartment where they are subsequently adsorbed or converted by a biofilm (Crespo *et al.*, 2004; Oehmen *et al.*, 2006). In the EMBR, the volatile organic matter in the wastewater penetrates through a dense organophilic membrane and then into the biomass in the other side of the membrane (Brookes and Livingston, 1995; Emanuelsson *et al.*, 2003). It should be noted that the EMBR and IEMBR allow the isolation of the microbial culture from the feed stream via a dense membrane barrier, which can avoid the contamination of the treated water with cells, organic matter, and excess carbon source. Ion-exchange membrane and extractive membrane, however, are expensive in comparison with the membranes often used in conventional MBRs. Hence, conventional MBRs are more viable for large-scale wastewater treatment. The IEMBR, MABR, and EMBR can be used in some special occasions. It can be observed that these MBRs are of different configurations and operating principles. Additionally, the transport of substrates in these MBRs is completed by different types of driving force. Detailed information of these MBRs can be found elsewhere (Crespo *et al.*, 2004; Syron and Casey, 2008).

Novel MBRs toward wastewater recycling

MBR + RO. To cope with the situation of water scarcity, investigators and engineers have developed a number of treatment methods, which aimed to either eliminate the micropollutants present in water or increase water supplies through a reliable reuse of wastewater (Shannon *et al.*, 2008). Of all the treatment methods for wastewater reuse, membrane-based technology has attracted much attention, which usually includes two processes (Arevalo *et al.*, 2009): conventional activated sludge (CAS) process followed by membrane filtration (CAS-MF) and MBRs. The former is mostly used for the upgrade of conventional wastewater treatment plants (WWTPs), whereas the latter is preferred for new commissioned plants. Typically, the WWTP effluent contains a pool of residues of biodegradation such as dissolved organic matter, which are also known as SMP. As a consequence, a posttreatment (e.g., ultrafiltration [UF]) is desirable to further remove parts of organic compounds present in WWTP effluent (van Hoof *et al.*, 1998; van der Graaf and Krmer, 1999; Duin *et al.*, 2000; Huang *et al.*, 2009; Zheng *et al.*, 2009). Another possible option is the direct reuse of MBR effluent, combining the biological treatment and membrane filtration in a single tank. Despite the presence of some dissolved species in MBR effluent, it can generally meet the requirement of irrigation purpose, toilet flushing, and possible industrial applications (Merz *et al.*, 2007). Crucially, the land requirement and capital cost of MBRs are much lower than CAS-MF processes, for example, MBR plant required half land of CAS-MF as reported by Cote *et al.* (2004). In some case, the MBRs could produce treated water with higher quality than CAS-MF system (Kent *et al.*, 2011).

Unfortunately, these low-pressure membranes (i.e., microfiltration and UF) cannot completely reject humic substances, virus, and waterborne pathogens, which can cause a number of public health problems such as cholera, diarrhea, and hepatitis (Rose *et al.*, 1996). At this point, RO is preferentially used as a posttreatment for low-pressure membrane-based processes. As such, a new MBR coupled with RO process has been recently applied for the safe reuse of wastewater (Shannon *et al.*, 2008). In the MBR + RO process, the RO membrane can remove the remaining compounds present in the MBR effluent (Comerton *et al.*, 2005; Tam *et al.*, 2007). On the other hand, MBR acts as a pretreatment to reduce fouling of RO membranes (Scholz *et al.*, 2005).

Owing to the enhanced biodegradation of pollutants by MBRs and the complete rejection of organic compounds by RO membranes, the MBR + RO process has an excellent performance for the removal of waterborne pathogens, disinfection byproducts, trihalomethanes (THM), and nitrate (Comerton *et al.*, 2005). For instance, RO membrane filtration can reject nitrate up to 88%–97% in tap water (Molinari *et al.*, 2001). Of particular interest is that although the residual ammonium in wastewater cannot be completely rejected by RO membranes because of the small molecular size, they can be totally converted to nitrate via nitrification in an MBR. As an example, RO can achieve an average nitrate removal of 93% during landfill leachate treatment by a full-scale MBR + RO process (Ahn *et al.*, 2002). Therefore, the combined MBR and RO treatment process could be technically viable for wastewater reuse.

It must be addressed that not all the organic compounds (e.g., polysaccharides, proteins, and humic substances) in sludge suspension could be rejected by MBRs; particularly the humic substances could pass the membranes more readily because of their small size (Meng *et al.*, 2009b). As a result, large-sized organic compounds play a significant role in MBR fouling; and the small-sized compounds, which are perhaps dominated by humic substances, would act to foul the subsequent RO membranes. At present, however, literature in relation to fouling mechanisms of the separate process in the MBR + RO process is limited. Additionally, there is a need to search for more effective approaches for RO fouling control, for example, replacing microfiltration membranes with UF membranes in the MBRs (Shannon *et al.*, 2008) to mitigate the organic loading of the following RO membranes, but it must be borne in mind that the discharge of the concentrated stream produced by RO can bring additional environment problems (Van der Bruggen *et al.*, 2003). In addition to RO filtration, disinfection is another option that can be used to improve the MBR effluent quality, for example, the use of hypochlorite (Boake, 2006) and granular ferric hydroxide (Ernst *et al.*, 2007). In fact, the implementation of posttreatment for the MBR-treated water strongly depends on the quality of MBR effluent and especially the requirements of end-users.

OMBR. In an OMBR, a forward osmosis (FO) membrane instead of microporous membrane is submerged in the bioreactor (Cornelissen *et al.*, 2008; Achilli *et al.*, 2009). The treated water is drawn from the sludge suspension across a semipermeable membrane by a draw solution containing salts (e.g., $MgCl_2$, NaCl, KCl, and NH_4HCO_3) that generates an osmotic pressure; it thus serves as a driving force for the OMBR. A pilot-scale study showed that fluxes of 3 and 7.2 L/m^2/h obtained at an osmotic pressure of 6 (0.12 M mixed salt of NaCl and $MgSO_4$ at a ratio of 3:1) and 24 atm (0.5 M NaCl), respectively (Qin *et al.*, 2009). Compared with the microporous membranes (e.g., microfiltration and UF), the semipermeable FO membranes have a higher rejection degree. For example, the OMBR was found to reject 99% of organic carbon and 98% of ammonium nitrogen, respectively (Achilli *et al.*, 2009), indicating that OMBR is of high interest for desalination. But, because of the inflow of treated water, the draw solution in an OMBR is gradually diluted. Therefore, OMBR process is usually followed by an RO filtration process, which is used to produce high-quality water and reconcentrate the draw solution (Achilli *et al.*, 2009). The dense FO membranes, which have a comparable structure as nanofiltration (NF) or RO membranes, can reject almost all organic substances and bivalent ions, so the subsequent RO system can be operated with higher flux because of the low organic loading (Cornelissen *et al.*, 2008). On the other hand, the FO membranes have lower fouling potential than microfiltration or UF membranes (Cornelissen *et al.*, 2008), which is probably due to the low driven force of the salt solution. The OMBR + RO process therefore is more technically and economically viable than the MBR + RO process.

However, the fact that the permeability of FO membranes is lower than that of RO membranes is a key barrier in the use of OMBR (Wang *et al.*, 2010). Different from the fouling mechanisms of microporous membranes used in conventional MBRs, the fouling of FO membranes mostly results from the presence of external and internal concentration polarization. The concentration polarization can reduce the osmotic driving force and then decrease the expected flux (McCutcheon and Elimelech, 2006; Xu *et al.*, 2010b). The concentration polarization is impacted by a number of factors such as hydrodynamics, sludge characteristics, and membrane structure. One approach to control the concentration polarization is to develop FO membranes with ultrathin active layer. Considerable attempts were performed with an aim to develop FO membranes with higher permeability. The work was based on either modification of currently used NF membranes or design of new membranes (Benko *et al.*, 2006; Wang *et al.*, 2007; Setiawan *et al.*, 2010). In addition, it should be pointed out that the concentration polarization in an OMBR may cause a more severe membrane scaling because of the increased concentration of ions on the membrane surface.

MDBR. The MDBR, which integrates activated sludge process with MD, was developed in Singapore (Phattaranawik *et al.*, 2008, 2009; Khaing *et al.*, 2010). In the MDBR, treated water is separated by "evaporation" mechanism, and the retention time of nonvolatile substances in the MDBR and the HRT are independent (Phattaranawik *et al.*, 2009). Because of the fact that the MD can only transfer volatile substances, the production of treated water with very low total organic carbon (TOC) levels and negligible salts is possible. A long-term experiment of 105 days showed that the salt rejection degree indicated by electrical conductivity was stable

(~99.75%) regardless of the variation of feedwater loading rates (Khaing *et al.*, 2010). As such, the MDBR can obtain wastewater reuse in one step, and thus it is comparable with the MBR + RO process. One noticeable feature of the MDBR is that the degradation of pollutants is completed by thermophilic bacteria because of the high operating temperature, for example, a temperature up to 50°C was reported by Phattaranawik *et al.* (2008). Practically, the high operating temperature is a challenge to the use of polymeric membranes, particularly with respect to the long-term operation, though the membranes were observed to be thermally stable in reported literature (Khaing *et al.*, 2010). As the MDBR is thermally driven (Phattaranawik *et al.*, 2008, 2009), additional energy is usually required. Therefore, the full-scale application of MDBR is viable only if waste energy is available. It implies that the process might be an alternative option for the treatment of some industrial wastewater, which is usually discharged with high temperature, such as distillery wastewater (Zhang *et al.*, 2006b).

In addition to the aforementioned novel MBRs (i.e., MBR + RO, OMBR, and MDBR), one more process worthwhile to mention here is nanofiltration MBR (Choi *et al.*, 2005, 2007), which enables the rejection of organic matter larger than ~200 Da and parts of ions. As a result, nanofiltration MBR can also generate permeate with high quality, for example, a dissolved organic carbon (DOC) concentration of permeate in a range of 0.5–2 mg/L and rejection degrees of monovalent and divalent ions in a range of 40%–60% and 70%–90%, respectively, were reported by Choi *et al.* (2007). However, the membrane deterioration as a result of biodegradation can lead to the increase of pore size and porosity and thus the decrease of membrane rejection degree (Choi *et al.*, 2005, 2007).

Technical innovations toward fouling mitigation

AsMBR. Air-sparging can significantly enhance the membrane permeation because of the effective use of bubbling for mitigation of concentration polarization and cake deposition (Cui *et al.*, 2003). In recent years, air-sparging has also been used in MBRs to control membrane fouling (Chang and Judd, 2002; Psoch and Schiewer, 2005a, 2006, 2008; Guglielmi *et al.*, 2008). Because MBRs themselves need aeration to provide oxygen for microbe respiration, the air-sparging technique offers an alternative to optimize the design of aeration modes. For a given AsMBR, air bubbles are injected into the lumen or outside of membranes to enhance permeate flux. The bubble motion in the lumen or module may generate a number of flow patterns. The flow patterns, depending on the value of void fraction, are defined as bubble flow, slug flow, churn flow, and annular flow (Cabassud *et al.*, 2001; Chang and Judd, 2002).

In a membrane tube, slug flow pattern is usually more effective for fouling control (Li *et al.*, 1997; Mercier *et al.*, 1997). Under this flow pattern, water film and air slug can remove fouling layer by generating shear stress (Psoch and Schiewer, 2005b). Because of this, there has been a strong trend toward the use of air-sparging in submerged MBRs (Chang and Judd, 2002; Ghosh, 2006). Experimental evidence suggested that air-sparging was very efficient to enhance critical flux during membrane filtration of sludge suspension (Yu *et al.*, 2003). The results of Psoch and Schiewer (2005b) showed that air-sparging could significantly increase membrane flux in a long-term operation of MBR. They found that when the liquid and gas velocities were fairly equal to each other, a high flux was achieved. Membrane module design also has significant impacts on the efficiency of air-sparging. Usually, a membrane module that can either make the bubbles stay in the shell with long time or induce a better air-lift flow pattern is favorable (Ghosh, 2006).

Considering the dual roles of aeration in MBRs (i.e., providing oxygen for microorganism metabolism and fouling control), optimization of air-sparing is required. An experimental study, for instance, showed that the integration of intermittent air sparing into an MBR can not only lead to the occurrence of denitrification, but also decrease the specific energy demand from 0.19 (for constant sparging) to 0.007 kWh/m^3 as well (McAdam *et al.*, 2010). Nevertheless, there are a couple of issues that need to be taken into consideration during the operation of AsMBRs. For instance, a high air-sparging intensity may cause floc breakage. Moreover, so far the investigation of AsMBRs was mostly based on lab-scale membrane modules; therefore, the design and standardization of membrane module for commercial use are not yet solved.

JLMBR. As dissolved oxygen (DO) concentration is the most important parameter affecting metabolic activity of sludge, the improvement of oxygen transfer kinetics is of great concern for the operation of MBRs. The mass transfer kinetics is usually quantified by the oxygen mass transfer coefficient, k_La. Generally, an improved mass transfer of a bioreactor could guarantee high removal rate of pollutants such as nitrification (Lazarova *et al.*, 1997). A jet loop reactor is basically composed of two concentric cylinders, of which the inner one is called "downcomer compartment" and the outer one is called "riser compartment" (Fig. 4). A two-phase nozzle is needed to disperse the gas delivered in. The high turbulence enables the disintegration of large microorganism aggregates, thus creating a large specific surface area. As shown in Fig. 4, the JLMBR is a compact biological treatment unit that requires

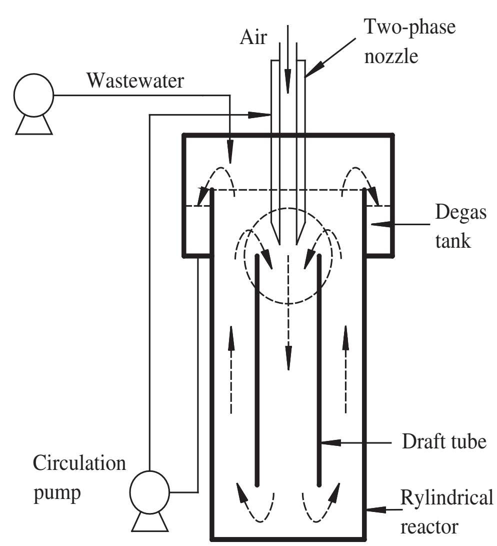

FIG. 4. Schematics of a jet loop bioreactor (drawn after Park and Lee, 2005).

much smaller footprint than CAS system (Farizoglu *et al.*, 2007).

As a result of high shear stress in jet loop reactor, the activated sludge is often found as nonflocculating (Farizoglu *et al.*, 2004, 2007; Farizoglu and Keskinler, 2006), which is troublesome for solid/liquid separation. On the other hand, MBRs are characterized by the high sludge concentrations, which result in high viscosities that limit oxygen transfer and subsequently require more energy for aeration (Rosenberger *et al.*, 2002; Drews *et al.*, 2005). A combination of membrane separation and jet loop bioreactor can not only realize good solid–liquid separation in the jet loop bioreactor, but also address the mass transfer problem in the conventional MBRs at considerable levels.

The enhancement of gas–liquid mass transfer in a JLMBR is achieved by optimizing two operating parameters: recirculated liquid flow rates and gas flow rates (Kouakou *et al.*, 2005). For example, Kouakou *et al.* (2005) reported that as the air and recirculated liquid superficial velocities were gradually increased from 0.013 to 0.019 m/s and 0.0056 to 0.011 m/s, respectively; the gas–liquid mass transfer coefficient was varied between 0.01 and 0.02 s^{-1}. It was also found that the square cross-sectional draft tube can obtain higher K_La than the circular draft tube (Farizoglu and Keskinler, 2007). The square draft tube could make air bubbles to stay in bioreactor for a long time, and thus the mass transfer between air bubbles and liquid was improved. A similar study performed by Lu *et al.* (2000) also stated that the irregularly geometrical region of draft tube would result in an increase in K_La. These findings indicate that the structure of jet loop bioreactor itself has strong influence on mass transfer coefficient.

Because of the improved mass transfer coefficient, the JLMBRs are expected to have higher organic removal rates (Park and Lee, 2005; Park *et al.*, 2005; Farizoglu *et al.*, 2007). Yildiz *et al.* (2005) found that the JLMBR had a COD removal efficiency of about 97% with a volumetric organic load of 2–97 kg COD/m^3/day. Farizoglu *et al.* (2007) observed that the removal efficiencies of COD, TN, and TP were 97%, 99%, and 65%–88%, respectively, at the SRT of 1.6 days and volumetric loading of 22.2 kg COD/m^3/day, 17–436 g TN/m^3/day, and 30–134 g TP/m^3/day. These evidences suggest that the high mass transfer coefficient in JLMBRs makes them possible to treat high-strength wastewater with high efficiencies. Another important feature of JLMBR is the effective control of membrane fouling (Yeon *et al.*, 2005). The bubbles and turbulence generated by the liquid jet in the JLMBRs can mitigate cake layer formation on the membranes. An attempt performed by Yeon *et al.* (2005) showed that the transmembrane pressure (TMP) of conventional MBR reached to 30 kPa after 1.5 h of operation, whereas the increase of TMP was negligible in the case of JLMBR. The optimum location of a membrane module was found at the bottom of the inside draft tube because of the presence of rotational flow pattern of the mixed liquor (Yeon *et al.*, 2005).

The JLMBRs have distinctive advantages such as the ability to treat high-strength wastewater, low fouling potential, low area requirements, and easy operation (Yeon *et al.*, 2005; Yildiz *et al.*, 2005). However, because of a high recirculation rate, it is expected that the JLMBRs might have higher energy consumption than conventional MBRs. Therefore, the benefits of JLMBRs for nutrient removal and fouling control have to be weighted against the potential of energy usage. More crucially, a too high recirculation rate of mixed liquor or aeration rate may lead to floc breakage, resulting in the production of more SMP, for example, as recirculation rate increased from 12 to 14 L/min, the concentrations of polysaccharides and proteins increased from 45 and 49 mg/L to 85 and 120 mg/L, respectively (Park *et al.*, 2005).

In addition to AsMBR and JLMBR, airlift MBR has also drawn increasing attention in recent years (Fan *et al.*, 2006; Futselaar *et al.*, 2007; Xu and Yu, 2008b). The airlift MBR with a side stream membrane filtration unit was commercially used by NORIT X-Flow. The membrane module is mounted vertically. A vertical orientation allows the cross flow to be maintained by both a circulation pump and injecting air at the bottom of the membrane module to circulate the sludge via an "air-lift" pump effect. The energy consumption of airlift MBR for toilet wastewater treatment was reported to be 0.32–0.64 kWh/m^3 by Fan *et al.* (2006). Noticeably, the configuration of airlift MBR seems to be much simpler than JLMBR, for example, the absence of two-phase nozzle. Yet all these MBRs, different as they seem, have one feature in common, that is, all of them were designed to make the best use of aeration and then to enhance the hydrodynamic conditions of membrane module. It indicates that a proper design of membrane module is another option for flux enhancement. A ladder-type flat membrane module with a certain inclined angle theta, for example, was designed and found to be effective for enhancing the intensity and collision frequency between air bubbles and membranes (Li *et al.*, 2009). Recently, other new modules such as helical membrane module reported by Liu *et al.* (2010a) and straw-like hollow fiber membrane module commercially used by PURON have also shown promising perspective in the development of antifouling MBRs. In fact, the development of antifouling MBRs is often related to the control of SMP or extracellular polymeric substances (EPSs). Some hybrid MBRs such as membrane coagulation/adsorption bioreactor and MABR (Tian *et al.*, 2008a, 2008b, 2010) and membrane electrobioreactor (Akamatsu *et al.*, 2010; Bani-Melhem and Elektorowicz, 2010) are representative examples. Fouling control via coagulation/adsorption and electric field has been discussed in detail previously (Meng *et al.*, 2009a).

Specialized microbes associated MBRs

Fungi MBR. Fungi are eukaryotic cells containing significantly more genes than bacteria, which have several advantages over bacteria, such as high resistance to inhibitory compounds (Guest and Smith, 2002). Some pollutants present in industrial wastewater (e.g., textile wastewater) are possibly toxic to bacteria, but they can serve as substrate for fungi. As such, fungi have been used to eliminate toxic organic compounds or heavy metals. Recently, a submerged membrane fungi reactor was developed by Guest and Smith (2002), Kim *et al.* (2004), and Hai *et al.* (2005, 2006, 2008a, 2008b), which was used to enhance textile wastewater treatment. Different from the microbes in conventional MBRs, fungi are responsible for pollutant degradation in this process. It was reported that the reactor obtained 97% TOC and 99% color removal from synthetic wastewater (TOC = 2000 mg/L; dye = 100 mg/L) at an HRT of 15 h (Hai *et al.*, 2006). Because of the complexity of dye-containing wastewater, the mixed culture in a fungi MBR is composed of fungi and bacteria (Hai *et al.*, 2008b). The abundance of fungi and bacteria in the MBRs strongly relies on the feed wastewater types, operating conditions, and membranes. The presence of bacteria may impair the enzymatic activity of fungi, which thus have a potential to decrease the removal rate of dyes (Hai *et al.*, 2009). On the other hand, avoiding or decreasing the washout of fungi enzyme is desirable to obtain a steady and excellent removal efficiency of dyes. The membranes including the fouling layer are expected to play a role in the rejection of fungi enzyme (Hai *et al.*, 2008b). In addition, some adsorbents such as powdered activated carbon can adsorb the enzyme and thus improve the MBR performance. Because of the unique characteristics of dye-containing wastewater and fungi, the fungi MBRs usually have more severe fouling than conventional MBRs. The fouling of fungi MBRs can be controlled by use of adsorbents, membrane fouling reducer, modification of membrane modules, etc. It is of interest to compare the fouling mechanisms of fungi and activated sludge. The findings of such work may help to develop more effective fouling control strategies.

Anammox MBR. In recent years, various MBRs were developed to upgrade currently used biological wastewater treatment technology. Of particular notice is the combination of MBR and anaerobic wastewater treatment processes such as anaerobic baffled reactor, upflow anaerobic sludge blanket, and expanded granular sludge blanket (Chu *et al.*, 2005, 2006; An *et al.*, 2008b, 2009; Pillay *et al.*, 2008). The increased awareness of climate change and global warming has boosted the use of anaerobic wastewater treatment process. In addition to the zero net CO_2 emission, the anaerobic MBRs have low energy consumption, and typically, they have high potential to generate bioenergy (e.g., H_2 and methane) (An *et al.*, 2008c; Lee *et al.*, 2009).

Among all the anaerobic processes, Anammox MBR is one interesting technology (Trigo *et al.*, 2006; van der Star *et al.*, 2008; Wang *et al.*, 2009; Zhang *et al.*, 2009). Anammox is a biological reaction in which ammonia is oxidized to nitrogen gas using nitrite as the electron acceptor under anaerobic conditions. The main limitation of this process is the low growth rate of Anammox bacteria. Thus, the problem of biomass washout with effluent should be avoided to increase biomass concentration. An alternative addressing this issue is to use membrane filtration. In fact, MBR is a promising tool for the cultivation of Anammox bacteria, in which the Anammox bacteria could be enriched with high purity and productivity (van der Star *et al*., 2008). Importantly, the decoupling of HRT and SRT in an MBR can allow the startup of the Anammox process with a high concentration of seeding biomass (Trigo *et al.*, 2006), which can thus shorten the startup period greatly. Based on microbial community analysis, it was found that Anammox bacteria can be gradually enriched during a long-term operation of the MBRs (Trigo *et al.*, 2006; Wang *et al.*, 2009). One interesting process recently reported is the integration of nonwoven MBR and Anammox, which was found to be of low cost and antifouling besides the role of Anammox enrichment (Ni *et al.*, 2010). An attempt showed that the MBR coupled with Anammox process could obtain a high nitrogen removal rate up to 710 mg/L/day with almost full nitrite removal (Trigo *et al.*, 2006). The Anammox bacteria grew in granules instead of flocs in this MBR, which are helpful for fouling control. Because of the different operating procedure between MBRs and conventional biological processes (i.e., activated sludge and biofilm), the Anammox bacteria in these bioreactors might be subject to different metabolism. Further investigation is needed to confirm such questions.

Summary

Recently, a variety of novel MBR processes have been developed, which are mainly aimed at enhancement of pollutant removal, fouling control, and decrease of operating cost. Air-sparging may provide a simple and cost-saving aeration mode in MBRs, as the aeration energy in MBRs is still considerably high so far. The JLMBRs can not only mitigate membrane fouling, but also enhance the mass transfer at considerable levels. It also implies that it is of high interest to develop novel membrane module to make the best use of aeration for fouling control. The use of membrane separation can also upgrade some currently used wastewater treatment processes (e.g., biofilm, Anammox, aerobic granular sludge, anaerobic granular sludge, and fungi). However, each MBR process has its own benefits and limitations, which should be weighed before being used for wastewater treatment. The MBR + RO, OMBR, and MDBR are potential alternatives to water reuse though their investment cost and operating energy are comparatively high. However, most of these novel MBRs are currently limited to lab-scale research stage, so there is a lot of work to do to bring academic research of these MBRs up to practical application. Anyway, the retrofit of MBR technology can strengthen the market competition of MBRs in the world and provide more options for wastewater treatment in the future.

Advanced Pollutant Elimination by MBRs

Owing to the good performance, MBRs have been extensively applied in the treatment of municipal and industrial wastewaters (Marrot *et al.*, 2004; Lesjean *et al.*, 2006; Liao *et al.*, 2006; Melin *et al.*, 2006; Yang *et al.*, 2006). In addition to this, the use of MBRs has been extended to drinking water purification, WWTP effluent reuse and recycling, emerging contaminant treatment, and heavy metal removal. In fact, the elimination of micropollutants from water bodies by MBRs has attracted much attention in recent years.

Enhanced biological nutrient removals

Activated sludge process is known as the most economical and practical technology for nitrogen removal. In activated sludge process, however, the number of nitrifying bacteria is able to increase only if their growth rate is higher than the removal rate through sludge wasting and discharge in the final effluent (Gerardi, 2002). Consequently, a longer SRT is required to increase the number of nitrifying bacteria. In view of this, the rejection of membranes in MBRs can not only prevent the nitrifying bacteria to be washed out, but also separate the HRT and SRT, which makes possible to operate the MBR with long SRT and thus allows operation at high sludge concentration.

The process relevant to anaerobic/anoxic/aerobic conditions are popularly used in MBRs for nitrogen removal (Wang *et al.*, 2005; Acharya *et al.*, 2006; Liang *et al.*, 2010). The membrane module is usually submerged in the aeration tank to obtain higher membrane flux. In addition, sludge recirculation is needed to enhance the nitrification and denitrification processes. Because of the fact that microbes responsible for nitrification and denitrification grow slowly, a longer SRT therefore would be preferable. For example, a study performed by Tan *et al.* (2008) suggested that compared with the SRTs of 5, 8.3, and 16.7 days, a better TN removal rate was achieved at the SRT of 33.3 days, which was due to the combined effect of high sludge concentration and low DO concentration in the recycled sludge. However, an extremely prolonged SRT would be harmful to the process of enhanced biological nutrient removal (EBNR). The results of Han *et al.* (2005) showed that the MBR obtained a higher TN removal rate at SRTs of 50 and 70 days (94% and 96%, respectively) than that at an SRT of 100 days (89%). This is attributed to the lower oxygen transfer and deficient substrate at the prolonged SRT. In general, the occurrence of denitrification requires considerable carbon sources (Choi *et al.*, 2008; Fu *et al.*, 2009), serving as electron donor. However, high denitrification rates could be obtained in postdenitrification systems even without dosing of any external carbon sources (Vocks *et al.*, 2005). Possibly, the enhanced biological phosphorus removal sludge could offer a part of carbon source for the postdenitrification (Vocks *et al.*, 2005). The results obtained in a previous investigation confirmed that the MBRs are reliable and attractive processes for nitrogen elimination, which mainly benefits from the role of membrane rejection or the uncoupling of HRT and SRT. Of particular importance is that the MBRs could endure high nitrogen loading rate because of the high sludge concentration.

In addition, the high sludge concentration in MBRs provides better condition for the occurrence of SND, which allows nitrogen removal in a single reactor without separation of the nitrification and denitrification in time or in space but requires adapted control strategies such as a lower DO concentration (Kim *et al.*, 2007; Weissenbacher *et al.*, 2007). Compared with conventional nitrification and denitrification, SND has a number of advantages, that is, shortened pathway, reduced requirements of aeration, carbon source, and alkalinity, and lower biomass yield (Chung and Bae, 2002). As low aeration rate or intermittent aeration is applied in the SND process, the low diffusion of oxygen may create an anoxic zone within the biological flocs where denitrification can take place. Therefore, it can be concluded that the SND process can be realized by two processes: sequencing batch MBR, which incorporates the advantage of sequencing batch reactor into MBR, and the MBR aerated continuously with low DO. Owing to the unique operating modes, these two MBR processes can achieve SND with high performance in a single bioreactor (Holakoo *et al.*, 2005; Zhang *et al.*, 2006a). In addition, inserting baffles into a normal submerged MBR can generate one high DO zone and one low DO zone in a single bioreactor, which are also expected to achieve high rate of SND (Kimura *et al.*, 2008; Meng *et al.*, 2008). We can notice that most of recent investigations attempted to create aerobic zone and anaerobic or anoxic zone either at macroscale level (i.e., in a single bioreactor) or at microscale level (i.e., within one sludge floc). These efforts also attempted to acclimatize the bacteria responsible for nitrification and denitrification to the ambient conditions.

Much attention has also been focused on enhanced phosphorous removal, which is able to remove phosphorous down to low levels at a relatively low cost. The enhanced phosphorous removal is completed by microbes such as polyphosphate-accumulating organisms (PAOs) and denitrification PAOs (DPAOs). The former only uses oxygen as the terminal electron acceptor, whereas the latter has the capacity to use either oxygen or nitrate (Zeng *et al.*, 2003). The DPAOs allow simultaneous phosphorous uptake and nitrate removal, saving the COD and aeration energy demand. Similar to the nitrogen removal, phosphorous removal also needs both aerobic and anaerobic conditions. Proper design of MBR configurations or optimization of operating modes can help to guarantee high removal efficiency of phosphorous, which probably achieve enhanced phosphorous removal (Adam *et al.*, 2002; Ahn *et al.*, 2003; Lesjean *et al.*, 2003). In addition, the research findings regarding the role of nitrate in phosphorous removal varied. Ahn *et al.* (2003) observed that the continuous introduction of nitrate into the anoxic zone by the internal recirculation inhibited the growth of PAOs. The results of Patel *et al.* (2005), on the contrary, suggested that phosphorous release in an anaerobic tank and phosphorous uptake in an anoxic tank were significantly enhanced by the presence of nitrate. The phosphorous removal in this study was possibly achieved by DPAOs. Anyway, the reported results revealed that the MBRs are an effective and promising technology for phosphorous removal as well.

In addition, the applications of MBRs are also extended to drinking water purification such as nitrate removal (Wasik *et al.*, 2001; Li *et al.*, 2003; Ergas and Rheinheimer, 2004; McAdam and Judd, 2007). Unlike wastewater treatment, drinking water purification by MBRs needs a much shorter HRT because of the low organic loading rate (OLR), for example, an HRT of 0.5 h has been previously reported (Tian *et al.*, 2008a, 2009). With respect to drinking water in particular, the quality of treated water is of high concern. Noticeably, the MBRs can effectively remove virus contained in water or wastewater bodies as a result of membrane sieving. The fouling layer on membranes can, to some extent, act as the second barrier for the virus (Wu *et al.*, 2010). For example, it was reported that the membrane with pore size of 0.22 μm, cake layer, and gel layer contributed to 1.7, 6.3, and 3.1 log, respectively, of virus removal (Lv *et al.*, 2006). It implies that employing membranes with smaller pore size such as UF and NF instead of microfiltration membranes would reject more viruses. Usually, addition of coagulants or adsorbents such as polyaluminium chloride and powered activated carbon (PAC) can enhance the microbial activity in the bioreactor and contribute to organic matter removal as well as to the mitigation of membrane fouling (Sagbo *et al.*, 2008; Tian *et al.*, 2010). In addition to DOC removal, PAC addition also helps to eliminate THM (Williams and Pirbazari, 2007).

With respect to the MBRs used for EBNR, one topic of concern is to study the role of membrane rejection on the metabolism of microorganisms that are responsible for phosphorous and nitrogen removals. Another issue of interest is the influence of EBNR process on sludge characteristics (e.g., EPS/SMP) and its consequence on membrane fouling. For example, it was reported that the fouling propensity of DPAO decreased by 45% after denitrification, but it increased for ordinary heterotrophic organisms (Kim and Nakhla, 2010). In fact, the occurrence of nitrification, denitrification, and phosphorous removal possibly impacts the generation and utilization of EPS/SMP. Additionally, the study and application of MBRs for drinking water purification are still focused on lab-scale experiments so far, and there are few full-scale applications. Although microbial contamination of water can be avoided, the retention of ions and low-molecular compounds by membranes is generally insufficient to satisfy the stringent drinking water criteria (Crespo *et al.*, 2004); therefore, either process modification or posttreatment is desired.

Removal of emerging contaminants

Hazardous compounds such as persistent organic pollutants (POPs) present in water bodies are of high interest. Typically, the presence of so-called "emerging" and "unregulated contaminants" such as endocrine disrupting compounds (EDCs), pharmaceuticals, and personal care products (PPCPs) has become a significant environmental problem (Snyder *et al.*, 2003). This kind of contamination may require more intensive treatments (Petrovic *et al.*, 2003; Xue *et al.*, 2010). In recent years, therefore, there is an increasing concern about the presence and environmental risk of EDCs, POPs, and PPCPs.

Because of the increasing strict discharge regulation, extensive treatment of emerging contaminants is imperative. However, these substances are refractory to biodegradation and are even toxic to microorganisms. In addition, adsorption of these compounds by sludge flocs and colloids can take place because of their hydrophobic/lipophilic nature. For example, sorption is the primary mechanism for the removal of 17α-ethinylestradiol (Clouzot *et al.*, 2010), so these compounds are expected to be easily discharged into environment along with the suspended bacteria clusters and colloids in the effluent of CAS processes. At this point, MBRs are likely to be a favorable option for the treatment of some emerging contaminants that exhibit high adsorption potential to sludge (Radjenovic *et al.*, 2009). However, the removal of some compounds such as bisphenol A is predominated by the mechanism of biodegradation instead of biosorption (Chen *et al.*, 2008). Based on a comprehensive study on 11 antibiotics, it was found that cefalexin, sulfamethoxazole, and sulfadiazine were mainly eliminated by biodegradation; however, trimethoprim, roxithromaycin, tetracycline, ofloxacin, ciprofloxacin, norfloxacin, and ampicillin were predominately removed by biosorption (Li *et al.*, 2010). Anyway, it can be noticed that the combination of biodegradation/biosorption and membrane rejection in one bioreactor can enhance the elimination of emerging contaminants, especially when compared with CAS processes. On the other hand, the retention of activated sludge in combination with long SRT can extend the contact time of microorganisms and pollutants. The results of Fatone *et al.* (2011) also showed that the actual removal degree of aromatic hydrocarbons had a logarithmic relationship with SRT. It also allows the development of specialized microorganisms capable of eliminating low-biodegradable pollutants (Bernhard *et al.*, 2006; Sipma *et al.*, 2010), resulting in an improved removal degree of emerging contaminants, for example, a degradation degree of 0.6% and 7.2% of 17α-ethinylestradiol by CAS process and MBR, respectively (Clouzot *et al.*, 2010). Because of the unique advantages of MBRs, they were increasingly used for the elimination of emerging contaminants present in water bodies (Wintgens *et al.*, 2002; Holbrook, 2003; Hu *et al.*, 2007; Chang *et al.*, 2008; Weiss and Reemtsma, 2008; Xu *et al.*, 2008a; Abegglen *et al.*, 2009). Satisfying performance of the MBRs was also observed (Table 2). A too short HRT of MBRs, however, may lead to lower removal degrees of EDCs than CAS plants (D'Ascenzo *et al.*, 2003; Hu *et al.*, 2007). Therefore, the operating conditions of MBRs should be optimized to obtain a satisfying elimination rate of emerging contaminants.

In summary, biosorption and biodegradation in combination with membrane separation were the major mechanisms underlying the removal of emerging contaminants by MBRs, which finally lead to a better performance of MBRs over CAS. The separate and synthetic effects of bio-elimination and membrane separation on the elimination of emerging contaminants yet remain unclear. More to the point, it is of high interest to know to what extent the MBRs are better than CAS processes for the removal of emerging contaminants. In fact, both processes can eliminate readily biodegradable compounds well (e.g., paroxetine, ibuprofen, and acetaminophen) (Bernhard *et al.*, 2006; Sipma *et al.*, 2010). In addition, the elimination of emerging contaminants may differ from one to

Table 2. Comparison of Conventional Activated Sludge Processes and Membrane Bioreactors for the Removal of Emerging Contaminants

	Target compounds	*SRT*	*HRT*	*Influent (mg/L)*	*Effluent (mg/L)*	*RE (%)*	*References*
MBR	BPA	350	8	0.1–20	ND	>93	Chen *et al.* (2008)
CAS		40	11	0.1–20	0.07–0.27	73–99	Chen *et al.* (2008)
MBR	BTri	26–102	7–14	12	4.6	61	Weiss *et al.* (2006)
	5-TTri			1.3	0.5	61	
	4-TTri			2.1	1.7	14	
CAS	BTri	15	18	12	7.7	37	Weiss *et al.* (2006)
	5-TTri			1.3	1.2	11	
	4-TTri			2.1	2.2	−6	
MBR	PCP	—	12	12	ND	>99	Visvanathan *et al.* (2005)
MBR	BPA	—	24	0.4–0.8	0–0.05	90	Nghiem *et al.* (2009)
	SMX			0.7–0.8	0.3–0.4	50	
MBR	BPA	5	60	5.2	1.3	74	Kim *et al.* (2009)
	2,4-DCP			4.1	0.91	78	

BPA, bisphenol A; BTri, benzotriazole; 5-TTri, 5-tolytriazole; 4-TTri, 4-tolytriazole; 2,4-DCP, 2,4-dicholorophenol; PCP, pentachlorophenol; SMX, sulfamethoxazole; RE, removal efficiency; ND, not detected; SRT, sludge retention time; HRT, hydraulic retention time; CAS, conventional activated sludge.

another depending on their affinity/toxicity to microorganisms, for example, acetaminophen and ketoprofen had much higher removal efficiencies than roxithromycin and sulfamethoxazole (Tambosi *et al.*, 2010). The competition effect between emerging contaminants and nutrients (i.e., ammonia, nitrate, phosphorous, and COD) during biodegradation should also be considered (De Gusseme *et al.*, 2009). The elimination of emerging contaminants cannot count on biosorption alone, as the activated sludge has finite capability to adsorb the emerging contaminants and the disposal of sludge is troublesome. Most importantly, the polar compounds such as pharmaceuticals have limited biosorption rate because of their hydrophilic nature (Sipma *et al.*, 2010). The ultimate goal for the elimination of emerging contaminants is to mineralize them or to transform them into harmless compounds. To this end, the development and cultivation of specialized microorganisms responsible for the elimination of emerging contaminants is of great significance. An example is the use of genetically engineered microorganisms for enhanced biodegradation of emerging contaminants (Liu and Huang, 2008; Liu *et al.*, 2008; Qu *et al.*, 2009). Additionally, the effective control of emerging contaminants in treated water calls for the development or application of posttreatment strategies (e.g., powder activated carbon and ozonation treatment) (Weiss *et al.*, 2006). Further, the emerging contaminants can impact the growth and metabolism of bacteria in the bioreactor, which can thus change the bacterial community structure.

Bio-elimination of heavy metals

Heavy metals are toxic substances that were released into environment because of human activities. Biosorption is an attractive method for the treatment of heavy metals, which is based on the ability of certain types of microorganisms that can accumulate heavy metals (Hawari and Mulligan, 2006b; Mack *et al.*, 2007). A major challenge to the biosorption is the selection of the most specific microorganisms in a large pool of available biomass. Up to now, most of the currently used biosorbents are suspended in the form of bioflocs. Thus, the second major problem in relation to the suspended flocs is the separation of biosorbents from treated effluent. To overcome these drawbacks, cell immobilization techniques (Hawari and Mulligan, 2006a) and fixed-bed column (Hawari and Mulligan, 2006b) have been developed and used for heavy metal removal. The cell immobilization techniques include cell-to-cell immobilization (i.e., anaerobic granular sludge and aerobic granular sludge) and cell-to-carrier surface attachment (i.e., biofilm). These processes have been proved to be effective in heavy metal removal (Costley and Wallis, 2000; Liu *et al.*, 2002; Hawari and Mulligan, 2006a; Nancharaiah *et al.*, 2006). As shown in Table 3, these approaches have their own limitations, for example, discharging of detached biofilms and unstability of the aerobic granular sludge.

In MBRs, unlike other processes mentioned earlier, not only the suspended flocs and treated effluent can be easily separated by membranes, but also the most predominant types of microorganisms can be enriched. As such, the MBRs have been interestingly used for the elimination of heavy metals (Innocenti *et al.*, 2002; Fatone *et al.*, 2007; Malamis *et al.*, 2009). For instance, a study performed by Innocenti *et al.* (2002) showed that the MBR had a good performance on the removal of Ag, Cd, Sn, Cu, and Hg. The results of Fatone *et al.* (2007) showed that the MBR was more effective than CAS process for the elimination of Cr, Cu, and Ni, but Cd and Hg were almost completely removed in both processes. These findings suggested that membrane rejection plays an important role in the removal of some heavy metals. It is of interest to understand how the heavy metals contact with sludge and the specific role of membrane rejection in the removal of heavy metals. Some heavy metals (e.g., Cd and Hg) may have higher affinity with biomass flocs, which can be well separated from the secondary liquid effluent through a secondary clarifier; some, however, are prone to bind soluble organic matter, which need a membrane separation to improve the permeate quality (Fatone *et al.*, 2007). As a result, EPS in either bound or soluble form should play an important role in the

Table 3. Comparison of Different Processes for the Biosorption of Heavy Metals

Processes	*Bioadsorbents mechanism*	*Advantages*	*Disadvantages*
Activated sludge process	Biosorption by suspended flocs	Low operation cost	(1) Discharge of heavy metal-containing biomass (2) Low biosorption capability
Fixed-bed column	Biosorption by fixed biofilm	(1) Low operation cost (2) Higher biosorption capability	Discharge of the detached biofilms
Anaerobic granular sludge	Biosorption by granular sludge	(1) Very low operation cost (2) Avoid wash-out of biomass	Production of odor by the anaerobic process is a problem
Aerobic granular sludge	Biosorption by granular sludge	(1) High adsorption capability (2) Avoid wash-out of biomass (3) Good sludge settle ability	Aerobic granular sludge instability
Conventional MBR	Biosorption by suspended flocs and followed by membrane rejection	(1) Avoid wash-out of biomass and colloids (2) Select the most promising bacteria	(1) Limited understanding of membrane fouling (2) Higher aeration cost
EMBR and IEMBR	Specific extraction by the membrane and followed by biofilm adsorption	(1) High selectivity of the dense membranes (2) Biomass retention (3) Low biomass yield rate	(1) Comparatively high membrane cost (2) High operating cost

EMBR, extractive membrane bioreactor; IEMBR, ion-exchange membrane bioreactor.

biosorption of heavy metal, but the ESP affinity also strongly depends on heavy metal itself. For example, Comte *et al.* (2006) found that the affinity of some metals with EPS was in the order of $Cu^{2+} > Pb^{2+} > Ni^{2+} >> Cd^{2+}$. Additionally, soluble EPS and bound EPS exhibit different behavior on the biosorption of heavy metals. With respect to Cu, Pb, and Ni, soluble EPS shows stronger biosorption capability than bound EPS. Nakhla *et al.* (2008) found that the contribution of soluble EPS to copper complexation in a MBR was 35%–43%. Clearly, based on the reported literature, the elimination of heavy metals is greatly attributed to the biosorption property of EPS, and the membrane rejection can further improve the quality of treated water by retaining most of the heavy metals adsorbed by colloids or sludge flocs.

In addition to conventional MBRs, IEMBR and EMBR have also been applied for the removal of heavy metals from water bodies. In an IEMBR, the fluxes of ionic mercury and arsenate through the ion-exchange membranes were reported as 2.5×10^{-8} and 2.7×10^{-8} mmol/cm^2/s, respectively (Crespo *et al.*, 2004). More previously, Chuichulcherm *et al.* (2001) found that over 90% of Zn ion in metal-containing wastewaters (250 mg-Zn/L) was removed in an EMBR using sulfate-reducing bacteria. These efforts further confirmed that the combination of membrane separation and bio-elimination is attractive for the removal of heavy metals, although the operating principles of these processes are different from those of conventional MBRs.

In MBRs, the high sludge concentration, selected bacteria, and colloid rejection by the membranes can enhance the removal of pollutants. As an emerging technology used for heavy metal removal, however, there is a need to know the fundamental mechanisms governing the elimination of heavy metals in a specific bioreactor such as MBR. The heavy metals are toxic to microorganisms, which can lead to death and decay of cells as well as release of EPS. Thus, the MBR fouling is another issue of high concern. To date, there are few reports dealing with membrane fouling in the MBRs used for the elimination of heavy metals. As expected, the combination of EPS with heavy metals could accelerate membrane fouling strongly, particularly inorganic fouling. Another key issue is that the disposal of sludge will cause additional environmental problem.

Summary

With the help of membrane separation, MBRs can achieve enhanced nitrogen and phosphorous removal (e.g., A/O MBR, A^2/O MBR, and sequencing batch MBR) or the operating conditions are optimized (e.g., low DO concentration). Recently, MBRs are also used for drinking water treatment, but a posttreatment is generally needed when the treated water is used for drinking purpose. A number of investigations also showed that MBRs can significantly enhance the elimination of emerging contaminants and heavy metals compared with CAS processes, which is generally attributed to the fact that (1) membrane separation can reject pollutants adsorbed by sludge flocs and colloids, and (2) MBRs can enrich specialized microorganisms capable of eliminating low-biodegradable pollutants.

Although considerable work has been performed by researchers, research focus on MBRs significantly differs. Figure 5 shows the annual publication of MBRs with different application purposes. As shown in Fig. 5, the research attempts not only varied in time, but also differed in topics. Taking the research of drinking water as an example, considerable work was done before 2005. However, the investigation focusing on use of MBRs for drinking water treatment was less in 2010. On the contrary, the research attempts on sludge handling seemed to be a hot topic recently. Of high interest is that the use of MBRs for treatment of emerging contaminants such as POPs, EDCs, PPCPs, and pharmaceuticals was always more popular. However, the study on heavy metals was less, which is possibly due to the fact that the heavy metal-contaminated water is much less than others.

Sludge Reduction

The wide application of activated sludge processes for wastewater treatment has aroused more and more sludge management problems, of which one significant problem is the cost of excess sludge treatment. It accounts for more than half of the total operating cost in WWTPs (Mahmood and Elliott, 2006), leading to an impetus to the reduction of sludge production. So far, several strategies have been developed for sludge reduction, which include lysis-cryptic growth, uncoupling metabolism, maintenance metabolism, and predation

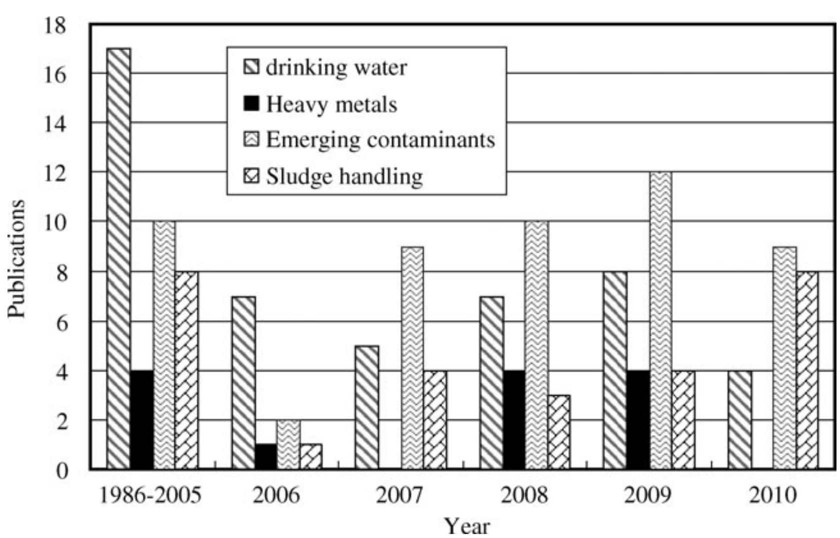

FIG. 5. Annual publication of MBRs for the treatment of drinking water, heavy metals, emerging contaminants, and sludge handling (i.e., sludge reduction, sludge thickening, and digestion) (according to ISI web of knowledge).

on bacteria (Ratsak *et al.*, 1996; Liu and Tay, 2001; Wei *et al.*, 2003). Being one of the most promising technologies, MBRs are also of high interest for sludge reduction. A kinetic model for sludge yield can be described as follows (Horan, 1989):

$$\frac{1}{Y}=\frac{1}{Y_G}+\frac{b}{Y_G}\cdot\frac{1}{\mu}$$

where b is endogenous decay coefficient (days^{-1}), μ is sludge specific growth rate (day^{-1}), and Y_G and Y are theoretical and observed sludge yields, respectively (g VSS/g COD). Sludge yield rate is in relation to microbial growth rate and endogenous decay rate (Delai Sun *et al.*, 2007).

Over a long-term of operation, Laera *et al.* (2005) found that the MBR reached a constant mixed liquor volatile suspended solid (MLVSS) concentration of 16–18 g/L at OLR below 0.1 g COD/g VSS/day and specific respiration rates of 2–3 mg O_2/g VSS/h, confirming that an equilibrium achieved between biomass growth and endogenous metabolism. Similarly, it was reported that zero sludge production could be achieved at high sludge concentrations (15–23 g/L) and with food to microorganism ratios as low as about 0.07 kg COD/kg mixed liquid of suspended solids (MLSS)/day in a pilot-scale MBR with complete sludge retention (Rosenberger *et al.*, 2000). This phenomenon was attributed to bacteria maintenance metabolism rather than the increase of protozoa and metazoa, which can prey on the bacteria. The finding of Mahmood and Elliott (2006), on the contrary, indicated that the relatively larger proportion of protozoa (ciliates and flagellates) and metazoa (rotifers and nematodes) retained by the membranes is one of the major reasons that lead to reduced sludge production. It should be pointed out that the combination of MBRs with other processes such as ozone (Hwang *et al.*, 2010), metabolic uncoupler (Lin *et al.*, 2010), Fenton oxidation (He and Wei, 2010), thermal pretreatment (Do *et al.*, 2009), and oxic-settling anaerobic process (Saby *et al.*, 2003; An and Chen, 2008a) could achieve a much better sludge reduction. For example, the results of Hwang *et al.* (2010) showed that the MBR-based ozone sludge reduction system could obtain 10%–20% higher degrees of biomass reduction over other reported ozone treatment methods. The good performance of MBRs on sludge reduction is mainly attributed to their unique characteristics over CAS, such as allowing prolonged SRTs and high sludge concentrations.

Under long SRTs and high sludge concentrations, it is expected to reach a situation in which the amount of the provided energy (i.e., feed water loading rate) equals biomass maintenance, implying that the influent carbon sources are predominantly utilized for metabolic purposes rather than cell growth (Pirt, 1965), thus minimizing sludge production. Moreover, as the viability of biomass decreases with an increase of SRTs (Olmos-Dichara *et al.*, 1997), a long SRT enables the growth of microorganisms that can prey on bacterial cells (Ghyoot *et al.*, 1999). Table 4 lists the reported results with regard to the impacts of SRT on sludge yield, endogenous decay, and sludge activity. It can be seen that as SRT increased or prolonged, the sludge yield rate and sludge activity significantly decreased, indicating that the production of activated sludge in MBRs can be reduced by prolonging SRT.

Another interesting process with regard to MBRs is the application of membrane filtration for thickening and digestion of waste activated sludge (Pierkiel and Lanting, 2005; Wang *et al.*, 2008c). To avoid the problems existing in the conventional methods used for sludge thickening and digestion (e.g., large footprint and low thickening efficiency), the membrane process for simultaneous sludge thickening and digestion (MSTD) was recently applied (Wang *et al.*, 2008b, 2008c; Wu *et al.*, 2009). Experimental results showed that about 80% MLSS destruction rate and 73% MLVSS destruction rate were achieved under an HRT of 1 day and a DO of 0.5–1.5 mg/L in the MSTD process (Wang *et al.*, 2008c). On the basis of observations of a pilot-scale MBR plant, Dagnew *et al.* (2010) found that the anaerobic MBR digester had a higher sludge destruction rate of 48% in comparison to 35.3% and 44% for two conventional processes, respectively.

In fact, operation of MBRs at a too long SRT and a high sludge concentration is unlikely practical when membrane fouling is taken into consideration. One problem associated with the long SRTs and high sludge concentrations comprises high sludge viscosity and poor oxygen transfer, resulting in increased aeration costs and severe membrane fouling, which requires frequent membrane cleaning and/or membrane replacement (Wei *et al.*, 2003; Wang *et al.*, 2008c). Therefore, the

Table 4. Impacts of Sludge Retention Time on Sludge Yield Y_G, Endogenous Decay *b*, and Sludge Activity SOUR

Wastewater	*HRT (h)*	*SRT (days)*	Y_G *(g VSS/g COD)*	*b (day^{-1})*	*SOUR (mg O_2/(g VSS/h))*	*References*
Domestic wastewater, COD = 95–400 mg/L	5	5	0.37	0.32	7	Huang *et al.* (2001)
		10	0.38	0.17	9	
		20	0.35	0.18	6–7	
		40	0.33	0.09	2–3	
		80	0.28	0.05	—	
Synthetic wastewater, COD = 300 mg/L	7.8	20	0.16	—	14.6	Lee *et al.* (2003)
		40	0.12	—	12.4	
		60	0.10	—	11.7	
Real wastewater, COD = 295 ± 116 mg/L	14	10	0.56	—	—	Innocenti *et al.* (2002)
		190	0.08	—	—	
		Prolonged	0.02	—	—	
Industrial wastewater, COD = 1000 mg/L	8	Prolonged	0.115	0.024	2.87	Delai Sun *et al.* (2007)

SOUR, specific oxygen uptake rate.

SRT should be optimized when both fouling control and sludge reduction are considered. Nevertheless, the membrane permeability could be acceptable in some MBRs used for sludge handling (Kim *et al.*, 2010). In addition, the membrane fouling could be controlled by optimization of operating parameters (Dagnew *et al.*, 2010), addition of cationic polymer (Eusebio *et al.*, 2010), use of ultrasound (Xu *et al.*, 2010a), etc. We can anticipate that the use of online ultrasound, ozone, or Fenton oxidation may play dual roles (i.e., enhanced sludge reduction or digestion and membrane fouling control) in the MBRs used for sludge handling.

Conclusions and Suggestions

In this article, recent research efforts in MBRs dealing with the development of new configurations and wider application of MBRs for pollutant elimination and sludge reduction have been reviewed. The main content of the present investigation and the critical research needs in the future can be summarized as follows:

(1) According to the market survey of several membrane companies, large-scale MBRs have been extensively employed for the treatment of municipal and industrial wastewater in the last decades. MBR market is currently experiencing a rapid growth and will keep the growth rate in the coming decades. To our knowledge, the main barriers to MBR implementation are (a) higher operating cost of MBRs, (b) higher investment cost, and (c) rapid decline of the permeation flux as a result of membrane fouling. In the future, therefore, research should still focus on development of novel membrane module or aeration modes to decrease aeration energy, development of low-cost or antifouling membranes or filters, and use of more effective fouling control strategies.

(2) Over decades of research, a number of novel MBRs including MBR+RO process, AsMBR, JLMBR, HFMBR, etc., have been proposed. The development of these novel MBR processes was mostly aimed at water/wastewater reuse, fouling control, high-strength wastewater treatment, and nutrient elimination. The diversification of MBR process has boosted the active academic research and wider application of MBRs, but potential limitations might exist for the scale-up of these MBRs. It therefore calls for more research to address the scientific and technical problems dealing with these novel MBRs and hence to improve their performance in full-scale application.

(3) In addition to nitrogen and phosphorous elimination, recent research efforts showed that MBRs are alternative options for wastewater reuse, emerging contaminant elimination, and heavy metal removal. It is of interest to know in what way the emerging contaminants and heavy metals are eliminated by MBRs, such as the biosorption or bioaccumulation of these compounds on sludge flocs and their biodegradation pathway. Analysis of microbial community structure is needed to further evaluate the impacts of emerging contaminants and heavy metals on bacterial community or to know the main microorganisms responsible for target pollutant elimination. Several attempts based on lab-scale MBRs have been performed to produce high-quality drinking water. The environmental risk of MBRs for drinking water treatment should be assessed.

(4) A noticeable advance is the use of MBRs for sludge reduction, sludge thickening, and sludge digestion. MBRs can minimize sludge production rate greatly as they allow high sludge concentration and long SRT. A major problem associated with MBRs for sludge reduction, sludge thickening, and sludge digestion is the severe membrane fouling caused by the high concentrations of sludge and organic matter. Therefore, it is desirable to study membrane fouling behavior further and propose more effective fouling control strategies.

Acknowledgment

The authors acknowledge the financial support by the Fundamental Research Funds for the Central Universities (No. 2010380003161541).

Author Disclosure Statement

No competing financial interests exist.

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