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Strategies for Mitigating MBR Membrane Biofouling

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ABSTRACT. Membrane biofouling is a roadblock to the application of membrane bioreactors (MBR) for wastewater treatment and reuse. Strategies for the mitigation of membrane biofouling have been extensively reviewed in this paper. The review was focused on feedwater pretreatment, modified membranes, suppression of the secretion and discharge of extracellular polymeric substances (EPSs) and soluble microbial products (SMPs), and novel MRB systems. The three identified novel strategies for mitigating membrane biofouling in an MBR for wastewater reclamation are lower EPS concentration by adding D-amino acids (D-AAs) and a cationic polymer flucculant; introducing a modified membrane in an MBR; and applying a novel algae-bacteria system. Experimental results have shown that membrane biofouling has been mitigated to some extent via these strategies.

Keywords: biofouling mitigation, MBR, EPS, SMPs, D-AAs, coagulant, modified membrane, algae-bacteria system, wastewater, reclamation

1. Introduction

Membrane biofouling is a roadblock to the application of membrane bioreactors (MBR) for wastewater treatment and reuse. Membrane biofouling is attributed to mineral precipitation, colloids and dissolved organic deposition, and the adhesion of bacteria (Guo et al., 2012). Biofilm formation, which starts with bacterial cells approaching and accumulating on the surface of the membrane, is the most critical factor in biofouling (Wang et al., 2010). Extracellular polymeric substances (EPSs) are insoleble substances discharged by bacteria, aiming to immobilize biofilm cells and allow intense interactions between bacteria (Razatos et al., 1998; Meng et al., 2009; Al-Juboori Yusaf, 2012; Liu et al., 2012). Polysaccharides, proteins, and eDNA are significant components of EPS. Soluble microbial products (SMPs) are another critical factor in biofilm formation (Wang and Waite, 2009; Hong et al., 2014). SMPs are soluble organic compounds released from biomass growth and decay associated with bacterial adhesion and biofilm formation. The membrane biofouling process is concluded as follows: bacteria initially adhere to the membrane surface, microcolonies are established, and then develop into a continuous biofilm by embedding in a self-produced matrix of EPS and SMPs. Once established, biofilms are very difficult to remove.

Membrane biofouling causes an increase in transmembrane pressure, which will either reduce water flux or increase energy consumption (Drews, 2010). Membrane biofouling leads to frequently membrane backwashing and chemical cleaning. It accelerates the membrane aging process and frequently requires membrane replacement. MBR biofouling can significantly increase MBR operating costs, especially the cost of membrane replacement. Therefore, strategies for preventing and controlling membrane fouling have been extensively studied worldwide. This review paper focuses on strategies for mitigating MBR membrane biofouling, including feedwater pretreatment to lower EPS concentration by adding D-amino acids (D-AAs) and a cationic polymer flocculant, introducing a modified membrane in an MBR, and applying a novel algae-bacteria system.

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2. Strategies for Mitigation of MBR Membrane Biofouling

2.1. Feedwater Pretreatment: D-AAs and a Cationic Polymer Flocculant

Pretreatment of feedwater to lower EPS concentration has been studied and applied for many years (Sadowska et al., 2021; Zhang et al., 2021). Several additives have been added into feedwater to change the physicochemical properties of the bacteria in the feedwater, rather than changing the membrane properties. Those additives provide explicit advantages and disadvantages. They are high efficiency, low costs, or nontoxic. However, sometimes it may lead to secondary pollution attributed to their by-products. Therefore, cost-effect and environmentally friendly additives for the pretreatment to suppress membrane fouling are urgently needed. In this section, the pretreatment using D-amino acids (D-AAs) and a high-efficiency cationic polymer flocculant, polydimethyldiallylammonium (PDMDAAC), are discussed in detail, from their antifouling performance to suppress membrane fouling mechanisms

L-amino acids are widely known as typical amino acids.

Its isomers D-amino acids, which are excreted from limited species of bacteria (Xing et al., 2015), can disturb other bacteria and their biofilm formation by affecting the metabolic process instead of killing them (Lam et al., 2009; Kolodkin-Gal et al., 2010; Xing et al., 2015). Due to the different structures of the cell wall, different types of bacteria, specifically gram-positive and gram-negative, have different interactions with different D-AAs (Glater et al., 1983; Glater et al., 1994). Whatever the nature of the medium, diverse D-AAs can inhibit biofilm formation (Meng et al., 2009).

The D-AAs are recently considered as a convinced method for membrane biofouling control (Wang et al., 2018). It proposes a novel solution to inhibit bacteria's initial adhesion and biofilm formation. D-tyrosine, D-tryptophan, and D-leucine are used to study the influence of different structures and functions of D-AAs by adding them to bacterial wastewater. It is worth mentioning that the productions and properties of EPSs and SMPs were assessed to evaluate the membrane biofouling.

The PES and PVDF membranes are used as the targets. After a series of pre-preparations, membranes' essential characteristics have been well understood. To clarify the function of EPS and SMPs, the quantities and properties of EPS and the effect of SMPs were analyzed. Gram-positive bacteria *B. subtilis* and gram-negative bacteria *P. aeruginosa* were used in batch-scale attachment experiments and filtration systems.

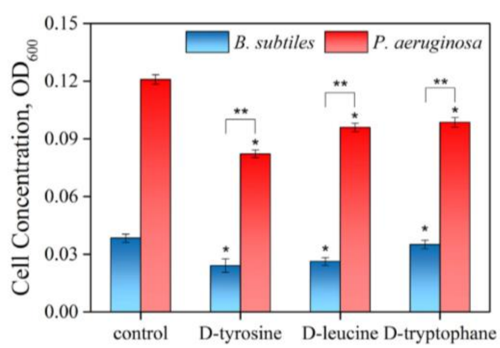

Figure 1. Effects of D-AAs on *P. aeruginosa* and *B. subtilis* attachment to the PES membrane. Reprinted with the permission from Wang et al. (2018).

Figure 1 shows that D-AAs notably decrease the initial bacterial attachment, varying from D-AAs types. Compared to the control group, D-tyrosine group has the most significant effect on cell concentration on the membrane surface. In addition, *B. subtilis* is more sensitive to D-AAs.

It is widely known that EPS production is a crucial factor in biofilm formation (Mansouri et al., 2010). Figure 2 indicates the effects of three D-AAs on two kinds of bacteria. The addition of D-AAs dramatically decreased the concentration of polysaccharides and proteins. Along with the increased concentration of D-AAs, the quantities of polysaccharides and proteins decrease linearly. *P. aeruginosa* produces more EPS production, which may result in adhesive interaction between bacteria and membrane surface due to the sticky nature of EPS (Hori and Matsumoto, 2010). This is consistent with the adhesion attachment experiment.

The quantitative analysis is conducted by 3D-EEM as well. The 3D-EEM indicates that protein-like substances have two peak waves marked as peak A, with proteins found to be one of the main components of EPS. Meanwhile, polysaccharide-like substances peak at specific wavelengths. That is, the fluorescence intensity represents the EPS concentration.

The potential of fouling by SMPs was measured in bench-scale dead-end filtration. Figure 4 shows the permeate flux changes. For 60 minutes filtration, the curve is almost identical with or without D-AAs. Therefore, D-AAs do not affect SMPs. Then a bench-scale dead-end filtration is processed to evaluate the biofouling potential.

First, the impacts of D-AAs on membrane flux are studied (Figure 5). There is no marked flux decline during the filtration of D-AAs, which means D-AAs have less potential to cause membrane fouling. The PVDF and PES ultrafiltration membranes are used in this experiment. Due to the rough and hydrophilic membrane surface, the PVDF membrane shows a higher risk of membrane fouling. The flux decreased by 93.0% (pristine PVDF membrane) and 87.2% (pristine PES membrane). When D-AAs exist, the permeate flux decline shows a slowing trend. D-AAs exhibit a more significant anti-fouling effect. *B. subtilis* (G+) has a peptidoglycan-rich cell wall, distinct from *P. aeruginosa* (G-). This is the reason that gram-positive bacteria are more sensitive to D-AAs.

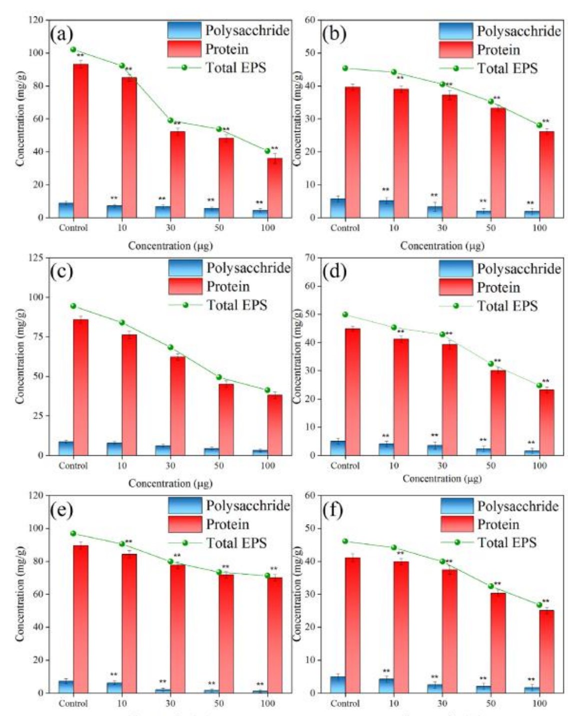

Figure 2. Effects of D-AAs on EPS production. (a) D-leucine on *P. aeruginosa*, (b) D-leucine on *B. subtilis*, (c) D-tyrosine on *P. aeruginosa*, (d) D-tyrosine on *B. subtilis*, (e) D-tryptophan on *P. aeruginosa*, and (f) D-tryptophan on *B. subtilis*. Reprinted with the permission from Wang et al. (2018).

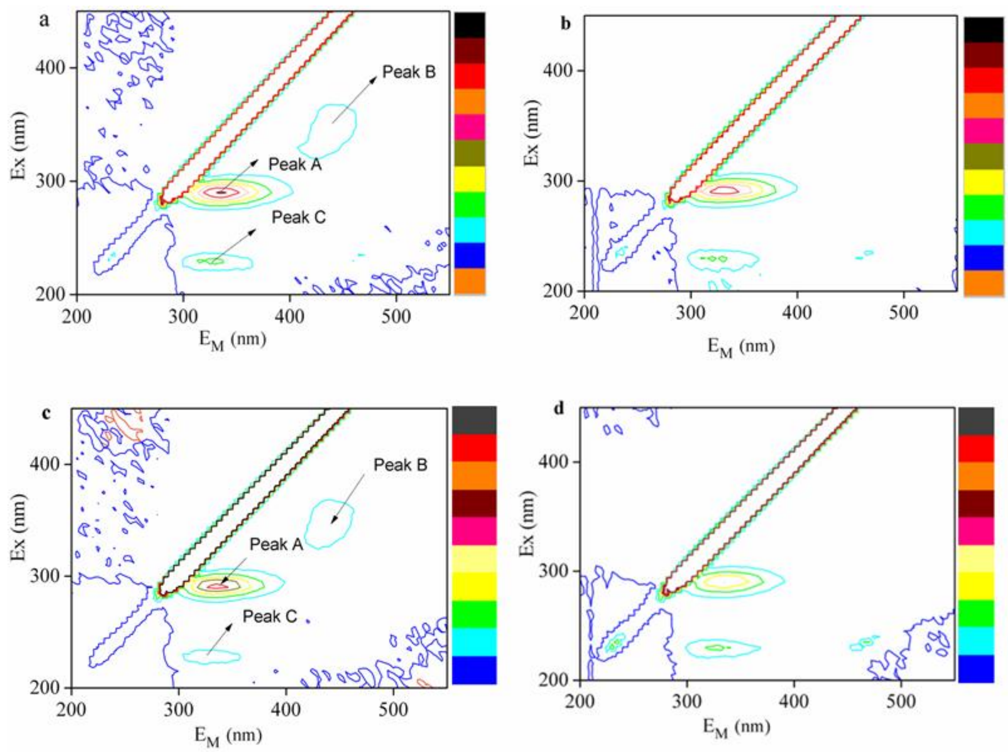

Figure 3. 3D-EEM of EPS under the condition of D-AAs and blank. Reprinted with the permission from Wang et al. (2018).

Finally, the permeate flux recoveries by physical and chemical backwashing are studied. The result shows that with the presence of D-AAs, it is near complete recovery with the chemical cleaning using NaClO. D-AAs indeed changed the membrane fouling to reversible fouling. It is worth mentioning that mixed D-AAs significantly affect any of the D-AAs in isolation.

A novel and nontoxic strategies mitigating membrane fouling by directly adding the D-AAs to the wastewater is shown by above results. Moreover, the other primary bacteria-produced contaminant SMPs are related to membrane fouling caused by microbial activity (Munz et al., 2007; Bugge et al., 2013). Flocculants such as granular activated carbon (GAC) and polymeric aluminum chloride (PAC) are added to enhance the antifouling characteristic (Zhao et al., 2011; Ding et al., 2014). However, flocculants have been proven to be harmful to nitrogen removal efficiency (Iversen et al., 2009; Guo et al., 2010). Recently, Polydimethyldiallylammonium (PDMDAAC) has been considered as a solid method in coagulation process due to its bridging mechanism and positive charge (Huang et al., 2015; Shen et al., 2017). Zhou et al. (2019) found that it can be applied as flocculants to aggregate sludge floc in MBR system, describing in the following paragraphs. The performance of MBR treatment is first studied via the removal efficiencies of the chemical oxygen demand (COD) and total nitrogen (TN). The final removal efficiencies of TN range from 86.8 to 91.8%, and the COD removal efficiency is 92%. The minimal dosage of PDMDAAC (90 $mg \cdot L^{-1}$) leads to the lowest transmembrane presure (TMP). This phenomenon means that membrane fouling has been mitigated at the slowest rate of rise.

The zeta potential is another crucial parameter for the evaluation of membrane fouling. Along with the continued addition of PDMDAAC, the zeta potential shows an increasing trend. Meanwhile, the sludge volume index (SVI) decreases when the concentration of PDMDAAC increases. It proves that the zeta potential is essential to large sludge floc formation, which is highly correlated to settling ability. The PDMDAAC is a cationic polyelectrolyte that can provide a more positive charge for sludge floc (Shen et al., 2017), through which it facilitated floc aggregation (Wang and Li, 2008). These joint sludge flocs are suitable for sludge settling.

Besides, an increased trend of EPSp is clearly shown with

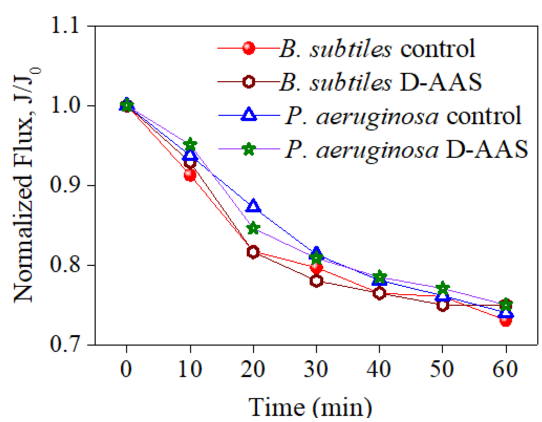

Figure 4. Effects of SMPs on membrane flux in filtration process. Reprinted with the permission from Wang et al. (2018).

the addition of PDMDAAC, which causes the increase trend of EPS products. However, the EPSc has an opposite trend showing slightly decreasing concentration comparing to control group. Meanwhile, the ratio of EPSp/EPSc increases indicating flocs aggregate. The concentration of SMPs decline significantly due to the presence of PDMDAAC. All of the changes of EPS and SMPs are regarded as the impacts of PDMDAAC on bacteria and sludge flocs. SMPs is produced by bacteria to defend harmful condition (Laspidou Rittmann, 2002; Lin et al., 2014). Once the impact-resistant granular sludge is established, the SMPs is secreted less (Seviour et al., 2010; Seviour et al., 2012).

Finally, pictures of membrane illustrate the thickness of cake layer is much thinner in R4 than control group (R1), showing that the biofilm was mitigated by PDMDAAC. These two studies demonstrates that D-AAs and cationic organic polymer coagulant can mitigate membrane fouling via interrupting EPS formulation and electrostatic interaction between flocs.

2.2. Membrane Modification

Membrane modification is a typical membrane fouling mitigating strategy (Wang and Waite, 2009; Wang et al. 2019; Oxley Livingston, 2022; Chen et al., 2023). With the modification of the membrane, the disadvantages of the pristine membrane have been overcome. For example, the surface of the membrane changes from hydrophobic to hydrophilic, which is exceptionally beneficial for antifouling. Besides, the changing zeta potential, surface roughness, and crosslinking some antibacterial groups can be altered as well. It shows the excellent antibacterial and antifouling performance when modifying some functional groups. In this section, graphene oxide-cellulose nanocrystal (GO-CNC) modified PVDF membrane is studied including relative material preparation, membrane modification, properties of modified membranes, and antifouling mechanism.

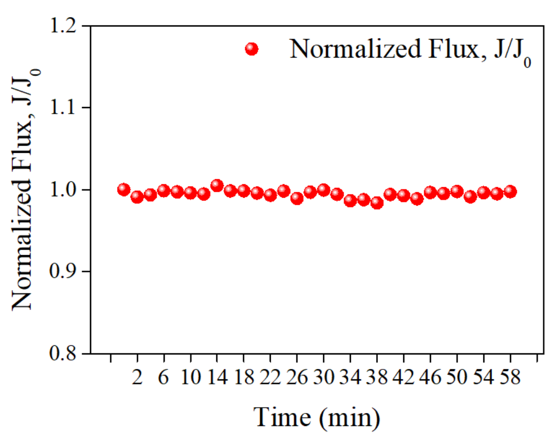

Figure 5. Effects of D-AAs on membrane flux. Reprinted with the permission from Wang et al. (2018).

In the past decades, graphene oxide (GO) has been applied in membrane fouling approaches (Liu et al., 2011; Zhao et al., 2014; Zeng et al., 2016; Meng et al., 2017; Zou et al., 2017; Abdel-Karim et al., 2018). It can be obtained when immobilizing GO nanosheets into the polymer matrix membranes. In addition, cellulose nanocrystal (CNC) is believed to develop a new membrane with several advantages: high permeate flux, high chemical resistance, and high hydrophilicity (Thakur Voicu, 2016). For its high-hydrophilicity, high-flux, and antifouling properties, one of the representative derivative nanomaterials GO-CNC has been considered as an effective antifouling membrane modification agent. By hydrogen bonding and intermolecular interactions, the GO-CNC composites will achieve when mixing GO and CNC.

A recent study performed by Lv et al. (2018) developed GO-CNC/PVDF membrane and its relative performance was evaluated. The mechanisms of this modified PVDF membrane (GO-CNC/PVDF) are studied. It was found that the chemical compositions and surface functional groups have been changed on the modified membrane. The pristine membrane has four distinct peaks corresponding to C-H and C-F stretching vibrations (Liu et al., 2015; Lv et al., 2017, 2018). The CH_2 peaks are retained on the modified membranes (Liu et al., 2015). The GO nanosheets have oxygen-containing groups and hydrogen bonds with the CNC, which are responsible for efficient bonding. However, no diffraction peak of GO is observed, which may be attributed to the coagulation effect.

The SEM images indicate that GO, CNC, and GO-CNC composites are wrinkled, stacked nanosheet micromorphology. Meanwhile, the observed smooth morphologies of modified membranes proved that hydrophilic CNC and GO-CNC accumulated on the membrane surface. Compared to pristine PVDF membrane, it can be seen that more pore channels have been shown in the modified membrane from the side images, which may enhance permeating flux and decrease resistance. It is attributed to the changes in thermodynamic instability of the casting solution caused by GO-CNC, leading to acceleration of the diffusion of non-solvent into the casting solution.

The lower water contact angle indicating high hydrophilicity is vital to mitigate membrane fouling (Meng et al., 2009; Meng et al., 2017). It shows that the oxygen-bearing functional groups and other hydrophilic groups from the CNC and GO-CNC may be responsible for the decreasing contact angle. Meanwhile, the higher zeta potential causes fewer foulants attaching to the membrane surface. It is worth mentioning that the pure water flux significantly increases after being modified with CNC and GO-CNC. It benefits from high hydrophilicity and developed membrane pore. Overall, the modified membrane has brilliant performance in membrane physicochemical properties.

Lv et al. (2018) also conducted a long-term experiment on the MBR system to test the performance of the modified membrane on membrane fouling. It was found that the GO-CNC/PVDF membrane had better antifouling performance as its TMP rising rate was much slower and the membrane lasted 73 days until the first chemical backwash. However, pristine and CNC/PVDF membranes were already chemically cleaned 2 ~ 3 times within the first 73 days. It was also found that the concentrations of COD and NH_4^+-N in the permeate separated by GO-CNC/PVDF, pristine and CNC/PVDF membranes mostly stayed the same.

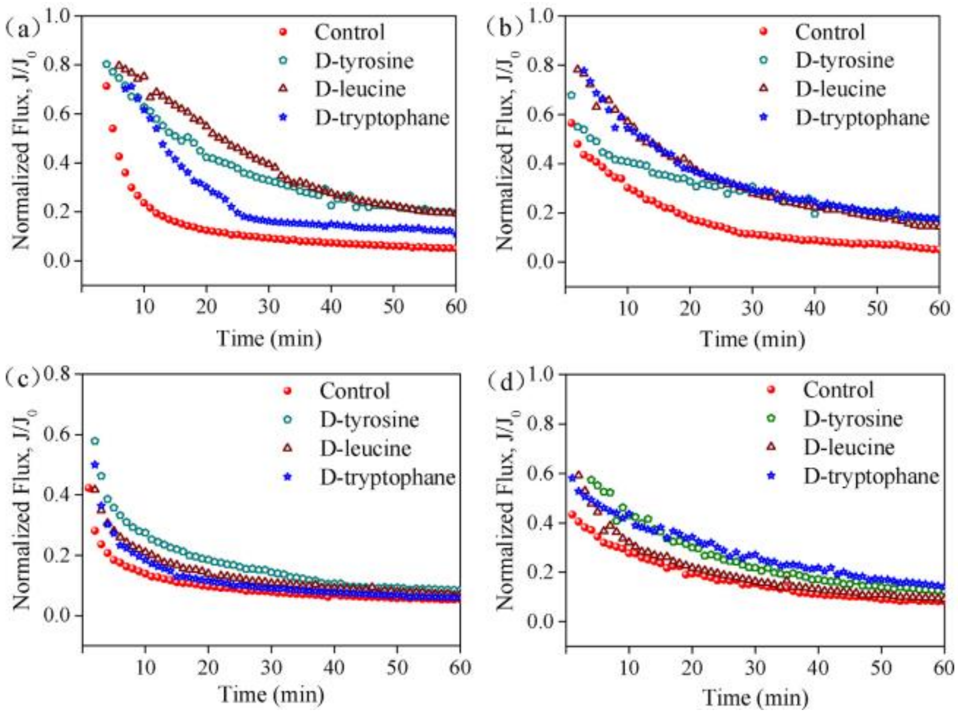

Figure 6. Effects of D-AAs on filtration process. (a) *B. subtilis* in the PVDF membrane, (b) *B. subtilis* in the PES membrane, (c) *P. aeruginosa* in the PVDF membrane, and (d) *P. aeruginosa* in the PES membrane. Reprinted with the permission from Wang et al. (2018).

This indicates that membrane modification may suppress membrane fouling but may not improve the treated water quality.

The EPS are regarded as one of the indicators of the sludge flocs and the physicochemical properties of microorganisms. The fouled membrane surface consists of charged proteins, polysaccharides and phospholipids (Hu et al., 2016; Sun et al., 2018a; Sun et al., 2018b). The modified membranes have less potential for membrane fouling. Due to the high hydrophobicity of proteins, the accumulated proteins are more extensive than polysaccharides and gradually form a fouling layer (Meng et al., 2009). Moreover, the GO-CNC can reduce membrane surface hydrophobicity by lowering protein adhesion.

A short-term experiment was conducted to further investigate membrane fouling mechanisms. It is evident that flux loss happens during the pristine membrane filtration and corresponding flux recovery ratio (FRR) values decrease, implying severe irreversible membrane fouling occurs (Meng et al., 2009; Iorhemen et al., 2016; Sun et al., 2018). However, the CNC/PVDF and GO-CNC/PVDF have better flux recovery after every backwashing with higher FRR. This indicates that the GO-CNC/PVDF membrane can effectively reduce fouling after cleaning (Sun et al., 2018). The GO-CNC/PVDF membrane has the highest resistance parameter Rc/Rf and lowest Rp/Rc, which means irreversible fouling can be removed more straightforwardly than the pristine membrane.

Three models have been introduced in this paper to better understand fouling mechanisms. The cake filtration-complete blockage model (CFCBM) fits the experimental data well for its coverage of the broadest range (Golbandi et al., 2013). In this model, cake formation and pore blockage can coincide (Golbandi et al., 2013). The suspended sludge particles can block membrane pores. In the early stage of membrane filtration, pore-clogging occurs. In other words, it is irreversible fouling. Although the cake layer can be removed by cleaning, the irreversible fouling in the pore cannot be cleaned by backwashing. This is the reason that TMP increases sharply. CNC/PVDF and GO-CNC/PVDF membranes can form compact cake layers due to the slow adsorption of containments. In addition, the increase in membrane hydrophilicity aids in removing to foulants from the pore. These remarkable properties reduce irreversible fouling.

The method mentioned in this paper is typical of membrane modification methods. These can be widely applied in wastewater treatment as well as in the study of membrane fouling mechanisms.

2.3. New MBR System

The traditional MBR system consists of a bioreactor and a membrane filtration unit. Due to its high efficiency, low cost, less sludge production, and small footprint, MBR has been widely used in wastewater treatment. However, membrane fouling is the most serious issue limiting the widespread use of MBR systems for wastewater treatment. The high sludge concentration was reported as the main reason for membrane fouling (Rosenberger Kraume, 2015). The consequences of membrane fouling include increased energy consumption and frequent membrane replacement. Two methods currently are inten-

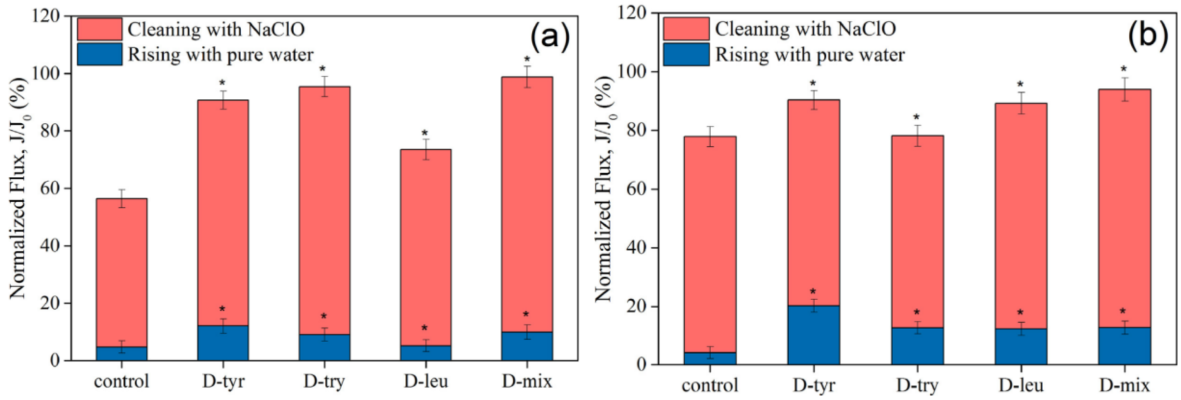

Figure 7. Effects of D-AAs on cleaning the PES membrane process (a) for *P. aeruginosa*, and (b) *B. subtilis*. Reprinted with the permission from Wang et al. (2018).

sively studied to suppress membrane fouling is he modifying membrane materials to improve membrane antifouling properties, and adding flocculants to the MBR (Ahsani et al., 2022; Choi, 2022).

The advanced bacteria-algal symbiotic systems are being studied intensively. In a symbiotic system, algae can provide an aerobic environment through photosynthesis and CO_2 through respiration. This autonomous system can achieve high contaminant removal efficiency at a low cost. Meanwhile, the microalgae system can effectively remove nitrogen and phosphorus (Decostere et al., 2016). This algal-bacterial wastewater treatment method has been put into practice in Spain and has achieved good performance in water treatment and disposal (Munoz and Guieysse, 2006). Based on a previous study, a novel algal-sludge bacterial MBR (ASB-MBR) system has been built to achieve excellent removal efficiency and the mitigation of membrane fouling (Huang et al., 2015). Due to aerobic conditions, algal bacteria have been introduced to the MBR system for antifouling. The EPS concentration has decreased in this combined system (Sun et al., 2018).

The Chlorophyll-α/VSS, representing the algae content/bacteria ratio, indicates the growing state of algae and bacteria. The balance growth occurs in the ASB-MBR system in long-term operation. Since pore size of membrane is smaller than that of the algae, the algae can be well retained and grown in the reactor. The algae and bacteria in the ASB-MBR system can build up a mutually beneficial relationship.

As for the removal efficiency, COD is 93.9% (C-MBR is 89.3%), NH_4^+-N is 95.7% (89.0%), TN is 30.2% (20.1%), PO_4^{3-} is 23.7% (15.2%). The ASB-MBR achieved excellent performance in removing contaminants and nutrients at a low mixed liquor suspended solid (MLSS) concentration. The TMP is measured to evaluate membrane fouling. ASB-MBR has one cleaning period, with TMP rising steadily compared to C-MBR. These results suggest that algae's presence reduced the membrane fouling rate.

The average particle size of ASB-MBR is 163.19 μm, which is smaller than that of C-MBR. The bigger particle size increases the permeate flux (Hwang et al., 2007). However, the results of this study differ from the previous study. In C-MBR, the excess filamentous bacteria cause a larger floc size. It may be related to less compact sludge flocs. Moreover, these flocs may tend to accumulate on the membrane surface.

The SMPs in ASB-MBR are higher than in C-MBR, possibly due to shorter SRT, which may lead to more SMPs (Pan et al., 2010). Furthermore, the polysaccharides and proteins are different between the two systems. Lower polysaccharides and higher proteins are shown in the ASB-MBR system. In the ASB-MBR system, the presence of algae might enhance the activated sludge and polysaccharide, which is easier to biodegrade by activated sludge (Zhang and Bishop, 2003).

Furthermore, SMPpr/SMPps increases due to high protein and low polysaccharides. A higher ratio led to a weak ability to form a cake layer that is easy to clean. The results of EPS are pretty different. The total EPS, protein, and polysaccharides are lower in the ASB-MBR system. It has been reported that a low EPS concentration decreases membrane fouling potential.

As for the mechanism, EPSs and SMPs produced impact the morphology of sludge flocs, which is a critical factor in cake resistance. The flocs in ASB-MBR are much closer to spheres, more inerratic and smoother. The improved morphology of cake flocs provides the benefit of decreasing resistance and mitigating membrane fouling. It is convinced according to above results that the algae-bacteria system ASB-MBR provides a potential method to solve membrane fouling.

3. Conclusions

An MBR has been used to treat greywater and wastewater. However, the MBR membrane biofouling is a roadblock to its wide applications in these areas. Therefore, there is an urgent need to develop and apply novel membrane biofouling prevention and control strategies. This paper extensively reviews three promising strategies for mitigating MBR membrane biofouling: feedwater pretreatment using D-AAs and a cationic polymer flocculant, membrane surface modification, and a new MBR system. Pretreatment using D-AAs and PDMDAAC is considered the most cost-effective and environmentally friendly strategy for membrane biofouling control because it can significantly change the physiochemical properties of bacteria or other contaminants and reduce the amount of EPSs and SMPs released into wastewater. Furthermore, GO-CNC/PVDF membrane is recommended for MBR membrane biofouling control because it is designed to form less biofilm and an incompact cake layer on the membrane surface and has outstanding antifouling performance with a lower chemical membrane washing frequency. Moreover, an advanced ASB-MBR system with an oxygen-provider algae-bacteria symbiotic system is recommended as one of the promising MBR membrane fouling suppression strategies because it is more suitable to treat a large amount of wastewater.

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