State-of-the-Art Review

Membrane Bioreactors for Nitrogen Removal from Wastewater: A Review

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Abstract: The application of membrane bioreactor (MBR) processes for conventional, municipal and industrial wastewater treatment [e.g., biological oxygen demand (BOD) reduction] is well established. The research and development of MBR processes for nitrogen removal is more recent. To date, no thorough review of MBR technology for nitrogen removal from wastewater has been carried out. The review presented here provides an overview of MBR process configurations for the removal of nitrogen based on conventional nitrogen-removal pathways (i.e., nitrification/denitrification) as well as alternative nitrogen-removal pathways, such as anaerobic ammonium oxidation (ANAMMOX). A wide range of system configurations have been explored for the application of MBR for nitrogen removal, including immersed or side-stream membrane configurations, single or multichamber processes, and the application of fixed and moving bed biofilms. Operating variables play an important role in controlling nitrogen removal and fouling, especially feed composition (particularly the carbon:nitrogen ratio), membrane characteristics, solids retention time, and hydraulic retention time. Modeling approaches for predicting nitrogen removal using MBR are evolving and are better able to represent key process differences in MBRs compared to conventional activated sludge. Although several challenges remain (e.g., membrane fouling, cost, and energy consumption), a number of opportunities exist (such as new reactor configurations, new microbial pathways, and development of a better understanding of process function through metaomic approaches) that may lead to the broader application of MBR processes for nitrogen removal from municipal wastewater in the future. **DOI:** 10.1061/(ASCE)EE.1943-7870.0001682.

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Introduction

The discharge of nitrogen to water bodies leads to a host of deleterious impacts, from contamination of groundwater to eutrophication and harmful algal blooms (HABs) in surface water (Camacho et al. 2015). As a result of the health effects of nitrogen, the USEPA has set limits for nitrogen in drinking water of 10 mg-N/L for nitrate and 1 mg-N/L for nitrite. The eutrophication of coastal waters, characterized by the excessive growth of plants and algae, depletes dissolved oxygen and smothers aquatic organisms. HABs release a host of toxic compounds, including saxitoxins, brevetoxins, and domoic acid, that can lead to serious health effects in both humans and animals (Davis et al. 2009).

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Major sources of nitrogen in aquatic systems include point sources (domestic wastewater treatment plants and industrial sources), nonpoint sources (agricultural discharges, stormwater runoff, and on-site wastewater treatment systems), and atmospheric deposition of nitrogen oxides (Vitousek et al. 1997). The relative contribution of nitrogen from point and nonpoint sources varies depending on local land-use patterns, adoption of advanced wastewater treatment, application of agricultural best management practices (BMPs), and other factors. In the Chesapeake Bay, agricultural runoff accounts for 40% of the nitrogen loading, according to the Chesapeake Bay Foundation. In the Great South Bay, Long Island, on the other hand, domestic wastewater contributes 55% of the nitrogen loading, whereas fertilizer use is responsible for only about 15% (Kinney and Valiela 2011).

In response to concerns over nitrogen and other nutrients in water bodies, limits on nutrient discharge have been enacted (Jarvie and Solomon 1998; USEPA 2009). A number of wastewater treatment plants in the United States are required to reduce nutrients through the National Pollution Discharge Elimination System (NPDES) permitting process. As part of this process, numeric limits are established for particular watersheds or receiving waters. In 2016, approximately 3% of the 16,860 permitted wastewater treatment facilities in the United States had numeric limits for nitrogen, and 10% of facilities had numeric limits for phosphate (USEPA 2016). An additional 4% of facilities had limits on both nitrogen and phosphorus. To comply with these limits, significant effort has been made to develop and implement biological nutrient removal (BNR) processes for wastewater treatment, examples of which include the Bardenpho process (and modifications thereof), sequencing anoxic/oxic batch reactors, and the modified Ludzack–Ettinger process.

Conventional BNR processes for nitrogen removal involve two main steps: (1) a nitrification step, carried out mainly by autotrophic nitrifying bacteria, in which ammonia and reduced nitrogen are converted to nitrate [Eq. (1)]; followed by (2) a denitrification step in which nitrate is reduced to nitrogen gas (N_2) by denitrifying bacteria with the addition of an external electron donor or with the residual biological oxygen demand (BOD) as the electron donor [Eq. (2)]

$$NH_4^+ + 2O_2 \rightarrow NO_3^- + H_2O + 2H^+ \quad (1)$$

$$5C_6H_{12}O_6 + 24NO_3^- + 24H^+ \rightarrow 12N_2 + 42H_2O + 30CO_2 \quad (2)$$

Although adoption of BNR processes has resulted in reductions in nitrogen loading to surface and groundwater, these processes are at least 25% more costly than conventional secondary treatment (Wilson et al. 1981), and potentially consume more energy and occupy a larger plant footprint. USEPA (2007) estimated that the cost to upgrade existing treatment plants for BNR ranges from \$588,000/mgd for facilities larger than 0.44 m^3/s (10 mgd) to close to \$7,000,000/mgd for facilities treating less than 0.44 m^3/s (1.0 mgd).

The membrane bioreactor (MBR) is a promising technology for developing new approaches to remove nitrogen from wastewater. For this application, MBRs typically consist of (1) a bioreactor zone containing microorganisms with functional genes that encode enzymes responsible for the biological transformation of nitrogen into harmless byproducts, and (2) a membrane filtration process either as a separate compartment or immersed in the bioreactor that separates the microorganisms and sludge from treated effluent. As an example, MBRs can be used to implement conventional nitrification/denitrification processes as described previously. In the nitrification zone of the bioreactor, first ammonia is converted into nitrite by a series of autotrophic microorganisms under aerobic conditions, and then nitrite is oxidized into nitrates by a separate population of microorganisms (Sliekers et al. 2002; Yun and Kim 2003). Subsequently, the bacterial community in the denitrification zone of the bioreactor converts the nitrates mostly into inert nitrogen gas under anoxic conditions (Sliekers et al. 2002). Increasingly, MBR approaches serve as a platform for the integration of novel microbial pathways and processes for the removal of nitrogen from wastewater, such as anaerobic ammonium oxidation (ANAMMOX) and simultaneous nitrification and denitrification (SND).

MBRs have a number of potential advantages over more-conventional treatment approaches. The coupling of membrane filtration with microbial processes eliminates the need for sedimentation after the bioreactor, reduces overall system footprint (MBRs have a footprint which is just 30%–50% that of more-conventional technology) (AMTA 2013), and improves effluent quality. For these reasons, MBR technology is gaining greater acceptance for secondary treatment of wastewater, which is focused on reducing BOD (Andersson et al. 2016; Judd 2008; Kraemer et al. 2012; Krzeminski et al. 2017). It is estimated that at least 34 new MBR facilities treating greater than 100,000 m^3/day of wastewater were placed into operation in 2019, with global MBR capacity exceeding 5 million m^3/day (Krzeminski et al. 2017). However, the full-scale application of MBR technology for nitrogen removal is less common than for secondary treatment. Although full-scale application of MBR for nitrogen removal is nascent, a great deal of promising research has focused on integrating membrane technology with BNR processes, with many new configurations and processes emerging. Despite promise, the MBR approach also faces a number of challenges before wider application can occur, for both secondary wastewater treatment and nitrogen removal. Some of the challenges include the high capital and operational costs, process intricacy, and membrane fouling (e.g., cake layer formation and pore clogging) (Kraemer et al. 2012).

To date, no review of MBR technology for nitrogen removal from wastewater has been carried out. The review presented here includes a summary of various MBR process configurations for removal of nitrogen using conventional mechanisms of nitrification and denitrification, followed by an in-depth discussion of several novel MBR configurations, in which nitrogen removal occurs via an alternative nitrogen-removal pathway. The sections that follow highlight the effects of operating variables on overall MBR performance. In addition, currently available modeling approaches for predicting nitrogen removal are discussed. Finally, we explore several of the future challenges and opportunities related to the broader application of MBR for nitrogen removal.

MBR System Configurations for Nitrogen Removal

Immersed versus Side-Stream MBRs

The side-stream membrane bioreactor (sMBR) and immersed-membrane bioreactor (iMBR) are the two major configurations of MBRs used in municipal wastewater treatment, based on the authors' experience. In an sMBR, the membrane module is external to the bioreactor, and the transmembrane pressure (TMP) and flow configuration create high cross-flow velocity along the membranes

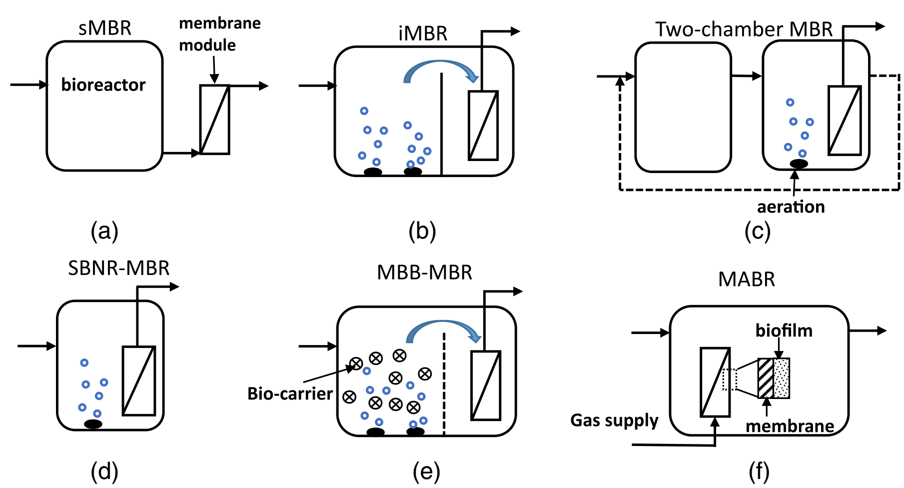

Fig. 1. System configurations of nitrogen-removal MBRs: (a) side-stream MBR; (b) immersed MBR; (c) two-chamber MBR; (d) simultaneous biological nitrogen-removal MBR; (e) moving-bed biofilm MBR; and (f) membrane-aerated biofilm reactor.

[Fig. 1(a)]. The cross-flow velocity also serves as the principal mechanism to prevent the deposition of foulants on the membrane and reduce cake layer formation (Judd and Judd 2011). sMBR has been used for drinking-water treatment and groundwater treatment (Yang et al. 2006). SMBR also has been used for industrial wastewater treatment, such as the treatment of pharmaceutical wastewater, landfill leachate, paper-mill effluent, and dairy wastewater (Andersson et al. 2016; Buer and Cumin 2010; Falahti-Marvast and Karimi-Jashni 2015). The membrane module in an iMBR, in contrast, is immersed in the bioreactor, and the treated water is withdrawn by a slight vacuum [Fig. 1(b)]. Rather than relying on cross-flow velocity, aeration typically is used to scour the surface of the membrane in the iMBR to reduce cake layer formation.

MBRs also may be categorized based on the membrane type or shape, including flat sheet (FS), hollow fiber (HF), and multitubular (MT). A FS MBR, for example, comprises single or multiple membrane sheets mounted to plates or panels. Water being treated passes through two adjacent membrane assemblies, and the treated water (permeate) flows through the channel provided by the plates/panels. A HF MBR consists of a bundle of hollow fibers (hundreds to thousands) installed in a pressure vessel. Water is applied to the outside of the fiber (outside-in flow). Finally, in a MT MBR, membranes are placed inside a support composed of porous tubes. Water flow passes from the inside to the outside of the tube (Judd and Judd 2011).

A survey of the membrane module design and operation over the last decade showed that iMBRs have been more widely accepted for domestic wastewater, whereas sMBRs have been used more for low-solid and high-strength wastewater flows (Buer and Cumin 2010; Gander et al. 2000). Although sMBRs may result in higher construction costs, the lower flux of immersed systems requires a higher membrane area (Gander et al. 2000). To date, the majority of MBR studies of nitrogen removal have applied immersed membrane modules, either in the main bioreactor (Krzeminski et al. 2017) or in a subsequent external tank to treat the effluent from the main bioreactor (Falahti-Marvast and Karimi-Jashni 2015).

Two-Chamber MBR

The sequencing anoxic/oxic MBR system is the most commonly used nitrogen removing MBR, based on the authors' experience. The system can be (1) a two-chamber configuration that consists of a clarification tank prior to the MBR with or without air supply and internal recycle [Fig. 1(c)]; or (2) a single chamber configuration, in which the clarification step is integrated into a single tank but separated from the aeration zone by baffles [Fig. 1(b)]. Baffles can create alternative aerobic/anoxic conditions in separate zones that facilitate nitrogen removal from domestic wastewater. Compared with the intermittently aerated MBR, in which the filtration operation is limited to the aeration period (Song et al. 2010) [Fig. 1(b)], a continuous MBR system with a separate anoxic zone for denitrification and a zone for aeration makes continuous filtration possible (Chae and Shin 2007; Song et al. 2010). The latter is a common approach in upgraded nitrogen-removal MBR systems in municipal wastewater treatment plants due to the simplicity of adding baffles to the existing bioreactor configuration rather than building a separate clarification tank prior to the MBR (Judd and Judd 2011; Kim et al. 2010). The process typically requires recycling of water/sludge from the aerobic zone to the anoxic zone. By changing the rate of sludge recycling, a high nitrogen-removal efficiency (>90%) can be achieved (Abegglen et al. 2008; Bracklow et al. 2010; Falahti-Marvast and Karimi-Jashni 2015; Kim et al. 2010; Perera et al. 2017; Song et al. 2010; Tan and Ng 2008).

A number of research papers demonstrate the effect of various operating factors on nitrogen removal using a two-chamber MBR configuration. Typical influent chemical oxygen demand (COD): nitrogen ratios (g-COD/g-N, i.e., C:N ratios) for effective nitrogen removal in a two-chamber MBR have been reported to be in the range 5:1–10:1 (Abegglen et al. 2008; Bracklow et al. 2010; Chen et al. 2010; Falahti-Marvast and Karimi-Jashni 2015; Kim et al. 2010; Tan and Ng 2008). A minimum C:N ratio of 3.5:1–4.5:1 is required to achieve adequate nitrogen removal. A higher nitrogen-removal rate (>90%) was observed when the influent C:N ratio increased to above 10:1 (Abegglen et al. 2008; Bracklow et al. 2010; Chen et al. 2010). External carbon addition may be applied to adjust the C:N ratio to facilitate efficient nitrogen removal using MBRs (Perera et al. 2017). Bracklow et al. (2010) found that when the COD of influent substrate to an MBR was switched from domestic wastewater to mono substrate (i.e., acetate), no significant change was observed in nitrogen removal. Wu et al. (2013) suggest that hydraulic retention time (HRT), recycle ratio, and dissolved oxygen have little effect on COD removal and nitrification (Wu et al. 2013). Song et al. (2010), however, demonstrated that when the HRT decreased below a certain level (6.5 days), insufficient nitrification causes a decrease in the overall nitrogen removal.

Simultaneous Biological Nitrogen-Removal MBR

A simultaneous biological nitrogen-removal (SBNR) MBR consists of a single bioreactor with only one chamber, in which periodic aeration occurs and the effluent is withdrawn through a membrane module. Unlike a two-chamber MBR, in which the separation of the biological zones leads to an increase in the total reactor volume, a SBNR-MBR does not possess defined anoxic zones; therefore the reactor configuration is simplified [Fig. 1(d)] (Daigger and Littleton 2014; Sarioglu et al. 2008). Hydraulic mixing and aeration provide cycling of the mixed liquor within the bioreactor, whereas diffusion resistance develops oxygen-sufficient and oxygen-deficient zones in the activated sludge flocs formed in the reactor (Judd and Judd 2011). The diffusion of substrates and the oxygen gradient between these zones enables SND to occur (Daigger and Littleton 2014). SBNR-MBR, in addition to the small footprint, offers several other advantages, including long solids retention time (SRT) to maintain the growth of the nitrification bacteria, flexibility of operating the system at various dissolved oxygen (DO) levels to facilitate SND, and simple system design and operation (Ahmed et al. 2008; Daigger and Littleton 2014; Hocaoglu et al. 2013; Hocaoglu et al. 2011a, b; Insel et al. 2014; Judd and Judd 2011; Sarioglu et al. 2009). However, nitrogen-removal efficiency by SBNR-MBR varies widely in the literature, and can range from 30% to 89% depending on operating conditions, such as the external carbon sources selected, sludge retention time, and dissolved oxygen levels (Ahmed et al. 2008; Hocaoglu et al. 2011a, b; Insel et al. 2014; Sarioglu et al. 2009).

Research over the last decade has elucidated the importance of DO, in particular, in controlling nitrogen-removal performance in SBNR-MBRs (Hocaoglu et al. 2013, 2011b; Sarioglu et al. 2009). Sarioglu et al. (2009) reported that a DO concentration of 1.8 mg/L could limit the diffusion of oxygen into the flocs and sustain SND, resulting in 40% nitrogen removal. Furthermore, their modeling predicted 85%–95% nitrogen removal when the DO concentration was maintained at a slightly lower value of about 1.5 mg/L (Sarioglu et al. 2009). Hocaoglu et al. (2011b) found that DO levels had a more significant effect on the denitrification step, in which the optimal DO concentration for nitrogen removal was 0.15–0.35 mg/L. Other modeling work predicted that when the DO level

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is kept at 0.2–0.3 mg/L, the SND process can contribute to an additional removal of 15–20 mg-N/L (Insel et al. 2014).

Very low DO levels, however, may affect process function during SBNR-MBR. When the DO level is below 0.3 mg/L, for example, it can trigger the growth of filamentous microorganisms as a side effect, which can lead to sludge bulking and can impair the sludge settling properties (Insel et al. 2014). At low DO levels, partial nitrification with denitrification also might occur within an SBNR-MBR process. Researchers have found that ammonium can be oxidized to nitrite and the resulting nitrite reduced to nitrogen gas by heterotrophic denitrification bacteria during SBNR-MBR treatment (Giraldo et al. 2011; Sarioglu et al. 2009). Incomplete nitrification in these studies was caused by low DO levels, high ammonium concentration, or inhibition from soluble microbial products (SMPs).

SRT is another critical factor controlling the nitrogen-removal performance in a SBNR-MBR. The SRT was found to have a direct effect on sludge age and granule/floc size and composition, which influences the nitrification and denitrification kinetics (Hocaoglu et al. 2011a). The SBNR-MBR process also can be integrated with a moving-bed biofilm (MBB) or a membrane-aerated biofilm reactor (MABR) (Hibiya et al. 2003; Hu et al. 2008) for nitrogen removal. The application of these two types of MBRs for nitrogen removal is described in the following sections.

Moving-Bed Biofilm Membrane Bioreactor

The moving-bed biofilm reactor (MBBR) is a technology that utilizes biofilm carriers to promote both suspended and attached growth biomass in mixed motion within a wastewater treatment basin. Importantly for nitrogen removal, these biofilm carriers provide more surface area which creates favorable conditions for the selective development of slow-growing microorganisms important for nitrification and denitrification (Lee et al. 2006). The high population density of bacteria not only promotes high biodegradation rates within the system, it also provides for higher treatment reliability and ease of operation (Dupla et al. 2006; Tang et al. 2016; Zhu et al. 2015). Commercialized plastic carriers (e.g., polyethylene) have been applied in municipal and decentralized wastewater treatment processes (Deguchi and Kashiwaya 1994; Rusten et al. 1997). Novel types of carriers, such as biodegradable polymer (PCL) carriers (Chu and Wang 2011; Pellegrin et al. 2011) that also serve as the source of carbon for denitrification, have been applied as well. Carriers that also serve as a source of carbon for denitrification simplify process controls and minimize the risk of under- or overdosing.

The moving-bed biofilm membrane bioreactor (MBB-MBR) is simply an MBBR that incorporates a membrane module (Ivanovic and Leiknes 2012) [Fig. 1(e)]. The MBB-MBR process retains a high biomass concentration within the reactor, leading to better nitrogen-removal efficiency at relatively small HRTs and less membrane fouling (Luo et al. 2015; Zhu et al. 2015). Other advantages of MBB-MBR for nitrogen removal include no requirement for recycling the activated sludge stream (Artiga et al. 2005), flexibility for increased loading (Rusten et al. 1997), less time for establishment of enriched microbial populations (Ivanovic and Leiknes 2012), and protection of the microbial community from disruption in cases of high substrate loading (Zekker et al. 2012). The coexistence of oxic and anoxic zones within the biofilm also can promote SND in the MBB-MBR (Yang et al. 2009b).

The most commonly reported configuration for nitrogen removal using MBB-MBR is a two-chamber system (Duan et al. 2015; Leyva-Diaz et al. 2015; Yang et al. 2009b; 2010), which includes the moving bed biotreatment unit and an immersed MBR unit, with a baffle wall between them. MBB-MBR has been used for the removal of nitrogen from high-strength wastewater (Zekker et al. 2012). Enhanced nitrogen removal (61%–82%) has been reported for MBB-MBRs, and mainly was attributed to the effect of SND (Leyva-Diaz et al. 2015, 2016; Luo et al. 2015; Yang et al. 2009b). Higher abundance of nitrifying bacteria in the microbial community was found in the carrier biofilms (Leyva-Diaz et al. 2015). Adhesion characteristics, such as roughness of the carrier surface, and protein and polysaccharide concentration, were found to be important in biofilm stability on the carrier surface (Tang et al. 2016). Microbial analysis showed that the microbial composition in the biofilm was significantly different from the suspended growth population (Leyva-Diaz et al. 2015; Tang et al. 2016; Zekker et al. 2012). Leyva-Diaz et al. (2015), for example, found higher abundance of ammonia-oxidizing bacteria (AOB), nitrite-oxidizing bacteria (NOB), and denitrifying bacteria in carrier biofilms compared with the suspended growth biomass (Leyva-Diaz et al. 2015). However, another study found no significant difference in the abundance of the functional species in the biofilm and suspended growth biomass (Reboleiro-Rivas et al. 2015).

Studies of membrane fouling in MBB-MBRs have shown contradictory results. Slower membrane fouling in MBB-MBRs was observed in studies in which low levels of SMPs were released compared with conventional MBRs (Lee et al. 2006; Leiknes et al. 2006; Liu et al. 2010; Luo et al. 2015). However, other studies found more severe membrane fouling in MBB-MBRs due to the formation of a thick cake layer on the membrane surface and the presence of more filamentous bacteria in the suspended solids (Lee et al. 2001; Yang et al. 2009a, b).

Membrane-Aerated Biofilm Reactor

The membrane-aerated biofilm reactor is another type of biofilm-based bioreactor, which contains both a biofilm and membrane in a single bioreactor [Fig. 1(f)]. Instead of using the membrane as a solid–liquid separation unit for the effluent, a hydrophobic permeable membrane is used to support biofilm growth and deliver a gaseous electron donor (e.g., hydrogen or methane) or an electron acceptor (e.g., oxygen) directly to the biofilm in MABR. This configuration can greatly improve the substrate utilization efficiency (Nerenberg 2016). In a nitrogen-removing MABR, air or oxygen is supplied to the reactor through the pores of the membrane directly to the biofilm without the formation of bubbles, providing up to 100% gas transfer (Brindle and Stephenson 1996; Casey et al. 1999; Mo et al. 2005). Various membrane module configurations, such as shell, tube, hollow fiber, and plate and frame, have been applied in MABRs (Brindle and Stephenson 1996; Casey et al. 1999; Downing and Nerenberg 2008). In addition, chemical modification of the membrane surface can significantly increase the gas flux, improve surface roughness, and increase the TN removal rate (Hou et al. 2013). The unique feature of MABRs is that the membrane does not serve as the filtration unit as in conventional MBRs and does not function to retain the biomass in the reactor (Judd and Judd 2011).

One advantage of using a MABR for nitrogen removal is the significant increase in nitrification rate due to interfacial oxygen mass transfer to the microorganisms (Brindle and Stephenson 1996; Brindle et al. 1998). Nitrifying bacteria, which grow much slower than BOD degraders, are more easily maintained in the biofilm. Because oxygen and nutrients are provided from two opposite sides of the biofilm, higher nitrification and denitrification activity can be achieved compared with conventional biofilm reactors due to the unique biofilm stratification and the high oxygen supply efficiency (Sun et al. 2010). Previous studies showed that during MABR startup, COD loading rates and intramembrane air pressures had little effect on ammonium oxidation rates, whereas the development of anoxic zones in the biofilm contributed to the observed increase in the denitrification rate (Satoh et al. 2004; Syron and Casey 2008). AOB mainly were distributed inside the biofilm layer, whereas denitrifying bacteria were distributed at the outside layer of the biofilm and in the suspended sludge (Downing and Nerenberg 2008; Gong et al. 2008; Hibiya et al. 2003; Hu et al. 2008; Satoh et al. 2004; Syron and Casey 2008). It therefore is important to control the biofilm layer thickness to maintain high oxygen transfer rates at the inner layer for effective nitrification, and low oxygen transfer rates at the outer layer allowing heterotrophic denitrification to occur (Casey et al. 1999). The biofilm layer thickness may be controlled through sloughing or through operating parameters such as the fluid velocity (Casey et al. 1999, 2000). Another advantage of MABRs is that efficient nitrogen removal can be achieved at low COD: total nitrogen (TN) ratios. A few studies reported 80%–100% TN removal in domestic wastewater treatment using MABRs at low COD:TN ratios (1–4) through sequential nitrification/denitrification by optimized oxygen supply (Downing and Nerenberg 2008; Hibiya et al. 2003; Hu et al. 2008; Terada et al. 2003). Hydraulic retention time also plays an important role in nitrogen removal within an MABR. Hu et al. (2008) found that nitrogen-removal efficiency decreased significantly when HRT was reduced. This result was due to the high organic loading rate and excessive growth of biomass on the membrane.

Small-Scale Systems

To date, only a limited number of studies have investigated the application of MBRs for small-scale, decentralized wastewater treatments. In contrast to larger systems, the design of small-scale, decentralized MBR systems generally must take into account greater fluctuations in wastewater flow and composition and environmental perturbations (Abegglen et al. 2008; Chong et al. 2013). Furthermore, reduction in operation complexity, process reliability, and energy consumption are critical challenges in the design for decentralized wastewater treatment (Tai et al. 2014; Verrecht et al. 2011). Membrane technologies have been used in source separation (Pronk et al. 2006; Udert and Wachter 2012) and in grey- and black-water treatment for single houses (Abdel-Kader 2013; Fountoulakis et al. 2016; Jabornig and Favero 2013; Li et al. 2009; Matulova et al. 2010; Pikorova et al. 2009). MBR has been utilized to achieve nitrogen removal from septic tank effluent (Abegglen et al. 2008; Ren et al. 2010; Verrecht et al. 2011; Wu and Englehardt 2016), and it has been applied to small decentralized communities (Atasoy et al. 2007; Chong et al. 2013; Tai et al. 2014; Verrecht et al. 2011). The high quality of MBR-treated effluent (e.g., elimination of pathogens) makes it more feasible to consider options for wastewater reuse (Chong et al. 2013; Jabornig and Favero 2013; Wu and Englehardt 2016). MBR studies focused on on-site wastewater nitrogen removal are summarized in Table 1.

The most common configuration of small-scale MBR systems for nitrogen removal is the immersed membrane bioreactor with intermittent aeration (Atasoy et al. 2007; Fountoulakis et al. 2016). Two-chamber MBRs, which provide for anoxic/oxic zones for nitrogen removal, also have been explored for applications in single homes or small clusters of homes (Abegglen et al. 2008; Chong et al. 2013; Ren et al. 2010; Verrecht et al. 2010; Wu and Englehardt 2016). Currently, however, there are only a few commercially available MBR systems; these include the BioBarrier-N (Bio-microbics, Lenexa, Kansas), the BusseGT (Busse, Binghamton, New York), and the SeptiMem in Membrane ClearBox (Huber Technology, Huntersville, North Carolina). All three of these

Table 1. Summary of MBR studies of nitrogen removal from on-site wastewater

Type of wastewater	Type of MBR	Membrane module	Membrane material	Scale	Membrane pore size	Influent TN (mg N/L)	Nitrogen removal efficiency	HRT	SRT	Reference
Real wastewater	Two-chamber	Flat sheet	N/A	1,500 L of each chamber	0.04 μm	100	90%	3 days	30–50 days	Abegglen et al. (2008)
Real wastewater	Three-tank in series	Flat sheet	PVDF	580 L/chamber	0.2 μm	214	33.5%	7.2 days	N/A	Pikorova et al. (2009) and Matulova et al. (2010)
Real wastewater	Two-chamber	Hollow fiber	PVDF	10.1 m^3 (anoxic chamber) 12.8 m^3 (oxic chamber)	N/A	81	N/A	1 day	35–50 days	Verrecht et al. (2010)
Real wastewater	Two-chamber	Flat sheet	N/A	N/A	0.2 μm	105	N/A	1–2 h	200 days	Chong et al. (2013)
Real wastewater	Two-chamber	N/A	N/A	4.15 m^3	0.04–0.06 μm	60	90%	2–3 days	12–24 months	Wu and Englehardt (2016) and Perera et al. (2017)
Real wastewater	Two-chamber	Filter bag	Nonwoven fabric	15 L	100 μm	43	33.6%–37.8%	8–12 h	N/A	Ren et al. (2010)
Grey and black water	Single-chamber	Microfiltration plate and frame	PEC	600 L	0.4 μm	Grey water (9) Black water (188)	92% for grey water 89% for black water	18 h (grey water) 36 h (black water)	50 days	Atasoy et al. (2007) and Abdel-Kader (2013)
Synthetic grey water	MBB-MBR	Hollow fiber	HDPE	200 L	0.4 μm	6.5	79%	24 h	N/A	Jabornig and Favero (2013)
Grey water	Single chamber	Flat sheet	N/A	1,000 L	0.04 μm	33	19%–45% with seasonal changes	N/A	N/A	Fountoulakis et al. (2016)

systems are immersed membrane bioreactors. No data are available regarding the number of MBR systems installed, but it likely represents a small fraction of the current residential on-site wastewater treatment market, based on our personal experience.

Similar to large-scale wastewater treatment systems, some small-scale treatment facilities applied internal recycle of sludge (30%–300%) and liquid to improve nitrogen removal (Abegglen et al. 2008; Chong et al. 2013; Perera et al. 2017; Verrecht et al. 2010). MBB-MBRs also have been used to enhance biomass concentration in a system designed for grey-water treatment (Jabornig and Favero 2013).

Both flat-sheet membrane modules and hollow-fiber membranes have been used for filtration (Abegglen et al. 2008; Chong et al. 2013; Fountoulakis et al. 2016; Jabornig and Favero 2013; Matulova et al. 2010) in small-scale MBRs for nitrogen removal. Various types of membrane materials have been used as well, including polyelectrolyte complex (PEC) (Atasoy et al. 2007), polyvinylidene fluoride (PVDF) (Matulova et al. 2010; Verrecht et al. 2010), microporous ceramic(Tewari et al. 2012), and nonwoven fabric bag (Ren et al. 2010). However, membrane fouling has not been well documented or characterized in small-scale MBR systems. Nitrogen-removal efficiency reported in small-scale MBRs varies widely from 19%–90% (Abdel-Kader 2013; Abegglen et al. 2008; Atasoy et al. 2007; Jabornig and Favero 2013; Matulova et al. 2010; Perera et al. 2017; Ren et al. 2010; Verrecht et al. 2010; Wu and Englehardt 2016).

The effect of operational factors, such as recycle ratio, loading, pH shocks, low temperature, and aeration pattern on nitrogen removal have been evaluated for small-scale systems (Matulova et al. 2010; Ren et al. 2010). Matulova et al. (2010) found that nitrification was inhibited due to the combined effects of high influent ammonium concentration (150 mg/L) and low temperature (below 11°C) (Matulova et al. 2010). Decreasing SRT also led to a decrease in TN removal due to incomplete nitrification (Verrecht et al. 2010).

Novel MBRs for Nitrogen Removal

Microalgae Membrane Bioreactor

Algae can grow in wastewater (Johnson and Admassu 2013; Markou and Georgakakis 2011) and the nutrients present in the wastewater can be assimilated to produce microalgae cells (Gao et al. 2016; Gao et al. 2014; Praveen et al. 2016). The resulting biomass potentially can be used to produce biodiesel, high-value chemicals, and/or agricultural products (Hoffmann 1998; Mata et al. 2010; Shaker et al. 2015), although these efforts still are under development and have not been commercialized. One of the challenges in using photobioreactors to treat wastewater is the dilute microalgae biomass maintained in the reactor, which may limit the treatment efficiency (Bilad et al. 2014; Gao et al. 2016, 2014). The advantage of using a microalgae membrane bioreactor (MMBR) is that it decouples the hydraulic retention time and biomass retention time (i.e., sludge retention time), which enables higher microalgae concentrations and makes downstream algae harvesting and treatment more efficient (Gao et al. 2014; Han et al. 2017; Marbelia et al. 2014; Mata et al. 2010; Tang and Hu 2016).

A typical MMBR configuration contains four major compartments: the main photobioreactor, a membrane module, a light-provision system, and a gas supplementation system [Fig. 2(a)]. In most MMBR studies, the membrane module was immersed into the wastewater to separate the effluent and the biomass (Gao et al. 2016, 2014; Wang et al. 2013a), although some studies separated the photobioreactor and the MBR as independent treatment units (Choi 2015; Wang et al. 2013a). Hollow-fiber and flat-sheet membranes commonly are used in different membrane modules for MMBRs (Choi 2015; Gao et al. 2014; Kumar et al. 2010; Marbelia et al. 2014; Tang and Hu 2016; Xu et al. 2015). Circulating water within the MMBR photobioreactor system was used to increase the mixing and improve the algal productivity and settleability (Choi 2015).

The effect of membrane materials for harvesting algae (i.e., batch versus continuous) were reviewed by Bilad et al. (2014). Membrane modules may be separate from the photobioreactor or directly integrated within the photobioreactor. A wide range of membrane materials have been utilized for harvesting microalgae separately from the photobioreactor, including cellulose acetate (CA), polyacrylonitrile (PAN), poly(ether sulfones) (PES), polyethylene terephthalate (PET), polypropylene (PP), polytetrafluoroethylene (PTFE), PVC, polyvinylidene fluoride (PVDF), and Al_2O_3 (Bilad et al. 2014). Integrated systems have utilized microfiltration (MF), ultrafiltration (UF), and dialysis membranes. One challenge with integrated systems is shearing of microalgae by bubbling systems used to introduce oxygen and control membrane fouling.

The use of MMBR has been well investigated in lab-scale experiments but has been examined only recently in pilot-scale experiments for removal of nitrogen from secondary sewage effluent (Gao et al. 2014, 2016, 2018; Han et al. 2017; Praveen et al. 2016; Tang and Hu 2016; Xu et al. 2015). Studies of using MMBR to treat primary effluent (i.e., without primary clarification) and synthetic domestic wastewater also have been reported (Choi 2015; Marbelia et al. 2014). In addition, MMBR studies using both pure algae cultures (e.g., *Chlorella vulgaris*) (Gao et al. 2014, 2016; Marbelia et al. 2014; Praveen et al. 2016; Xu et al. 2015) and mixed algae cultures (Babaei et al. 2016) have been examined with respect to nitrogen-removal performance.

Other studies explored a mixed bacteria-microalgae inoculum and demonstrated enhanced nitrogen removal. However, the complicated intraspecies relationship among algae and bacteria made it difficult to run the system at a steady state (Babaei et al. 2016; Choi 2015; Han et al. 2017; Sukacova et al. 2015). There also are reports showing more-effective nitrogen removal using MMBR than using

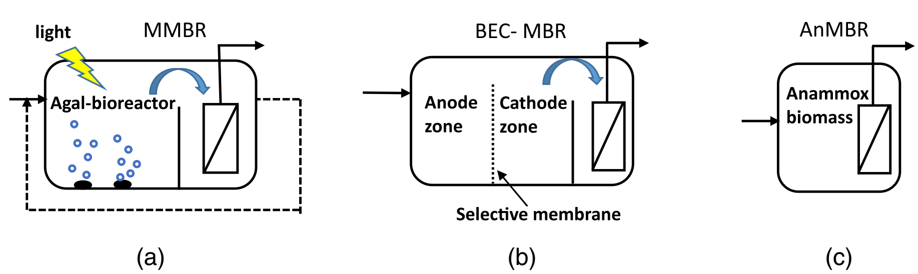

Fig. 2. System configurations of novel MBRs for nitrogen removal: (a) microalgae MBR; (b) bioelectrochemical MBR; and (c) anaerobic MBR for ANAMMOX growth.

a photobioreactor or MBR alone (Choi 2015; Gao et al. 2014, 2016; Tang and Hu 2016). Choi et al. (2015) combined an optical panel photobioreactor (OPPBR) and a MBR to treat wastewater with an average TN concentration of 40 mg-N/L. Over 96% TN removal was achieved compared with other reported studies that used the OPPBR or MBR process separately (Choi 2015). Other studies found that the nitrogen uptake rate was strongly controlled by the dilution rate (Gao et al. 2014; Marbelia et al. 2014; Xu et al. 2015). A higher concentration of microalgae could be obtained at short HRT in the reactor (Xu et al. 2015).

Bioelectrochemical Membrane Bioreactor

Microbial electrochemical technology has been applied in wastewater treatment. This technology is promising for its potential to produce electricity through the microbial metabolism of wastewater (Logan et al. 2015). The bioelectrochemical (BEC) membrane bioreactor (BEC-MBR) is an approach that incorporates the benefits of membrane processes, electrochemical processes, and biological processes to treat various types of wastewater along with electricity production. In a BEC-MBR, the microbial fuel cell (MFC) unit is integrated with a membrane bioreactor [Fig. 2(b)]. The membrane unit can be installed externally to the MFC unit for liquid–solid separation (Hou et al. 2017; Li et al. 2017; Ma et al. 2015) or it can be incorporated into the MFC cathode chamber (Li et al. 2014a; Tian et al. 2014; Zhou et al. 2015) [Fig. 2(b)]. The anode and cathode compartments can be integrated in a single chamber (Huang et al. 2017; Ma et al. 2015; Malaeb et al. 2013; Tse et al. 2016; Wang et al. 2011, 2013b; Yu et al. 2011; Zhou et al. 2015; Zuo et al. 2015) or they can be separated by a selective membrane, such as forward osmosis (FO) membranes (Hou et al. 2017; Nakhate et al. 2017), cation exchange membranes (Li et al. 2014a), proton exchange membranes (Tian et al. 2014), or a stainless-steel separator (Zhang et al. 2014a).

In a conventional BEC-MBR, organic carbon degradation takes place in the anode chamber (anoxic zone), whereas the cathode chamber (aerobic zone) is aerated to facilitate oxygen reduction on the cathode surface and generate electricity. However, more energy may be consumed than is recovered from wastewater due to the aeration process (Nakhate et al. 2017). Nitrogen removal from BEC-MBRs has been studied in modified system configurations to promote autotrophic and heterotrophic denitrification in the cathode chamber. To achieve this goal, MFC units have been incorporated into aerobic MBRs (Li et al. 2014a; Tian et al. 2014, 2015; Wang et al. 2011, 2013b; Zhou et al. 2015), anaerobic MBRs (Hou et al. 2017; Tian et al. 2014), and membrane photobioreactors (Tse et al. 2016). With proper configuration and operating conditions, SND can be successfully achieved in the cathode chamber (Zhou et al. 2015). Biofilms were developed on the surface of various types of cathodes. This process enabled autotrophic denitrifying bacteria to occupy the inner biofilm and utilize the electrode as the electron donor to reduce nitrate/nitrite where nitrification bacteria were dominant in the outer layer of the biofilm and the bulk medium that could oxidize ammonium (Ma et al. 2015; Wang et al. 2011; Zhang et al. 2014b; Zhou et al. 2015). In lab-scale BEC-MBR studies, various materials have been investigated to serve both as a cathode for redox reactions and as a filtration membrane, including electrically conductive ultrafiltration or microfiltration membranes (Huang et al. 2017; Malaeb et al. 2013), nonwoven cloth separators (Wang et al. 2013b), reduced graphene oxide (GRO)/polypyrrole-modified polyester cathode membrane (Liu et al. 2013), carbon microfiltration membranes (Zuo et al. 2015), and stainless-steel mesh with biofilm on the surface (Wang et al. 2011). Various TN removal efficiencies have been reported (10.3%–100%) in BEC-MBR studies, depending on the performance of the biological nitrogen removing processes (Li et al. 2014a; Tian et al. 2014, 2015; Wang et al. 2011, 2013b; Zhou et al. 2015).

A major challenge of using BEC-MBR for nitrogen removal is to obtain high power production and minimize membrane fouling. In previous studies, the maximum power density reported was in the range 0.6–6.8 W/m^3 (Li et al. 2017; Malaeb et al. 2013; Tian et al. 2014; Wang et al. 2011; Zhou et al. 2015), which still is lower than the neutral power requirement. However, the overall energy cost of the system can be lower than conventional MBRs (Li et al. 2014a; Ma et al. 2015). In BEC-MBRs with anode and cathode compartments separated by a membrane, only water and ions can pass through the selected membrane, which results in less membrane fouling in the subsequent membrane separation unit (Nakhate et al. 2017; Tian et al. 2014). Previous studies also have demonstrated that membrane fouling can be alleviated by coupling MBRs with MFCs (Li et al. 2017; Ma et al. 2015; Tian et al. 2014; Zhou et al. 2015). Dissolved oxygen concentration in the cathode chamber is critical for nitrate reduction. Denitrification efficiency increased significantly when oxygen concentration was lower, but too low a DO concentration inhibits nitrifying bacteria and limits the nitrogen removal (Yu et al. 2011; Zhang et al. 2014b).

BEC-MBR performance also is sensitive to temperature changes. Power production in the BEC-MBR was negligible at low temperatures (<10°C–15°C) (Ma et al. 2015). In addition to the operational limitations (reactor design, electrodes material, and HRT), the characteristics of wastewater [i.e., BOD, pH, temperature, total dissolved solids (TDS), and nitrate and phosphorus concentrations] are key factors that affect the power production from a MFC unit, which in turn determines its economic viability (Logan et al. 2015; Nakhate et al. 2017). The high cost of membranes and the energy demand for aeration are two other limiting factors for large-scale implementation of BEC-MBR in wastewater treatment (Logan et al. 2015; Malaeb et al. 2013; Nakhate et al. 2017).

Anaerobic MBR

An anaerobic MBR (AnMBR) operates without oxygen supply. This technology appears to be suitable for treatment of high-strength wastewater, such as wastewater from the food industry and landfill leachate (Lin et al. 2013). Although AnMBR offers a few advantages over conventional aerobic processes, such as lower energy requirement, less biomass production, and generation of valuable biogas (e.g., methane), more serious membrane fouling has been observed in AnMBR treatment than in aerobic MBRs (Dvorak et al. 2016; Lin et al. 2013; Skouteris et al. 2012). Most AnMBR studies have been conducted in lab-scale systems (Skouteris et al. 2012), although a few case studies reported the application of AnMBR for domestic wastewater treatment at the pilot-scale (Saddoud et al. 2006; Skouteris et al. 2012). Dvorak et al. (2016) reported the implementation of AnMBR for industrial wastewater treatment.

Most AnMBR studies for municipal wastewater treatment focused on COD removal and biogas production from high COD–strength wastewater, whereas negligible total nitrogen removal was achieved (Lin et al. 2013) because the conventional nitrogen-removal process requires anoxic and aerobic zones. Complete autotrophic nitrogen removal over nitrite (CANON) could be a promising solution for nutrient removal using an AnMBR (Zhang et al. 2013). Lab-scale studies have shown MBR to be a suitable experimental setup for the operation of the CANON process (Lin et al. 2013). In this process, ammonium first is oxidized to nitrite by AOB [Eq. (3)]. Subsequently, nitrite and the remaining ammonium are converted to nitrogen gas by anaerobic ammonium-oxidizing bacteria (AnAOB) [Eq. (4)]. This anaerobic ammonium oxidation process is an innovative technological advancement in nitrogen removal from wastewater. The abundance and activity of NOB, which consumes nitrite, should be reduced in the microbial community, and the enrichment of AnAOB was found to be critical for a successful CANON process (Lin et al. 2013). The combined partial nitrification, ANAMMOX, and simultaneous nitrification and denitrification (SNAD) process also has been applied to remove COD and nitrogen simultaneously from wastewater using intermittently aerated MBRs (Abbassi et al. 2014; Wang et al. 2016). Abbassi et al. (2014) found that MBR with higher ANAMMOX biomass was more robust in terms of nitrogen removal compared with conventional MBR when aeration time decreased. Stoichiometric models for AnMBR indicated that over 76% TN was removed by ANAMMOX, whereas 19% of the TN removal was due to heterotrophic denitrifiers. Furthermore, 95% of COD (in the form of acetate) was removed by heterotrophic denitrifiers (Wang et al. 2016)

$$NH_4^+ + 3/2O_2 \rightarrow NO_2^- + H_2O + 2H^+ \tag{3}$$

$$NH_4^+ + NO_2^- \rightarrow N_2 + 2H_2O \tag{4}$$

Another application of AnMBR for nitrogen removal is to grow microbial communities dominated by anaerobic ammonium oxidizing bacteria (AnAOB). van der Star et al. (2008) reported that high purity of AnAOB (>97%) in the biomass could be enriched in an AnMBR (van der Star et al. 2008). The system start-up with MBR appeared to be more effective at shorter periods compared with a sequencing batch reactor (SBR) for the enrichment of ANAMMOX bacteria due to the higher sludge retention time achieved in the MBR (Suneethi and Joseph 2011; Tao et al. 2012). Wang et al. (2009) reported a start-up time of 16 days for ANAMMOX activity enriched from aerobic activated sludge using an immersed AnMBR (Wang et al. 2009). ANAMMOX granules were formed, and the increased granule size, from 287 to 896 μm, potentially can decrease membrane fouling, thus increasing the operation circle of the anaerobic MBR (Li et al. 2014b). However, in the lab-scale autotrophic nitrogen-removal studies, the influent usually was synthetic wastewater that contained ammonium and nitrite/nitrate but did not contain any organic carbon (Li et al. 2014b; Lin et al. 2013; Wang et al. 2016), which is not representative of a real wastewater stream. In addition, a single-stage MABR has been reported to achieve successful CANON by controlling the vertical and horizontal microenvironment (Hibiya et al. 2003). Successful partial nitrification and ANAMMOX also have been observed in a single-stage MABR by controlling the dissolved oxygen at low levels (<1 mg/L) (Gong et al. 2007, 2008). Table 2 summarizes the research on MBRs focused on nitrogen removal from wastewater.

Effect of Operating Variables on Performance

Based on the review of specific technologies above, the main system parameters and operating variables that affect nitrogen removal using MBRs are illustrated in Fig. 3. This section discusses how these parameters affect performance.

Feed Composition

The feed composition plays an important role in controlling nitrogen removal using MBRs. In MBRs that rely on the conventional nitrification and denitrification process, the carbon:nitrogen ratio is essential to facilitate effective nitrogen removal. Organic carbon is consumed by (1) aerobic heterotrophs as the electron donor to break down BOD; and (2) heterotrophic denitrifiers as the electron donor coupled with nitrate/nitrite reduction to nitrogen gas [Eq. (2)]. Based on stoichiometry, a carbon:nitrogen ratio of at least 5:1 is needed for complete nitrogen removal (Rittmann and McCarty 2001). If the carbon:nitrogen ratio is too low, an external source of carbon needs to be added to facilitate complete nitrogen removal (Chae and Shin 2007). Various types of carbon sources have been evaluated in MBR studies for nitrogen removal, such as acetate, propionate, glucose, methanol, and biodegradable polymers (Ahmed et al. 2008; Chu and Wang 2011). Nitrogen-removing technologies that utilize alternative microbial pathways, such as ANAMMOX or CANON, however, do not rely on heterotrophic denitrification and therefore are not dependent on the C:N ratio.

Importantly, the feed composition also can affect the formation of N_2O during nitrification and denitrification processes. The formation of N_2O is an important consideration during biological nitrogen removal because N_2O has 310 times more greenhouse effect than carbon dioxide. In general, N_2O can be produced by ammonia-oxidizing bacteria during nitrification or due to incomplete removal during heterotrophic denitrification (Sabba et al. 2018). Tsushima et al. (2014) examined N_2O emissions from a number of different full-scale wastewater treatment technologies and found that emissions from plants using MBR for nitrogen removal had lower N_2O emissions than conventional activated sludge plants. Sabba et al. (2018) found that the carbon:nitrogen ratio in the feed to the denitrification process is an important operating variable, with low carbon:nitrogen ratios favoring the formation of N_2O. The complexity of microbial pathways for nutrient removal (i.e., multiple microorganisms, species, genes, and enzymes), however, makes it difficult to develop quantitative assessments of gaseous emissions and general conclusions about how different operating conditions may influence emissions.

Membrane Characteristics

Membrane characteristics and fouling play an important role in the performance of MBRs. This topic has been extensively researched and was reviewed elsewhere (Drews et al. 2006; Krzeminski et al. 2017; Meng et al. 2017; Pollice et al. 2005; Wang and Wu 2009). A few studies have focused on the role of membrane characteristics on nitrogen-removal performance using MBR. Ghyoot et al. (1999) compared the efficiency of a polymeric and a ceramic MBR in terms of the removal of nitrogen from sludge reject water generated from the sludge dewatering process. They found that the polymeric PES membrane fouled rapidly, whereas the ceramic membrane retained its high flux for a prolonged period. Chung et al. (2014) studied the effect of backwashing of ceramic membranes in an MBR system designed for the removal of nitrogen from wastewater. They found that sufficient nitrogen removal was achieved by the MBR and the ceramic membranes could retain their structure and efficiency during the periodic backwashing due to their rigidity. Interestingly, Jiang et al. (2009) found that a ceramic membrane also can catalyze the decomposition of N_2O on the membrane surface, which could reduce the release of this climate change gas during MBR. Despite several advantages of ceramic membranes, such as resistance to extreme operating conditions (temperature, acidity, and alkalinity) and long life span, the high cost of ceramic membranes hinders their widespread application in wastewater treatment when economic considerations are a major constraint (Tewari et al. 2010).

Although MBRs show great promise for the efficient removal of nitrogen from wastewater, membrane fouling remains a major challenge that hinders widespread application (Meng et al. 2009). A few studies focused on membrane fouling during nitrogen removal using MBR. One study showed that although the nitrogen-removal efficiency of a mixed liquor MBR was higher than a fixed biofilm

Table 2. Summary of nitrogen removing membrane bioreactors reviewed in this study

Type of MBR[a]	Scale	Membrane module configuration	Nitrogen removal mechanism	Nitrogen removal efficiency	Critical operation factors	Advantages	Major limitations	Reference
Two-chamber MBR	Lab Pilot Full	Flat sheet Hollow fiber	Denitrification (anoxic zone) Nitrification (oxic zone)	Moderate to high (>90%)	A minimum C:N ratio needs to be maintained Sufficient HRT needs to be maintained for nitrification	High nitrogen removal efficiency Easy to upgrade from conventional activate sludge system	May need external carbon May need internal sludge recycle to improve removal efficiency	Chae and Shin, (2007), Abegglen et al. (2008), Tan and Ng (2008), Song et al. (2010), Kim et al. (2010), Bracklow et al. (2010), Wu et al. (2013), Falahti-Marvast and Karimi-Jashni (2015), and Perera et al. (2017)
SBNR-MBR	Lab Pilot Full	Hollow fiber	Nitrification occurs in the oxygen-sufficient zone of the bioflocs, Denitrification occurs in the oxygen-deficient zones of the flocs	Varies widely (30%–90%)	DO concentrations SRT has a direct effect on the granule/floc size and composition	Small foot print Long SRT Simple system design Can be integrated with MBBR or MABR	May need external carbon source addition Intermittent aeration pattern is critical for efficient nitrogen removal	Hocaoglu et al. (2011a), Hibiya et al. (2003), Hu et al. (2008), Sarioglu et al. (2009), Hocaoglu et al. (2011b), Hocaoglu et al. (2013), Giraldo et al. (2011), Daigger and Littleton (2014), and Insel et al. (2014)
MBB-MBR	Lab Pilot	Hollow fiber	Biofilm formed on the surface of the carriers are responsible for nitrification and denitrification	Moderate to high (62%–82%)	Characteristics of the bio-carrier surface Biofilm stability on the carrier surface Microbial community structure in the biofilm	Retain high biomass concentration Higher treatment reliability and ease of operation No requirement for recycling Flexibility of loading	Some studies reported more severe membrane fouling compared with conventional MBRs	Artiga et al. (2005), Lee et al. (2006), Leiknes et al. (2006), Dupla et al. (2006), Yang et al. (2009a, b), Liu et al. (2010), Ivanovic and Leiknes (2012), Luo et al. (2015), Zhu et al. (2015), and Tang et al. (2016)
MABR	Lab Pilot	Hollow fiber[b]	Nitrification occurs at the inner part of the biofilm on the membrane with efficient oxygen transfer, and denitrification occurs at the anoxic zones in the biofilm.	Moderate to high (up to 100%)	Optimize the oxygen supply Control the biofilm layer thickness Sufficient HRT needs to be maintained for efficient nitrogen removal	Improved substrate (e.g., oxygen) utilization efficiency Efficient nitrogen removal at low C:N ratios Potential SND Energy cost savings Potential to be integrated with AnMBRs	Biofilm management is needed to maintain high flux and efficient nitrogen removal Membrane fouling from excess biomass accumulation is a concern	Brindle and Stephenson (1996), Casey et al. (1999), Satoh et al. (2004), Syron and Casey (2008), Sun et al. (2010), Hou et al. (2013), Downing and Nerenberg (2008), and Nerenberg (2016)
MMBR	Lab Pilot	Flat sheet Hollow fiber	Nitrogen is assimilated by algae for biomass production	Moderate to high (up to 96%)	HRT has a strong impact on the nitrogen uptake rate Microbial community structure (pure algae, mixed algae or mixed bacteria–algae) has a strong impact on the nitrogen uptake rate	Decouples HRT and SRT, which enables higher microalgae concentrations for harvesting	Difficult to maintain the system at steady state due to the complexity of the intraspecies relationship among algae and bacteria	Mata et al. (2010), Kumar et al. (2010), Wang et al. (2013a), Gao et al. (2014), Marbelia et al. (2014), Choi (2015), Xu et al. (2015), Tang and Hu (2016), Gao et al. (2016), and Han et al. (2017), and Tang and Hu (2016)

Table 2. (*Continued.*)

Type of MBR[a]	Scale	Membrane module configuration	Nitrogen removal mechanism	Nitrogen removal efficiency	Critical operation factors	Advantages	Major limitations	Reference
BEC-MBR	Lab	Electrically conductive UF/MF membranes Nonwoven cloth separators, and so forth	Autotrophic and heterotrophic denitrification in the cathode chamber	Various (10.3%–100%)	Low DO in the cathode chamber is critical for nitrate reduction. The system performance is sensitive to temperature change Wastewater characteristics has a direct impact on power production	Potential electricity production Potential lower energy cost compared with conventional MBR Potential to lower membrane fouling	High cost of the membrane Energy demand for aeration	Malaeb et al. (2013), Wang et al. (2013b), Tian et al. (2014), Li et al. (2014a), Ma et al. (2015), Zhou et al. (2015), Huang et al. (2017), Li et al. (2017), and Nakhate et al. (2017)
AnMBR	Lab	Hollow fiber	Complete autotrophic nitrogen removal over nitrite (CANON) ANAMMOX and denitrification (SNAD)	Low to moderate (88%)	Minimize the organic carbon in the influent, which inhibits the autotrophic bacteria growth Very low DO in the reactor is critical for maintaining the ANAMMOX bacteria	Lower energy requirement Less biomass production Potential to decrease membrane fouling Ideal to grow AnAOB biomass Potential to be integrated with MABR	Lab scale studies focused on nitrogen removal from synthetic wastewater (no organic carbon) Hard to achieve steady performance when treating real wastewater	Hibiya et al. (2003), Gong et al. (2007), van der Star et al. (2008), Gong et al. (2008), Wang et al. (2009), Suneethi and Joseph (2011), Tao et al. (2012), Zhang et al. (2013), Lin et al. (2013), Li et al. (2014b), Abbassi et al. (2014), and Wang et al. (2016)

[a]Because iMBR has been more widely accepted for domestic wastewater treatment, this table focuses on various types of immersed-membrane bioreactors.

[b]In a MABR, a hydrophobic permeable membrane is used to support the biofilm growth and to deliver gaseous electron donor or electron acceptor directly to the biofilm; the membrane does not serve as the solid–liquid separator.

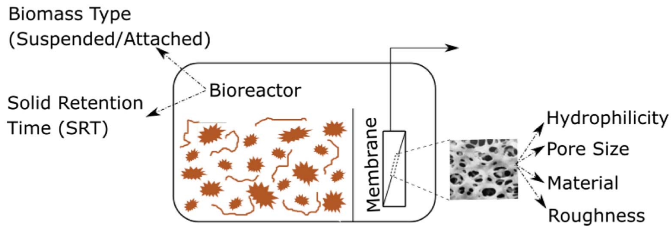

Fig. 3. Operating variables that affect the nutrients removal performance and membrane fouling in a nitrogen-removing MBR.

MBR, there was no considerable difference in the fouling behavior of the membranes operated in parallel under similar conditions (Liang et al. 2010). Furthermore, an investigation of the fouling behavior during nitrogen removal of a conventional MBR system (CMBR) and a moving-bed membrane bioreactor (MBMBR) showed that the CMBR had lower fouling (Yang et al. 2009b). This behavior was attributed to the difference in the bacterial community in the two reactors, in which a higher concentration of filamentous bacteria was present in the MBMBR system and resulted in greater fouling (Yang et al. 2009b). Nonetheless, it was shown that the biofilm in the MBMBR exhibited higher nitrogen-removal efficiency (Yang et al. 2009a).

HRT and SRT

The SRT is an important factor affecting COD removal, nitrogen removal, and membrane fouling in nitrogen-removing MBRs, because the SRT can change the properties of the mixed liquor, including viscosity, biomass concentration, composition of the microbial community, granule size, and cell surface properties (Hocaoglu et al. 2011a). In two-chamber nitrogen removing MBRs and SBNR-MBRs, SRTs typically are in the range 20–50 days to facilitate complete nitrification and denitrification and to reduce the frequency of sludge handling (Abegglen et al. 2008; Bracklow et al. 2010; Insel et al. 2014; Kim et al. 2010; Tan and Ng 2008). However, when the SRT is too long (e.g., 60 days), the granules formed in the system may break apart and cell decay may decrease nitrification/denitrification kinetics, resulting in less-effective nitrogen removal (Hocaoglu et al. 2011a). On the other hand, when the SRT is maintained at a low level (<10 days), nitrifying bacteria may be washed out, because they are slow growers, and the nitrogen-removal efficiency is decreased due to the incomplete nitrification (Verrecht et al. 2010). In MBB-MBRs, SRT has less effect on the nitrogen-removal performance and can be maintained at less than 10 days because the biomass growth mainly develops on carriers and no biomass recycling from the membrane tank to the MBBR is required (Leyva-Diaz et al. 2015; Liu et al. 2010). Mannina et al. (2017a, b, 2018, 2019) studied the formation of N_2O in a MBR for nitrogen removal and found that lower SRT favors N_2O formation.

SRT also has a significant impact on the extent and characteristics of membrane fouling. Membrane fouling in a sequencing bioreactor at different SRTs (30–100 days) showed that the critical flux decreased with increasing SRT (Van den Broeck et al. 2012). The higher membrane-fouling rate with increasing SRT was attributed to greater concentration of foulants and higher fluid viscosity. However, at very low SRTs (10 days), the COD removal and nitrification rates are drastically reduced.

Modeling MBR Systems

The modeling of nitrogen-removing MBR systems can enable the optimization of system performance, and thus is a useful tool for reducing cost (Verrecht et al. 2010). The model structure for nitrogen-removing MBRs usually is based on activated-sludge models (ASMs), which are robust and dynamic ways to simulate activated sludge–based wastewater treatment processes (Henze et al. 2000). Since the first version, ASM1, was developed in the 1980s, various improved versions of the ASM models have been developed (Fenu et al. 2010). Early attempts to model MBR systems simply transferred the ASM model to the MBR process for process design, effluent characterization, oxygen demand estimation, and sludge production prediction (Cosenza et al. 2013; Fenu et al. 2010). Although ASMs have been applied to model nitrogen removal using MBRs, a number of factors must be considered (Table 3) (Fenu et al. 2010). The significant differences in SRT, mixed liquor suspended solids (MLSS), and degree of aeration for MBRs, in comparison to conventional activated sludge (CAS) processes, must be accounted for when modeling MBR systems. Furthermore, additional effort must be undertaken to model SMP and extracellular polymeric substance (ESP) in order to model nitrogen-removal performance and to predict the extent of membrane fouling.

To model nitrogen-removing MBRs, nitrification kinetics based on autotrophic nitrifying bacteria (Jiang et al. 2005) and heterotrophic denitrification were incorporated in the model structures (Sarioglu et al. 2008). To better predict EPS/SMP production and its effect on membrane fouling (Chen and Cao 2012; Fenu et al. 2010; Mottet et al. 2013; Zuthi et al. 2013), modifications have been applied to the ASM models, including (1) the separation mechanism of the MBR (Fenu et al. 2010), (2) the standalone EPS and/or SMP models (Zuthi et al. 2013), and (3) ASM extensions incorporating EPS/SMP concepts (Drews et al. 2007; Fenu et al. 2010). The most frequently reported platforms for modeling nitrogen-removing MBRs include ASM1 (Sarioglu et al. 2009), ASM 2d (Verrecht et al. 2010), ASM2dSMP (Perera et al. 2017), BioWin model (Chong et al. 2013), and ASM3 in Simba (Abegglen et al. 2008).

The aforementioned modeling structures typically include two major parts: (1) biological models which focus on the bulk

Table 3. Considerations in applying ASMs to MBRs for nitrogen removal compared with conventional activated sludge (Fenu et al. 2010)

Parameter	Conventional activated sludge (CAS)	MBRs for nitrogen removal
SRT	Lower SRT in CAS	Higher SRT affects microbial community composition, as well as kinetics of nitrification and denitrification
MLSS	Lower MLSS	MLSS is higher for MBR
Aeration	Aeration used for biodegradation of organic carbon and nitrification	Aeration also used for fouling control, resulting in greater turbulence which reduces floc size and mass transfer processes
SMP/EPS	Not retained in the bioreactor; not considered in basic ASMs	Accumulation of SMP in the MBR by membranes affects fouling as well as biological processes; size distribution and characteristics (protein versus polysaccharide composition) of SMP/EPS may be different than those of CAS
Fouling	Not applicable	Additional models required to predict impact of SMP on membrane fouling

suspended biomass, such as carbon and alkalinity feed, the effect of various aeration patterns on aerobic/anoxic growth of heterotrophs and autotrophs, microbial decay, and hydrolysis of entrapped organics (Perera et al. 2017; Sarioglu et al. 2009); and (2) physical models which focus on the cake layer formation on the surface of the membrane module during the suction and backwashing phases (Di Bella et al. 2008; Mannina et al. 2010). The integrated models have been applied to predict nitrogen removal in various MBR configurations, such as two-chamber MBRs, simultaneous biological nitrogen-removal MBRs, and moving-bed biofilm MBRs (Abegglen et al. 2008; Cosenza et al. 2013; Perera et al. 2017; Sarioglu et al. 2009; Tan and Ng 2008). The kinetic parameters were calculated based on lab-scale and field-scale data and were applied to the modeling structure to predict and optimize the system performance of nitrogen removal (Cosenza et al. 2013). In a simultaneous nitrification/denitrification MBR system, it was reported that when MLSS was below a certain level (16,000 $\mathrm{mg\,L^{-1}}$), nitrogen removal essentially was controlled by denitrification, and when the MLSS level increased above the threshold, the rate-limiting mechanism shifted to nitrification (Sarioglu et al. 2009). In two-chamber MBRs, the model simulation results indicated that decreasing membrane aeration and SRT were most beneficial for total energy consumption, and increasing the return sludge ratio improved TN removal (Abegglen et al. 2008; Verrecht et al. 2010).

Kinetic modeling has been applied successfully to predict the overall performance of small-scale MBR systems and has shown that the sludge recycle ratio is another key operational factor affecting nitrogen-removal efficiency (Abegglen et al. 2008; Chong et al. 2013; Perera et al. 2017; Verrecht et al. 2010). One study suggested that the MBR system could be more robust to hydraulic shock loads than is biofiltration technology for small community wastewater treatment (Chong et al. 2013).

Challenges and Opportunities

Although significant progress has been made in the research and development of MBR technology for nitrogen removal from wastewater, significant challenges remain for the widespread implementation and adoption of this approach (Fig. 4, Panel A). For one, the operational control of MBR systems can be challenging, especially as new microbial pathways are implemented, pathways that may require a narrower range of operating conditions to be successful (e.g., ANAMMOX or SND). The application of SND, for example, is effective only within a narrow range of carbon:nitrogen ratio and dissolved oxygen concentration, as discussed previously. Other factors related to wastewater streams, such as variations in feed composition and flow rate, also make operational control of MBRs challenging. In addition, the use of MBR for small-scale or decentralized wastewater treatment can be particularly difficult due to large changes in wastewater generation and composition at these scales (e.g., weekend versus weekday, vacations, and so forth).

Another challenge facing greater adoption of MBR technology for nitrogen removal is cost. Although the cost of MBR technology has declined in recent years, MBRs remain more expensive than competing processes (Krzeminski et al. 2017). The installation and maintenance of membranes remains a costly aspect of MBRs. Fouling of the membranes increases the pressure requirements, which results in greater energy use and higher operating costs. The need for air scouring to prevent or reduce fouling also adds to the energy consumption. In general, the specific energy consumption of MBRs, even for larger plants, remains greater than 1 $\mathrm{kWh/m^3}$, whereas more conventional processes such as activated sludge operate at less than 1 $\mathrm{kWh/m^3}$ (Lesjean et al. 2011). The energy consumption for smaller or decentralized MBRs serving less than 2,000 people can be above 3 $\mathrm{kWh/m^3}$ (Lesjean et al. 2011). The need for chemical cleaning of membranes also contributes to the greater cost of MBRs.

Understanding and preventing membrane fouling in MBR systems remains a particularly difficult challenge. The complexity of MBR systems for nitrogen removal, in terms of microbial community structure, soluble product composition, and potential for biofilm formation, make prediction of membrane fouling particularly difficult. Although significant progress has been made to understand fouling in such systems (Meng et al. 2017; Wang and Wu 2009), additional research is needed to gain further insight and enable the translation of these insights into process improvements to minimize or eliminate fouling. Fortunately, progress continues to be made in identifying and developing new membrane materials (e.g., nanocellulose and graphene oxides) with novel functionalities (e.g., biomimetic or self-cleaning) and/or surface properties (superhydrophilic) that reduce fouling. Nanocellulose, for example, is superhydrophilic, which prevents fouling by biomolecules (Hadi et al. 2019). Incorporating nanomaterials (e.g., silver or TiO_2) with specific fouling-mitigation potential, either catalytic or antimicrobial, may be beneficial. New membrane cleaning approaches (Meng et al. 2017) also will help spur improvements in MBR technology and the wider adoption of this approach for nitrogen removal. An emerging approach for membrane cleaning is mechanically assisted air scouring using granular media, such as activated carbon and latex beads. New approaches for enzymatic cleaning using proteases and cellulases are also emerging. The use of polysaccharide-degrading bacteria may hold promise for reducing difficult-to-degrade biopolymers in the biocake layer or soluble microbial product. Novel electrically assisted fouling mitigation processes, in which techniques such as electrocoagulation and electrophoresis are integrated into the MBR system, also are being explored.

New and more robust approaches for sensing gaseous emissions and probing microbial community function (Ju et al. 2017; Narayanasamy et al. 2015; Vanwonterghem et al. 2014) are being

A. Challenges

- Membrane fouling
- Gas emissions (e.g., N_2O, CH_4, and CO_2)
- Low temperatures during winter
- Varying feed flow/composition, especially for onsite systems
- Operational control
- Energy consumption
- Cost
- Pharmaceuticals and personal care products

B. Opportunities

- New sensors to enable better process control and understanding
- Bioprocess models for design and control
- Novel membrane materials to reduce fouling
- Improved understanding through meta-omic approaches and data analytics
- Development and application of novel microbial pathways
- New reactor configurations

Fig. 4. Current challenges (Panel A) and future opportunities (Panel B) for the application of MBR for nitrogen removal from wastewater.

developed and implemented. New approaches in metagenomics provides insight into the key microbial pathways occurring during treatment and how metabolic processes vary across different wastewater treatment systems. Metaproteomics and metatranscriptomics offer the potential for elucidating the actual microbial processes occurring during nitrogen removal and how these processes are related to the microbial community structure. Yu and Zhang (2012) elucidated nitrification activity during wastewater treatment by analyzing gene expression using cDNA, despite also finding low numbers of nitrification-related genes. Coupling these omic approaches with real-time nitrogen dynamics (Huang et al. 2019) will enable a better understanding of the magnitude of such emissions and how these emissions are related to process operation.

It also is important to highlight the changing nature of wastewater discharges, and in particular, the presence of pharmaceuticals and personal care products. The occurrence of these compounds in wastewater is changing rapidly as new chemicals, products, and therapies are developed (Deng et al. 2012; Slater et al. 2011). The impact of these compounds on microbial processes during wastewater treatment is just starting to be understood (Amin et al. 2006; Deng et al. 2012; Slater et al. 2011; Zhang et al. 2016). Furthermore, our understanding related to the ability of wastewater treatment systems, and MBR systems in particular, to remove these compounds from wastewater also is evolving (Deblonde et al. 2011; Jones et al. 2005; Onesios et al. 2009; Radjenović et al. 2009; Wang and Wang 2016).

The flexibility of the MBR in terms of process design and the integration of a variety of microbial communities and other complementary pathways suggests improvements to this technology can be made to at least partially address the challenges noted previously. MBR technology not only offers a platform for the development of nitrogen-removal processes, but also may provide approaches for nutrient recovery (Huang et al. 2015; Johir et al. 2011; Sutton et al. 2011; Yan et al. 2018). Johir et al. (2011) demonstrated the recovery of nutrients using MBR coupled with ion exchange. They demonstrated recovery of 85% and 95% recovery of phosphate and nitrate, respectively. Huang et al. (2015) demonstrated the recovery of struvite using an osmotic MBR. As materials and resources become more constrained in the future, MBRs hold promise for the development of approaches which can more effectively utilize wastewater as a resource of water, nutrients and energy. A more holistic evaluation of MBR, using tools such as life cycle assessment (LCA) and technoeconomic analysis (TA), will help guide future development and application of MBR for nitrogen removal or recovery from wastewater. A number of LCA's and TA's have been conducted for MBRs, and suggested that MBRs produce higher effluent quality than CAS but require greater energy (Bertanza et al. 2017; Krzeminski et al. 2017), but none have been carried out to date specifically for nitrogen-removal applications.

Data Availability Statement

No data, models, or code were generated or used during the study.

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