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Review on MBR Technologies for Emerging Pollutant Removal from Wastewater and Their Associated Antifouling Strategies

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ABSTRACT. It was necessary to reclaim water from wastewater to tackle water scarcity issues. However, it was difficult to treat wastewater for resue purpose through conventinal treatment technologies due to the wastewater contains various emerging contaminants. Membrane bioreactors (MBRs) were promising techniques to reclaim wastewater, which hybrid activity sludge and membrane technologies. Although it was a challenge to eliminate the emerging contaminants efficiently through conventional MBRs due to specific chemical structures of these chemicals, more and more novel hybrid MBRs were applied to the removal of emerging contaminants. The evolution of MBR systems for treating emerging pollutants was summarized in this review. In addition, the process of biofouling on membranes and the development of relevant antifouling technologies were investigated. Besides, the perspectives of MBR systems on the application of emerging pollutant treatment were provided, which would help support the research and development of technologies in the field of water reclaiming in the future.

Keywords: MBR, emerging pollutants, EPSs, SMPs, antifouling

1. Introduction

According to the Sustainable Development Goals Report 2022, above 10% of the global population (beyond 0.7 billion people) lived in countries with high and critical levels of water stress in 2019 (UN, 2022). Such an increasingly serious water stress was caused by underlying effects of multiple factors (i.e., those related to anthropogenic activities and climate change) and their complicated interactions. Water reclamation was a great adaptation for this intractable water-stress issue through the development of alternative sources to enhance water security, sustainability and resilience, such as reusing treated wastewater for irrigation, particularly in rural areas. Thus, relevant standards of reusing water were established based on the categories of use by international organizations and countries (Becerra-Castro et al., 2015). However, emerging organic pollutants, including persistent organic pollutants (POPs), pharmaceutical and personal care products (PPCPs), veterinary medicines, endocrine-disrupting chemicals (EDCs), carbon-based nanomaterials, and micro-/nano-plastics, were detected in the effluents of wastewater treatment plants (WWTPs) with activated sludge process over the world (Radjenovic et al., 2009). This indicated that these pollutants, in particular PPCPs and EDCs, could pass through the conventional processes without efficient treatment, and thus causing health and environmental risks due to such emerging pollutants are capable of degradation resistance, long-range transport, and bioaccumulation, as well as endocrine disruption within central nervous, reproductive, and immune systems (Kumar et al., 2022). Figure 1 exhibits performance and risks of water reclamation from emerging pollutant-containing wastewater through conventional WWTPs.

Compared with conventional activated sludge process, membrane bioreactor (MBR) integrated membrane filtration and biodegradation processes in one system, which has stronger treatment capacity, smaller space requirement, and slighter sludge production (Lin et al., 2012). Removal efficacies of emerging pollutants were investigated, which are various with pollutant species and membrane porosities. Citalopram and metronidazole could be removed above 91% through the membrane with 0.1 μm pore size (Llorens-Blanch et al., 2015). However, the removal efficacy of fenoprop was only 69%, even MBR equipped with nano-size membrane (Ghoshdastidar and Tong, 2013). In addition, membrane fouling was an issue of long-term MBR operation to reduce membrane performance and increase maintenance cost. Significant decrease in specific flux in response to increase in trans-membrane pressure (TMP) indicated membrane fouling existed in MBR where biopolymers, colloids, sludge flocs, and particles are deposited on the membrane surface, and thus clogging the pores (Wang and Wu, 2009). Backwash and chemical cleaning measurements were necessary

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to recover the MBR system. Recently, advanced MBR systems and cutting-edge antifouling technologies were developed to deal with these drawbacks during the treatment of emerging pollutants.

Therefore, the objective of this review is to summarize advance MBR systems and associated antifouling technologies for enhancing emerging pollutant treatment. In detail, it entails (1) evolution examination of advanced MBR systems for treating emerging pollutants; (2) analysis of biofouling formation on membrane during MBR operation and further investigation of relevant antifouling technologies; (3) perspectives of MBR systems on the application of emerging pollutant treatment, which would help support research and development of technologies in the field of water reclaiming in the future.

2. Emerging Pollutants

Emerging pollutants were chemicals which have health and environmental risks. Effluents from agricultural fields, municipal wastewater treatment plants, and manufacturing of medicines, petroleum products, electrical devices, textiles and others were main sources of these pollutants (Daughton, 2001). For example, there were various emerging pollutants in domestic wastewater due to a large number of consumptions in personal care, pharmaceutical, cosmetic and inflammable products (Luo et al., 2014a). Among these products, pharmaceutical wastes including antibiotic drugs, biologics, diagnostic agents, nutraceuticals, fragrances, sunscreen agents, etc., were received the most of attention. A survey on removal efficiency of emerging pollutants through WWTPs was reported (Deblonde et al., 2011). Effluent concentration of emerging pollutants ranged from 0.007 to 56.63 g/L with corresponding removal rate from 0 to 97%. The removal rate of phthalates was beyond 90%, while that of antibiotics varied from 50 to 71%. Anti-inflammatory and beta-blockers were the most difficult to be decomposed. The removal rate of such compound was of 30 ~ 40%. It was noteworthy that removal rate of pharmaceuticals, such as tetracycline and codeine was below 10%.

Most of emerging pollutants were not regulated under any current environmental laws and posed significant risks in human health and ecosystems. Inhalation, ingestion, and dermal absorption were three major exposure routes for these pollutants. The emerging pollutants could induce a large range of acute and long-term toxic effects (e.g., immunotoxicity, neurological disorders, and cancers) on human health and ecosystems (Maeng et al., 2013). Such toxic effects would complicate since the interactions of various exposure conditions related to pollutant species, intensity, duration, and frequency.

3. Evolution of Membrane Bioreactor Systems

3.1. Conventional MBRs

Recently, a number of MBRs were focused on removing emerging pollutants, which integrate biodegradation and filtration, as well as other processes (e.g., adsorption, advanced oxidation, and electrochemical processes) (Torre et al., 2015). Generally, as shown in Figure 2, biodegradable emerging pollutants were decomposed by relevant microbes in MBRs, while refractory pollutants were adsorbed on sludge or colloids depending on their hydrophobicities (Xiao et al., 2019). On the one hand, the emerging pollutants with electron-donor groups (e.g., hydroxyl and amide groups) could facilitate higher efficacy of biodegradation than those with electron-acceptor groups (e.g., carboxyl and halogenated ones). On the other hand, the colloids attached hydrophobic pollutants could be further retained by membrane. Hereby, the pore size of membrane was another key factor to retain trace pollutants. However, the smaller pore size the membrane had, the more energy the MBR system required, and thus undesired membrane fouling phenomena were more inclined to be formed. Consequently, side stream membrane bioreactor (SSMBR) was proposed to separate filtration and biological processes into two individual reactors, which is different with one-reactor MBR, such as immersed membrane bioreactors (IMBR) or submerged membrane bioreactors (SMBR) (Kraume and Drews, 2010). Figure 3 illustrates the configurations of MBRs. In addition, MBR-based hybrid technologies were developed to help remove such pollutants for improving effluent quality.

Generally, various membranes, including microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) were equipped in MBRs. It was reported that removal rates of ranitidine and ofloxacin were above 80% through the treatment of MBR coupled with MF membrane, which are higher than those obtained through conventional activated sludge process (CAS) (Radjenovic et al., 2007). The sludge composed of microbes could be blocked in mixed liquid phase where solid retention time is extended to facilitate removal of biodegradable pollutants. It was noteworthy that the hydrophobicity of pollutants governed their removal efficiencies. For example, removal efficiencies of fenoprop and diclofenac (both of them were hydrophilic) were less than 20% in an MBR with 0.4 μm hollow fiber membrane, while those of salicylic acid and gemfibrozil were above 95% (Tadkaew et al., 2011). Both of fenoprop and diclofenac contained chlorine, a strong electron-acceptor group, and thus were significant resistant to biodegradation (Nguyen et al., 2013a). Compared with CAS, MF-flat sheet MBR and UF-hollow fiber MBR were exhibited better performance in pharmaceutically active compounds removal (Radjenović et al., 2009). The removal rate of naproxen was increased from 70% in CAS to about 90% in both MBRs, while ibuprofen and acetaminophen could be removed completely in either CAS or MF/UF-MBRs. These indicated that hydrophobic interaction between these drugs and sludge matrix was a dominant effect rather than electrostatic repulsion between them, and thus adsorbed pollutants on sludge matrix were blocked in MF/UF MBRs. As a consequence, increased sludge concentration and extended solid retention time facilitated specific microorganisms to biodegrade their favoured pollutants. However, many researchers reported that carbamazepine concentrations in the effluents of MBRs were similar to or even higher than the influent ones due to hydroxylated forms of carbamazepine were increased through microbe-initiated deconjugation process (Beier et al., 2011; Ooi

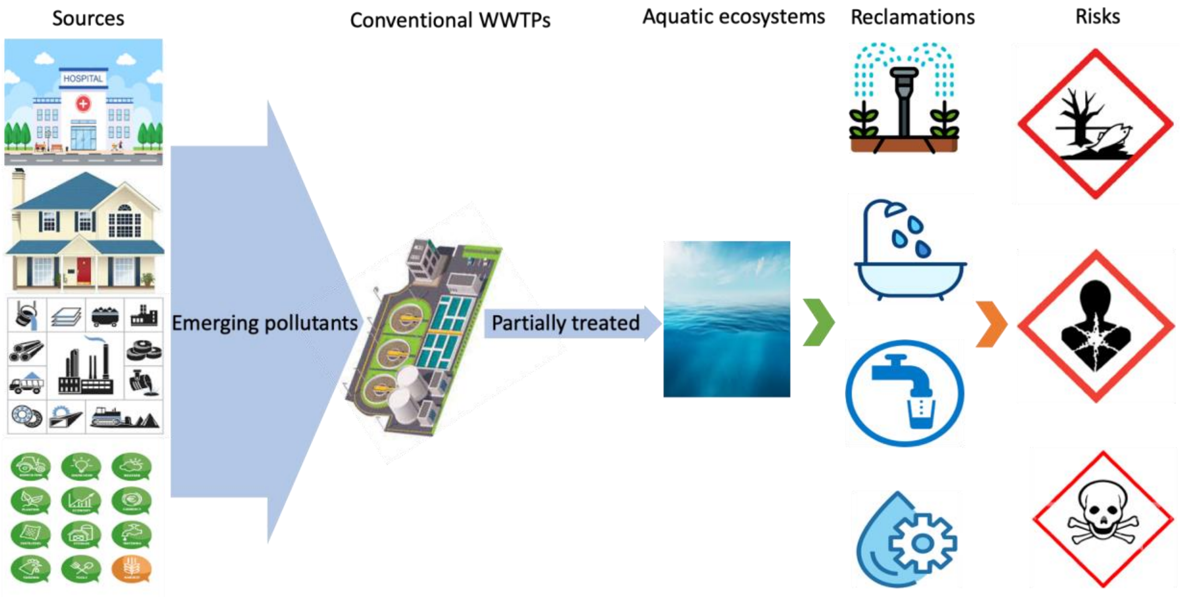

Figure 1. Performance and risk of water reclamation from emerging pollutant-containing wastewater through the treatment of conventional WWTPs.

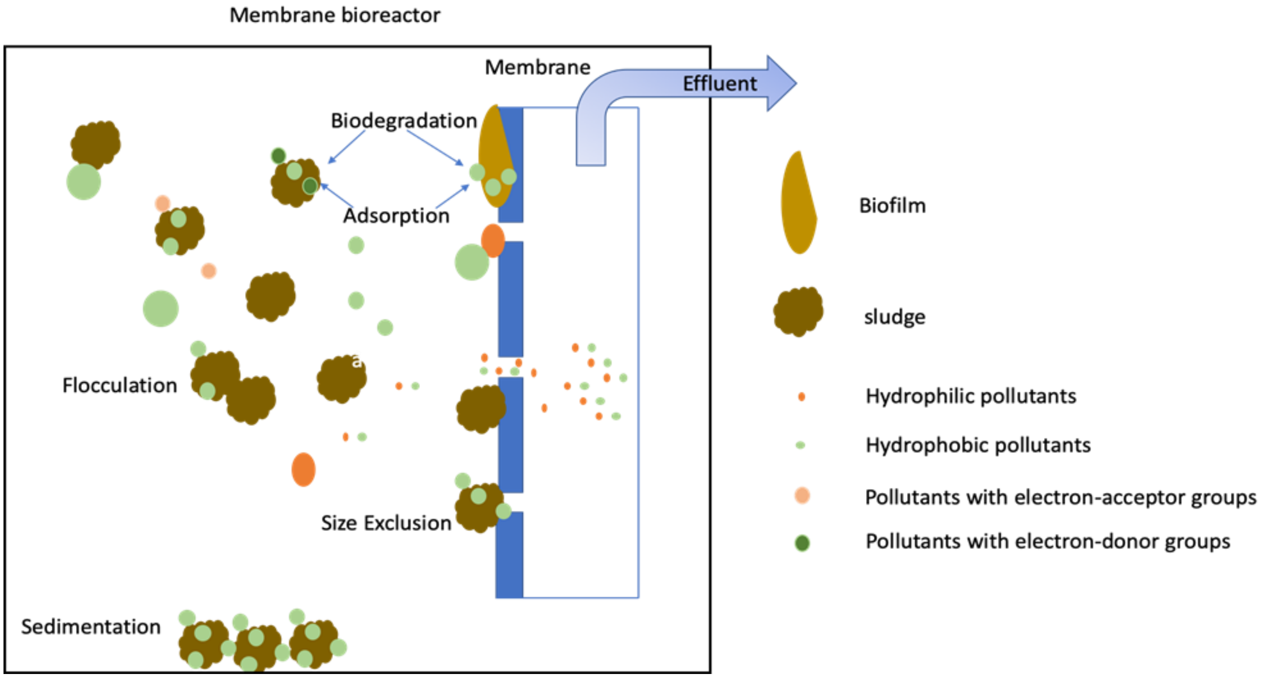

Figure 2. Fate and transport of emerging pollutants in conventional MBRs.

et al., 2018). On the other hand, amide group of carbamazepine was a strong electron-acceptor group to eliminate its biodegradation (Fan et al., 2014). In addition, loratadine, one of antihistamines, could also not be treated efficiently by MF/UF-MBRs since its desorption on sludge matrix (Arcanjo et al., 2022). These drawbacks of MF/UF-MBRs were amplified in decentralized wastewater treatment facilities, particularly in remote areas, due to the fluctuation of influent quality and quantity (Hube and Wu, 2021). NF-MBR could remove emerging pollutants better than MF/UF-MBRs due to its filtration of lower molecular weight cut-off (MWCO) membrane. It was reported that NF-MBR could eliminate above 90% of emerging pollutants from textile wastewater (Dharupaneedi et al., 2019). However, associated fouling was inevitable resulting from the accumulation of protein, humic acid, and other smaller size impurities on membrane surface.

3.2. Hybrid MBRs

Recently, hybrid MBRs were developed to enhance the removal of emerging pollutants (Figure 4). Thus, a configuration based MBR, moving bed membrane bioreactor (MBMBR) was developed, which combines moving bed biofilm (MBBR) and membrane filtration in one reactor. The organic loading of such an MBMBR was 10 ~ 15 times higher than that of other MBRs, while the corresponding HRT of MBMBR was 10 ~ 15 times less than that of others (Sombatsompop et al., 2006). For MBBRs, biocarriers provided a high surface area for the growth of microbes and these microbes formed biofilms attached on these biocarriers (Barwal and Chaudhary, 2014). Such biofilms contained various microbes with individual growth rates and conditions, resulting in aerobes and anaerobes (e.g., nitrifiers and denitrifies) could be available in one reactor (Sipma et al., 2010). Hence, the range of removal targets was expanded with the development of MBBRs. For example, the removal rates of iohexol and diatrizoate in MBBR were 79 and 73%, respectively, which have about 20% increases in those of MBR (Casas et al., 2015). Furthermore, the removal rates of naproxen, primidone, salicylic acid, and 17 β-estradiol were 81.1, 83.5, 91.1, and 96.2% in an MBBR with the biocarriers of polyurethane sponge pieces (Luo et al., 2014b). The MBMBR was developed based on MBBR which has adequate solid retention time and thus allow specific microbes to play relevant roles in biodegradation. For instance, biodegradation efficiency and kinetic rate of BPA and BHT in MBR were enhanced by 7 and 44%, respectively, due to the enhanced nitrification of enriched nitrifying microbes on biofilm (Boonyaroj et al., 2017).

Osmotic membrane bioreactor (OMBR) integrated biological treatment and forward osmosis (FO) in one reactor. In this system, the filtration of FO was driven by osmotic pressure gradient, such that a draw solution with high concentration (relative to that of the feed solution), was fed to induce a net flow of water through the membrane into the draw solution, thus effectively separating the feed water and its solutes (Meng et al., 2009). Compared to RO, such a filtration was an energy-saving process. Studies on investigation of this new system for treating emerging pollutants were conducted. It was reported that all of 12 antibiotics could be rejected above 75% by FO membrane of OMBR (Srinivasa Raghavan et al., 2018). Except to the FO rejection, there were also biodegradation and adsorption on activated sludge as significant removal pathways, depending on pollutant species. In this study, the removal of ciprofloxacin and roxithromycin was relied on the biodegradation, while that of ofloxacin and roxithromycin was attributed to the adsorption. In order to obtain high-quality treated water for reclaiming, the OMBR should be combined with MF/RO as a post-treatment unit to recover water from permeate contained draw solutes. It was reported that OMBR-MF could remove above 90% of caffeine and 89% of atenolol, while removal rate of atrazine was only below 40% in such an OMBR-MF (Pathak et al., 2018). In addition, organic draw solute could provide stronger driven force for the OMBR-MF than inorganic one, resulting in better removal rate of organic pollutants. In another study, the application of RO improved OMBR performance significantly so that 99% of emerging pollutants with hydrophilic and bio-refractory features were removed through OMBR-RO (Li et al., 2018).

Except to the combinations with advance membrane configurations, other processes, including UV oxidation, electrochemical process, and additives had combined with MBR to help enhance the performance of emerging pollutant removal (Dhangar and Kumar, 2020). UV oxidation could decompose bio-refractory and hydrophilic organics, whereas its capacity could be eliminated by a large number of suspended solids resulting in light shielding. MBR followed by UV oxidation (MBR-UV) could tackle this issue through membrane filtration; and the organics of permeate could be further decomposed through UV oxidation (Yang et al., 2022). It was reported that the removal efficiency of fenoprop, ketoprofen, salicylic acid, carbamazepine and metronidazole could be higher than 85% through MBRUV treatment (Nguyen et al., 2013b). It was noteworthy that the removal efficiency of carbamazepine was 90% in MBR-UV, while it was only removed 32% in mere MBR treatment. UV oxidation was a promising post-treatment for polishing the removal of such bio-refractory organics. Recently, electrochemical and MBR processes were hybrid (eMBR) to improve the removal of organics and the antifouling of membranes (Ensano et al., 2019). Electrocoagulation promoted small-size (i.e., sub-micron) suspended matters to form larger agglomerations which are thus separated from the permeate of MBR through precipitation. It was reported that the removal rates of carbamazepine, diclofenac and amoxicillin through eMBR treatment were increased 25.26, 25.16 and 27.58%, respectively, compared with MBR (Ensano et al., 2019). Besides, granulated activated carbon filter was applied to polish the permeate of MBR, and thus removed above 90% of pharmaceuticals (Mousel et al., 2021). Although this approach had lower energy consumption and less secondary pollutants than MBR-UV and eMBR systems, the life of carbon filter was limited, and it was needed to be replaced or regeneration for sustaining expected capacity. Furthermore, additives, such as coagulants, were investigated to enhance the performance of MBR. Polyaluminium chloride (PAC) and chitosan were added into MBR to form coagulants with PPCPs (Park et al., 2018). As a result, the PAC could increase removal efficiency to 17 ~ 23%, while the chitosan exhibited a slight effect on PPCP removal. In addition, the coagulants formed by PAC and chitosan could rise membrane permeability by 2.3 and 2.8 times, respectively, which could also ease membrane fouling caused by small-size particles.

4. Progress of Emerging Antifouling Technologies

4.1. Membrane Fouling

Although the MBRs had excellent performances in removing emerging organic pollutants from wastewater, the membrane fouling was an inevitable issue during treatment, resulting in the decline of membrane flux and the increase of energy consumption. During the treatment of emerging organic pollutants, organic/inorganic pollutants (i.e., organic/inorganic fouling), suspended solids (i.e., colloid fouling), and soluble microbial products (SMPs) and extracellular polymeric substances

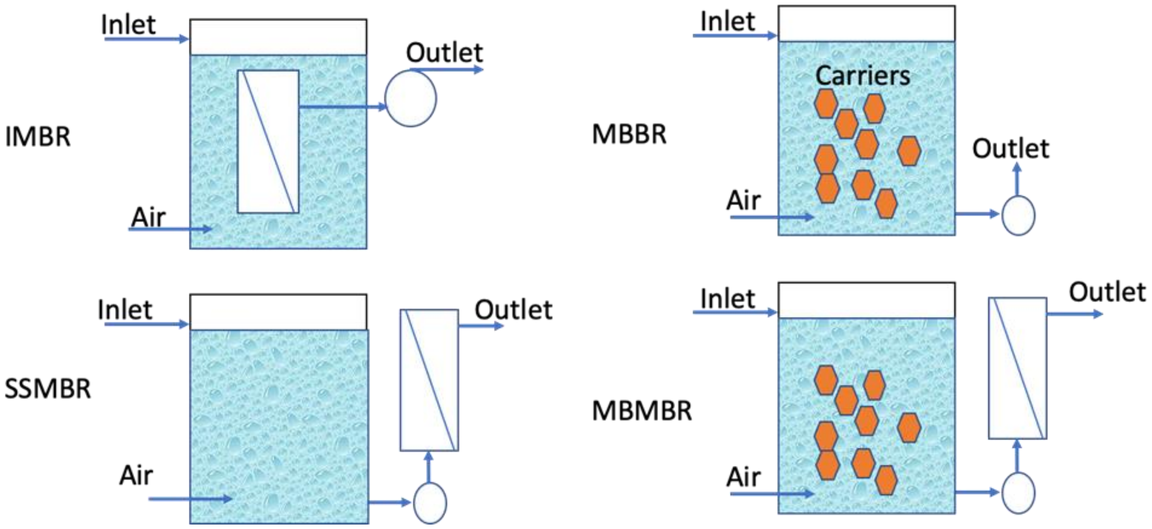

Figure 3. Schematic diagram of conventional MBRs.

(EPSs) (i.e., biofouling) could accumulate on membrane and thus caused associated pore blocks (Chu and Li, 2005). Among these fouling phenomina, biofouling was dominant one. However, conventional physical and chemical methods could not deal with the biofouling efficiently. As shown in Figure 5, the biofouling formation was initiated by conditioning film adhered on membrane followed by the transport of suspended cells to membrane, the production of SMPs and EPSs by bacteria and thus the formation of biofilm, and the expansion of biofilms on membrane (Cui et al., 2021). The biofilm contained SMPs and EPSs could provide the protection of covered cells from antibacterial effects, resulting in the reduction of permeability (Liu et al., 2012). Consequently, physical methods, such as backwashing and vibrating, could not scrub such biofilms thoroughly; while chemical methods, such as corrosive reaction, could give rise to membrane unforeseen damages (Liu et al., 2021). Therefore, emerging antifouling technologies, such as membrane modification of nanomaterials, polymers and their composites, were developed based on the characteristics of interface between membrane and foulants (Figure 6).

4.2. Microorganism Analyses for Biofouling

The accumulation of microbial cells could be visualized through high-resolution observation techniques such as scanning electron microscopy (SEM), fluorescence microscopy confocal laser scanning microscopy (CLSM), atomic force microscopy (AFM), and direct observation through the membrane (DOTM) (Meng et al., 2009). Both DOTM and CLSM were extensively used to analyze membrane biofouling. It was reported that a DOTM was employed to observe the interactions between the bioflocs and the membrane surface (Zhang et al., 2006a). The images showed that the bioflocs could move across the membrane surface by rolling and sliding. More recently, CLSM was a powerful approach for imaging membrane biofouling, which can not only identify the deposited cell, but also present the 3D structure of the fouling layer. Bacterial distribution on the membrane surface was visualized through CLSM; it was found that bacteria were widely accumulated on the fouled membrane (Ng et al., 2006). The combination of CLSM and image analysis could visualize or quantify the architecture of biocake lay characterized the biofilm structure and analyze its effect on membrane permeability in MBR for dye wastewater treatment. It was found that the capability of membrane filtration was strongly associated with the structural parameters of the biofilms. The visualization of biofouling through these technologies could help understand deposition process of the cells and the microstructure or architecture of the cake layer.

In addition, a few investigations were performed to study the microbial community structures and microbial colonization on the membranes in MBRs. The microbial community structures could be analyzed through microbiology methods such as polymerase chain reaction denaturing gradient gel electrophoresis (PCR-DGGE) and fluorescence in-situ hybridization (FISH). It was reported that the microbial communities on membrane surfaces could be very different from the ones in the suspended biomass (Zhang et al., 2006b). The results help provide a list of bacteria that might be the pioneers of surface colonization on membranes. It was observed that microbial communities in a fullscale submerged MBR for treating the effluent from primary sedimentation tank of a municipal wastewater treatment plant (Miura et al., 2007). It was also reported that the microbial communities on membrane surfaces were quite different from those in the suspended biomass. In this study, both of FISH and 16S rRNA gene sequence analyses revealed that a specific phylogenetic group of bacteria, the Betaproteobacteria, probably played a major role in development of the mature biofilms, which led to severe irremovable membrane fouling. It was reported that g-Proteobacteria more selectively adhered and grew on membranes than other microorganisms, and the deposited cells had higher surface hydrophobicity than the suspended sludge (Piao et al., 2006). The deposition of cells could be selected by high shear stress induced by aeration. Although parts

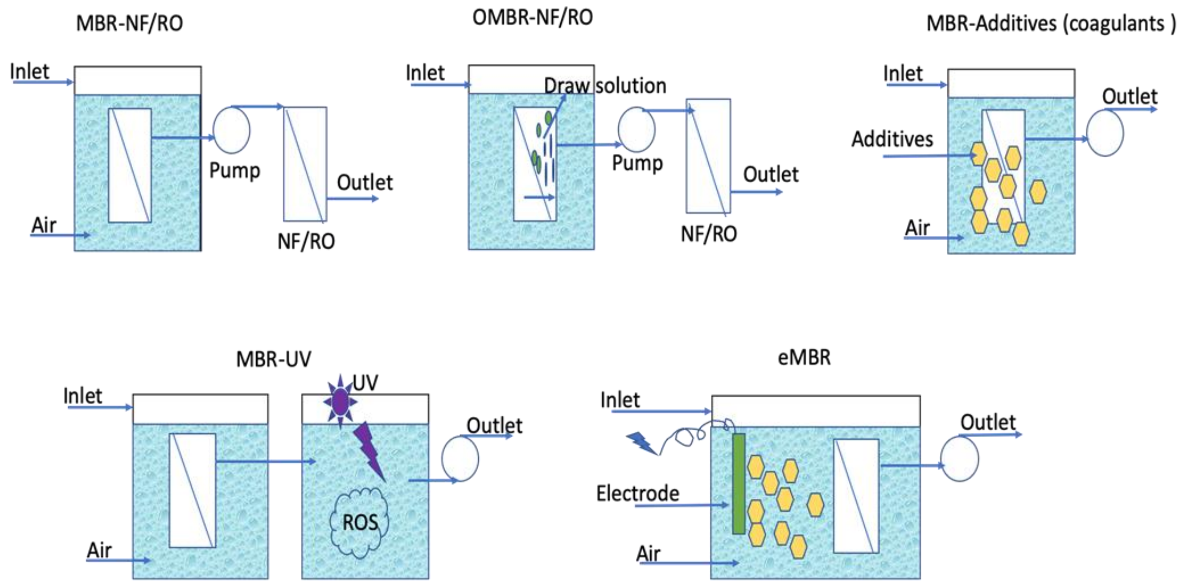

Figure 4. Schematic diagram of hybrid MBRs.

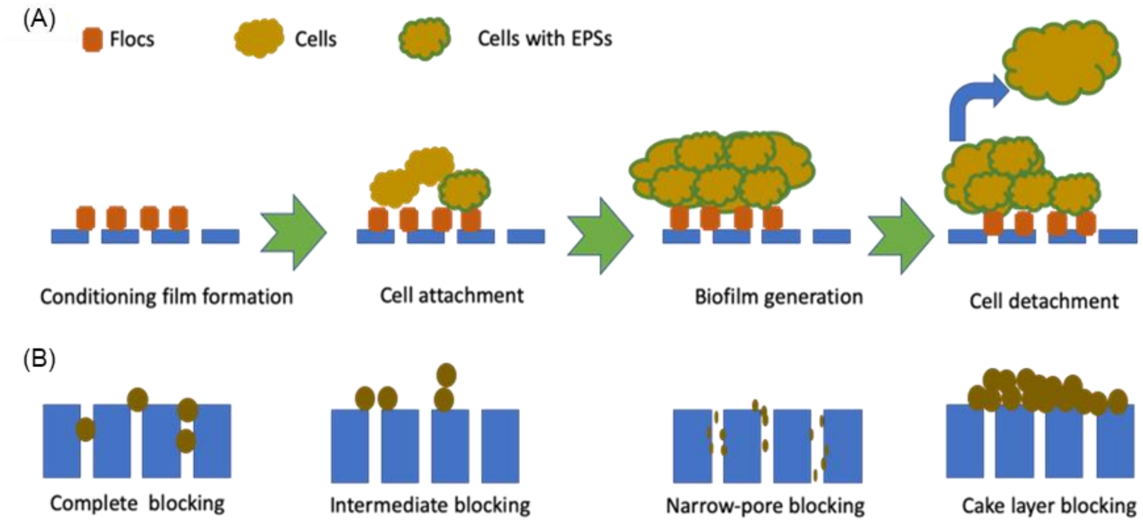

Figure 5. Biofouling formation on membrane (A) biofouling formation stages and (B) biofouling types.

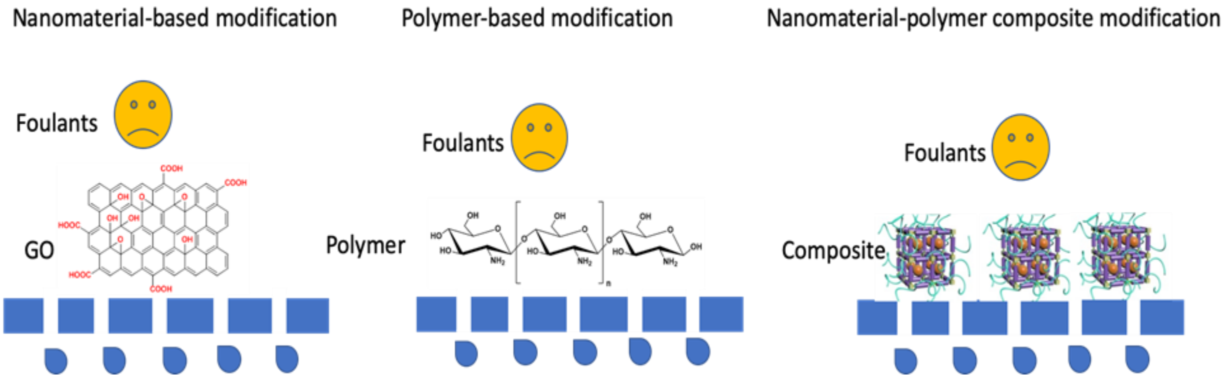

Figure 6. Surface modification strategies for membrane antifouling.

of cells would be detached easily by the shear stress, while residual ones still adhered to membrane surface tightly. The affinity of cells to membranes determined selective deposition of the cells. In addition, microbial community structure varied with the temporal variation under the anoxic condition. Hence, the bacteria in the sludge played an important role in membrane biofouling.

4.3. Nanomaterial-based Membrane Modification

The modification of membrane in terms of increasing hydrophilicity, promoting porosity, enhancing microbial inhibition, and enriching negative zeta potential. Nanomaterials with multifunction properties based on their nano-scale structures were applied to modify the surfaces of membranes to empower them (Jhaveri and Murthy, 2016). Carbon-based nanomaterials such as carbon nanotubes (CNTs) and graphene oxides (GOs) were adopted to decorate the membrane surface to intensify membrane permeability and inhibit microbial growth. It was reported that water permeability of membrane which was modified by polyethylene glycol (PEG) functionalized CNTs was 4 times higher than pristine one (Khalid et al., 2018). In addition, protein-affinity resistance of modified membrane was promoted remarkably due to the increase in the hydrophilicity of membrane, leading to prolonged duration of MBR operation. Similarly, GO-cellulose nanocrystal (GO-CNC) was coated on polyvinylidene fluoride (PVDF) membrane to activate antifouling capacity of MBR (Lv et al., 2018). The EPS accumulation on GO-CNC@PVDF membrane of MBR was scarcer, and thus the TMP increased gently during wastewater treatment. Consequently, operation duration of this modified MBR was extended three times longer than pristine MBR. According to the results of physiochemical analyses, GO-CNC@PVDF membrane had stronger hydrophilicity, higher porosity, and larger negative zeta potential, resulting in effects on the resistance of EPSs attaching. Functional groups on GO facilitated the membrane surface with such a stronger negative zeta potential that the accumulation of negatively charged fluctuates was prevented. Besides, nano-oxides were also added to modify the membrane. TiO_2/polypropylene membrane was synthesized for enhancing MBR which shows larger flux than the pristine one due to improvements in both hydrophilicity and porosity (Bae and Tak, 2005). Similar effects were found on zinc oxide (ZnO) doped membranes during MBR operation. It was observed that MBR with ZnO-polyvinyl chloride (PVC) membrane exhibited thinner biocake layer (i.e., about 5 times slimmer than that of pristine MBR system) with corresponding the extension of operation duration (i.e., about 2.4 times longer than that of pristine MBR system) (Alsalhy et al., 2018). Except to hydrophilicity, porosity and zeta potential, ZnO also had antibacterial capacity to eliminate cell growth on the membrane (Vatanpour et al., 2021). Furthermore, the performance of MBR with Fe_3O_4 nanoparticles doped membrane was investigated. The flux of such a MBR was 70% larger than pristine MBR with corresponding 70% reduction in resistance (Mehrnia et al., 2021). The foulants could be detached from membrane through its vibration involved Fe_3O_4 nanoparticles under a magnetic filed (Noormohamadi et al., 2020).

4.4. Polymer-based Membrane Modification

Polymers were adopted to modify membrane for antifouling through blending with bulk membrane or coating on the membrane surface. Copolymerization was employed to integrate monomer with raw film material to form a new material for further membrane casting. It was reported that polypropylene-sulfonamide copolymer membrane could inhibit protein adsorption due to propylene amine groups was involved in copolymerization (Hester et al., 2002). In another study, polyoxymethylene methacrylate was grafted on to PVDF, forming a new membrane with stronger hydrophilicity than pristine one (Hester et al., 2002). On the other hand, surface modification, including coating, plasma assisting, and grafting, were also investigated for membrane antifouling. Hydrophilic polymer was coated on PVDF UF, inducing flux recovery rate of this new membrane could reach above 90% (Kim et al., 2015). However, such a simple coating method was not durable due to the modified molecules could be effortlessly detached from membrane surface. Plasma modification, particular low temperature plasma, could introduce various polar groups on the membrane without causing a high-temperature damage (Kang et al., 2001). Besides, surface grafting covered an expected layer on membrane through chemical bonding between membrane surface and polymer chain. For example, dimethyl aminoethyl acrylate (DMAEMA) was grafted onto polysulfide (PSF) MF to form PSF-g-PDMAEMA membrane (Du et al., 2020a). In addition, acrylic acid was grafted onto polypropylene film through UV radiation to form a hydrophilic film (Wang et al., 2015). Moreover, sulfobetaine methacrylate, pentaerythritol monoester, and 2-hydroxyethyl methacrylate were grafted onto polypropylene MF/hollow fiber membranes to reduce water contact angle and increase flux significantly (Du et al., 2020b). It was noteworthy that the water flux was increased with the amount of polymer grafted monotonically. For instance, the water flux of polypropylene hollow fiber membrane grafted by sodium styrene was increased with the grafting amount till grafting ration was 9.0% (Wang et al., 2015).

4.5. Nanomaterial-Polymer Composite Membrane Modification

Amino functionalized SiO_2 nanoparticles and polyelectrolyte sodium alginate were assembled on PVDF membrane through plasma-assisted functionalization, forming a super-hydrophilic membrane with an excellent antifouling performance in flux recovery ratio (Zhao et al., 2015). In another study, a membrane with silver metal-organic framework (Ag-MOF) coating had stable antifouling properties due to durable Ag depletion and powerful antimicrobial features of Ag-MOFs (Yuan et al., 2022). In addition, silver-silicon dioxide (Ag-SiO_2) nanoparticles could be coated on PVDF membrane homogeneously, and thus promoted the resistance of EPSs accumulation and biofilm formation on membrane during MBR operation for treating pharmaceutical wastewater (Ahsani et al., 2020). Bismuth dimercaptopropanol (BisBAL), a low-toxic alternative of Ag, was doped on hollow fiber membrane to reduce EPSs and SMPs attachment through its antimicrobial feature (Yavuz et al., 2019). It was observed that biocake was loosen on such a membrane so that water flux was decreased slowly. Furthermore, polycitrate-alumoxane (PC-A) nanoparticles were coated on membrane to increase hydrophilicity due to hydrophilic hydroxyl groups on PC-A were involved in (Pirsaheb et al., 2019).

5. Conclusions and Perspectives

Nowadays, available water resources have been scarce with the development of industries and growth of population. It is necessary to reclaim water from wastewater. However, wastewater could not be treated to the standards of reuse purpose through conventional treatment technologies due to wastewater contain various emerging contaminants. MBRs are promising techniques to reclaim wastewater, which hybrid activity sludge and membrane technologies. Although it is a challenge to eliminate the emerging contaminants efficiently through conventional MBRs due to specific chemical structures of these chemicals, more and more novel hybrid MBRs have been applied to the removal of emerging contaminants, such as advanced membrane-assisted MBR, MBBR, MBMBR, OMBR and other combinations. However, membrane fouling issues, particularly biofouling, are still inevitable during MBR operation. EPSs and SMPs are two dominant compounds released from microorganisms, forming biofilms on membrane and thus resulting biofouling during MBR operation. Therefore, multiple biofouling methods have been developed to promote MBR performance, such as nanomaterial-based, polymer-based, and composited modification methods. The hydrophilicity and antimicrobial capability of membrane are empowered significantly through these methods.

Based on review in the development of MBR technologies and associated antifouling strategies, the perspectives are provided as followed:

(1) Appropriate Hybrid MBR technologies should be selected based on characteristics of wastewater and microorganism community. Because biodegradation of pollutants, microbial growth, and fouling controlled by molecular features of emerging pollutants and surface characteristics of membranes. DO, pH, temperature and other factors are involved in pollutant removal and biofouling. Hence, these factors should be concerned before MBR applied. Particularly, the effects of these factors and their interactions on emerging pollutant removal are complicated, which should be analyzed through factorial experimental design.

(2) Mathematic tools should be utilized to project, stimulate, and forecast biofouling. Because the procedure of biofouling could be revealed by experiments and expressed as mathematical formulas. However, there are uncertainties involved in the pollutant removal and biofouling processes of MBRs. Stochastic methods should be applied to improve the accuracy of stimulation. In addition, it is necessary to develop modelling approaches to tackle dynamic, uncertain and complicated issues in MBR processes to achieve real-time control for MBR systems.

(3) Novel membrane materials and modules should be developed. Microbial growth on the membrane is an importance factor of MBR fouling formation. How to decrease attached of microorganisms and increase permeant flux are needed to be researched. In addition, advanced modules should also be developed to improve antifouling efficiency through self-cleaning and anti-biocake, such as vibrating membrane and forward osmosis membranes. Besides, cutting-edge analysis methods should be applied to mechanism exploration of membrane fouling and associated antifouling processes, such as proteogenomics and synchrotron-based technologies.

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