REVIEW ARTICLE

Ceramic membrane fouling mechanisms and control for water treatment

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HIGHLIGHTS

- The fouling is summarized based on ceramic membrane performance and pollutants.
- The current research methods and theoretical models are summarized.
- The membrane fouling control methods and collaborative technology are reviewed.

GRAPHIC ABSTRACT
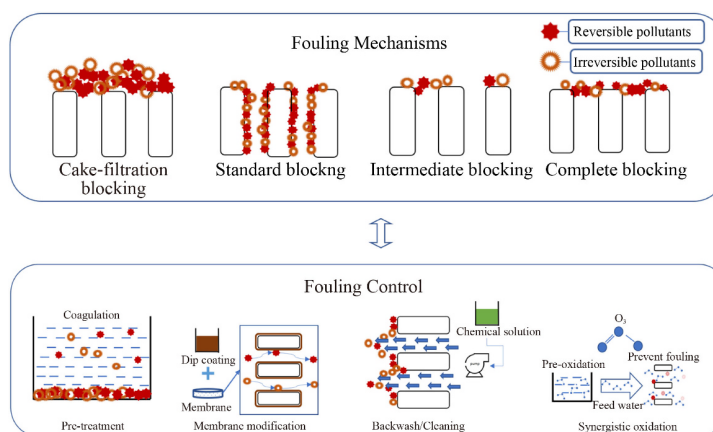
ABSTRACT

Membrane separation, as an important drinking water treatment technology, has wide applications. The remarkable advantages of ceramic membranes, such as chemical stability, thermal stability, and high mechanical strength, endow them with broader prospects for development. Despite the importance and advantages of membrane separation in water treatment, the technique has a limitation: membrane fouling, which greatly lowers its effectiveness. This is caused by organics, inorganic substances, and microorganisms clogging the pore and polluting the membrane surface. The increase in membrane pollution greatly lowers purification effectiveness. Controlling membrane fouling is critical in ensuring the efficient and stable operation of ceramic membranes for water treatment. This review analyzes four mechanisms of ceramic membrane fouling, namely complete blocking, standard blocking, intermediate blocking, and cake filtration blocking. It evaluates the mechanisms underlying ceramic membrane fouling and summarizes the progress in approaches aimed at controlling it. These include ceramic membrane pretreatment, ceramic membrane surface modification, membrane cleaning, magnetization, ultrasonics, and nanobubbles. This review highlights the importance of optimizing ceramic membrane preparation through further research on membrane fouling and pre-membrane pretreatment mechanisms. In addition, combining process regulations with ceramic membranes as the core is an important research direction for ceramic membrane-based water treatment.

1 Introduction

In recent decades, membrane technology has become widely used in water treatment field, which can be attributed to continued improvements in the permeation efficiency, stability, and eco-friendliness of membrane. The common membrane modules on the market can be divided into two categories based on material, namely, organic (i.e., polymer membrane) and inorganic (i.e.,

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ceramic membrane) (Fig. 1). Inorganic membranes have partially replaced the position of organic membranes in the water treatment market due to an array of advantages over organic membranes. Specifically, since the 1990s, inorganic membrane technology has developed more rapidly than its organic counterparts, at an annual growth rate of 30%–35%.

Inorganic ceramic membranes outperform organic membranes in terms of chemical stability, mechanical strength, pore size distribution, separation effectiveness, service life, and anti-fouling capacity. However, in terms of cost, organic membranes are cheaper than inorganic membranes. This makes them more preferred during the initial investment of a project, but in practical scenarios, the production capacity, energy consumption, and chemical consumption of organic membranes are considerably affected by changes in water quality and temperature; they also require high-frequency backwashing and chemical cleaning. Additionally, organic membranes are also more impacted by physical damage, making it difficult to maintain a continuous and stable operation within their service life. Moreover, the application of ceramic membrane technology is no longer limited to the treatment of difficult wastewater, such as during printing and dyeing, metallurgy, and the chemical industry. Table 1 shows the recent applications of ceramic membranes in different water treatment scenarios. In recent years, large-scale projects such as municipal water treatment and seawater desalination have also begun to use ceramic membranes to replace organic membranes.

Although ceramic membranes are less prone to fouling than organic membranes, it nevertheless poses a significant barrier to their use in water treatment. In this study, mechanisms underlying ceramic membrane fouling are reviewed. Our discussion includes various solutions against fouling, such as process combination, membrane surface modification, and optimization of membrane operating conditions. The membrane fouling can be effectively controlled, extending the cleaning cycle and service life of ceramic membranes (Sakamoto et al., 2022). Additionally, we also review the advantages of ceramic membranes, including their high-efficiency filtration. Finally, we conclude by addressing the future of membrane technology in terms of research avenues and applicability. Ceramic membrane fouling could be attributed to a set of complex physical and chemical functions. These are determined not only by the properties of the membrane itself, but also by the properties of the filtered solution. Therefore, understanding the mechanism underlying the formation of membrane fouling is essential to devise ways to effectively control it.

2 Causes and mechanisms of ceramic membrane fouling

Membrane fouling has always plagued the long-term stable operation of membrane systems. The most effective way to solve membrane fouling is to understand its formation and its drivers. Membrane fouling was defined as a phenomenon in which membrane pores are blocked by pollutants, resulting in a decrease in the amount of water filtered or a decline in water quality. Specifically, fouling is often caused by the physical and chemical interaction of particles, colloidal particles, or solute macromolecules in the feed liquid in contact with the membrane. Due to concentration polarization, the concentration of some solutes on the membrane surface exceeds their solubility, which results in their adsorption
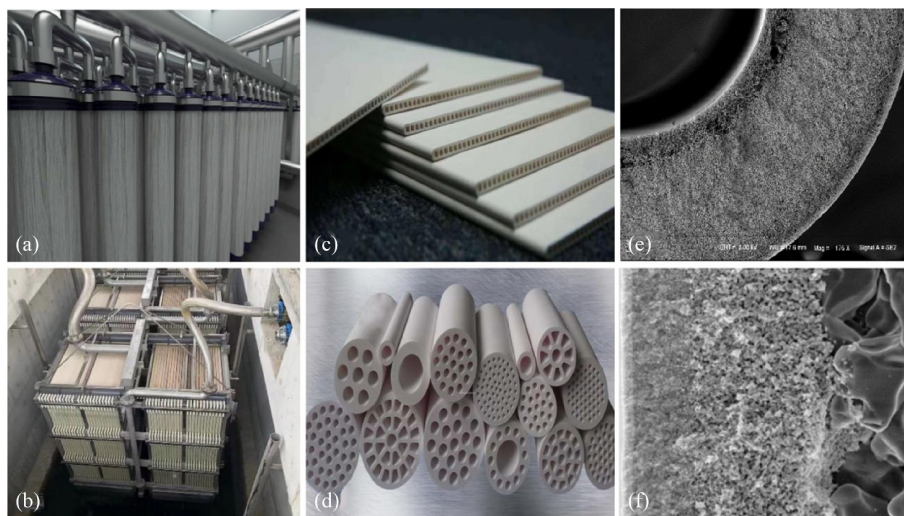

Fig. 1 Common inorganic ceramic membranes and organic membranes (ultra/microfiltration membranes) in the market (Source: original). (a) Hollow fiber organic membrane, (b) flat organic membrane, (c) plate inorganic ceramic membrane, (d) tubular inorganic ceramic membrane, (e) organic membrane section, (f) ceramic membrane section.

Table 1 Applications of ceramic membranes in different water treatment scenarios

Region	Project name	Application field	Main impurities in water	Operation time	Process capability (m³/d)	Treatment goal	Reference
Alberta, Canada	Industrial application of ceramic nanofiltration membrane for water treatment in oil sands mine	Oily wastewater treatment	Salts, oil, high concentration of iron, surfactants, naphtha, hexane and other light hydrocarbons	2019	21600	Reduce ion concentration, Total Suspended Solids (TSS) and Total Organic Carbon (TOC)	Motta Cabrera et al. (2021)
Bursa Organized Industrial Zone, Turkey	Pilot scale study of hot water recovery and reuse with ceramic nanofiltration in textile fatory	Printing and dyeing wastewater, high temperature wastewater	Turbidity, color, chemical oxygen demand (COD), suspended solids, conductivity, high pH, salinity	2020	Not specified	Remove COD, TOC, color, and hardness	Ağtaş et al. (2020)
Wuxi, China	A pilot MBR–NF for treating antibiotic production wastewater	Antibiotic production wastewater from pharmaceutical company	High concentrations of organic compounds: NH_4^+-N, spiramycin and new spiramycin	2015	0.52	Remove TOC, NH_4^+-N, TP	Wang et al. (2015)
Ohio, USA	Ovivo applies plate ceramic membrane from Japan in the largest MBR wastewater plant in the Canton, Ohio, USA	Municipal sewage treatment	The main pollutants are COD, ammonia nitrogen, total nitrogen, suspended solids, total phosphorus	2017	159000	Remove COD, ammonia nitrogen, total nitrogen, suspended solids, total phosphorus	Gwi (2017)
Saudi Arabia	A desalination plant in Saudi Arabia will apply CFM Systems® (flat sheet ceramic membrane)	Seawater desalination	High salt content, suspended solids, algae	2022	110000	The seawater pretreatment meets the requirements of stable reverse osmosis inflow	Li et al. (2020)
Japan	The longest running large-scale ceramic membrane based WTP in Japan, which is operational for >16 years	Water supply and treatment	/	2005	100000	The effluent meets the process operation requirements	Asif & Zhang (2021)
Singapore	Singapore PUB Choa Chu Kang Waterworks using ceramic membrane from Japan Metawater built the largest seawater desalination plant on earth. The ceramic membrane has an estimated service life of 20 years	Seawater desalination	High salt content, suspended solids, algae	2019	160000	Remove suspended solids and algae	Atkinson (2019)

and precipitation on the membrane surface and in the membrane pores due to mechanical action, eventually causing reduced or blocked membrane pores. The permeation flux reduces, and the separation characteristics deviate from the original design; this does not meet the production requirements. Fouling has been classified into different types. Reversible fouling is fouling that can be removed using physical backwashing, whereas irreversible fouling can only be eliminated using chemical cleaning (Kimura et al., 2006; Psoch and Schiewer, 2006).

2.1 Causes of ceramic membrane fouling

The most direct factors leading to membrane fouling are tied to properties of the filtration feed and the general performance of the membrane. The pollutants in the feed solution directly act on the membrane pore and membrane surface during filtration; this is an important factor influencing membrane pollution. In addition, the membrane's hydrophobicity, electrical status, and pore size directly affect the membrane fouling rate.

2.1.1 Substances that cause membrane fouling

When the type and nature of pollutants are used as the classification criteria, the substances that cause membrane fouling can be divided into three categories: organic, inorganic, and microbial pollutants (Asif and Zhang, 2021). Please see Fig. 2 for the detail classification.

2.1.1.1 Organic pollution

Organic matter in slightly polluted water sources can be further divided into natural organic matter (NOM) and synthetic organic chemicals (SOCs).

NOM refers to the macromolecular organic matter produced by the decay and decomposition of animals/plants in the natural cycle. This is the major organic component in surface water and groundwater. Natural organic matter in water sources usually exists in the form of suspended matter, colloidal matter, and dissolved matter. It affects not only the efficiency of water treatment processes, but also the color, odor, and regrowth of microorganisms in the membrane separation system. Notably, 85%–90% of NOM is the humic
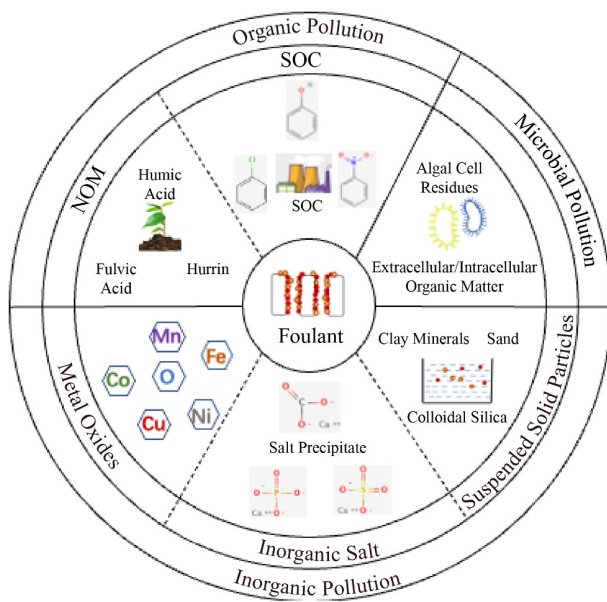
Fig. 2   Membrane foulant (source: original).

substance (HS), including fulvic acid (FA), humic acid (HA) and humin (HU) (Her et al., 2003). Humic acid, which has the highest proportion in humic substances, contains three organic functional groups: carboxyl group (COOH), hydroxyl group (OH), and carbon-oxygen double bond (C = O). It is a high molecular organic acid composed of aromatic hydrocarbons and various functional groups. It has good physiologic activity and functions such as absorption, complexation, and exchange. Furthermore, it is the main pollutant in the ultrafiltration process. During ultrafiltration experiments, Schäfer et al. (2000) observed that humic acid can cause a 78% decrease in membrane flux, while fulvic acid can only cause a 15% decrease in flux. Most SOCs consists of toxic and harmful organic substances; for example, phenols, nitrobenzenes, chlorobenzenes, phthalates, pesticides, polychlorinated biphenyls, polybrominated diphenyl ethers, etc. SOCs are commonly found in industrial wastewater, such as printing and dyeing wastewater, chemical wastewater, and oil wastewater. Wastewater from modern industrial production usually contains a large amount of SOCs, which is difficult to decompose. If the pretreatment is insufficient, it will cause serious fouling of the membrane.

Furthermore, parameters such as hydrophilicity, hydrophobicity, molecular shape, and the molecular size of organic molecules will affect the intensity of their pollution (Zhu et al., 2009). Cho et al. (2000) proved that hydrophilic organic components, mainly composed of non-humic acid components (mainly proteins, amino acids, and polysaccharides), can cause the irreversible pollution of ultrafiltration membranes. However, Gong et al. (2023) found the model and actual protein fractions only led to light membrane fouling, implying that proteins were not mainly responsible for membrane fouling at the

early stage of ceramic membrane microfiltration. Mustafa et al. (2016) systematically studied the influence of inorganic ions on DOM pollution of Ceramic membrane for the first time. In this study, surface facial mask contamination is determined by the whole membrane surface chemistry rather than the unique membrane hydrophilicity.

2.1.1.2   Microbial pollution

Microbial contamination is a type of organic contamination by definition. However, due to its complex nature and intractability, and to distinguish it from organic chemicals, most literature have classified microbial contamination separately and believe that biofouling is the most harmful to ceramic membranes, and it is difficult to control (Meng et al., 2017; Li et al., 2020; Asif and Zhang, 2021). Since the organic and inorganic substances accumulated on the surface of the membrane can provide a particular physicochemical and nutritional environment for the growth of microorganisms, such an environment is conducive to the reproduction of microorganisms. A large number of microorganisms are likely to form a layer of biofilm on the surface of the membrane, resulting in irreversible blockage and reduction in water flux.

Algae- and microbial-derived organic matter are endogenous organic pollutants. The extracellular organic matter, intracellular organic matter, and algal cell residues released by algae can cause severe membrane pollution (Qu et al., 2012). Chiou et al. (2010) further analyzed the functional groups on the surface of algal cells, and identified polysaccharides and amino sugars. In addition, membrane critical flux decreased as the concentration of the polysaccharides increased, indicating that polysaccharides are the major algal membrane components responsible for fouling.

2.1.1.3   Inorganic pollution

In water treatment, inorganic pollutants mainly include chemical pollution, solid particle suspended matter pollution, metal oxide pollution, and inorganic salt scale pollution caused by pretreatment. Inorganic fouling is also an important cause of membrane fouling, which can not only reduce the flux of the membrane, but also increase the pressure, reduce the quality of the membrane effluent, and even shorten the service life of the membrane.

In the water treatment process, in order to ensure the quality of water entering the membrane, some chemicals are added in the pretreatment, such as polyaluminum, polyiron, PAM, and other coagulants. A study showed excess aluminum-contained coagulant could cause an increase in inorganic membrane fouling (Gabelich et al., 2006).

Solid particle pollution refers to the presence of

undissolved suspended solids in water. These solid pollutants may be the solid particles contained in the raw water, the waste debris remaining in the pipeline engineering, or the filter material leaked from the pretreatment, etc. The leaked material would primarily be clay minerals, colloidal silica, metal oxides, and salt precipitates. Due to their large size, these pollutants end up trapped on the membrane surface to form membrane fouling or cause membrane damage during filtration (Mustafa et al., 2018).

In some instances, water can have a high hardness and contain cations with the potential to cause membrane fouling. Cations like metal ions could come from the iron or aluminum flocculants added during the coagulation or flocculation process, from the device pipelines and tanks due to corrosion or oxidization by the air (i.e., Ca^{2+}, Mg^{2+}, Ba^{2+}, etc). Anions are also included in this water (i.e., HCO_3^{-}, CO_3^{2-}, SO_4^{2-}, PO_4^{3-}, etc). As those cations/anions accumulate on the surface of the membrane, they can easily reach saturated solubility, forming inorganic salt scales. During the treatment of municipal daily wastewater, calcium carbonate, calcium sulfate, and barium sulfate are found on the membrane surface and result in membrane fouling (Xu et al., 2020). Calcium carbonate and calcium sulfate were also found on the membrane surface in the treatment of contaminated underground water and antibiotic pharmaceutical waste-water (Wei et al., 2009). Among them, calcium sulfate and barium sulfate are not easy to remove and need to be pre-treated by precipitation or pH adjustment. Otherwise, the pollution can become irreversible.

2.1.2 Influence of membrane properties on membrane fouling

Membrane properties also have a significant impact on membrane fouling. The pore size of the membrane determines its ability to retain substances. The substances which size is larger than the pore size of the membrane will be retained, and that size is smaller than the pore size of the membrane will generally pass through the pores of the membrane, but will also deposit in the pores of the membrane due to adsorption, resulting in pollution. At the same time, the electrical properties and hydrophilicity/hydrophobicity of the membrane will affect the adsorption capacity of the membrane to pollutants.

2.1.2.1 The relationship between membrane fouling and membrane pore size

The surface characteristics of the membrane and the relative size of the membrane pores are important factors that affect the mechanism and degree of fouling. For example, a ceramic membrane with the roughest surface and the largest pore size shows the most severe fouling in membrane bioreactor applications (Jin et al., 2010), in

which the membrane surface properties play a key role in membrane fouling. Huang et al. (2023) reached a similar conclusion in their study of relationship between ceramic membrane fouling and pore size. However, in their study, continually decreasing the ceramic membrane pore size could also lead to more severe membrane fouling in the membrane bioreactor. Huang and his team believe in optimizing pore size is a key factor to prepare ceramic membrane.

Stoller used a nanoparticle size analyzer to study the effect of particle size in water on membrane fouling (Stoller, 2009). The results showed that particulate pollutants with a ratio of particle size to membrane pore size between 0.1 and 10 can block membrane pores, whereas particulate pollutants with a ratio of more than 10 to the pore size of membrane pores are more likely to cause cake-filtration blocking.

2.1.2.2 Effect of membrane surface charge on membrane fouling

The chargeability of the membrane surface is an important factor in membrane fouling. By changing the chargeability of the membrane surface and the interaction between the membrane and the retained substances, the formation of membrane fouling can be effectively controlled, improving the permeability of the membrane. Moritz et al. (2001) studied the effect of charge properties of the membrane surface on the permeate flux; in this study, the charging properties of TiO_2, α-Al_2O_3, ZrO_2, γ-Al_2O_3 ultrafiltration membranes were characterized using the flowing potential method. This approach allowed for an investigation into the permeation flux of polyethylen-eimine and glucose under different pH values. The experimental results showed that the change of permeate flux depends on the charge on the membrane surface and the charged state of the particles in the solution. The same charge on the membrane and the particles in the feed solution can reduce the formation of a fouling layer on the membrane surface and improve the permeate flux of the membrane. If the particles in the membrane and the stock solution have different charges, when the stock solution passes through the membrane, it will be attracted by the charges outside the electric double layer, adsorbed and deposited inside the membrane pores, resulting in the pore size becoming smaller or blocked, culminating in membrane fouling (Wiesner et al., 1989).

2.1.2.3 The effect of membrane hydrophilicity/hydrophobicity on membrane fouling

The hydrophilic/hydrophobic properties of membranes are generally characterized by their contact angle. A contact angle > 90° and < 90° indicates that the membrane is hydrophobic and hydrophilic (Law, 2014), respectively. The smaller the angle, the better the

hydrophilicity and the higher the corresponding anti-pollution performance. Ceramic membranes are mainly prepared from metal oxides (i.e., Al_2O_3, TiO_2, ZrO_2, etc.). General ceramic membrane materials are hydrophilic materials. The performance of the ceramic membrane and the hydroxyl groups in the membrane's pores fuel its hydrophilicity. Modification of the membrane surface through an appropriate method can enhance the hydrophilicity of the membrane and improve the permeability and anti-fouling properties of the membrane (Gentleman and Ruud, 2010; Fan and Xing, 2013). Compared to those with polymeric membranes, anaerobic ceramic membrane bioreactors exhibited higher water flux, higher organic elimination, and lower membrane fouling. A significant amount of biomass was rejected; however, the highly hydrophilic surface of the Al_2O_3 ceramic membrane was primarily responsible for the reduction of membrane fouling, through inhibiting the hydrophobic attraction of membrane-foulants (Jeong et al., 2018). Fig. 3 summarized membrane properties that can effect membrane fouling.

2.2   Ceramic membrane fouling mechanism

2.2.1   Four types of membrane fouling mechanisms

When the aqueous solution containing different pollutants passes through the ceramic membrane, the pollutants will accumulate in the membrane pores or on the membrane surface due to various physical or physicochemical effects such as sieving, adsorption, hydrogen bonding, and electrostatic effects. The membrane fouling mechanism can be classified as cake-filtration blocking, complete blocking, standard blocking, and intermediate blocking (Asif and Zhang, 2021). Elucidating the

Fig. 3   Membrane fouling effected by membrane properties (source: original).

mechanisms of membrane fouling enables designers to effectively identify the leading underlying causes and deploy countermeasures to extend the service life of the membrane. The mechanisms are shown in Fig. 4.

2.2.1.1   Complete blocking

Complete blocking occurs when the size of the pollutants is bigger than or similar to that of the membrane pores, resulting in a blockage of the membrane pores.

2.2.1.2   Standard blocking

Standard blocking occurs when the size of the pollutants is smaller than that of the membrane pores, which leads to the pollutants accumulating in the membrane pores through adsorption, culminating in a decrease of the pore size and the permeation flux.

2.2.1.3   Intermediate blocking

Intermediate blocking occurs when the pollutants are larger than the pore size. This results in only a portion of membrane pores being sealed by the pollutants, while other pollutants accumulate on top of each other and do not completely block the membrane pores.

2.2.1.4   Cake-filtration blocking

Cake-filtration blocking occurs when pollutants accumulate on the membrane surface in a permeable cake layer. As the filtration process continues, the cake layer increases its thickness and becomes denser.

2.2.2   Theoretical models of ceramic membrane fouling

Theoretical models of membrane fouling are widely used to determine important parameters during membrane operation. These parameters will be used to determine the dominant membrane fouling mechanism. Researchers and manufacturers can formulate membrane fouling control guidelines using this data, thereby extending membrane life. The majority of theoretical models, including the combined pore blockage-cake filtration model, the laws of constant pressure filtration, the resistance-in-series model, and the unified membrane fouling index model, were created using organic membranes. Despite being originally developed for organic membranes, these models can be used for ceramic membranes (Lee et al., 2013). The differences of membrane materials cause variations in the dominant fouling mechanisms, even under the same operating conditions. Kim et al. (2020) used Hermia's model and found that the polymeric membrane was in proximity to all four models, including the complete pore blocking. The ceramic membrane had a greater propensity toward adhering to the cake filtration model when processing algae-containing waters. This
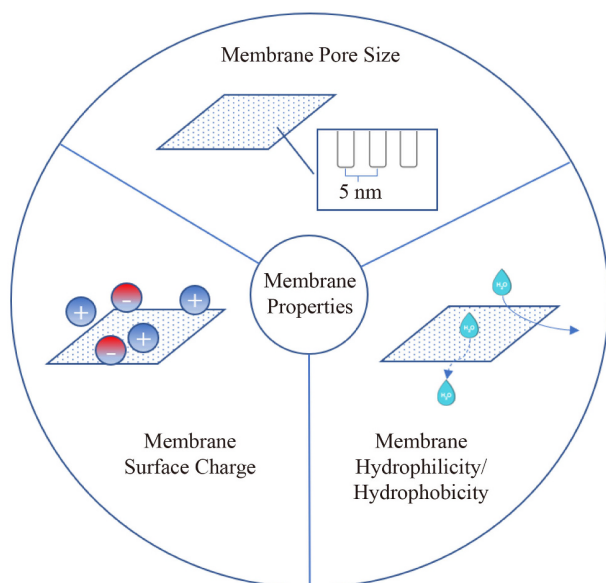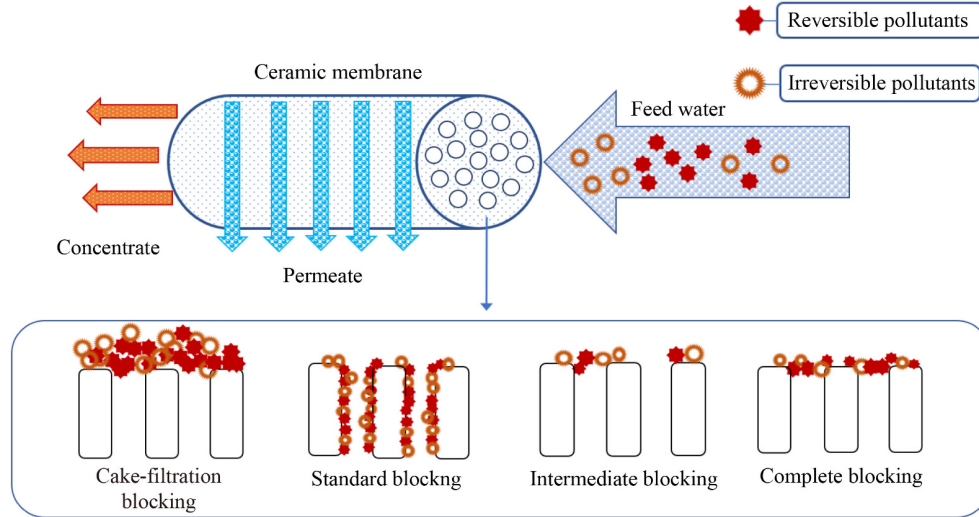
Fig. 4 Membrane fouling mechanisms (source: original).

could be attributed to the hydrophilicity of the polymeric membranes and the relative hydrophobicity of the ceramic membranes. Therefore, the knowledge of organic membranes will not be enough to explain some of these dynamics, making it necessary to further study the theoretical model of ceramic membrane fouling.

As ceramic membranes are used more extensively, the research on fouling of ceramic membranes in different scenarios is becoming more in-depth, and a large number of related models have been developed. The following subsection introduces only two theoretical models that are widely used in the study of ceramic membrane fouling.

2.2.2.1 Resistance-in-series model

Based on the blocking theory, the resistance of each part that may cause membrane fouling is analyzed to obtain the membrane fouling mechanism. The determination of membrane resistance can be calculated and analyzed according to Darcy's Law filtration model, and the membrane flux can be expressed as the ratio of driving force to resistance (Eq. (1)):

$$J = \frac{\Delta P}{\mu R_t}, \tag{1}$$

where J is membrane flux (m^3/(m^2·s)); ΔP is transmembrane pressure difference (Pa); μ is viscosity (Pa·s); and R_t is the total resistance at moment "t" (m^{-1}) (Eq. (2)).

$$R_t = R_m + R_{cp} + R_{pr} + R_{cr} + R_{if}, \tag{2}$$

where R_t is the total resistance; R_m is the intrinsic membrane resistance (m^{-1}); R_{cp} is the resistance due to concentration polarization (m^{-1}); R_{pr} is the cake and pore deposit resistance removable by backwash (m^{-1}), i.e., physically removable fouling; R_{cr} is the chemically reversible resistance (m^{-1}); and R_{if} is the chemically irreversible resistance (m^{-1}).

2.2.2.2 Hermia's Model

Hermia's Model is a blocking model based on four fouling mechanisms, which are used to evaluate membrane pollution caused by different water matrices with complicated compositions. The model is provided below (Eq. (3)):

$$\frac{d^2 t}{dV^2} = k \left(\frac{dt}{dV} \right)^n, \tag{3}$$

where t is the filtration time (s) and V is the total filtered volume (m^3); k and n are two model parameters with varying values or dimensions. In them, n characterizes the filtration model, with $n = 0$ for cake filtration, $n = 1$ for intermediate blocking, $n = 3/2$ standard blocking, and $n = 2$ for complete pore blocking. From constant pressure filtration tests, t and V are determined. The primary pollution process can be determined by plotting the data as d$^2 t$/dV^2 vs. dt/dV curves (Ho and Zydney, 2000).

This model's ability to precisely identify a number of idealized yet related fouling mechanisms (or non-ideal boundary conditions) that can be explained using straightforward mathematical expressions and summed using this straightforward equation is one of its distinguishing features (Huang et al., 2008).

2.2.2.3 Membrane fouling mechanisms under different application scenarios

Many studies on membrane fouling are based on the above two models. See Table 2 below.

3 Ceramic membrane fouling control

Controlling ceramic membrane fouling is an effective

way to prolong membrane life and reduce operating costs. The mitigation of membrane fouling in the membrane separation process can be improved in the following aspects: First, the properties of the feed solution can be changed and controlled through pre-membrane pretreatment, to improve the type, charge characteristics, and molecular structure of the organic matter contained in it, which can cause rapid membrane fouling. Second, membrane fouling can be alleviated by improving membrane hydrophobicity, roughness, surface potential, and catalytic performance through membrane modification. Third, the degree of membrane fouling can be reduced by exploring effective physical and chemical cleaning methods for membranes. Fourth, the concentration polarization on the membrane surface and the formation speed of the filter cake layer can be alleviated by adjusting the membrane operating parameters, such as flow rate, operating pressure, and operating temperature. The membrane filtration operation conditions can be improved by combining different processes, such as advanced oxidation and nanobubbles, to alleviate membrane pollution.

3.1 Pretreatment methods

To reduce the likelihood of pollutants attaching to and blocking membrane pores, different pretreatments can be used to improve the removal efficiency of pollutants, reduce their content, improve water quality, and alleviate ceramic membrane pollution. Fig. 5 shows the common pre-treatment of ceramic membrane.

For reducing the pollution and damage of the membrane caused by particulate impurities in the water, a membrane grid with a pore size of 0.1–0.5 mm can be set up in front of the membrane for interception, removing large particles from the water prior to the water being processed by the membrane. Coagulation process can effectively remove colloidal particles in water and reduce membrane fouling. When the mechanical cleaning was combined with pre-coagulation using PACl, the formation

of a fouling layer could be efficiently controlled (Sakamoto et al., 2022). The organic and inorganic substances in the water are adsorbed on the flocs to prevent their adsorption or precipitation in the membrane pores, resulting in the narrowing or blocking of the membrane pores.

In view of the influence of natural organic matter on membrane fouling, the use of an activated carbon adsorbent to absorb dissolved organic matter in water is also recommended. At the appropriate dosage, the adsorbent provides a new interface for the pollutants in the water, and the pollutants accumulate on the surface of the adsorbent through physical and chemical adsorption, thereby reducing the interaction of the pollutants with the membrane surface and reducing the membrane fouling load. Oh et al. (2007) devised an integrated strategy using activated carbon and ceramic membrane to treat polluted river water. The results showed that the water purification efficiency of the PAC-ceramic membrane integrated process is significantly higher than that of the traditional ozone-activated carbon process. The removal rates of UV_{260} and DOC are 90.3% and 80.2%, respectively. The production potential of halomethanes decreased to below 0.5 mg/L and 8.8 μg/L, respectively, and neither bacteria nor virus was detected. Konieczny and Klomfas (2002) showed that both granular activated carbon (GAC) and PAC adsorption pretreatment delayed the decline of ceramic membrane flux, and the total fouling resistance, reversible and irreversible fouling resistance of ceramic membranes were reduced to varying degrees.

In recent years, many studies on pre-ozonation before membranes have demonstrated that pre-ozone can use the strong oxidizing properties of ozone to inhibit the growth of microorganisms in water and reduce the pollution of ceramic membranes; or change the nature of organic pollutants, causing the macromolecular organic matter fouled by the membrane to be oxidized and degraded into small molecular substances, thereby alleviating the organic pollution of the ceramic membrane. Cheng et al. (2016) studied the effect of ozone pre-oxidation on

Table 2 Membrane fouling mechanism models under different water

| Membrane type	Treated water	Model type	Research background	Reference
Ceramic	Seawater	Hermia's Model	Membrane fouling is controlled by a combination of several blocking mechanisms, with cake-filtration blocking contributing the most to the reduction of permeation flux. And with the increase of membrane surface flux, the decrease of membrane flux tends to slow down.	Ma et al. (2010a)
Ceramic	Simulated surface water	Hermia's Model	Membrane surface retention and membrane pore adsorption were the mainly removal routes. The first and third stages of membrane fouling mainly caused by complete blocking, and the second stage was mainly controlled by standard blocking. The study found that humic acid would cause both the pore blocking and the fouling of the membrane surface when turbidity was present.	Yang et al. (2021)
Organic	Drinking water	Resistance-in-series model + Hermia's Model	A combination of pore-blocking and cake-filtration blocking is the main mechanisms responsible for membrane fouling.	Xing et al. (2021)
Organic	High alkalinity organic wastewater	Resistance-in-series model + Modified Hermia's Model	In membrane fouling in high salinity organic wastewater, Al^{3+} is closely related to irreversible membrane scaling.	Cai et al. (2022)
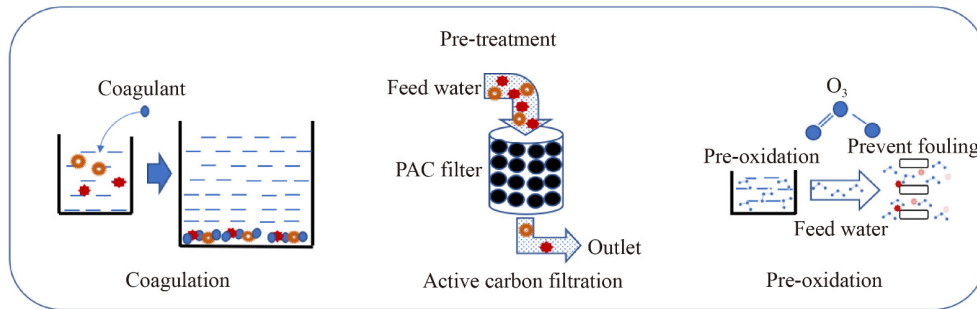
Fig. 5   Pre-treatment.

organic matter fouling of UF ceramic membranes. It was found that under lower ozone dosage, the molecular mass distribution of humic acid and sodium alginate gradually shifted to the low molecular mass range. Membrane fouling was effectively reduced, but the concentration of organic matter in the effluent increased significantly. Song et al. (2018) also found that pre-ozonation with 1 mg/L and 2 mg/L O_3 reduced membrane fouling by 33% and 55%, respectively, in synthetic water containing bovine serum albumin, respectively.

In terms of preventing membrane inorganic fouling, pretreatment is the most researched approach. Chemical methods such as alkali precipitation, adding scale inhibitors, and ion exchange can be used; additionally, physical methods such as water electrolysis, magnetization, ultrasonic waves, and radio frequency irradiation can also be used (Baker and Judd, 1996; Khalil et al., 1999).

3.2   Membrane modification

Improvement of membrane performance can also be achieved through the regulation of the membrane production process. Modification of membranes can alter membrane properties such as surface roughness, hydrophilicity, hydrophobicity, and surface potential, which in turn affect membrane fouling. The application of nanoparticles to the modification of inorganic ceramic membranes can provide a high degree of control over membrane fouling and yield desirable structural properties. Fig. 6 summarized the ceramic membrane modification and the outcome.

Firstly, studies have shown that increasing the membrane surface roughness will increase the adhesion rate of membrane pollutants on the membrane surface and thus aggravate membrane fouling. Therefore, the membrane surface roughness can be reduced through membrane modification to slow down the membrane fouling. At present, the modification methods of inorganic ceramic membranes mainly include the dip coating method, pulsed laser deposition method, layer-by-layer method, and vacuum filtration (Lu et al., 2016; Guo et al., 2018).

Secondly, membrane fouling can be controlled through

the preparation of modified materials. Combining an advanced oxidation process with catalytic ceramic membrane filtration modified by nano metal oxide particles can catalyze the oxidation of organic membrane pollutants on the membrane surface (Kim and Van Der Bruggen, 2010). There are still some technical difficulties to be overcome in modifying ceramic membranes by nano-metal oxide particles, and the problem of loading and fixing the nanoparticles used for modification on the ceramic membrane remains to be solved. Using linker molecules or organic binders to form chemical bonds between nanoparticles and ceramic membrane supports can somewhat solve this problem. For example, amino groups can be embedded on the membrane surface to form covalent bonds with silver atoms to achieve membrane modification (Lv et al., 2009). Ceramic membranes can be immersed in hydroxyapatite (HAP) sol for uniform distribution to stabilize TiO_2 nanoparticles for modification (Ning et al., 2009). The modification of ceramic membranes via the sol-gel method can also solve the adhesion problem of modified nano-metal oxide particles on the membranes. Byun et al. (2011) used the catalytic properties of metal oxides to load iron oxide and manganese oxide nanoparticles on the surface of a UF ceramic membrane by layer-by-layer coating and sintered at a high temperature to make a modified catalytic ceramic membrane. Compared with the unmodified ceramic membrane (the filter layer is made of titanium dioxide), it is found that the flux recovery and TOC degradation of the manganese oxide modified ceramic membrane is significantly better than that of the iron oxide modified when ozone and ceramic membrane are used to treat surface water, and the number of coating layers has a certain influence on the catalytic performance. Ma et al. (2010b) prepared silver-titania/hydroxyapatite/alumina photocatalytic ceramic membranes. Under UV light irradiation, the removal rate of humic acid and the anti-pollution performance of the catalytic ceramic membranes were effectively improved. When treating surface water, the removal of trace organic pollutants is apparent, and the membrane flux is also improved.
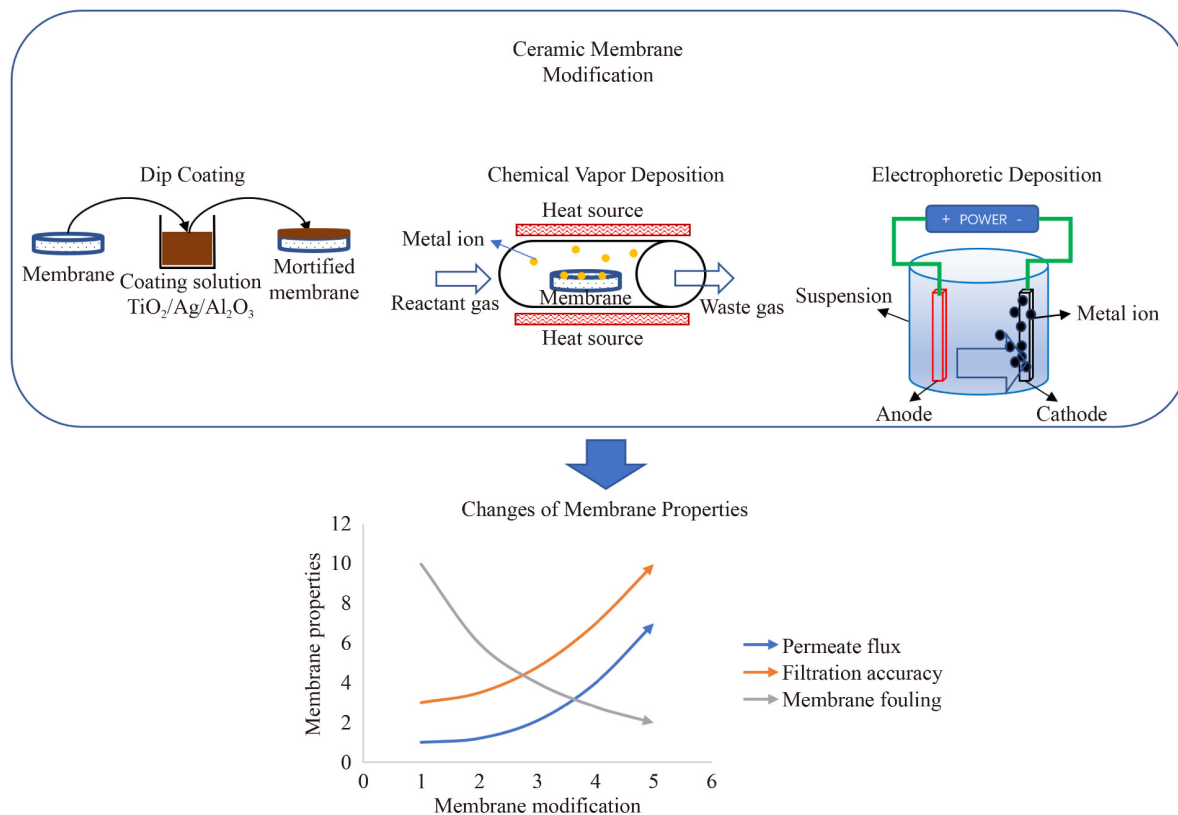
Fig. 6   Ceramic membrane modification and outcome (Source: original).

3.3   Membrane cleaning

Membrane cleaning refers to the process of separating pollutants from the membrane surface and restoring membrane flux. To prevent long-term pollutants from chemically interacting with the membrane, which will shorten its service life and reduce the high operating cost caused by frequent membrane replacement, the ceramic membrane must be cleaned regularly. The cleaning methods can be divided into categories: physical and chemical cleaning.

3.3.1   Physical cleaning

Physical cleaning generally refers to the removal of contaminants by hydraulic or mechanical means (Weerasekara et al., 2014; Marti-Calatayud et al., 2018). Physical cleaning is a cleaning method that does not use any chemicals to peel off pollutants on the membrane surface. It mainly removes the reversible pollutants (i.e., colloidal particles, suspended solids, etc.) adsorbed on the membrane surface and membrane pores by physical methods. The primary advantages of the physical method are that it does not introduce new pollutants and that it has simple cleaning steps.
    Common ceramic membrane physical cleaning

methods are as follows: backwash, gas-liquid mixing vibrating cleaning, mechanical cleaning, ultrasonic cleaning, and electric cleaning (Zhou et al., 2010).

3.3.1.1   Backwash

During filtration through the ceramic membrane, bacteria, organics, suspended solids, and other impurities are trapped. These gather on the membrane surface. After a period of time, these impurities affect the membrane performance, cause serious membrane pollution, and even affect the service life of the membrane. Backwash can effectively remove the impurities in the membrane and ensure the stability of membrane operation. The backwash liquid generally adopts the product water of ceramic membrane or water of better quality (NF/RO product water).

3.3.1.2   Gas-liquid mixing vibrating cleaning

Blowing compressed air into the inner cavity of the membrane module, along with the permeate from the backwash, makes the air bubbles and water flow oscillate on the surface of the ceramic membrane, shaking off or washing away the contaminants attached to the outer surface of the membrane (Ullah et al., 2021).

3.3.1.3 Mechanical cleaning

The tubular ceramic membrane module can use soft foam plastic balls and sponge balls to clean the inner pressure membrane tube and mechanically remove pollutants. This method is suitable for the cleaning of membrane surfaces with organic colloids as contaminants.

3.3.1.4 Ultrasonic cleaning

Ultrasonic cleaning entails using ultrasonic waves to generate turbulence, cavitation, and vibration in water to remove membrane pollution. This approach is effective and fast. Ultrasonic cleaning of the membrane is effective in certain cases, especially for objects that do not meet the requirements for conventional cleaning methods and have complex geometric shapes (Qasim et al., 2018).

3.3.1.5 Electric cleaning

Electric cleaning implies applying an electric field on the ceramic membrane to move charged particles or molecules in the direction of the electric field. Applying an electric field within a certain time interval without interrupting the operation enables removing particles or molecules from the interface (Anis et al., 2022). This reduces the concentration polarization and increases the flux of the ceramic membrane. However, this method has a limitation; it requires the use of conductive films and special membrane devices equipped with electrodes.

3.3.2 Chemical cleaning

Physical cleaning cannot fully restore the membrane flux, and this approach is only effective in the initial stage of contamination of the ceramic membrane. Beyond this stage, chemical cleaning is required to remove irreversible pollution. The chemical cleaning entails using chemical agents to remove pollutants. This approach can effectively remove the gel layer and pore-blocking pollution (Garmsiri et al., 2017). Generally speaking, chemical cleaning mainly relies on the interaction between chemical cleaning agents and pollutants to remove pollutants from the membrane surface (Garmsiri et al., 2017). In this approach, the effectiveness of the cleaning agent is important, but its chemical stability,

cost, and safety must also be considered. Cleaning agents can affect the presence of contaminants on the membrane surface in different ways, resulting in changes in the morphology of the contaminants (i.e., swelling and compaction) or changes in the surface chemistry of the deposits to remove contaminants from the membrane. Possible reactions between contaminants and cleaning agents include hydrolysis, peptization, saponification, dissolution, dispersion (suspension), and chelation (Madaeni et al., 2001; Weis et al., 2003). Commonly used cleaning agents are acids, alkalis, oxidants, surfactants, chelating agents, etc. (Al-Amoudi and Lovitt, 2007). Table 3 shows the types of chemical cleaning agents and their functions.

Membrane cleaning should not only effectively remove membrane pollutants and restore flux, but also prevent damage to the membrane itself. Cleaning is generally performed when the membrane performance is lower than the expected permeability, typically around 10%, or when the feed pressure is increased by about 10%, and the differential pressure is increased by 15%–50%.

3.4 Influence of ceramic membrane structure, design, and operational parameter optimization on membrane fouling

In addition to the above factors that affect membrane fouling, the structural design of ceramic membrane modules and the optimization of operating parameters can also help delay membrane fouling.

3.4.1 Influence of ceramic membrane structure on membrane fouling

Bai et al. (2018) studied the influence of flow rate and pressure distribution on the flux of each component of the ceramic membrane equipment through experiments; the results of these experiments were used to perform CFD calculations. The components were optimized by widening the inlet or introducing a reverse flow baffle in the liquid accumulation chamber; this was done so that the hydraulic distribution was more uniform and the dead angle was reduced. Using turbulence promoters and properly designed flow channel structures can increase the flow rate of ceramic membranes, reduce the thickness of the boundary layer, and delay membrane fouling.

Table 3 Types of chemical cleaning agents and their functions (Gruskevica and Mezule, 2021; Ullah et al., 2021)

Types	Specific agents	Cleaning principle
Acids	strong acid: HCl, HNO$_3$ weak acid: H$_3$PO$_4$	Dissolves inorganic precipitates, acidifies macromolecular hydrolyzed acids, adjusts pH
Alkalis	strong alkali: NaOH, KOH weak alkali: Na$_2$CO$_3$	Hydrolysis and dissolution of organic pollutants, alkaline protein hydrolysis, pH adjustment
Oxidizing agents	NaClO, H$_2$O$_2$, O$_3$	Oxidize organic matter, sterilize
Surfactants	SDS, CATB	Dispersed/suspended sediment
Chelating agents	EDTA	Suppresses the catalytic decomposition of metal ions

3.4.2 Influence of operating parameter optimization on membrane fouling

When the ceramic membrane equipment is filtering the filter material under different operating parameters, the pressure difference caused by the membrane fouling increases, and the membrane cleaning cycle is also different. To slow down the membrane fouling rate, generally, by increasing the membrane cross-flow flow rate and increasing the mass transfer coefficient, the trapped solute can be taken away by the water flow in time, and the formation of concentration polarization on the membrane surface can be delayed. Another option is adjusting the operating pressure to prevent increase in the thickness and density of the precipitation layer. In addition, appropriately increasing the water temperature of the feed liquid, accelerates molecular diffusion and increases the filtration rate; in addition, reducing the pressure difference on both sides of the membrane or the concentration of the feed liquid can alleviate concentration polarization.

3.5 Synergistic antifouling technologies

3.5.1 Oxidation by O_3

Ozone is a strong oxidant, in addition to being used as a disinfectant and improving the biodegradability of organic matter, it can also be combined with the water treatment membrane filtration process to reduce membrane fouling in the treatment of drinking water (Schlichter et al., 2004; Zhang et al., 2013). Ceramic membranes have strong chemical stability and can withstand severe oxidative stresses in the presence of ozone and hydroxyl radicals (Karnik et al., 2005). Studies have shown that the integrated process of ozonation and ceramic membranes effectively mitigated algal enrichment membrane fouling in the treatment of drinking water (Yu et al., 2016). Ozone can not only effectively oxidize refractory organic pollutants, but it can also facilitate sterilization, decolorization, and deodorization of membrane effluent. In the case of inorganic ceramic membrane filtration coupled with ozone oxidation, a higher permeation flux was observed. *In situ* ozonation can alleviate membrane fouling when the ozone dosage is 2.0–2.5 mg/L; under the test conditions, the working cycle time of the membrane can be doubled (Zhang et al., 2013). Kim et al. (2008) studied the effects of ozone dosage and hydraulic conditions on the flux of UF ceramic membranes. The study showed that the higher the cross-flow rate on the membrane surface, the greater the ozone dosage and the smaller the transmembrane pressure difference. The slower the membrane flux declines, the lighter the fouling.

3.5.2 Advanced oxidation process

Scholars have also conducted research on advanced oxidation process pretreatment, such as ultraviolet/ hydrogen peroxide (UV/H_2O_2). Zhang et al. (2014) used UV/H_2O_2 oxidation pretreatment to alleviate the fouling of ceramic membranes caused by algal organic matter. The study found that compared with ACH coagulation pretreatment, UV/H_2O_2 pretreatment exerts a similar mitigation effect on the total fouling resistance of the membrane. Irreversible pollution is more serious, mainly because the small molecular organics generated during the oxidation process will block the membrane pores, resulting in more extensive and irreversible pollution. However, UV/H_2O_2 oxidation pretreatment removes microcystins, while coagulation pretreatment has been limited in this regard. Oxidation pretreatment significantly improves the removal of disinfection by-product precursors and trace organic pollutants in water and can alleviate membrane fouling to a certain extent.

3.5.3 Nanobubble technology

An air nanobubble (NB) with a diameter of less than 200 nm has a relatively long life in liquids. It can effectively reduce membrane fouling because introducing a gas-liquid two-phase flow enhances the surface shear rate and creates better fluid dynamics near the membrane surface. Air nanobubble can effectively reduce membrane pollution owing to its high mass transfer efficiency. The most recent water treatment method that efficiently prevents or reduces membrane fouling is membrane filtration combined with air NB (Wu et al., 2008). NB significantly cleans the membrane, with very few negative effects, by feeding air NB into the membrane holder. Consequently, air NB could be regarded as a membrane cleanser. Reduced utilization of chemicals like antiscalants is the main economic benefit of air NB applications. Furthermore, membrane life-cycles can be increased by feeding air NB into the membrane, thus reducing the cost of water treatment (Ghadimkhani et al., 2016).

4 Conclusions

Membrane fouling is unavoidable; however, it can be controlled by collectively improving the pre-treatment process, membrane modification, membrane cleaning, structural design of membrane modules, and optimizing operating conditions with anti-fouling technologies to extend the service time of ceramic membrane as much as possible. Current research on ceramic membrane fouling mainly focuses on organic fouling, but little is known about biological fouling and the interaction between biomass and membranes. Therefore, future studies should concentrate on elucidating the biofouling of ceramic membranes and developing practical defenses for long-term use.

To prevent membrane pollution, the control technology must be integrated with other aspects, such as low energy consumption, drug consumption, and cost. In addition to improving existing membrane modifications, the development of new membrane materials is a worthwhile research effort. For example, if the antibacterial and anti-fouling properties of the membrane can be improved, the degree of membrane fouling can be greatly reduced. If the acid and alkali resistance of the membrane can be improved, the damage of the membrane during chemical cleaning can be reduced. The above measures are of great significance to control membrane fouling and reduce operating costs.

With the development of new technologies, the pollution problem associated with ceramic membranes will eventually be addressed in a more effective manner, allowing for ceramic membranes to play a more critical role in water treatment in the future.

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