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Effect of Different Nanomaterials on Antifouling and desalination performance of reverse osmosis membranes: A Review

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ABSTRACT

Thin-film nanocomposite (TFN) reverse osmosis (RO) membranes offer immense potential for revolutionizing water purification and desalination but face limitations including fouling, scaling, and high energy consumption. To address these challenges, researchers have explored incorporating various additives into TFN membranes. This review highlights the efficacy of organic and inorganic additives in overcoming these limitations and enhancing membrane performance by various additives, both organic and inorganic, into thin-film nanocomposite (TFN) reverse osmosis (RO) membranes has shown promise in overcoming the restrictions of possible nanoparticle leaching and improving membrane performance. In particular, the use of metal-organic frameworks (MOFs) has demonstrated significant potential. MOFs, such as ZIF-8, UiO-66, and MIL-101, have been successfully incorporated into TFN membranes, resulting in improved water flux, salt rejection, and antifouling characteristics. The unique features of MOFs, including high water permeability, broad surface area, and water stability, make them suitable for membrane modification. Additionally, the employment of zeolites, such as NaY and NaA, has been shown to enhance the permeate flux of TFN RO membranes without compromising salt rejection. Furthermore, the incorporation of non-organic additives, such as silica nanoparticles and halloysite nanotubes (HNTs), has demonstrated the potential to improve the thermal stability, permeate flux, and antifouling properties of TFN membranes. For instance, the inclusion of HNTs in the TFN membrane resulted in a greater flux compared to the pristine Thin Film Composite (TFC) membrane while maintaining a comparable salt rejection rate. Similarly, the integration of silica nanoparticles into TFN RO membranes has shown promise in improving their thermal stability, permeate flux, and salt rejection. However, careful control of nanoparticle concentration and dispersion is necessary to avoid agglomeration and maintain optimal membrane performance. Therefore, the incorporation of these additives into TFN RO membranes holds great potential for enhancing their performance and overcoming key challenges in membrane technology. Further research and development in this area are essential to unlock the full potential of these advanced membrane materials.

Keywords: Desalination, Reverse osmosis membrane, SWCNT; MWCNT, Quantum Dots, Metallic additives, Nano composites

1. INTRODUCTION

Although approximately Water covers 70% of the Earth's surface, only 2.5% of this is freshwater, with just 1% being easily accessible [1]. This limited availability of fresh water poses a significant challenge, particularly for the 40% of the global population number residing in arid regions or islands where freshwater scarcity is prevalent [2]. Moreover, the rise in drought occurrences worldwide, the vulnerability of conventional water resources to climate change, and the overexploitation of existing sources have led to an increased reliance on desalination technologies [3]. Numerous factors such as economic, environmental, technical, social, and political issues are preventing the successful implementation of these technologies. The water market's recent addition only highlights the intricate challenges we need to overcome in the 21st century. Experts predict that by 2025, around two-thirds of the global population may experience water scarcity. [4]. Necessitating the establishment of effective policies by governments to ensure equitable access to water for the most vulnerable communities, while also meeting the demands of industrial and domestic users [5].

The goal of the United Nations' 2030 agenda is to guarantee access to water and sustainable management of sanitation facilities for everyone. [6]. To achieve this goal of sustainable development, various water management strategies, such as decarbonized desalination and improvements in irrigation systems, are crucial [7]. A diverse range of desalination and water treatment technologies is tailored to different water types. However, researchers are actively working towards reducing the energy consumption associated with these processes [8].

Table 1. Comparison of Energy Use for Different Water Supply Alternatives

Alternative Water Supply	Technology	Energy Use (kWh/m3)	Reference
Conventional surface water treatment	Physical treatments; coagulation	0.2–0.4	[9]
Reclamation of water	--	0.5–1.0	[10]
Treatment of waste water	Filtration, coagulation, and/or biological treatments	0.2–0.67	[10]
Indirect potable reuse	--	1.5–2.0	[9]
Desalination of brackish water	BWRO	0.8–2.5	[11]

Water Desalination in the Pacific Ocean	SWRO	2.5–4.0	[9]
Seawater	SWRO	2.58–8.5	[12]

Over 20 distinct technologies are now used in seawater desalination [13]. Despite the many available technologies for global water production, only a handful are commonly used. Commercial desalination processes fall into three main categories: thermal processes, membrane separations, and emerging technologies. The most notable thermal processes are multi-effect distillation (MED) and multi-stage flash distillation (MSF). Meanwhile, reverse osmosis (RO) is the most widely used technology in membrane processes. [14], [15]. MED technology accounts for 7% of total installed capacity, while MSF technology accounts for 18% and is widely used in large-scale desalination plants that produce both water and electricity; on the other hand, RO technology is mostly used in smaller plants for freshwater production and accounts for 69% of total installed capacity globally [8]. Nanofiltration (NF) is a membrane technique that adds 3% to the whole desalination process. Other membrane technologies that account for 2% and 1% of the total are electrodialysis (ED) and reverse electrodialysis (EDR). [8].

Desalination through reverse osmosis (RO) has emerged as a highly efficient and economically feasible process. Noteworthy advancements in flow devices and energy recovery mechanisms, coupled with progress in membrane materials, integration with other desalination technologies, and incorporation of renewable energy sources, have significantly contributed to the enhancement of this technology. Moreover, the optimization of RO systems through the application of artificial intelligence techniques has emerged as a crucial aspect in this field.

The initial commercialization of reverse osmosis (RO) desalination was carried out by Loeb & Sourirajan in 1964 [16]. Since then, significant advancements have been made, establishing RO as the leading technology in desalination operations. The versatility of RO stems from its ability to separate water without the need for evaporation. In comparison to thermal technologies, RO exhibits relatively low energy consumption [17], [18]. Furthermore, it offers high flexibility to operate under varying salinity conditions, requires minimal space, and is user-friendly and easily automated [15], [19]. RO plants have a current installed capacity exceeding 60 Mt/day, and their annual growth rate ranges between 10% and 15%. These plants' combined energy consumption amounts to 100 TWh/year. [20]. Reverse osmosis (RO) units come in various sizes for commercial use. They range from small-scale household applications with a capacity of 0.1 m3/day to large-scale industrial and municipal systems with a capacity of up to 900,000 m3/day. [21]. Figure 1 depicts the typical configuration of rolled membranes commonly used in seawater desalination processes.

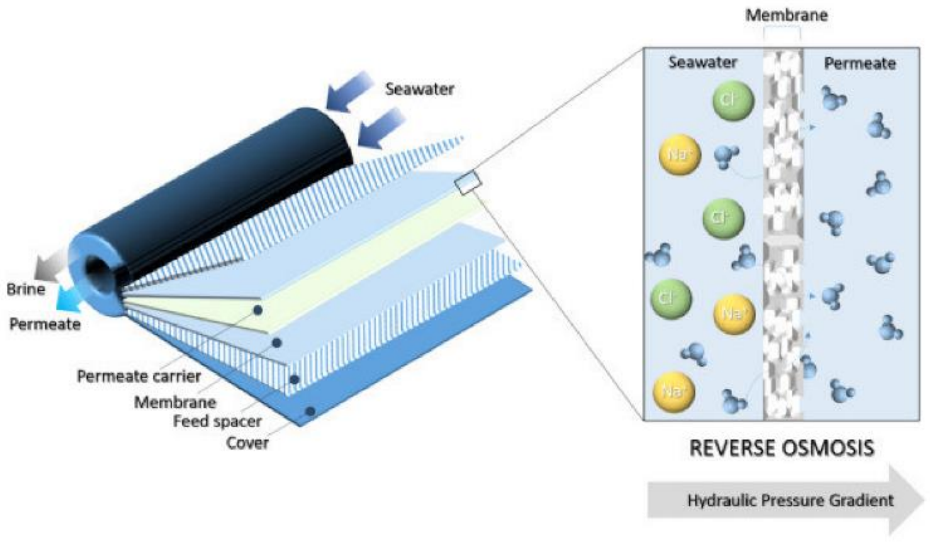

Fig.1 is an essential component of reverse osmosis (RO) systems. The driving force principles of RO, illustrated in the inset of the figure, are also crucial for the operation of these membranes, adapted from [22].

2. Role of Nano-Structured Membrane Materials

There has been significant progress in the field of nanotechnology in recent years, with a move from solely academic research to actual applications in the commercial industry. This progress has resulted in the creation of innovative nanotechnology-based membranes that outperform traditional technologies and give additional benefits such as increased selectivity, catalytic activity, and fouling resistance. Nanotechnology has been used to improve conventional ceramic and polymeric membrane materials in a variety of ways to achieve these advances. [23]. There are now various types of nanotechnology membranes available. Advanced membranes such as zeolite and catalytic nanoparticle-coated ceramic membranes, hybrid inorganic-organic nanocomposite membranes, bio-inspired nanotechnology membranes, bio-hybrid immobilized enzyme membranes, bio-hybrid magnetic-responsive membranes, aquaporin membranes, vertically aligned nanotube membranes, and isoporous block copolymer membranes are examples of these. [23]. Nanoscale functional materials have been identified as efficient instruments for water treatment applications [23]. To improve the functionality of reverse osmosis (RO) membranes, a method called thin-film nanocomposite (TFN) membrane has been developed. This involves adding nanomaterials to the polyamide (PA) membrane's selective layer to create a new composite membrane. By adding nanomaterials to the dense layer of PA, the surface charge of the interfacial polymerized layer is enhanced, which leads to higher selectivity due to charge interaction and increased throughput of porous membranes. The concept of TFN RO membrane was first introduced by researchers as in [24], Porous Sodium zeolite A (NaA) nanomaterials were incorporated into the PA layer through interfacial polymerization (IP) by an unidentified party to create a nanocomposite membrane. This resulted in a higher permeate flux compared to the original TFC membrane while maintaining the same level of selectivity. Since then, different types of both porous and nonporous nanomaterials have been used in preparing TFN RO membranes. For instance, [25] incorporated silica and zeolite nanomaterials into the PA layer over a polysulfone (PSF) substrate, resulting in a TFN RO membrane with higher permeate flux and enhanced mechanical stability. Other nanomaterials, such as silver [26], graphene oxide [27], [28], carbon nanotubes (CNT) [29], [30], multi-walled carbon nanotubes (MWCNT) [31], silica [32], [33], metal oxides [34], TiO2 [35], metal-organic frameworks (MOF) [36], alumina [37], [38], and zeolites [24], [39]–[44] have also been utilized for Nano-enhanced RO membranes. The way nanomaterials are distributed is determined by whether they are hydrophilic or hydrophobic. Incorporating nanomaterials can enhance the physical attributes of the membrane, such as its mechanical, chemical, and thermal stability. The effectiveness of TFN membranes for transportation and separation depends on several factors, including the size of the nanomaterials, their chemical properties, surface hydrophilicity, the number of nanomaterials in the PA layer, and the thickness of the film. Additionally, interfacial polymerization has been utilized to add nanomaterials and create TFN RO membranes in the form of hollow fiber modules. [45]–[47]. Low-pressure TFN RO membranes can be developed using interfacial polymerization [48]. In this review, we will explore the advancements and efforts made in recent years to improve Nano-enhanced RO membranes. [49]. Furthermore, the potential use of TFN RO membranes in desalination, their market viability, and possibilities for extra improvement are also explored.

3. Approaches for preparing polymer reverse osmosis membranes enhanced with Nano-fillers.

Membranes made from polymers and nanoparticles can be classified into four categories depending on the location of the nanomaterials and the structure of the membrane. These categories are: [50]: a. Conventional nanocomposite or mixed matrix membrane (MMM), b. Thin film composite (TFC) with a nanocomposite substrate, c. Thin film nanocomposite (TFN), and d. The nanocomposite is located at the surface of the membrane. These different configurations and locations of nanomaterials play a crucial role in enhancing the performance of polymer-based Nano-enhanced reverse osmosis membranes. By carefully designing and preparing these membranes, researchers can optimize their selectivity, permeability, and fouling resistance, leading to improved water purification processes. The different configurations of these membranes are depicted in Figure 2, where spheres are used to represent nanoparticles, nanotubes, nanofibers, or Nanosheets.

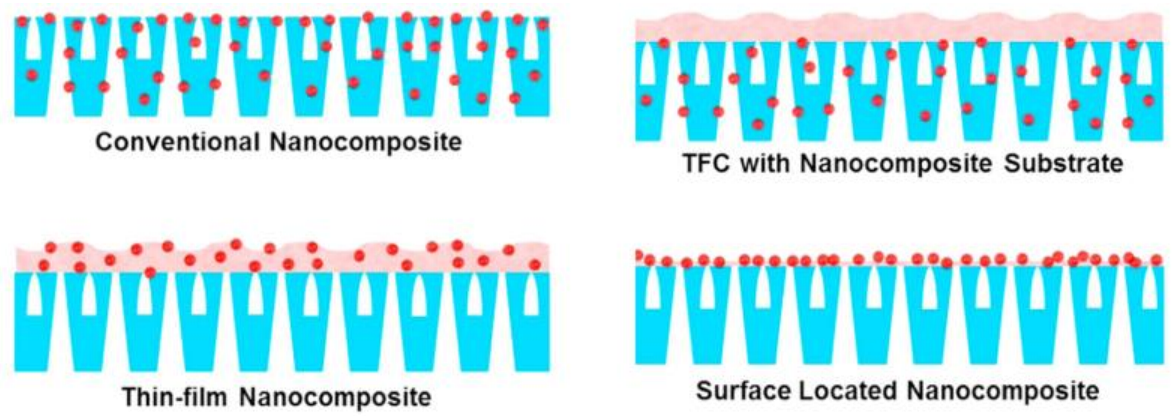

Fig 2. Nanocomposite membrane types adapted from [12]

3.1. Nanocomposite thin-film composite substrate

There is a growing interest in using nanocomposite substrates in the field of thin-film composite (TFC) membranes. These substrates include nanomaterials into the polymer substrate, which is then used to construct TFC membranes using an interfacial polymerization (IP) method. There has been little research into the development of reverse osmosis (RO) membranes utilizing this technology [51], [52]. However, some studies have shown that TFC membranes with nanocomposite substrates have higher permeate flux than pure TFC membranes. This enhancement in performance can be attributed to the membrane's improved hydrophilicity as a result of nanoparticle incorporation in the substrate. Another method for creating thin-film composite RO membranes is to add cellulose nanofibers to polymer substrates and then perform interfacial polymerization [53]. This approach is primarily used to lessen the effects of internal concentration polarization in the manufacture of forward osmosis membranes.

3.2. Thinfilm nanocomposite

Thin-film nanocomposite (TFN) membranes are created by integrating nanoparticles into a thin layer of polyamide (PA) using an interfacial polymerization (IP) technique. The solubility of the nanomaterials determines whether they are distributed in an organic or aqueous phase. TFN membrane features such as selectivity, permeability, and fouling resistance are principally determined by the PA thin layer [54]. The physical properties of the membrane, such as hydrophilicity, porosity, and charge density, can be modified by inserting nanoparticles into this layer, resulting in increased permeability and selectivity. Nanoparticles, nanotubes, and nanofibers, which are routinely used in traditional nanocomposite membranes, can also be used to create TFN membranes. When compared to typical thin-film composite (TFC) membranes, the TFN approach has the potential to improve the performance of reverse osmosis (RO) membranes. Figure 3 depicts a general production procedure for TFN membranes employing the IP technique between a solution of m-phenylenediamine (MPD) and trimesoyl chloride (TMC) [55].

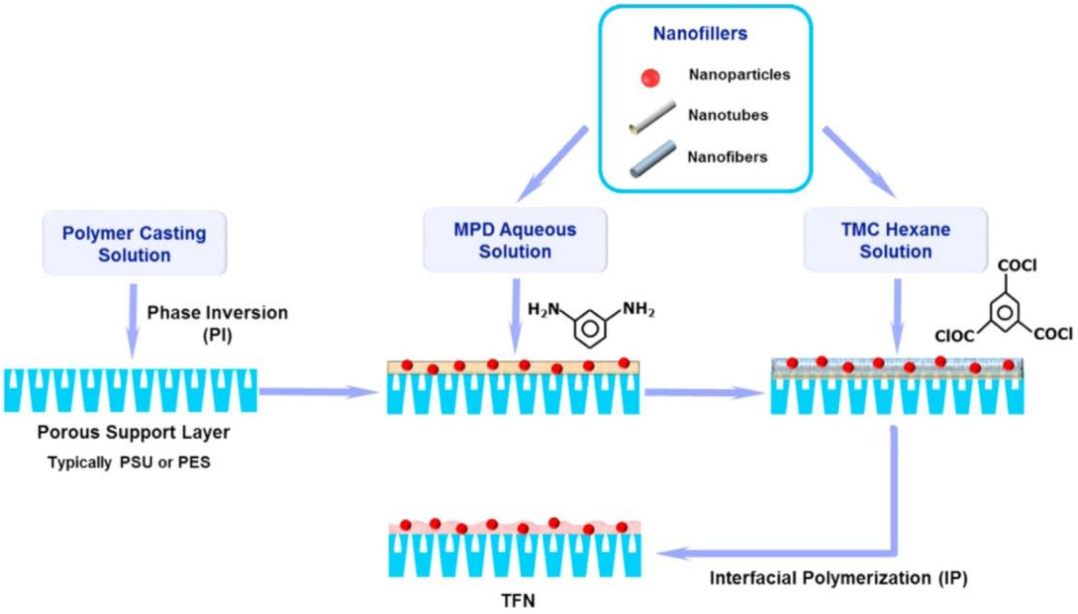

Figure 3 [55], IP process that involves the formation of a thin film on the surface of the support membrane. This is achieved by immersing the membrane in a solution containing the aqueous phase and the organic phase, which contains the monomer and the nanoparticles. The aqueous phase usually consists of an aqueous solution of a diamine, while the organic phase consists of an organic solvent containing a diacid chloride adapted from [55].

The IP procedure can be repeated several times to thicken the polyamide layer and improve the performance of the TFN membrane. Following the IP procedure, the membrane is usually treated to a post-treatment step that includes rinsing with a suitable solvent to remove any leftover reactants or by-products.

3.3. Nanocomposite located at membrane surface

The membrane's performance, particularly in terms of separation and antifouling properties, depends not only on its structure, porosity, and thickness but also on its surface characteristics, such as charge density, pore size, roughness, and hydrophilicity. Modifying the surface of a membrane can significantly improve its effectiveness in water treatment applications. Surface-located nanocomposite membranes have been developed to modify the membrane's surface without changing its intrinsic structure. Various techniques, such as chemical grafting, coating/deposition, and self-assembly, have been used to prepare these surface-located nanocomposite membranes. These methods utilize unique properties such as bonding force and bonding process to create the desired surface modifications. However, there are only a few studies that have reported on the preparation of reverse osmosis (RO) membranes using this approach. Table 1 summarizes the RO membranes that have been developed using surface-located nanocomposite membranes. For instance, TiO2 nanoparticles have been attached to particular surfaces of the membrane through H-bonding and coordination interactions in a self-assembly process. [56], [57]. When TiO2 nanoparticles are added to the surface of a membrane, it can improve its hydrophilicity and make it more effective at fighting microbes. In the same way, silver nanoparticles can also be added to the surface of the membrane to make it more antimicrobial. To achieve this effect, the membrane surface absorbs silver ions, which are then reduced using either chemical agents or light irradiation. [58]–[61]. An alternative method involves the use of copper nanoparticles that are encapsulated in positive-charged polyethyleneimine (PEI) and then attached to the negatively charged surface of a membrane through electrostatic attraction. This approach boosts the antimicrobial and antifouling capabilities of the membrane. [60]. One method of improving RO membranes is through multiple coatings of nanomaterials using the layer-by-layer assembly technique. This involves incorporating the coatings onto the membrane surface through electrostatic attraction, hydrogen bonding, and/or chemical bonding. However, this technique may result in potential nanoparticle leaching from the membrane surface. Nonetheless, layer-by-layer assembly has been proven effective in enhancing the thermal stability, selectivity, and chlorine resistance of RO membranes. [59]. The assembly process for creating a graphene oxide (GO) membrane is depicted in Figure 3, showcasing the layer-by-layer technique. [59].

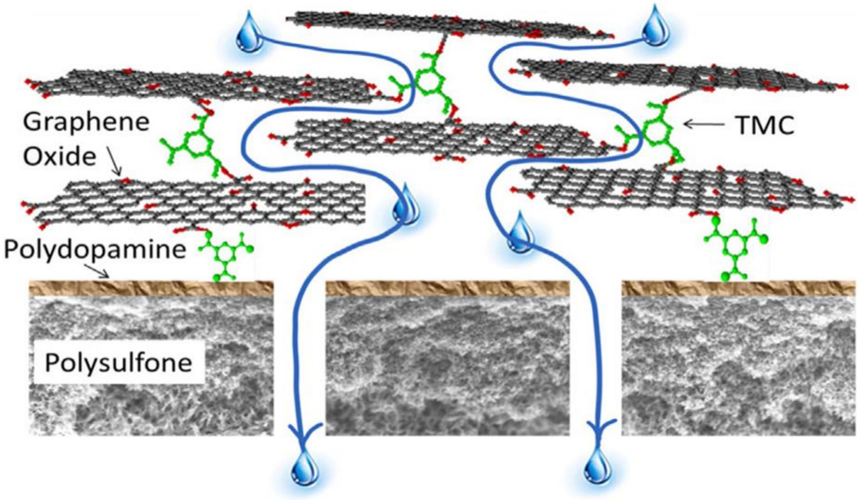

Figure 4 depicts the chemical grafting procedure used to connect silver nanoparticles (AgNPs) to a polyamide (PA) thin-film composite (TFC) membrane, In order to reduce membrane biofouling, the AgNPs and TFC membrane undergo covalent bonding. The figure's source has been adapted from a research article [59].

Table 2. Performance of Nanocomposite insertion at the membrane surface.

Method of fabrication	Nanomaterial	Pressure (bar)	Flux (LMH)	NaCl Removal (%)	Performance enhancement	References
Selfassembly	TiO2	15.5	129.25	96	Hydrophilicity, fouling resistance, antimicrobial activity	[62]
		15.5	24.49	96.6		[63]
Adsorption reduction	Ag	55.5	54.17	95	Antimicrobial activity	[64]
		10	40	96		[65]
Electrostatic attraction	Cu	27.6	69	98.6	Antimicrobial activity	[66]
Layer-by layer assembly	CNTs	15.5	13.6	92.5	Thermal stability, chlorine resistance	[67]
	GO	3.44	68.8	60	Selectivity, chlorine resistance	[68]
		15.5	12.5	97.1		[69]
Chemical grafting	CNTs	27.6	38.64	-	Antimicrobial activity	[70]
	Ag	20.7	69.4	93.6	Antimicrobial activity	[71]

4. Nanocomposites for reverse osmosis membranes

Currently, TFN membranes are becoming more popular compared to other nanocomposite-based RO membranes. Various hydrophilic nanomaterials, including GO, CNT, metal, and metal oxide, are being used to create TFN membranes. This section will discuss the use of different nanoparticles in the development of RO membranes, their production methods, applications, and performance.

3.4. Conventional nanocomposite or mixed matrix membrane (MMM)

Nanocomposite membranes and mixed matrix membranes (MMMs) are typically created using the phase inversion method. This involves adding nanomaterials, such as inorganic, organic, and biomaterials, to a polymer solution. These types of membranes have been found to enhance membrane structure, and physical and chemical properties, and even possess antibacterial and antifouling properties. MMMs, specifically, have been commonly utilized in microfiltration and ultrafiltration applications due to their porous structure. Recent research has demonstrated the potential benefits of both MMMs and conventional nanocomposite membranes. For Example in a study by [72], a conventional nanocomposite reverse osmosis (RO) membrane was developed by adding graphene oxide (GO) Nano sheets to a cellulose acetate (CA) polymer matrix. The presence of GO Nano sheets improved the desalination performance of the CA-based RO membranes.

4.1. Carbon Based

Various carbon-based nanoparticles, including carbon nanotubes (CNTs), graphene oxide (GO), reduced-GO, and carbon quantum dots (CQD), have been incorporated into thin-film nanocomposite (TFN) reverse osmosis (RO) membranes for water treatment purposes Table 2 [73]–[79]. CNTs, in particular, have gained attention due to their ability to facilitate rapid water transport and exhibit antifouling properties [74], [75]. The smooth inner wall of CNTs allows for efficient water molecule transport, making them superior to other Nano-porous materials [74], [75]. Studies have shown that CNTs with the appropriate diameter can enhance ion rejection and improve water transport through the membrane [74]. Nanocomposite membranes have been created using multi-walled carbon nanotubes (MWCNTs) because of their distinctive physicochemical properties. [80]. Incorporating MWCNTs into nanocomposite membranes has been found to enhance water permeability, salt rejection, antifouling properties, and antimicrobial characteristics [76]. In the fabrication of antifouling TFN RO membranes, MWCNTs are doped into the polyamide (PA) layer during the interfacial polymerization (IP) process [80]. The addition of MWCNTs (15.5 wt. %) in the PA layer significantly improves the membrane permeability, antifouling properties, and chlorine resistance, resulting in a permeate flux more than two times higher than that of the pristine PA membrane [80]. Another study demonstrated the antifouling and low-protein adhesion properties of MWCNT-incorporated PA RO membranes, which revealed enhanced antifouling properties related to pristine PA membranes [78]. The addition of MWCNTs to the PA layer led to a smoother membrane surface, reducing the interaction between foulants and the membrane surface and improving the antifouling mechanism [78]. Furthermore, the PA membranes based on MWCNT demonstrated comparable water permeability and salt rejection to other currently available PA membranes, indicating its potential for water treatment applications [29]. Incorporating carbon nanotubes (CNTs) into the PA layer of TFN RO membranes has also been found to improve surface features and antifouling capabilities, resulting in a 30% increase in water flux and a 30% reduction in energy consumption [29]. CNT-based TFN RO membranes were shown to have lower specific energy consumption (SEC) than other commercial RO membranes, showing their potential for energy-efficient water treatment [29]. Furthermore, it has been demonstrated that incorporating CNTs into the PA layer of TFN RO membranes improves water transport through the membrane [79].

To create a thin-film nanocomposite reverse osmosis (TFN RO) membrane, single-walled carbon nanotubes (SWCNTs) are modified using chainlike zwitterion groups [29]. These zwitterion groups present in the SWCNTs introduce both negative and positive charges on the membrane surface by integrating into the polyamide (PA) layer. As a result, the membrane exhibits enhanced permeability, antifouling properties, and salt rejection [29].

4.1.1. Graphene oxide

In recent times, scientists studying membranes have become increasingly interested in Graphene embedded membranes. This is because of their exceptional water transport abilities in graphene Nano channels, as well as the charge imparted by Graphene. These nanomaterials are widely used in membranes for water treatment, particularly in desalination. Furthermore, graphene based nanomaterials possess antifouling and antimicrobial traits, which are utilized to create TFC membranes [81], [82]. Graphene oxide (GO) can be prepared using the Hummers or Staudenmaier techniques [83]. GO has superior hydrophilicity and can easily form suspensions in water, making it a highly promising material for creating nanocomposite MMM. [84] [85]. GO can be utilized in preparing TFN PA RO membrane for desalination. Many studies have reported the successful preparation and application of TFN RO membrane by incorporating GO in the PA layer. The antifouling and antimicrobial properties of GO have been utilized in TFN membranes to reduce biofouling [82]. Furthermore, the mechanical strength of membranes can be increased by adding GO to the polymer matrix. Various studies have demonstrated that incorporating GO into the PA layer can enhance the permeability of the TFN RO membrane. [81] A TFC RO membrane with high permeate flux, anti-biofouling properties, and chlorine resistance has been reported. To create the PA nanocomposite membrane, they used IP between MPD and TMC solution, with an optimized concentration of GO (40 ppm) included in the MPD solution. The addition of GO in the PA layer increased the membrane's water flux and antifouling properties while maintaining similar salt rejection as the pristine TFC membrane. The inclusion of GO also affected the membrane's surface roughness, surface charge, hydrophilicity, and PA layer thickness, all contributing to enhanced membrane performance. TFN membrane containing GO showed improved antifouling properties, likely due to a smoother surface, altered surface charge, and increased hydrophilicity [86]–[88]. The permeate flux of the TFC membrane based on GO increased from 9.18 L/m^2h to 32.5 L/m^2h. The salt rejection of both TFC membranes, with or without GO, is similar. [89] Developed GO-embedded TFC membrane for water desalination. To create these composite membranes, a process called IP is used between solutions of MPD and TMC. To prepare the nanocomposite membrane, a specific concentration of GO (100 ppm) is added to the MPD solution. The interaction between MPD and TMC during the IP process and the addition of GO in the PA layer can be seen in Fig. 10.8. When GO is incorporated into the PA layer, it increases the membrane's hydrophilicity and improves it's permeate flux. [90]Developed a GO-enhanced PA TFN membrane by IP process among aqueous MPD and organic TMC-GO solution. During the IP process, they mixed the GO Nanosheets with TMC-hexane solution and adjusted the concentration of GO from 0 to 0.02 wt. %. Their findings revealed that including up to 0.015 wt. % GO in PA enhances the permeability of the membrane. This is because GO inclusion in the PA layer improves hydrophilicity, resulting in increased permeate flux. The hypothesized mechanism of GO TFNC membrane is shown in Fig. 5, where it is evident that the interlayer spacing within the GO Nano sheets may aid in water passing through the PA thin-film layer, leading to enhanced flux. Studies have shown that GO-incorporated PA TFN membranes can enhance antifouling, antimicrobial, physicochemical properties, and transport properties, making them ideal for desalination applications. Raval and Das proposed a novel approach to bind GO over the polyamide layer by post-treatment. They treated the polyamide membrane with sodium hypochlorite solution and then subjected it to an aqueous suspension of GO, anchored by crosslinking with N-hydroxysuccinimide. This approach did not require any changes to the membrane-making process but imparted GO by post-treatment [91].

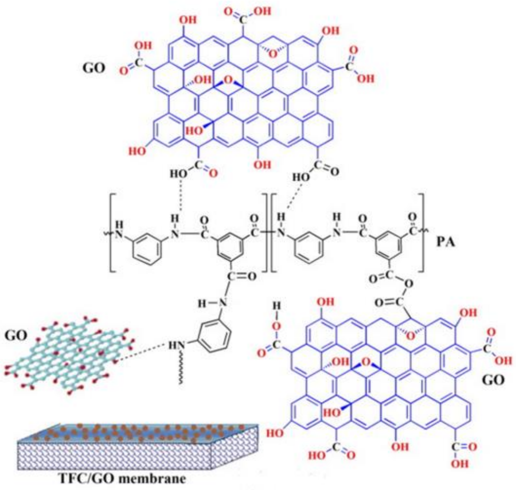

Fig 5. Shows how the graphene oxide (GO) incorporated into (PA) layer. This image was adapted from [89].

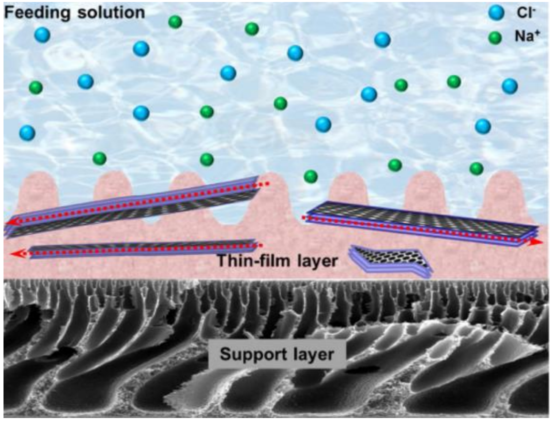

Fig 6. Shows how the graphene oxide (GO) thin film nanocomposite (TFN) membrane works. This image was adapted from [91].

4.1.2. Quantum Dots

Quantum dots (QDs) have emerged as a promising material for the preparation of thin-film nanocomposite (TFN) reverse osmosis (RO) membranes owing to their unique properties Some of the desirable qualities for certain materials include being small in size, biocompatible, hydrophilic, and having functional groups on the surface [92]. In recent studies, graphene oxide quantum dots (GOQDs) and carbon quantum dots (CQDs) have been incorporated into the polyamide (PA) layer of TFN membranes to enhance their performance [92]–[94]. For instance, GOQDs were dispersed in a 2-methyl-1,3-propanediol (MPD) solution and then mixed with a trimesoyl chloride (TMC) solution to form a TFN membrane via an interfacial polymerization (IP) process [92]. The GOQD/MPD suspension was filtered onto a polysulfone (PSF) substrate to create a cushion layer, and a PA layer was formed on top of it. The resulting GOQD-embedded TFN membrane exhibited improved permeate flux and solute rejection compared to a pristine thin-film composite (TFC) membrane. Moreover, the inclusion of GOQDs in the PA layer enhanced the membrane's antifouling and chlorine resistance properties [92]. Similarly, CQDs were dispersed in an MPD solution and then reacted with a TMC solution to prepare a nanocomposite TFN membrane [93]. The incorporation of CQDs in the PA layer increased the permeate flux without compromising the solute rejection. This enhancement was attributed to the interaction between the carboxyl groups of CQDs and the amine groups of the MPD solution, as well as the formation of covalent bonds between the remaining carboxyl groups of CQDs and the terminal acyl chloride groups of TMC during the IP process [93].

In another study, nitrogen-doped GOQDs (N-GOQDs) were used as nanomaterials to improve the performance of TFN RO membranes [94]. The N-GOQDs were incorporated into the PA layer, resulting in a threefold increase in permeate flux compared to a pristine PA membrane, while maintaining comparable salt rejection. The concentration of N-GOQDs in the PA layer was varied from 0 to 0.1 wt. %, and it was observed that higher concentrations of N-GOQDs enhanced the hydrophilicity of the membrane, thereby increasing the permeate flux. Additionally, the incorporation of N-GOQDs improved the thermal stability of the membrane [94]. These studies demonstrate the potential of QD-incorporated TFN RO membranes for various applications, including desalination, due to their improved performance in terms of permeate flux, solute rejection, antifouling properties, chlorine resistance, and thermal stability [92]–[94].

Table 2. This table shows how carbon-based nanoparticles are employed in the interfacial polymerization approach to make thin-film nanocomposite reverse osmosis membranes.

Filler material	Quantity of NPs wt. %	Pressure bar	Flux LMH	Enhancement	NaCl elimination (%)	References
MWCNTs	0.1	50	71	Permeate flux, antifouling, chlorine resistance	90	[80]
	0.1	50	6.5	Hydrophilicity, permeate flux, antifouling, salt rejection	99.7	[78]
	0.001	15.5	51.15	Permeate flux, antifouling	97	[79]
	0.005	15	25.9	Hydrophilicity, permeate	98.1	[95]

				flux, antifouling, salt rejection		
	0.7	10	11.4	Permeate flux, permeate flux, salt rejection	97.04	[96]
CNT	0.1	7	7	Permeate flux	96	[97]
	0.00375	15.5	51.3	Hydrophilicity, permeate flux	98.5	[74]
	0.002	15.5	44	Hydrophilicity, permeate flux	95.4	[73]
Carboxy-functionalized MWCNT	0.1	16	28	Hydrophilicity, permeate flux, antifouling, chlorine resistance	More than 90	[84]
Zwitterion functionalized single-walled CNT (SWCNT)	0.2	36.5	48.5	Hydrophilicity, permeate flux, salt rejection	98.6	[29]
GO	0.0038	15.5	16.5	Hydrophilicity, permeate flux, antifouling, chlorine resistance	99.3	[81]
	0.01	15	39	Hydrophilicity, permeate flux, antifouling chlorine resistance	>=97	[89]
	0.06	20	31.8	Hydrophilicity, permeate flux, antifouling chlorine	98.8	[98]

				resistance		
p-minophenolmodified GO	0.005	15	23.6	Hydrophilicity , permeate flux, salt rejection	99.7	[99]
Carbon dots(CD)	0.02	15.5	88.7	Hydrophilicity , permeate flux, antifouling, salt rejection	98.8	[93]
Graphene oxide quantum dots (GOQD)	0.01	16	37.4	Hydrophilicity , permeate flux, antifouling chlorine resistance	98.8	[92]
Nitrogen-doped GOQD (N-GOQD),	0.02	15	24.9	Hydrophilicity , permeate flux	93	[94]
Na-CQDs	1	15	64	Hydrophilicity , permeate flux, antifouling	98.6	[100]
Sulfonic decoration on GOQD (S-d-GOQD)	0.5	15	88.35	Hydrophilicity , permeate flux, salt rejection	97.1	[101]
Graphite carbon	0.004	16	73.4	Hydrophilicit, permeate flux, antifouling, salt rejection	99.04	[102]
Graphitic carbon nitride	0.005	15	91.8	Hydrophilicity , permeate flux, antifouling	98.1	[103]

4.2. Metallic and Metallic oxides-additives

4.2.1. Metal Oxides

Thin-film nanocomposite (TFN) reverse osmosis (RO) membranes have been improved by incorporating metal oxides like zinc oxide (ZnO) and alumina (Al2O3). ZnO nanoparticles have proven beneficial in modifying membranes due to their antibacterial and photo-catalytic properties. When included in TFN membranes, ZnO nanoparticles improve their fouling resistance and increase their hydrophilicity, resulting in better permeability. Ghoul et al.[104] made a high flux antifouling TFN RO membrane by incorporating ZnO nanoparticles in the polyamide (PA) layer using the interfacial polymerization (IP) process. Rajakumaran et al. [105] The effects of incorporating different nanoparticles in TFN membranes were explored, and it was discovered that TFN membranes with GO-ZnO nanoparticles had higher permeability and hydrophilicity than pure membranes.

Alumina nanoparticles have also been used to improve the antifouling and hydrophilicity features of TFN RO membranes. Saleh and Gupta [106] incorporated Al2O3 nanoparticles into the PA layer of TFN membranes and observed improved salt rejection performance, increased hydrophilicity, and enhanced antifouling characteristics. Jeong et al. [24] reported similar results with the zeolite incorporation in the PA layer of TFC membranes.

The incorporation of metal oxide nanoparticles, such as ZnO and Al2O3, shows promise as a method for modifying TFN membranes to increase their performance in reverse osmosis applications. A summary of these nanoparticles can be found in Table 3.

4.2.2. Silver and Copper Nanoparticles (Metallic additives)

Silver (Ag) nanoparticles have excellent antimicrobial properties and have been increasingly used in the preparation of antifouling membranes [107]. Various approaches, including as chemical fixing and covalent bond formation, have been employed to embed Ag nanoparticles into the PA layer of TFC membranes. [108]. Ag nanoparticles have been successfully embedded within the PA layer of the TFC membrane through irreversible binding and interfacial polymerization while in situ. [107]. This results in Ag nanoparticle-embedded TFC membranes with superior antimicrobial activity compared to pristine membranes [109].

Copper (Cu) nanoparticles also possess remarkable antimicrobial properties and are a more cost-effective option related to the silver nanoparticles effect [66], [110]. Copper (Cu) nanoparticles are frequently employed in membrane technology to improve fouling resistance and limit the formation of dangerous microbes [111], [112While the literature on the use of Copper nanoparticles in reverse osmosis (RO) membrane applications is limited, one study reported the incorporation of Copper nanoparticles into a polyamide TFC membrane using the dip-coating approach [66]. A new method was created to attach Copper nanoparticles with biocidal properties onto TFC RO membranes, improving their ability to fight bacteria. [113]. Furthermore, it has been demonstrated that incorporating copper oxide (CuO) nanoparticles into the polyamide layer of TFC membranes improves water permeability and antifouling capabilities without impairing salt rejection [114].

Table 3. Interfacial Polymerization (IP) Process Properties of Thin-Film Nanocomposite (TFN) Reverse Osmosis (RO) Membranes with Metal and Metal Oxide Nanoparticles.

Filler material	Quantity of NPs (wt.%)	Pressure (bar)	Flux (LMH)	enhancement	NaCl rejection (%)	References
Silver		27.5	58.3	Antifouling	98.64	[115]
		20	50	Permeate flux, salt rejection	99.1	[109]
		15.5	52.91	Hydrophilicity, permeate flux, salt rejection	99.18	[108]
		15.5	31	Antifouling, permeate flux, and hydrophilicity	99.4	[116]
Copper		27.5	82.5	Permeate flux	98.31	[113]
	1	20.7	45.12	Hydrophilicity, permeate flux, antifouling	97.4	[114]
Titanium dioxide (TiO2)	0.015	15.5	40.14	Hydrophilicity, permeate flux,salt rejection	99.72	[117]
	0.0125	15.2	24.3	Hydrophilicity, permeate flux, antifouling	97.7	[118]
Titanate nanotubes (TNT)	0.05	15	36.74	Hydrophilicity, permeate flux,salt rejection, antifouling	96.53	[119]
Zinc oxide (ZnO)	0.009	15.5	48	Hydrophilicity, permeate flux, antifouling, chlorine resistance, rejection of salts	99	[104]
	0.03	20	25.6	Hydrophilicity, permeate flux, antifouling, chlorine resistance, rejection of salts	99.3	[105]
	0.02	20	23.85	Hydrophilicity, permeate flux, antifouling	97	[120]

Alumina (Al2O3)	1	10	4	permeate flux, Hydrophilicity, antifouling, salt rejection	88	[106]
MOF-based-ZIF8	0.4	15.5	52	permeate flux, Hydrophilicity, antifouling, salt rejection	99.5	[54]
	0.4	15	34.5	Hydrophilicity, permeate flux, salt rejection	99.4	[121]
	0.2	15.5	61.2	Permeate flux, salt rejection	99.2	[122]
	0.005	16	17.6	Hydrophilicity, permeate flux	99.8	[123]
	0.15	20	52.2	Hydrophilicity, permeate flux, salt rejection	98.6	[124]
MOF-based UiO66	0.05	15.5	56.9	Permeate flux, Hydrophilicity, salt rejection	99.35	[125]
MOF-based UiO66-NH2	0.02	20	40	chlorine resistance, Hydrophilicity, permeate flux, salt rejection	99.2	[126]
MOF-based MIL101	0.05	16	36	Hydrophilicity, permeate flux, salt rejection	99.1	[125]
MOF-based MIL125	0.3	20.7	74.9	Hydrophilicity, permeate flux, salt rejection	>98.5	[126]
MOF-based PCN-222	0.01	17.2	5.8	Permeate flux	94.5	[127]
Zirconium metal 2 organic cages (Zr-MOCs)	0.06	15.5	22.79	Hydrophilicity, permeate flux	94.7	[128]

4.3. Hybrid materials

Metal-organic frameworks (MOFs) are a new class of hybrid materials that consist of inorganic metal clusters or centers connected by organic linkers [129]. These materials have porous structures in one, two, or three dimensions, making them suitable as porous fillers in membrane technology. MOFs have unique features such as high surface area, high porosity, variable pore size, and easy surface functioning. MOFs can also be incorporated into polymeric membranes for liquid separation applications. Because of the strong interactions between the organic linkers in MOFs and the polymer matrix, MOF-incorporated polyamide (PA) membranes offer advantages such as customizable pore structure and size, as well as better polymer compatibility [93], [130].

Because of their high water permeability, broad surface area, and water stability, zeolitic imidazolate frameworks (ZIFs) have been employed for membrane modification in liquid-separation processes. For example, [121] ZIF-8 nanoparticles were introduced into the PA layer of a thin-film nanocomposite reverse osmosis (RO) membrane, resulting in a 162% increase in water permeate flux when compared to a pristine PA membrane. Aljundi [121] It was discovered that adding 0.4 weight percent of ZIF-8 to the PA layer improved the membrane's hydrophilicity and increased permeate flux without altering ion rejection. Furthermore, TFN membranes containing ZIF-8 nanoparticles demonstrated superior antifouling characteristics as compared to ordinary PA membranes. The size of the ZIF-8 particles inserted in the PA layer also influences TFN membrane performance. [121].

Other MOFs, such as Zr-based UiO-66 and Cr-based MIL-101, have also been utilized in TFN RO membrane preparation due to their good water stability and pore size characteristics. Liu et al. [131] When UiO-66 was added to the PA layer of a TFN membrane, it increased water flux by 50% compared to a TFC membrane while preserving equivalent salt rejection. Because of its good water stability and increased pore size, MIL-101 has been widely used in membrane modification for water treatment [125]. Incorporating 0.05 wt. % MIL-101 in TFN membranes has been shown to enhance water flux by 44% compared to pristine TFC membranes, with salt rejection above 99%.

Titanium-based MOFs, such as MIL-125, have also been utilized in the development of TFN membranes. Kadhom et al. [126] MIL-125 and UiO-66 nanoparticles were mixed in TFN membranes and shown to improve permeate flux and salt rejection compared to virgin TFC membranes. These MOF nanoparticles may be used to improve TFN RO membranes for water purification, particularly in desalination procedures. More research is needed, however, to identify chlorine-resistant MOFs and develop antifouling membranes.

4.4. Non-Organic additives

4.4.1. Silica

Nanoparticles made of Silica (SiO2) have been incorporated into the creation of thin-film nanocomposite (TFN) reverse osmosis (RO) membranes. This integration aims to improve the membranes' performance by enhancing their thermal stability and permeate flux. [132]. The advantages of silica nanoparticles include their low cost, thermal stability, and chemical resistance. In one experiment, the researchers incorporated silica nanoparticles into the TFN membrane's polyamide (PA) layer. The findings demonstrated that the concentration of silica nanoparticles may be changed to alter the pore size of the PA layer [132]. Another study incorporated SiO2 nanoparticles into the PA layer of the TFN membrane for high-pressure desalination applications. It was found that the permeate flux increased with the concentration of SiO2 nanoparticles, reaching a maximum of 50 L m22 h21 [133]. However, the salt rejection initially increased with the concentration of SiO2 nanoparticles but decreased after a certain concentration, possibly due to nanoparticle agglomeration within the PA layer [133].

In order to improve the effectiveness of TFN RO membranes in blocking salt and increasing water flow, hydrophobic fluorinated silica nanoparticles were incorporated during the preparation process [134]. For the interfacial polymerization (IP) process, these nanoparticles were disseminated in an organic phase. The salt rejection rose with the concentration of fluorinated silica nanoparticles up to 0.1 wt. %, but after that decreased [134]. This decrease in salt rejection was attributed to the hydrophobic nature of the nanoparticles. Additionally, the water flux continuously decreased with increasing nanoparticle loading due to the hydrophobicity of the fluorinated silica nanoparticles [134].

A new technique has been suggested for adding SiO2 nanoparticles to the PA layer of TFN membranes. This involves using a process called in situ IP, which combines aqueous amine and silicon tetrachloride (SiCl4) solutions. [135], by using this technique, the membrane's water permeability has greatly improved without sacrificing its salt rejection capabilities [135].

Overall, the incorporation of silica nanoparticles into TFN RO membranes has shown promise in improving their thermal stability, permeate flux, and salt rejection. However, careful control of nanoparticle concentration and dispersion is necessary to avoid agglomeration and maintain optimal membrane performance.

4.4.2. Halloysite (aluminosilicate)

Numerous research on the adoption of Halloysite Nanotubes (HNTs) in the manufacture of Mixed Matrix Membranes (MMMs) for Ultrafiltration (UF) or Nanofiltration (NF) applications have been done. However, there has been little research on the use of HNTs in Thin Film Nanocomposite (TFN) Reverse Osmosis (RO) applications. Ghanbari et al. [136] created a TFN membrane with high flux and antifouling capabilities using hydrophilic HNTs. The TFN membranes were created utilizing the Interfacial Polymerization (IP) process, which distributed HNT nanoparticles in the organic phase. The inclusion of HNTs in the TFN membrane resulted in a greater flux compared to the pristine Thin Film Composite (TFC) membrane while maintaining a comparable salt rejection rate of 95.6%, according to the researchers. The higher water flux can be attributed to the membrane's increased hydrophilicity due to the presence of HNTs. Furthermore, the inclusion of HNTs in the TFN membrane's Polyamide (PA) layer boosted its antifouling capabilities.

4.4.3. Zeolite

The employment of zeolites for changing reverse osmosis (RO) membranes has increased significantly in recent years. This is mostly due to their chemical stability and ion rejection capacity [137], [138]. One method of enhancing the performance of thin film nanocomposite (TFN) RO membranes is by incorporating zeolites. Dong et al. [139] created a high flux TFN RO membrane by combining m-phenylenediamine (MPD) and trimesoyl chloride (TMC) in an interfacial polymerization process with the insertion of NaY zeolite into the polyamide (PA) layer. The NaY zeolites enhanced permeate flux without impacting salt rejection, according to the researchers. The TFN membrane displayed twice the flux after post-treatment as compared to the original NaY-doped TFN membrane. Other research has demonstrated that integrating zeolites into TFN RO membranes can improve permeability and affect interfacial parameters such as charge density, hydrophilicity, pore structure, antibacterial capabilities, chemical stability, and mechanical stability. Huang et al. [40] investigated the effects of incorporating NaA nano-zeolites into the PA layer of TFN RO membranes using an IP process. They found that TFN membranes with zeolites dispersed in organic solutions exhibited better performance compared to those with aqueous dispersed zeolites.

4.5. Organic additives

4.5.1. Cellulose

One of the major drawbacks of thin-film nanocomposite (TFN) reverse osmosis (RO) membranes is the possible leaching of nanoparticles into both the retentate and permeate streams, which might have negative environmental consequences [140]. To address this issue, scientists investigated the addition of cellulose nanocrystals (CNCs) to the polyamide (PA) layer to develop TFN membranes. CNCs are tiny rod- or needle-shaped particles formed by acid hydrolysis of natural cellulose. Because of their negative surface charge and huge surface area, they are suited for use in membranes. CNCs are also environmentally friendly, biodegradable, and non-toxic, making them ideal for the development of TFN membranes.

In a study conducted by Asempour et al. [141] a new type of membrane for desalinating brackish water was created by adding 0.05-0.1 wt. % of CNCs to the PA layer, resulting in a two-fold increase in flux compared to a regular TFC membrane. The new membrane also showed antifouling properties, which is very beneficial for membrane applications. Various techniques can be used to modify the surface of CNCs. [142]. For instance, the surface of CNCs can be altered through 2, 2,6,6-Tetramethylpiperidine-1-oxyl (TEMPO)-mediated oxidation [143]. Smith et al. [144] developed a high-flux reverse osmosis membrane by integrating into the PA layer cellulose nanocrystals treated with CNCs and TEMPO oxidation.

Finally, the integration of cellulose nanocrystals (CNCs) in thin-film nanocomposite (TFN) reverse osmosis (RO) membranes shows promise in overcoming the restrictions of possible nanoparticle leaching and improving membrane performance. TFN membranes were effectively produced with CNCs, resulting in better flux and antifouling properties.

Further research into surface modification techniques for CNCs may lead to ever more sophisticated TFN membranes for water desalination and other membrane applications.

Table 3. Nanoparticle Comparison in Thin-Film Nanocomposite Reverse Osmosis Membrane Prepared by Interfacial Polymerization (IP)

Filler material	Quantity of NPs (wt.%)	Pressure (bar)	Flux (LMH)	Performance enhancement	NaCl rejection (%)	Reference
Silica (SiO2)	0.04	17	21.3	Permeate flux, salt rejection	91.1	[145]
	0.1	44	50	Hydrophilicity, permeate flux, antifouling	95	[133]
Silica SBA-15	0.1	15	74.17	Hydrophilicity, permeate flux, salt rejection	98.5	[146]
Alkyl capped silica	0.1	15.5	55.3	Permeate flux, salt rejection	99.6	[147]
Halloysite nanotube (HNT)	0.05	15	36	Hydrophilicity, permeate flux, antifouling	95.6	[136]
Zeolite A nanocrystals	0.4	12.4	17	Hydrophilicity, permeate flux, antifouling, salt rejection	93.9	[24]
Sodium zeolite Y (NaY)	0.75	15.5	74	Hydrophilicity, permeate flux, salt rejection	98.8	[139]
NaA nanozeolites	0.1	16	35	Permeate flux, salt rejection	97.5	[40]
Zeolite (NaX) nanocrystals	0.2	12	14.6	Hydrophilicity, permeate flux, salt rejection	96.4	[39]
S-beta zeolite	0.05	20.7	65.25	Hydrophilicity, permeate flux, salt rejection	97.33	[148]

Cellulose nanocrystals (CNC)	0.1	20	63	Hydrophilicity, permeate flux, antifouling	97.8	[141]
Laponite nanoclays (NC-LAP)	0.3	20	54	Hydrophilicity, permeate flux, salt rejection	98.18	[149]
Boron nitride nanosheets	0.02	15.5	64	Hydrophilicity, permeate flux, salt rejection	96.4	[150]
Twodimensional (2D) MXene Ti3C2Tx	0.015	16	40.5	Hydrophilicity, permeate flux, antifouling, chlorine resistance		[151]

5. Challenges facing membrane technology

Significant research is necessary to enhance the efficiency of TFN membranes. There are several obstacles that membrane technology currently faces;

- Fouling: The accumulation of particles, solids, or organic matter on membranes can reduce their efficiency and lifespan. Developing effective strategies to prevent fouling is a major challenge.

- Scaling: Deposits of minerals and salts on membrane surfaces can decrease their performance. Finding ways to control and prevent scaling is important for maintaining membrane efficiency.

- Membrane degradation: Membranes can degrade over time due to chemical attack, high pressure, or harsh conditions. Enhancing membrane durability and stability is crucial for long-term performance.

- High energy consumption: Membrane processes often require significant energy inputs, especially in reverse osmosis. Researchers are working on reducing energy consumption without compromising performance.

- Cost-effectiveness: Membrane technology can be expensive, particularly for large-scale applications. Finding ways to lower costs and improve the economic feasibility of membrane processes is a challenge.

- Environmental impact: The production and disposal of membranes, as well as the chemicals used in membrane processes, can have environmental consequences.

Developing sustainable and environmentally friendly membrane technologies is a challenge.

- Scaling up: While many membrane technologies have been successful at the laboratory scale, scaling up to industrial or commercial applications can be challenging. Developing efficient and cost-effective manufacturing processes for large scales is a key challenge.

6. Conclusion

In conclusion, hybrid materials such as metal-organic frameworks (MOFs) and zeolitic imidazolate frameworks (ZIFs) have shown great potential in enhancing the performance of thin-film nanocomposite (TFN) reverse osmosis (RO) membranes. These materials offer unique features like high surface area, high porosity, variable pore size, and easy surface functioning, making them suitable for incorporation into polymeric membranes. MOF-incorporated polyamide (PA) membranes provide advantages such as customizable pore structure and size, as well as better polymer compatibility. ZIFs have been employed for membrane modification in liquid-separation processes, resulting in increased water permeate flux and improved antifouling properties.Silica nanoparticles have also been incorporated into TFN RO membranes, showing promise in improving their thermal stability, permeate flux, and salt rejection. However, careful control of nanoparticle concentration and dispersion is necessary to avoid agglomeration and maintain optimal membrane performance. Cellulose nanocrystals (CNCs) have been considered as an environmentally friendly and biodegradable additive for TFN RO membranes, resulting in improved flux and antifouling properties. Further research into surface modification techniques for CNCs may lead to more sophisticated TFN membranes for water desalination and other membrane applications. Despite the significant progress in enhancing TFN membrane performance, there are still challenges to overcome, such as fouling, scaling, membrane degradation, high energy consumption, cost-effectiveness, and environmental impact. Addressing these challenges will require continued research and development in the field of membrane technology.

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