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A Mini Review on Recent RO Membrane Surface Modification Methods for Different Purposes

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Freshwater scarcity is becoming increasingly widespread, particularly in coastal countries and territories. Many advanced technologies have been introduced to desalinate seawater into potable water, including multi-effect distillation (MED), multi-stage flash distillation (MSF), vapor compression distillation (MVC), and membrane filtration technologies, such as reverse osmosis (RO), nanofiltration (NF), and electrolysis (ED), which allow water to pass through while retaining salt and other minerals. RO filtration technology is commonly used owing to its advantages, such as high salt removal ability, wide application range, and high-purity output water. The different water qualities and operating conditions cause the filtration performance and lifespan of RO membranes to decrease gradually over time. This mini review provides an overview of the different methods for modifying RO membrane surfaces for various purposes, including fouling resistance, chlorine resistance, improved separation performance, and microbial resistance, to prolong the lifespan and separation performance of RO membranes. The technical advantages and limitations, as well as the feasibility of RO membrane surface modification methods that can be scaled up on an industrial scale, are clarified. Finally, prospects and research gaps in RO membrane surface modification methods are discussed to guide further research on RO membranes.

1. Introduction

Reverse osmosis (RO) technology is being progressively applied in the water treatment industry, especially for filtering salty water into potable or domestic water. A spiral-wound RO module is composed of curled filter-membrane clusters. Each filter membrane assembly consisted of thin composite membranes made of polyamide, permeating carrier material, and feed water carrier stacked on top of each other and wrapped around a central clean water recovery tube. The outer surface of the filter core is covered with a thin layer of plastic film or a mixture of epoxy resin and glass fiber to protect and enhance the mechanical properties of the filter core. The RO process begins when pressurized water is forced through a semipermeable membrane. This membrane has extremely small pores, typically approximately 0.0001 µm in size. These pores are sufficiently small to allow water molecules to pass through but effectively block larger contaminants such as salts, minerals, and other impurities (Ochando-Pulido et al., 2017). Polyamide films are commonly synthesized on polysulfone support substrates on non-woven layers. Interfacial polymerization is an important step in the synthesis of thin-film composites, occurring at the interface between the support substrate and active layer. The m-phenylenediamine (MPD) solution is poured onto the polysulfone support layer to form a thin layer before polymerization, with the trimesoyl chloride (TMC) solution disperse in an organic solvent (hexane or heptane). The reaction between monomers leads to the formation of a thin polyamide (PA) layer, which serves as the active layer of the RO membrane, and durability is one of the top criteria of concern.
Modifying the surface of the active layer prevents the deposition of contaminants on the membrane surface, enhances filtration performance, and increases the chlorine resistance and antibacterial ability of the RO membrane, while still controlling membrane surface properties, such as separation performance and water flux. Many studies have described diverse techniques for surface modification of PA membranes using chemical and

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physical agents (surface coating, deposition, and grafting). The technical limitations of the RO membrane manufacturing process and possible methods to overcome these limitations have not been clearly stated. Many mechanisms have been reported to improve the fouling resistance of RO membranes, such as the modification of the substrate (before interfacial polymerization), integration of additives (during interfacial polymerization), and modification of the membrane surface (post-interfacial polymerization) for inorganic, organic, colloidal, and biological fouling (Maria da Gloria et al., 2019). Among them, post-surface modification using inorganic nanomaterials such as Ag, Cu, SiO_2, TiO_2, graphene oxide nanosheets, organic polymers, and combining of various materials has been widely used.

In this mini review, recent trends in RO membrane surface modifications for various purposes are listed, including antifouling, chlorine resistance, enhanced filtration performance, and antibacterial properties to prolong the lifespan of RO membranes. Technical limitations and advantages can be exploited to modify RO membranes.

2. RO membrane surface modification methods

2.1 Fouling resistance

One common method to improve the fouling resistance of RO membranes is to reduce the interaction between dirt and the surface of PA layer by reducing the surface negative charge of PA layer. A summary of recent studies on membrane surface modification methods with different agents for enhancing the antifouling of RO membranes is presented in Table 1. Zwitterions (also known as dipolar ions) of SO_3^- and COO^- incorporating Ag^+ ions (zwitter-Ag) have been used by Yi et al. (2019) for enhanced fouling resistance of RO membranes. This molecule contains functional groups with both positive and negative charges, the overall molecule is electrically neutral. The water contact angle of the thin-film composite (TFC) after zwitterion treatment decreased significantly, indicating that the surface became more hydrophilic, leading to an increase in the permeability of the membrane surface. Implantation of the zwitter-Ag enhances the negative surface potential of the polyamide layer. This conflicts with previous reports of improved fouling resistance because of the enhanced negatively charged surface, which increased the fouling interactions. This work also showed excellent resistance to 1,000 ppm bovine serum albumin (BSA) at pH 7.4, and resistance to *E. coli* and *S. aureus* bacteria. Surface modification using this method showed that separation performance did not change significantly (Yi et al., 2019). Park et al. used silane agents, including 3-aminopropyltrimethoxysilane (APTMS), chlorotrimethoxysilane (CTMS), and 3-mercaptopropyltrimethoxysilane (MPTMS) for coating on RO membrane surface by sol gel process to enhance antifouling. This modification resulted in the higher antifouling of the RO membrane to casein owing to the hydrophilic and uncharged surfaces. The flux recovery ratio (FRR) of the original RO membrane was 75.5 %, increasing in modified RO membrane with APTMS, CTMS, and MPTMS were 98, 92.2, and 88.3 %. The salt rejection rate of all membranes was still above 97.4 % (Park et al., 2019). This approach is considered feasible when commercialized, with chemicals that are easy to find, popular and a simple but highly effective process.

A novel modification method, using layer-by-layer (LBL) engineering to increase RO membrane fouling resistance with sodium alginate/Cu^{2+} hydrogel by Xia et al. Owing to the excellent hydrophilicity and the controllable thickness of hydrogel layer in LBL method, water molecules can pass through easily. RO membrane has a water flux of 71.40 LMH, higher than pristine RO membrane with 192.97 % while still retaining the ability to remove salt. The flux recovery rates of the modified RO membrane in the antifouling test are 26.44 % for BSA, and 29.14 % for dodecyl trimethyl ammonium bromide (DTAB), which is enhanced compared to unmodified RO. Furthermore, combining with sodium alginate/Cu^{2+} hydrogel, RO membrane also demonstrated bactericidal activity against gram-positive *S. aureus* and gram-negative *E. coli* (Xia et al., 2020). Sodium alginate-based hydrogels are often reported to have the ability to swell under environmental conditions such as temperature and pH. The suitability of this material in RO membranes should be considered, especially the mechanical strength that must be ensured under the operating conditions of the RO membrane.

Kavaiya et al. reported RO membrane surface modification using the organic polymers, poly (ethylene glycol) diacrylate (PEGDA) and ethylene glycol dimethacrylate (EGDMA). For the PEGDA modification, the maximum permeation flux of 48.7 %, and salt rejection increased to 3.43 %. For the EGDMA modification, the maximum permeation flux and salt rejection increased by 46.13 % and 3.08 %, compared to the unmodified RO membrane. The presence of poly (ethylene glycol) chains on the membrane surface creates a more hydrated and slippery surface, discouraging the adhesion of foulants such as organic matter, salts, and particulates. Fouling of the PEGDA-treated RO was significantly lower, and the removal efficiencies for zinc and chromium were also higher. EGDMA can crosslink with other polymer chains when exposed to suitable conditions (e.g., UV light or heat), forming a network structure on the membrane surface (Kavaiya et al., 2022). This crosslinked network can provide mechanical stability and chemical resistance to the modified membrane while also affecting its surface

properties. Modification of the RO membrane surface by cross-linking increases its selectivity for certain metal ions.

Table 1: Summary of surface modification methods of RO membrane for antifouling

Modification materials	Modification methods	Key results	Refs.
Zwitter-Ag complexes	Surface grafting	- Enhanced surface charge to 68-71 %; - Decreased water contact angle from 62.5 º to 45 º, more hydrophilic; - Lower fouling adsorption of BSA (1,000 ppm, pH = 7.4), improved resistance of *E. coli* and *S. aureus;*	(Yi et al., 2019)
APTMS, CTMS, MPTMS	Sol–gel process	- Higher of water fluxes and salt rejection. - Lower water contact angles of APTMS- and CTMS-RO with 36.08 º and 45.94 º, compared to pristine RO of 50.31 º but higher with MPTMS-RO of 71.52 º; - Higher surface charge of APTMS- and CTMS-RO, more negative charge of MPTMS-RO; - Enhanced water fluxes of APTMS-RO, decreased for CTMS-, and MPTMS-RO; - Improved fouling resistance to casein, less change of salt rejection; - Higher FRR of modified RO compared to pristine RO.	(Park et al., 2019)
Sodium alginate/Cu^{2+} hydrogel	Layer-by-layer	- Reduced water contact angle, more hydrophilic; - Increased water flux to 71.40 LMH, 192.97 % higher than pristine RO; - Unchanged salt rejection; - Higher FRR of the modified RO with 26.44 % for BSA, and 29.14 % for DTAB; - Modified RO had a potential against *E. coli* and *S. aureus* bacteria.	(Xia et al., 2020)
PEGDA, EGDMA	Surface crosslinking	- Increased water fluxes of PEGDA- and EGDMA-RO with 48.7 % and 46.13 %; - Decreased water contact angle of PEGDA-RO from 61 º to 36.63 º; - Increased salt rejection of PEGDA- and EGDMA-RO with 3.43 % and 3.08 %.	(Kavaiya et al., 2022)

2.2 Chlorine resistance

The major challenge for RO membranes is not only maintaining membrane properties but also being susceptible to fouling and chlorine attack over time during operation. Exposure to chlorine can cause deterioration of RO membranes, which can react with the polymer matrix of the membrane, leading to chemical degradation, loss of mechanical strength, and increased susceptibility to fouling and scaling. Most RO membranes are designed to resist low moderate chlorine concentrations commonly found in municipal drinking water supplies (typically less than 1-2 ppm). A summary of the different surface modifications used for chlorine resistance is presented in Table 2.

Wang et al. developed a nanocomposite RO membrane incorporated with two-dimensional MXene $Ti_3C_2T_x$ via embedding and interfacial polymerization. The results indicated that water permeability increased to a maximum value of 2.53 $L.m^{-2}.h^{-1}.bar^{-1}$, and the salt rejection reached 98.5 %. The $Ti_3C_2T_x$ modified RO membrane exhibited a high salt rejection of 97.1 % in a chlorine resistance test at 10,000 ppm·h owing to the surface formation between the functional groups of $Ti_3C_2T_x$ and active chlorine, which effectively prevented chlorine attack on the RO membranes (Wang et al., 2020). The RO membrane is also combined with β-cyclodextrin-activated graphene oxide nanosheets (β-CD-f-GO) by Matshetshe et al. (2021), forming via amide bond between ethylenediamine-grafted β-cyclodextrin and graphene oxide. The β-CD-f-GO-modified RO membrane outperformed the unmodified RO membrane in terms of water flux and antibacterial activity, by 38 % and 45.9 %. The nanocomposite membrane also exhibited antifouling properties with a lower rate of 26.2 % and a higher

flux recovery rate of 88.3 %, compared to pristine RO membranes of 46.8 % and 74.1 % (Matshetshe et al., 2021). These methods open new approaches for the surface modification of RO membranes to improve the membrane lifespan under operating conditions with high free chlorine concentrations. The process of incorporating $Ti_3C_2T_x$ or β-cyclodextrin-decorated graphene oxide into the RO membrane must ensure uniform dispersion in the MPD solution before interfacial polymerization. The cost of synthesizing and integrating these materials into a membrane can significantly increase the overall manufacturing cost of RO membranes. The stability and long-term durability of this modified membrane under operating conditions require careful consideration of factors, such as additive leaching, chemical bond stability, and performance variability, which can affect the reliability and longevity of the membrane.

Table 2: Summary of surface modification methods of RO membrane for chlorine resistance

Modification materials	Modification methods	Key results	Refs.
MXene $Ti_3C_2T_x$	Embedding/ Interfacial polymerization	- Increased water flux from 1.74 to 2.53 ($L.m^{-2}.h^{-1}.bar^{-1}$); - Unchange salt rejection; - Lower flux decline value of 11.11 %; - Higher chlorine resistance.	(Wang et al., 2020)
β-CD-f-GO	Embedding/ Interfacial polymerization	- Decreased water contact angle, enhanced water flux of 38 %; - Unchange salt rejection significantly; - Higher fouling resistance of BSA and chlorine resistance; - Increased against *E. coli* bacterium to 45.9 %.	(Matshetshe et al., 2021)

2.3 Performance enhancement

Different surface modification methods for the RO membrane to enhance the separation performance are listed in Table 3. Seyyed Shahabi et al. reported a modification method for RO membrane, using hydrophilic graphite carbon nitride nanosheets (g-C_3N_4) with functional groups OH, COOH and SO_3H, resulted in a smoother RO membrane surface (82 %). The addition of functionalized g-C_3N_4 nanosheets significantly enhanced the permeability of the RO membrane while maintaining the separation performance, with a maximum pure water flux of 91.8 $L{\cdot}m^{-2}{\cdot}h^{-1}$, 54 % higher than that of the RO membrane combined with g-C_3N_4 nanosheets without functionalization (Seyyed Shahabi et al., 2020). The limitation of this method is complex, using sulfuric acid in the synthesis process has many limitations when applied on an industrial scale.

Khoo et al. reported a novel modification of RO surface membranes by plasma-enhanced chemical vapor deposition (PECVD) to enhance fouling resistance and desalination performance. The PA layer surface was treated with aniline monomer and oxygen gas (O_2) for 15 s and 60 s. The water permeability of the RO membrane modified with aniline (PANI) was 5.57 $L{\cdot}m^{-2}{\cdot}h^{-1}{\cdot}bar^{-1}$, and the RO membrane modified with O_2 exhibited a higher water permeability of 6.64 $L{\cdot}m^{-2}{\cdot}h^{-1}{\cdot}bar^{-1}$. After plasma treatment with O_2, the water contact angle decreased from 88.39 ° to 79.46 °, salt rejection increased by 4.2 % for NaCl and 2.6 % for Na_2SO_4. In addition, the O_2 modified RO membrane has a high flux recovery rate of 96 % and antifouling ability of 76.5 %, which is higher than that of the original RO membrane owing to the reduction of polar groups on the surface of the conductive PA layer to reduce membrane resistance (Khoo et al., 2021). The drawback of this method is that it requires a complex reactor and consumes considerable energy. In addition, aniline has been reported to adversely affect human health and the environment.

Shi et al. used organic agents such as dimethyl carbonate (DMC) and tannic acid (TA), that were combined to fabricate high-performance RO membranes. The addition of DMC enhanced the leaf structure on the surface of PA and TA caused the PA active layer to become denser and thinner. RO membranes modified with TA and DMC (TA-RO@DMC) have higher filtration performance, compared to pristine RO membranes, TA alone (TA-RO), or DMC (RO@DMC). Modification with TA and DMC gave the RO membranes a high salt rejection of 99.03 % and a water flux of 64.2 ± 1.8 $L.m^{-1}.h^{-2}$ at 15.5 bar. In addition, chlorination was used to enhance the functionality of the membrane. After chlorination, TA-RO@DMC showed more excellent performance than RO@DMC due to the PA active layer being more compact, which reduced the impact of free chlorine on the PA surface. After chlorination, TA-RO@DMC further enhanced water flux and salt rejection to 90.5 $L.m^{-1}.h^{-2}$, and 99.25 % (Shi et al., 2020). The method has outstanding advantages when using clean, easy-to-find solvents and simple conversion processes, and the synthesized RO membranes have a separation efficiency of more than 99 %.

Table 3: Summary of surface modification methods of RO membrane for performance enhancement

Modification materials	Modification methods	Key results	Refs.
OH, COOH and SO_3H functionalized g-C_3N_4	Incorporating/ Interfacial polymerization	- Lower water contact angle with under 67 º, more hydrophilic; - Improved water flux with functionalization of g-C_3N_4; - High salt rejection from 96 to 98 %; - BSA resistance over 96 %.	(Seyyed Shahabi et al., 2020)
Aniline and O_2	Plasma enhanced chemical vapor deposition	- Decreased water contact angle, more hydrophilic; - Increased NaCl and Na_2SO_4 rejection by 4.2 % and 2.6 %; - Higher FRR from 79.8 to 96.2 %, compared to pristine RO of 76.5 %.	(Khoo et al., 2021)
TA and DMC	Co-solvents	- Decline of water flux using DMC over 1.5 % (w/v); - Higher water flux of 64.2 ± 1.8 $L.m^{-1}.h^{-2}$, combining TA and DMC; - Higher salt rejection over 99 %.	(Shi et al., 2020)

2.4 Antibacteria

A reported trend to increase the antibacterial capacity of RO membranes is to integrate materials or coatings that inhibit bacterial growth without affecting the membrane filtration efficiency, as demonstrated in Table 4. Zhang et al used a p-aminophenol-modified graphene oxide (mGO) combined with PA surface of the RO membrane by interfacial polymerization. mGO-RO membrane had a improved water flux of 23.6 $L.m^{-2}.h^{-1}$, and higher NaCl rejection of 99.7 %, which was 24.5 % higher than that of the pristine RO membrane. mGO-RO membrane can kill 96.78 % and 95.26 % of gram-negative *E. coli* and gram-positive *S. aureus* bacteria (Zhang et al., 2020). Zhao et al. studied tannic acid (TA)-functionalized carbon nanotubes (CNT@TA) by integrating silver nanoparticles into the porous structure of CNT@TA (Ag-CNT@TA). Subsequently, Ag-CNT@TA was introduced into the PA layer through polymerization to create a high-performance nanocomposite RO membrane. After modification, the water permeability was 4.81 $L.m^{-2}.h^{-1}.bar^{-1}$, which was 49.8 % higher than that of unmodified RO, while maintaining a NaCl rejection of 99.3 %. This nanocomposite membrane showed resistance to *E. coli* and anti-fouling (BSA) properties due to the presence of TA coating and silver nanoparticles in the structure of the CNT, maintaining the antibacterial properties of the membrane. In addition, the outstanding compatibility between CNT and PA layer helps stabilize the performance of the modified RO membrane (Zhao et al., 2021). Nanomaterials have great potential for improving the antibacterial abilities of RO membranes, stability and effectiveness of these materials under RO membrane operating conditions such as temperature, salinity, or pH have not yet been elucidated.

Table 4: Summary of surface modification method of RO for antibacteria

Modification materials	Modification methods	Key results	Refs.
mGO	Incorporating/ Interfacial polymerization	- Lower water contact angle of 48.2 °and more hydrophilic; - Improved water flux to 28.3 $L.m^{-2}.h^{-1}$, salt rejection of 99.9 % at 0.005 wt% mGO; - Enhanced resistance to *E. coli* and *S. aureus* bacteria of 96.8 % and 95.3 %.	(Zhang et al., 2020)
Ag-CNT@TA	Interfacial polymerization	- Increased water permeability to 49.8 %; - Unchange salt rejection remarkably; - Resisted to BSA fouling and *E. coli* bacterium.	(Zhao et al., 2021)

3. Conclusions and future prospects

Polyamide is an important active layer in the RO membrane structure. The membrane surface is easily fouled by impurities and bacteria during operation and free chlorine attacks, causing damage, degradation, and a

reduced lifespan of RO membrane. This review summarizes recent trends in the modification of RO membrane surfaces to improve fouling resistance, chlorine resistance, enhanced filtration performance, and antibacterial properties to provide an overview of possible methods that can be applied in the RO membrane manufacturing industry.

In the future, the aspects that need to be further explored to find suitable materials for RO membrane surface modification without affecting membrane water flux and performance, such as not use toxic and expensive agents and solvents such as sulfuric acid, inorganic nanomaterials, and aniline to replace cheaper and cleaner agents for commercial production; the ability to apply complex technologies such as plasma chemical vapor deposition, which is difficult to implement in actual production and consumes more energy. The modification process must be simplified; assess the toxicity level, exposure level, and impact of RO membranes incorporating nanomaterials during operation and discharge into the environment; effect of modified RO membranes on various organic, inorganic, biological, and colloidal fouling.

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