Membrane fouling and control in algae-rich water treatment

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Abstract. Microfiltration and ultrafiltration membrane technology has now become one of the important methods for algae-rich water treatment due to its better solid-liquid separation effect and microbial retention capacity, while the existence of membrane fouling has been restricting the full-scale promotion and application of low-pressure membrane technology. The main foulants causing membrane fouling in algal-rich water include algal cells, algal debris and algal organic matter AOM (EOM and IOM). In this paper, the mechanism of membrane fouling caused by algal foulants in MF/UF for high algal water treatment is analysed in terms of both individual and composite fouling. Both individual and composite effects of algal foulants can lead to severe membrane fouling, in which algal cells and algal debris mainly dominate the accumulation of filter cake, while AOM mainly leads to membrane pore blockage. Membrane fouling is exacerbated by compounding effects, with severe reversible and irreversible fouling caused by filter cake formation and membrane pore shrinkage. This paper also discusses several common membrane fouling control methods, including pretreatment (oxidation, coagulation, combined oxidation-coagulation and adsorption) and membrane modification, and it is found that these several membrane fouling control methods can achieve good results.

1 Introduction

Surface water is the major source for drinking water production all over the world. In recent decades, eutrophication of reservoirs and lakes has become an worldwide puzzle due to the discharge of pollutants and nutrients such as nitrogen and phosphorus. And as a result, there have been more and more algal bloom events in the past years. Algal blooms represent a primary threat to the security of this kind of water resources. Under suitable environment, algae in water can form cell colonies through cell division. These algal cells are generally difficult to be preyed by zooplankton, so they exhibit strong competitive growth advantage. Meanwhile, since most algal cells in surface water do not possess the solid cell shell, they often suffer from the issue of cell breakage, which further deteriorate the water quality of the eutrophic water bodies.

With rapid development of membrane technology, low-pressure membranes such as ultrafiltration (UF) and microfiltration (MF) are increasingly applied to treat algae-rich water, due to the low treatment efficiency and high chemical dosage of traditional treatment approaches including coagulation, sedimentation, air flotation, centrifugation, etc. The pore size of UF and MF membranes is significantly smaller than algal cells, therefore, these membranes can effectively retain algal cells with low energy consumption, which has currently become the main process for algae-rich water treatment. However, membrane fouling will inevitably occur during the UF/MF treatment of algal-rich water. Serious membrane fouling increases the frequency of physical/chemical cleaning, thus causing the difficulties in system maintenance and reducing the lifespan of the membrane. The membrane fouling is mainly attributed to pore shrinkage, blockage and formation of the cake layer. The main foulants in algae-rich water include algal cells, algal debris and algal organic matter AOM (EOM and IOM), both individually and jointly, causing membrane fouling with different degrees and different mechanisms. This paper comprehensively overviewed the UF/MF membrane fouling mechanisms and fouling control methods for the treatment of algae-rich water based on the relevant articles published in the literature, with the purpose to deepen the understanding of algae fouling behavior and promote the successful operation of low-pressure membrane systems for efficient algae-rich water treatment.

2 MF/UF membrane fouling analysis

2.1 Algal cell fouling

Algal cell foulants include intact algal cells and algal debris. Although algal cells are diverse, since the cell size of algal cells and most of the algal debris are substantially larger than the pore diameter of UF and MF membranes, they can be almost completely retained and accumulated near the membrane surface, forming a cake layer. For some small sized algal debris, they might also enter the membrane pores, resulting in irreversible membrane pore blockage. Generally, as the concentration of algal cell
solution increases, the rate of decrease in specific flux of the membrane increases significantly. With the progress of filtration, the number of algal cells deposited on the membrane surface increases, the cake layer becomes thicker and less reversible, thus causing serious membrane fouling.

The interfacial interaction between algal cell and membrane surface is an important fundamental governing the membrane fouling mechanism. Investigating the adhesive and cohesive free energies according to XDLVO theory, which represent the initial foulant-membrane interaction and later foulant-foulant interaction, respectively. Although the algae cell caused the fastest flux decline, algal debris had higher negative adhesive free energy and cohesive free energy, indicating the debris would be easier to attach to the membrane surface causing irreversible fouling. On the other hand, although the adhesive free energy of algal debris was comparable to algal organic matter, the relative flux reduction caused by adsorption of algal debris was substantially lower than algal organic matter. The reason was that the cells and algal debris are too large to penetrate into membrane pores, while the organic matter will block the membrane pores.

Electrostatic force is one of the relevant factor influencing interaction energies. The algal debris possessed lower zeta potential as compared with the algal cell. The decrease of zeta potential had been attributed to the disruption of cell structure and release of intracellular organic matter (IOM). The lowered electrostatic repulsion might lead to much easier adhesion of algal debris to the membrane surface than algae cells, and promote their aggregation on membrane surface. Therefore, algal debris cells generally caused more serious membrane fouling and fouling irreversibility than algae cells.

The membrane fouling caused by algal cells and cell debris is featured by secondary dramatic decline of the flux, which is associated with the cake layer compression. This could be attributed to the compressibility of the algal cake layer deposited on membrane surface. For cake layer-induced membrane fouling, it was noticed that the fouling extent was significantly augmented as the cake layer thickness increased. As algal cells are much larger than the membrane pores, if they are incompressible, it is difficult for the cake layer to generate excessive filtration resistance on the membrane. In addition, the larger the concentration of algal cell solution, the earlier the second decrease in specific flux appears, which is mainly due to the fact that the larger the concentration of algal cell solution, the faster the growth of cake resistance, and therefore the compression of the cake layer also appears earlier. Babel and Takizawa [1] revealed three different stages for cake resistance development, defined as slight increase, exponential increase and linear increase, respectively. In the initial stage, algal cells attached to membrane surface with the progress of filtration, while no significant resistance could be observed due to the low number of cells deposited. As the amount of algal cells on the membrane increased, the cake layer was formed and cell compression occurred, with the cake compactness rising to the maximum level in a short time. After the stage, the cake resistance increased with the thickness of the algal cake linearly. Liu [2] et al. found that under higher transmembrane pressure (TMP), the cake layer resistance of Microcystis aeruginosa would be dramatically increased, which was attributed to the more severe deformation and compression of the cake layer at higher TMP. In other words, low TMP condition may facilitated the formation of a loose cake layer of algal cells on membrane surface with substantially decreased compression extent and filtration resistance, thus deceasing the physical/chemical cleaning and operation energy requirements. For example, for UF membrane filtration, due to the smaller pore size possessing higher membrane resistance, the lower actual flux causes a low TMP, which contributes to a gentle, loose deposition of algal cells. In contrast, this phenomenon was not observed in MF membrane systems because their high flux and high TMP resulted in the deposition of a tightly accumulated initial cake. From this viewpoint, optimization of TMP by simultaneously considering the cake compressibility and permeate flux is very important for the sustainable operation of low-pressure membrane systems.

2.2 Algal organic matter fouling

A large number of studies have been conducted on the membrane fouling mechanisms of algal-rich water, and found that algal organic matter (AOM) is another dominant foulant. AOM consists mainly of algal cell-released extracellular organic matter (EOM) and algal cell-broken or dead-released intracellular organic matter (IOM). Some studies have demonstrated that microbes can cause substantial fouling on UF and MF membranes through releasing extracellular polymers (EPS) [3,4]. Similar to EPS, the composition of both EOM and IOM contains proteins, polysaccharides and humic-like substances, indicating the algal AOM (EOM and IOM) could adversely impact the membrane filtration performance. For UF and/or MF, the algal cells generally caused reversible fouling, i.e. the fouling that can be effectively removed by hydraulic cleaning; While AOM causes larger flux decreases than algal cells due to the fact that AOM forms larger membrane pore blockages and less porous cake, resulting in higher irreversible membrane fouling [5], which could only be removed by chemical cleaning.

AOM consists of a variety of organic compounds, among which peptides/proteins and polysaccharide compounds are the main components. It has been demonstrated that algal cultures contain more than 50% of biopolymers such as polysaccharides and proteins in AOM, with the remainder consisting mainly of difficult-to-degrade organic compounds such as humic acid-like and/or low molecular weight acidic and neutral compounds [6]. Peptides and proteins contain a large number of polar and charged functional groups, posing a high potential for membrane fouling. Proteins and polysaccharides of AOM are widely considered to be responsible for serious fouling problems in membrane filtration processes. The organic substances causing irreversible fouling cover all molecular weight ranges, and most AOMs are composed of a large proportion of
The characteristics of AOM are influenced by the algal growth phases. The hydrophilic fraction (HPI) was the main fraction during the whole lifetime; while the hydrophobic (HPO) and hydrophilic (TPH) fractions gradually appear after the exponential phase. Therefore, algal AOM possesses both hydrophilic and hydrophobic fractions. However, it has been found that for cyanobacteria, green algae and diatoms, the hydrophilic component of AOM predominates in both the exponential and stationary phases \cite{8}. This observation is highly consistent with that by Namguk \cite{9}, who found the AOM is characterized by high HPI fraction, low DOC/DON, low SUVA, and abundant proteins and polysaccharides. Compared to the stable phase, AOM at the early exponential phase had significantly less high MW organic components (e.g., biopolymers (>20,000 Da) and humic-like substances (500-20,000 Da)) but contained more medium MW components (i.e., blocks, 350-500 Da), low MW acids, and low MW humic substances (<350 Da). In addition, stationary phase AOM contains significantly more humic-like substances, which can be deposited by hydrophobic interactions with the membrane surface \cite{10}, and large amounts of high MW humic-like substances may lead to enhanced membrane fouling effects. Therefore, the fouling potential of AOM shows a stationary phase > exponential phase.

Generally algal EOM and IOM differ in molecular weight distribution and hydrophobicity. Small molecules may penetrate into the membrane pores causing blockage, while the macromolecular fraction is mainly concentrated on membrane surface and then form a cake layer. Therefore, there existed some differences between EOM and IOM in terms of membrane fouling. The MWs of EOM and IOM exhibit three main peaks corresponding to biopolymers (MW of 100-1000 kDa), humic substances (MW of 2500 Da), and low molecular weight acids and building blocks (MW of 1000 Da). In addition, EOM has a higher peak area for high MW biopolymers than IOM. In the study by Li \cite{11} et al. it was found that the MW ranges of IOM were <1 kDa, 40-800 kDa and >800 kDa accounting for 27%, 42% and 31%, respectively. In contrast, the major EOM peaks are located in the 1-100 kDa range and accounting for 85%. In addition, the average MW of IOM was higher than that of EOM. This difference may be attributed to the discrepancy between the algae species and culture periods in the two studies. Usually, Low MW EOM is primarily negatively charged, which results in more electrostatic repulsion than size repulsion, thus mainly contributing to reversible fouling. While the large MW fraction of EOM is more likely to cause cake fouling layer due to size rejection. IOM with MW over 30kDa macromolecular components are dominated by biopolymers such as proteins and polysaccharides, and have a relatively high negative cohesive surface free energy with a higher tendency of self-aggregation, which may form a more dense cake layer. Relatively small polysaccharide molecules irreversibly block or shrink and compete for membrane pores in the initial stages of fouling, resulting in most of the proteins in the IOM can only be deposited on the membrane surface (more reversible). Therefore, during the IOM filtration process, the cake formation and membrane pore blockage control the formation of fouling.

Furthermore, the hydrophobic organics composed typically of non-polar molecules tend to move to the interface of the aqueous phase. As a result, hydrophobic organics prefer to adhere to the membrane, and the hydrophobicity will promote foulant-membrane interactions and aggravate the irreversible adhesion of organics to the membrane surface. For hydrophilic organics, which typically contain a large number of polar functional groups, which can form hydrogen bonds with water molecules, resulting in a stronger affinity for water than hydrophobic organics. Therefore, the hydrophilic organics have a lower tendency to adhere to the membrane. However, when the hydrophilic organics form a cake layer on the membrane surface, they can create more resistance to water flow than hydrophobic organics owing to stronger foulant-water interactions. EOM is characterized by strong hydrophilicity, usually the hydrophilic components may enter the interior of the membrane, causing pore blockage. Similarly, although the hydrophobic and hydrophilic components account for 23.9% & 66.5% in EOM and 15.3% & 78.5% in IOM, respectively, EOM is more hydrophilic than IOM, which stemmed from that he assumed that cyanobacteria-related proteins have stronger hydrophobicity. In contrast Pivokonsky \cite{12} also found that both the EOM and IOM of cyanobacteria, diatoms and green algae in the exponential and stable phases are dominated by HPI substances. The high peptide/protein content in the IOM contributes to reduce the hydrophobicity of the IOM, resulting in more hydrophilic IOM than EOM for cyanobacteria, diatoms and green algae, which stems from treating peptides/proteins as strongly hydrophilic.

### 2.3 Combined fouling

In algae-rich water treatment, when algal cells rupture, many algal debris are formed and large quantities of IOM are released. At the same time, intact algal cells release EOM as they grow. The coexistence of different algal
foulants results in a composite fouling that is more serious than the individual foulants. Permeation loss during filtration of algae-rich water initially comes from cells and algal debris, while AOM may fill the voids in the filter cake layer, thus further reducing the flux. As shown in Figure 1, the pristine membrane undergoes pore clogging and shrinkage during the initial filtration stage. Algae cells and AOM will be deposited on the membrane surface, and a portion of the AOM will enter the interior of the membrane pores. As filtration continues, the accumulation of algal cells and AOM near the membrane increases and forms a cake layer, and more and more AOM enters the interior of the membrane pores, leading to more severe pore shrinkage. At the same time, algal cells have a tendency to compress, which reduces the porosity of the filter cake layer and further aggravates membrane fouling [13]. According to Qu [14] et al. the foulants of algal cells + algal debris, EOM + IOM and algal cells + EOM + IOM all exacerbate membrane contamination. Generally, in truly algal-rich water, different algal-derived foulants coexist. In order to determine the mechanisms of individual and combined fouling, four classical filtration models can be used and a schematic of the filtration models is provided in Fig. 2 [15]. The $R^2$ values (as shown in Table 1) indicate the relevance of the different membrane fouling mechanisms. In the case of separate fouling by algal foulant, the main manifestation is standard clogging, whereas in the case of multiple composite fouling, standard clogging and filter cake formation predominate.

<table>
<thead>
<tr>
<th>Feed solution</th>
<th>Complete blocking</th>
<th>Standard blocking</th>
<th>Intermediate blocking</th>
<th>Cake filtration</th>
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</thead>
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<tr>
<td>EOM</td>
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<td>0.999</td>
<td>0.959</td>
<td>0.986</td>
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<tr>
<td>IOM</td>
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<td>0.997</td>
<td>0.909</td>
<td>0.975</td>
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<tr>
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<td>0.999</td>
<td>0.951</td>
<td>0.993</td>
</tr>
<tr>
<td>Debris</td>
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<td>0.999</td>
<td>0.981</td>
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</table>

3 MF/UF membrane fouling control strategies

3.1 Oxidation pre-treatment

Ozone oxidation can be effective in changing the characteristics of foulants, thus reducing membrane fouling. When ozone acts on the algal solution, it will adsorb onto some of the algal cells due to the decomposition of the algal cells and EOM, which will increase the negative charge on the surface of the algal cells and further enhance the electrostatic repulsion effect, which will help the structure of the filter cake to become looser, thus mitigating the reversible fouling to a certain extent. As shown in Figure 3, ozone oxidation significantly reduced the hydrophobic fraction, which is usually considered as the main component leading to membrane fouling, thus weakening the thickness of the filter cake layer [16]. With the increase of ozone dosing, the rate of TMP increase gradually decreased, and the membrane fouling resistance was reduced, especially the hydrodynamic reversible fouling resistance.

Potassium permanganate is a mild oxidizing agent, which has also been widely studied in the pretreatment of algal-rich water. On the one hand, the mild oxidation provided by KMnO$_4$ can inactivate algal cells; on the other hand, the MnO$_2$ generated by oxidation adsorbs on the surface of algal cells, lowering the (-) zeta potential of the cell surface and attenuating the electrostatic repulsion between algal cells, making them easier to aggregate. In addition, the adhesion of MnO$_2$ particles likewise promotes cell aggregation, significantly improves their
coatings and deactivating viable algae, thereby enhancing coagulation by altering zeta potential, destroying organic caused by algae-rich water. simultaneously, to better mitigate membrane fouling oxidation and coagulation of algae-rich water oxidization-coagulation pretreatment can realize the metal ion concentration. Therefore, the combined causes further rupture of algal cells or too high residual better control of membrane fouling. By simply adding mitigate part of the membrane fouling, but not achieving those formed by aluminum-based coagulants, and iron-based coagulants are more resistant to stress than aluminum salt flocs tend to break up in the combination coagulation efficiency of polymerized aluminum (PACl) on algae harvested at different growth stages, and found that at optimal dosages, especially at the exponential and stabilization stages, more than 90% of the algal removal rate. In addition to aluminum-based coagulants, excellent membrane fouling control can also be achieved with iron-based coagulants. Flocs formed by iron-based coagulants are more resistant to stress than those formed by aluminum-based coagulants, and aluminum salt flocs tend to break up in the combination of coagulation and membrane filtration. It is shown that iron salt coagulation is more favorable for algae removal and thus for subsequent membrane fouling control.

3.2 Coagulation pre-treatment

The main components of high molecular weight biopolymers in AOM are polysaccharides and proteins. Polysaccharides and proteins are rich in carboxyl and hydroxyl groups, which have a strong binding capacity with trivalent metals and therefore form larger size complexes during coagulation. Tang et al. investigated the coagulation efficiency of polymerized aluminum (PACl) on algae harvested at different growth stages, and found that at optimal dosages, especially at the exponential and stabilization stages, more than 90% of the algal removal rate. In addition to aluminum-based coagulants, excellent membrane fouling control can also be achieved with iron-based coagulants. Flocs formed by iron-based coagulants are more resistant to stress than those formed by aluminum-based coagulants, and aluminum salt flocs tend to break up in the combination of coagulation and membrane filtration. It is shown that iron salt coagulation is more favorable for algae removal and thus for subsequent membrane fouling control.

3.3 Combined oxidation-coagulation treatment

Conventional oxidation and coagulation are also the prevailing methods of membrane fouling control in algae-rich water treatment. Although oxidation and coagulation mitigate part of the membrane fouling, but not achieving better control of membrane fouling. By simply adding more oxidant or coagulant to improve the removal rate of algal cells and their metabolites is not ideal, and also causes further rupture of algal cells or too high residual metal ion concentration. Therefore, the combined oxidation-coagulation pretreatment can realize the oxidation and coagulation of algae-rich water simultaneously, to better mitigate membrane fouling caused by algae-rich water.

Pre-oxidation processes can effectively assist coagulation by altering zeta potential, destroying organic coatings and deactivating viable algae, thereby enhancing the removal of algae and other suspended particles during subsequent settling or filtration. The oxidant will be activated by metal ions in the combined treatment and its products play an positive role in membrane fouling control. When KMnO₄ is used as an oxidant, KMnO₄ will generate manganese dioxide (MnO₂) in situ, and MnO₂ facilitates an increase in particle concentration to accelerate flocculation kinetics and promotes flocculation by adsorption onto other particles, in addition, MnO₂ particles can contribute to increase the porosity and decrease the compressibility of the cake layer. Similarly, the activation of persulfate (PMS) leads to the production of SO₄⁻ and -OH, which can induce the generation of -OH, whereas the oxidation of SO₄⁻ and -OH leads to the mineralization of some low MW organic matter also contributes to the reduction of AOM and alleviates its fouling of membrane modules. In the presence of the iron or aluminum, et al. pre-oxidation may destroy the metal ion complexes, leading to in-situ coagulant production, moreover, in-situ produced coagulant has better coagulation effect. For example, the in situ formed Fe³⁺ by Fe²⁺ oxidation can act as a procoagulant to neutralize the negative charge on the cell surface and promote the settling of algal cells. Moreover, the in situ formation of Fe³⁺ has a larger reaction surface area than the pre-formed Fe³⁺, which can improve the sedimentation properties of flocs and contribute to the effective removal of algae. Nowadays, numerous studies have demonstrated the contribution of oxidation-assisted iron-based coagulation and oxidation-assisted aluminum-based coagulation in membrane fouling mitigation, and both are superior to the treatment effect when oxidation or coagulation alone is used.

3.4 Adsorption pre-treatment

When adsorbent is fed into a membrane filtration system, adsorbent particles accumulate on the membrane surface and rapidly form an adsorbent layer. As shown in Figure 5, larger foulants are retained by the particulate layer, and when smaller adsorbent particles are used, the retention of foulants is enhanced, and pore clogging in the particulate layer will be the primary fouling mechanism for flux reduction. When larger adsorbent particles are used, more foulants pass through the adsorbent layer, resulting in the formation of a gel layer on the membrane surface. As the most common adsorbent, PAC has achieved better membrane fouling control. PAC particles have more adhesion sites, leading to greater biodegradation potential. Also, PAC has better adsorption of certain proteins and significantly reduces the amount of protein foulants in solution. When PAC enters the membrane system, the deposition of PAC particles weakens the algal cell layer on the membrane surface. Since the diameter of most PAC particles is much larger than the membrane pore size, the PAC layer of the membrane has a relatively loose structure and high porosity, which will help to improve the permeability of the membrane.
3.5 Modification of membranes

Hydrophobic membranes are susceptible to fouling by adsorption, deposition, and irreversible attachment of extracellular and hydrophobic macromolecules (e.g., proteins, polysaccharides, and lipids) present in surface water, wastewater, and microalgal media, whereas hydrophilic membranes have better resistance to fouling than hydrophobic membranes. Therefore, hydrophilic membranes should be considered first in the filtration of algae-rich water. As shown in Fig. 6 for hydrophilic surface coated membranes with tightly hydrated surfaces, the hydrophilic surface forms a layer of water on the membrane surface, which restricts the space for the attachment of pollutants and thus resists their deposition on the membrane surface, which manifests itself as a mechanism of fouling resistance [28].

At present, there have been many studies on the modification of hydrophilic membrane preparation. One is the modification of the membrane surface by coating technology to improve the hydrophilicity of the membrane. By coating the surface of PVDF and PET membranes with functional coating materials such as hydrophilic polyvinyl alcohol (PVA) polymer, the PVA coating makes the membrane surface more hydrophilic with a nearly 64% reduction in contact angle, and the adsorption fouling caused by hydrophobic macromolecules in the algal culture solution will be greatly decreased [29]. Similarly, by coating the PVDF/LiCl membranes with NaY zeolite and ZCC, the membrane contact angle could be reduced to 74.41° and 69.21°, respectively [30]. The second is the modification of membranes by surface grafting method, and this approach has also been well applied. The modification of polysulfone (PSF) surfaces by graft polymerization of acrylic acid (AA) surfaces induced by atmospheric pressure air plasma (Air APP) surface treatment and the generation of superhydrophilic layers of branched aniline oligomers (BAO) grafted on membrane surfaces by diazo-induced grafting both resulted in a significant reduction in fouling resistance. Moreover, the modified PSF membrane has high surface hydrophilicity and larger negative zeta potential compared with the original PSF membrane, while the water contact angle of BAO-PES modified membrane is reduced to 0, exhibiting excellent stable permeability and anti-fouling properties. Besides there are quite studies of hydrophilic modifications based on induced phase separation techniques. Using the non-solvent induced phase separation (NIPS) technique [31] or the thermally induced phase separation (TIPS) process [32] for modifying PES membranes, both resulted in smaller static contact angles, higher water fluxes and excellent anti-fouling properties when filtrating algal solutions. The hydrophilic modification of the membranes also achieved better anti-fouling performance in algae harvesting with high permeate flux and almost 100% algae retention.

4 Conclusion

This paper mainly focuses on the analysis of membrane fouling problems occurring during the treatment of algae-rich water by MF/UF low-pressure membrane system, and proposes several membrane fouling control strategies with better effect and wider application. The conclusions are as follows:

(1) Algae cells and algal debris have large physical size and compressibility, resulting in the fastest flux decrease. While AOM fills the voids in the filter cake layer, the flux decline is significantly intensified. In the case of compound fouling, the flux decline will be further exacerbated.

(2) After oxidative pretreatment, most of the algal cells and AOM were inactivated, and the number of foulants associated with severe membrane fouling was reduced, thus reducing the membrane fouling caused by algal foulants.

(3) The addition of appropriate coagulants to the algal solution results in an increase in the particle size of the organic matter, and these aggregates can form a porous cake layer that improves membrane permeability.

(4) Adsorbents reduce the resistance to irreversible fouling by adsorbing small fractions of substances, and large molecular weight substances, in the presence of adsorbents, increase the permeability of the cake layer formed on the membrane surface. In addition, a portion of the adsorbent adheres to the membrane surface, resulting in a relatively loose adsorbent layer with large porosity, which positively affects the membrane permeate flux.

(5) By hydrophilic modification of the membrane, the hydrophilic membrane forms a hydration layer with water
molecules, weakening the interaction between the membrane and the fouling, which can effectively reduce the fouling.

References


