A review on different nanomaterials-based hollow fiber membranes for water desalination

Abstract

This paper intends to review the state of hollow fiber membranes embedded with different nanomaterials--carbon nanotubes (CNTs), graphene oxide (GO), zeolites, aquaporin (AQP) for water desalination. An ultra-strong polymeric hollow fiber membrane was mainly discussed in details. In addition, characteristics of these nanomaterials and their application as well as performances in desalination after incorporated with hollow fiber membranes were also briefly discussed. It can be seen that the use of these nanomaterials in hollow fiber membranes for reverse osmosis (RO), membrane distillation (MD), forward osmosis (FO), pervaporation, organized the main body of the reported project in this review. Apart from that, a hypothesis that to combine ultra-strong polymeric hollow fiber membranes with these nanomaterials was proposed. According to our best knowledge, only few articles have worked on studying nanomaterials-based hollow fiber membranes for desalination.

1. Introduction

Water is one of the most essentials of life and how to sustainably supply it is a huge and major challenge for humans as the growing population and rising standards of living bring significant consumption of them. In developing countries, the infrastructure to supply water and manage wastewater in many cases simply does not exist, and billions of people have no access to basic services because of economic and social obstacles.[1] According to the data of 2015, there were still over 840 million people who cannot drink safe water, and approximately 2.3 billion people did not have basic sanitation services. In sub-Saharan Africa and central and southern Asia, the situation is most severe.[2] Developed countries have established systems for producing and delivering water to their citizens, but these systems often waste resources and discharge harmful pollutants. In many places, water and sanitation infrastructure has exceeded its planned time limit for construction, posing significant challenges in maintaining the expected water quality and reliability.[1] Much of the country's water and wastewater treatment infrastructure was built in the 1970s and 1980s.[3] The average age of US water-network pipe is 45 years old, moreover, some cast-iron pipes have been used for more than a century old.[4] In the United States, typically 14 to 18 percent of total daily treated potable water is lost through leaks and some water systems are reported that water-loss rates exceed 60 percent.[3]

Therefore, water scarcity is a huge challenge globally. However, as we all know that 70% of earth is covered by ocean, it is the maldistribution of freshwater resources that contribute to water scarcity. Only 3% of the whole water on the earth is freshwater. And only small fraction of the freshwater is easily available since glaciers and deep underground are the largest reservoirs which are unreachable normally. Clearly, seawater is a big treasure from which we can get endless water. Considering that conventional water treatment processes are not suitable for seawater because of high salinity of seawater. Thus, desalination came out and solved the problem. Honestly, desalination has been mature to supply safe drinking water in a lot of places such as deserts and remote area.[5] Among 4 main categories of seawater desalination processes which are thermal energy, mechanical energy, electrical energy, and chemical energy, self-fouling, huge power consumption, need of intense resources and economical issues limited the widespread applications of these technologies.[6]

Speaking of economical issues, hollow fiber membrane is widely applied to wastewater treatment plants and other industrial factories. And the characteristics of hollow fiber membrane are the reason why it is popular in industry. First, the pressure resistance of hollow fiber membrane mainly depends on the ratio of inner diameter to outer diameter, and has nothing to do with the thickness of tube wall. So, the pressure resistance of hollow fiber membrane is very good. Second, hollow fiber membrane does not need support and the process of using hollow fiber membrane is simple with no phase change and energy saving. Third, the membrane module can be made into any size and shape with strong applicability. Fourth, the effective membrane area per unit volume is large with high filling density and good permeability. Fifth, the application range of hollow fiber membrane is wide and it is suitable for the separation of a variety of macromolecular organic matter and inorganic matter.[7] Notably, Liang et al. developed ultra-strong polymeric thin film composite (TFC) hollow fiber membranes with exceptionally high hydraulic burst pressures of up to 110 bar, indicating high pure water permeance and a NaCl rejection of about 98%. This work provides a new idea for preparing super-strong TFC hollow fiber membranes for water treatment and desalination.[8]

In order to resolve the stated obstacles of water desalination, a lot of researchers have developed some novel technologies. In recent years, developments of nanomaterials have attracted much attention in water purification systems.[9] Nanotechnology is promising to provide high performance, economical water and wastewater treatment solutions because of the highly efficient, modular, and multifunctional processes of nanotechnology. And it has the potential to not only conquer challenges of existing water treatment technologies, but also develop new methods which are economically applied to expending the utilization of unconventional water resources.[10]

The definition of nanomaterials is that this material should be smaller than 100nm in at least 1 dimension. When material at this scale, they often show some novel size-dependent properties which are different from their large counterparts. And a lot of these nanomaterials have been explored and identified as suitable material for membrane filtration. These include magnetic nanoparticles (NPs), noble metal NPs, carbon nanotubes (CNTs), nanoscale metal oxide, nanofibers, zeolites, aquaporin (AQP), graphene, etc. Qu Alvarez et al. concluded Table 1 to show the current applications and properties of nanomaterials in membranes and membrane processes.[9]

Because of favorable features of large surface area, long range of porosity, superior thermal and electricity conductivity, and exceptional mechanical strength and stiffness, carbon-based nanomaterials (CNMs) such as CNTs have especially attracted much attention of scholars and researchers.[11,12] It has been improved that the novel properties of CNTs and graphene oxide (GO) constitute an important potential breakthrough in water desalination. In previous research studies of the transport properties of CNMs, the facts that smooth and frictionless graphitic walls of CNTs and rapid sorption-desorption mechanism could facilitate rapid transport of water molecules have been commonly found. Thus, high flux separation performance can be offered.[13,14] Nevertheless, zeolites, as another new versatile, unique and low cost nanomaterial , they had also been widely employed in seawater desalination.[15–23] Zeolite is a potential nanomaterial with high surface area because of its porous structure with many channels and cavities.[24] The pores sizes of zeolites are between the diameter of hydrated salt ions and water molecules, which is favorable for zeolites to remove ions from saline water, contributing to exceptional performance in water purification.[25,26]

Table 1 Current applications and properties of nanomaterials.[9]

Application	*Representative nanomaterials*	*Desirable nanomaterials properties*	*Enable technologies*
Membranes and membrane processes	Nano-zeolites	Molecular sieve, hydrophilicity	High permeability thin film nanocomposite (TFN) membranes
	Nano-Ag	Strong and wide-spectrum antimicrobial activity, low toxicity to humans	Anti-biofouling membranes
	CNTs	Antimicrobial activity	Anti-biofouling membranes
		Small diameter, atomic smoothness of inner surface, tuneable opening chemistry, high mechanical and chemical stability	Aligned CNTs membranes
	AQP	High permeability and selectivity	AQP membranes
	Nano-TiO2	Photocatalytic activity, hydrophilicity, high chemical stability	Reactive membranes, high performance TFN membranes
	Nano-magnetite	Tuneable surface chemistry, superparamagnetic	Forward osmosis (FO)

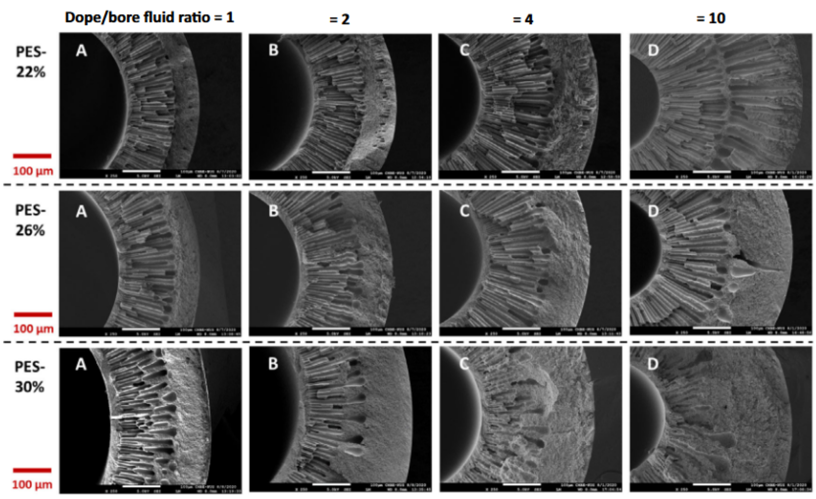
Furthermore, because of AQP proteins, biological membrane becomes the most effective way for water transport across an osmotic pressure gradient.[27] AQP are water channel proteins and they are bounded with phospholipid cellular membrane. They only allow water molecules to penetrate through the membrane and keep ions species out the membrane.[23] In the last decade, an increasing number of researchers have studied to develop an artificial membrane to mimic the natural cellular membrane since the discovery of AQP.[28,29] Pendergast & Hoek et al. thought that bio-inspired membrane - aquaporin-based biomimetic membrane (ABM) was able to offer the best opportunity for revolutionary performance because it had opened up an exciting direction in water desalination so that it could provide perfect selective channels for water with extraordinary osmotic permeability.[30]

Researchers had reported some literature reviews on the aforementioned nanomaterials for water desalination.[10,13,31–33] However, all these reviews only concentrate on types of nanomaterial for a specific water desalination technology and the contents of the article basically consist of the characteristics of nanomaterials, their preparation and various endeavors for exploring the nanomaterial in a specific water desalination technology and their performance, future perspectives of the technology for desalination and water reuse. It is not found that a literature gave an overview of different nanomaterials-based hollow fiber membranes for various water desalination technologies. Therefore, the objective of this paper is to review the state of the art of different nanomaterials-based (CNTs, GO, zeolites, AQP) hollow fiber membrane for water desalination. Furthermore, we also introduce ultra-strong polymeric hollow fiber membranes for saline dewatering and desalination.[8] Finally, the conclusions and future perspectives are proposed.

2. Ultra-strong polymeric hollow fiber membrane

Osmotically assisted reverse osmosis (OARO) process is a promising technology to achieve high water recovery (e.g., >50%). The mechanism of OARO is that when water penetrates the membrane, it can be driven by the pressure which is lower than the actual osmotic pressure because a stream with a no more than salinity keeps sweeping in the permeate side. However, in the process of OARO, the burst pressure of membrane still limits the water recovery.[34,35] Therefore, a membrane with a strong structure and efficient properties is significant for OARO process.

Herein, Liang et al. designed ultra-strong inner-selective thin-film composite (TFC) hollow



fiber membranes to realize the requirements of strong and efficient membranes with high water permeability of 3 litre/(m^2 h bar) and a salt rejection of 98%. According to the results of experiments, high polymer concentration of the spinning dopes could increase the mass transport resistance as well as lower the water permeability of the membranes. As Fig. 1 showed, A, B, C, D represent the simplified code names of PES hollow fiber membranes with specific ratio of dope to bore fluid flow rate as shown on top of the figure. And notably, the number of the finger-like macro-voids in the cross-section decreases while the sponge-like microstructure increases as dope/bore fluid ratio and PES concentration both increase.[8]

Fig. 1 FESEM morphologies of selected cross-sections of the as-spun PES hollow fiber membrane substrates.[8]

Moreover, adjusting the inner diameter and wall thickness of hollow fibers can tune the dimension of them and with this configuration, this kind of membrane would be simple to subsequently scale up and produce. Fig. 2 highlighted FESEM morphologies of representative PES hollow fiber substrates which were spun at the dope to bore fluid ratio of 10 as well as compared them spun from different PES concentrations. And we can see that the hollow fiber (P22-D) spun from a dope containing a low PES concentration of 22 wt% has a dual-layer structure of finger-like macro-voids, while the others (P26-D and P30-D) possess only one layer of finger-like macro-voids. Liang et al. explained that the nascent hollow fiber substrate had a dual-layer structure of finger-like macro-voids as result of it spun from a dope with a low viscosity, leading to non-solvent intrusion from both the internal and external coagulants.[8]

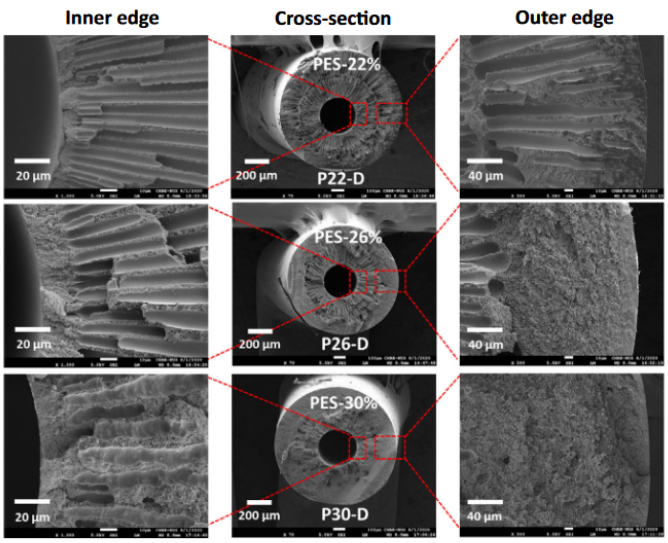


Fig. 2 FESEM morphologies of representative PES hollow fiber substrates.[8]

As shown in Fig. 3a, b, when feed sides were at a given salt concentration, the water flux and water permeability of the three optimal membranes follow the order of PT22-D > PT26-D > PT30-D. This is because the structural parameters of the three membranes are in the order of PT22-D < PT26-D < PT30-D. And with smaller structural parameters, the internal concentration polarization (ICP) effect would be smaller.

Fig. 3c, d depicted the effects of the operating pressure on water flux and water permeance. It was interesting to find that the water permeance displays a V-shape trend in Fig. 3d. The reason why the trend in water permeance declined firstly and then increased may come from the combined effects of both ICP and membrane expansion at high pressure. The water flux increased with the increasing of operating pressure, which contributed to an ICP diluting effect. Thus, the water permeability declined.[8]

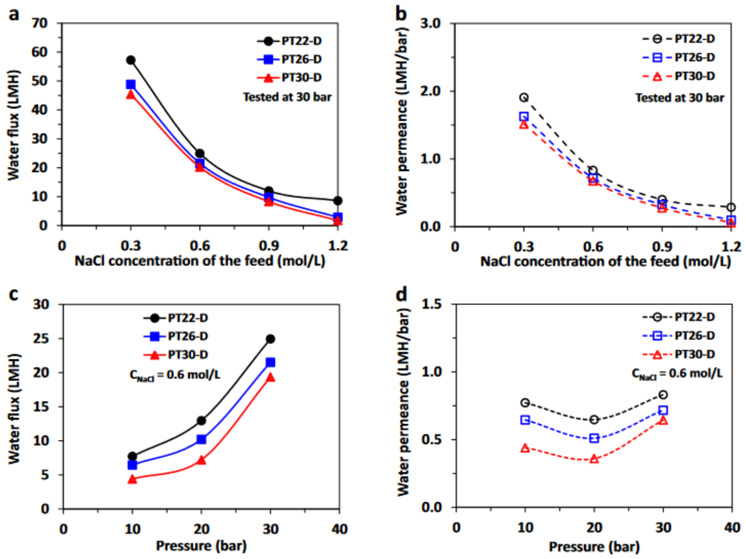


Fig. 3 The OARO performances of the optimal PES-TFC hollow fiber membranes.[8]

3. Carbon-based nanomaterials

3.1 Carbon nanotubes

CNTs are allotropes of one-atom-thick carbon sheets. And this carbon sheets can be rolled into long, hollow cylindrical nanostructure to make CNTs. The diameter of CNTs can be as small as 1 nm and the length can be up to several centimeters. Among any known material, CNTs have the highest ratio of strength-to-weight because the type of bond holding the carbon atoms in CNTs is very strong. Single CNTs are naturally arranged into "cords" that are connected by van der Waals forces, which is also called π stacking. And the electrical charge of CNTs can move freely with the electron localization as result of the hexagonal lattice of carbon. On the other hand, it was reported that CNTs have exceptional adsorptive properties against both chemical and biological contaminants such as heavy metals, phenols, organic chemicals, and various types of natural organic matter (NOM). Herein, CNTs are promising to be applied to water and wastewater treatment such as desalination on account of their superior properties (e.g., high strength, low weight and special electrical and adsorptive properties).[36–41]

Fan et al. found that with electrochemical assistance, superhydrophobic CNT (SCNT) hollow fiber membrane showed high water flux and salt rejection, and antifouling during membrane distillation (MD). Fig. 4 is the mechanism of the electrochemical assistance (EA). When the membrane works as cathode, electrostatic repulsion exists between the negatively polarized SCNT hollow fiber membrane and negatively charged NOM. The repulsion forces probably contribute to the enhanced antifouling ability under negative polarization. And as Fig. 5a showed, the SCNT hollow fiber membrane presented a stable flux of 30 kg m^{-2} h^{-1} after 36 h operation under negative polarization (voltage of −0.5 V), much higher than that (25.5 kg m−2 h−1) under open circuit, indicating that SCNT hollow fiber membrane had good fouling resistance. From Fig. 6, it could be judged that CNT hollow fiber membrane had highest water flux and salt rejection (lowest conductivity).[42]

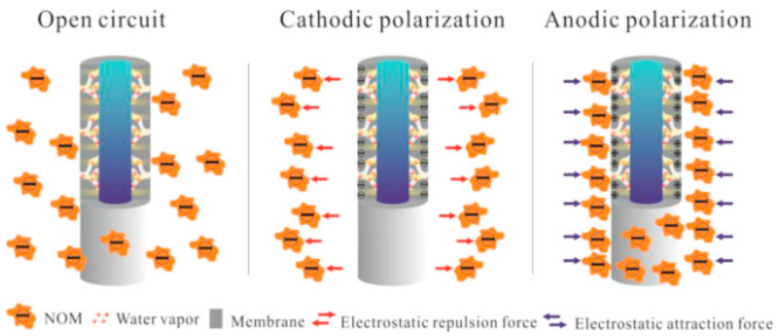


Fig. 4 Mechanism of electrochemically assisted membrane distillation.[42]

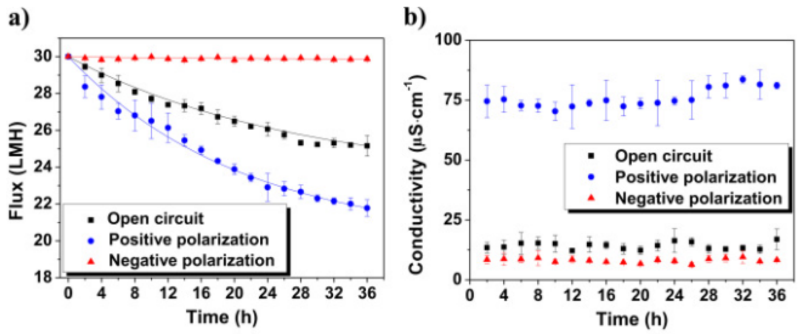


Fig. 5 (a) Normalized flux, (b) desalted water for the superhydrophobic CNT hollow fiber membrane under open circuit, negative polarization and positive polarization for 36 h operation.[42]

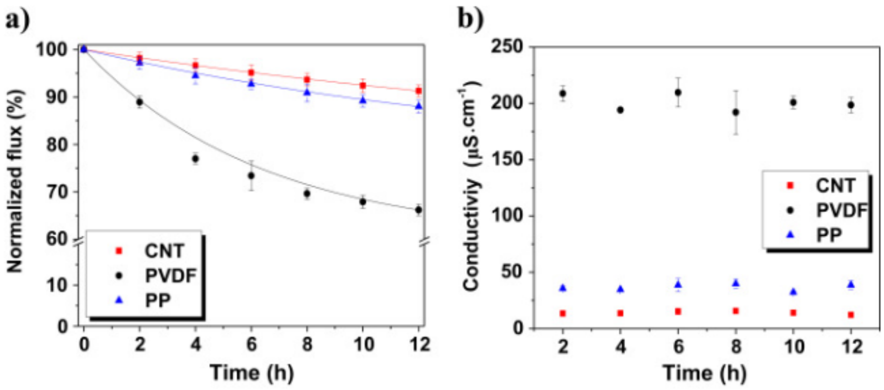


Fig. 6 (a) Normalized flux and (b) conductivity of desalted water for the PVDF, PP and superhydrophobic CNT hollow fiber membrane. (Feed solution: 3.5 wt% NaCl, 50 mg L−1 NOM, 60 mM Na2SO4, and 20 mM CaCl2).[42]

In 2019, Fan et al. added a reduced GO (RGO) active layer on the foundation of CNT hollow fiber membrane and applied it to FO. And it showed low internal concentration polarization as result of high porous and hydrophilic substrate and high water flux and low reverse salt flux during forward osmosis process.[43]

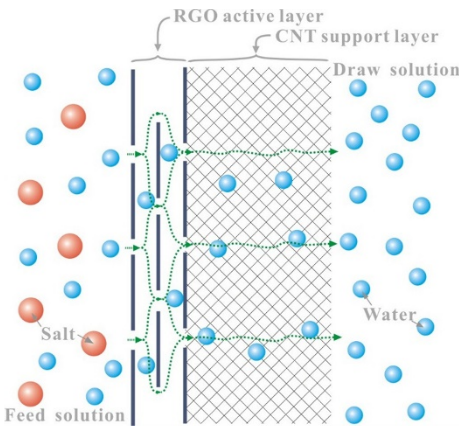


Fig. 7 Model of RGO/CNT hollow fiber membrane.[43]

3.2 Graphene oxide

Graphene is a robust but flexible membrane providing essentially infinite possibilities because of the modification or functionalization of its carbon backbone. And GO offers potential to produce chemically modified graphene (CMG) on the ton scale. About 150 years ago, GO was first prepared and since then, it had become a precursor to offering the potential of cost-effective, large-scale production of graphene-based materials.[44]

Huang et al. synthesized a novel GO-polyimide (GO/PI) hollow fiber membranes and evaluate its performance on desalination by pervaporation. It was proved that the GO/PI hollow fiber membranes showed exceptional water permeance and salt rejection because the homogeneous dispersion of GO nanosheets in the PI matrix could form continuous and robust membranes. As Fig. 8 showed, the water flux is 11.5 kg m^{-2} h^{-1} as well as the ion rejection is over 99.8% at 75 °C, which indicated the high separation performance for desalination of the GO/PI hollow fiber membrane.[45]

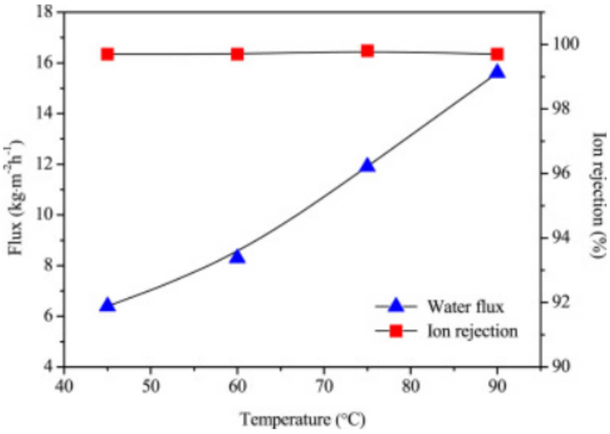


Fig. 8 Water flux and ion rejection of the GO/PI hollow fiber membrane as a function of the operation temperature for desalination of 3.5 wt% seawater by pervaporation.[45]

Goh et al. designed a GO surface deposited poly(amide-imide)–polyethyleneimine (PAI–PEI) hollow fiber membrane which could not only perform up to 86% higher permeability in pure water as result of the small hydrodynamic resistance of GO nanosheets but also shorten the cross-linking time of membrane from original 90 min to 30 min due to the less excessive PEI cross-linking. It could be seen from Fig. 10a, c that both pure water permeability coefficient and salt permeability coefficient of the membranes with GO were lower than those without GO, implying that GO nanosheets worked as effective barriers. And as shown in Fig. 10b, MgCl2 salt rejection of the membranes with GO was correspondingly higher than those without GO.[46]

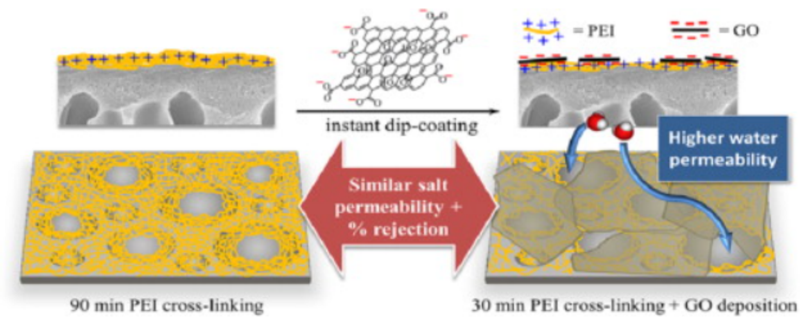


Fig. 9 Mechanism of GO deposited PAI_PEI hollow fiber membranes.[46]

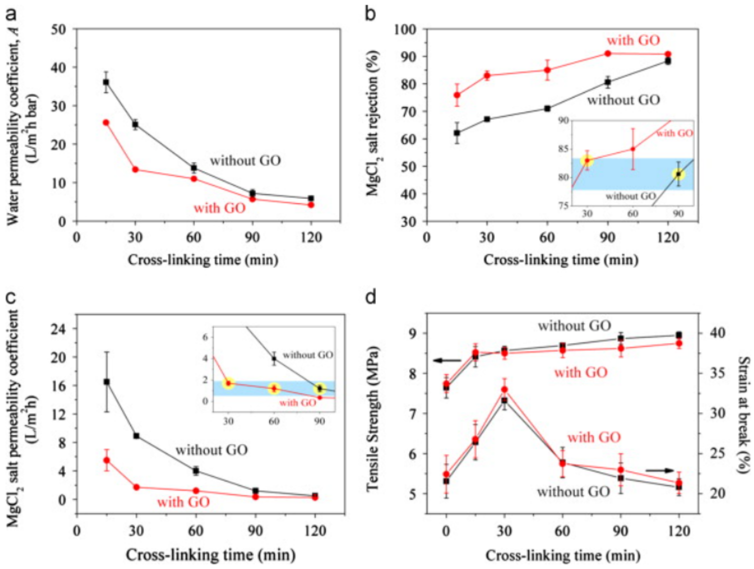
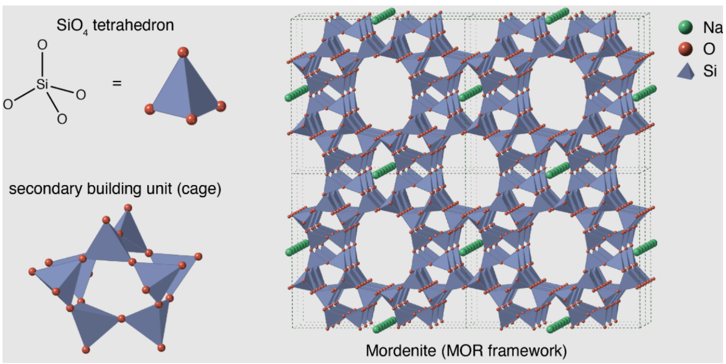


Fig. 10 Results showing (a) pure water permeability coefficient; (b) MgCl2 salt % rejection and (c) salt permeability coefficient of the respective hollow fiber membranes (with and without GO) at different cross-linking times. The inserts highlight similar results between PAI-PEI(90) and GO_PAI_PEI(30) membranes; (d) Mechanical properties of the PAI UF substrate and its respective modified hollow fiber membranes at different cross-linking times.

4. Zeolites

Zeolites are microporous, crystalline and aluminosilicate materials which contain alkali and alkali-earth metals. As shown in Fig. 11, the structure of zeolites is a 3D tetrahedral framework and each oxygen atom is shared by two tetrahedra. The zeolites framework has cavities and channels inside, which allows the resident ions easily to drift and makes molecules freely get into and out of the structure. And this process is a cation exchange. These unique physical and chemical properties make zeolites a novel surface selectivity, leading to high adsorption ability,



sieving capability, and ion exchange property. Thus, zeolites are potential to be employed widely for seawater desalination.[10]

Fig. 11 Microscopic structure of a zeolite framework

Makhtar et al. had incorporated A-type zeolite membranes into porous glass hollow fibers for desalination. When it was in the preparation of porous glass hollow fibers, phase inversion and sintering technique were employed. And yttria stabilized zirconia (YSZ) was added to improve porosity so that pure water permeability could increase. As shown in Fig. 12, it is proved that the solute fluxes for 5,000 and 10,000 ppm NaCl salt solutions were 24.45 and 17.86 L m^{-2} hr^{-1} in the experiments of OARO, respectively. And the salt rejections were both up to 98%.[47]

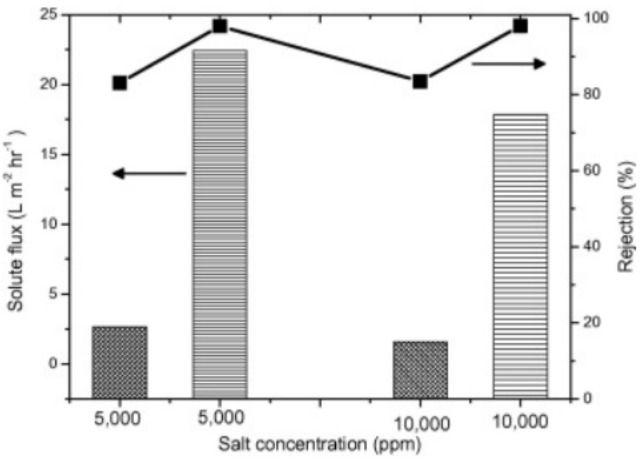


Fig. 12 Water permeability of RO-NaCl , OARO-NaCl for zeolite membranes, prepared using in-situ hydrothermal method for 12 h at 120 °C.[47]

5. Aquaporin

AQP is the best selective material contributed by nature. Actually, researchers have applied AQP into membrane processes for desalination for a decade. Initially, researchers just incorporated AQP-containing liposomes (proteoliposomes) or polymersomes onto the surface of membrane substrate straightforward to form a dense layer. It was predicted that this dense layer could act as a selector to increase salt rejection and the substrate could be responsible to stand the applied pressure just as the structure of TFC membrane. Simultaneously, AQP could facilitate water molecules to transport, leading to high water flux. However, the external environment can easily affect the activity of the AQP and it was a huge challenge to scale up the production of a large defect-free selective layer via this way.[48]

Li et al. synthesized an AQP-based hollow fiber composite membrane (Fig. 13). The process of the preparation of the AQP-based hollow fiber composite membrane was different from the methods before. First, AQP-containing proteoliposomes were fixed on the inner surface of the hollow fiber membrane substrate. Then AQP-containing proteoliposomes were coated by a polyamide layer formed by a non-gas-assisted interfacial polymerization process.[48]

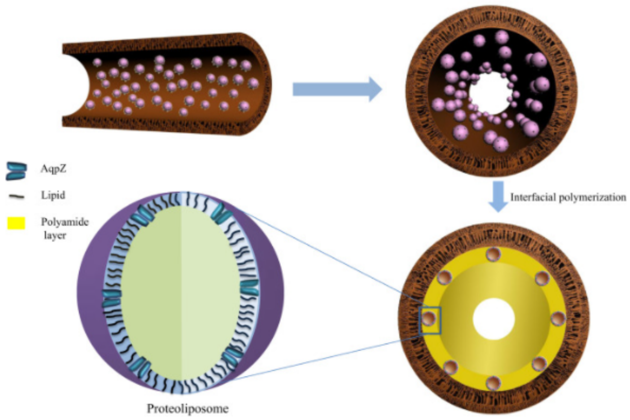


Fig. 13 Schematic of preparation of AQP-based hollow fiber membrane (not to scale).[48]

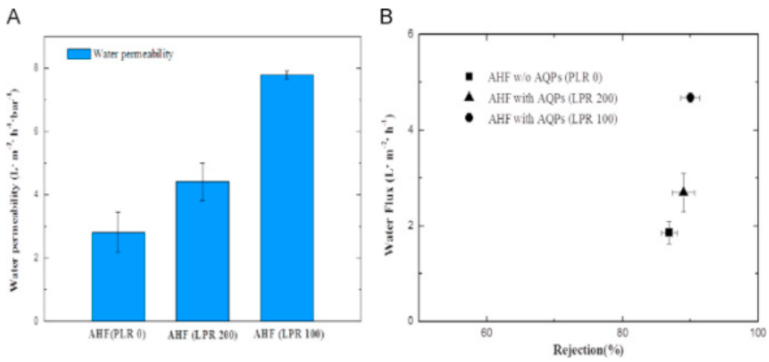


Fig. 14 (A) Pure water permeability of AQP-based hollow fiber (AHF) membranes with different AQP covering densities (PLR 0, LPR 200, LPR 100); (B) Water flux and salt rejection of AHF membranes with different AQP covering densities (testing conditions: 1 bar, 500 ppm NaCl). The AHF with (PLR 0) refers to the hollow fiber membrane with liposomes.[48]

As shown in Fig. 14a, the pure water permeability of the membrane embedded with proteoliposomes (LPR 200) was increased by 60% compared to the control group embedded with liposomes only. As shown in Fig. 14b, even under low pressure, all membranes in test showed high salt rejections (87–90%). Moreover, the pure water permeability was higher than the water flux gained from salt water at the same applied pressure (1 bar). indicating that the osmotic pressure effect existed.[48]

Ren et al. evaluated the FO performance of a unique hollow fiber membrane from Aquaporin A/S, Denmark. This kind of membrane was incorporated with AQP inside. And according to their results of a series of tests, they concluded that this kind of membrane was qualified to be applied to FO desalination. As shown in Fig. 15, the AQP hollow fiber membrane performed very well in both FO and pressure retarded osmosis (PRO) for the high water flux and the low reverse salt flux. Moreover, the AQP hollow fiber membrane showed excellent salt rejection in PRO.[49]

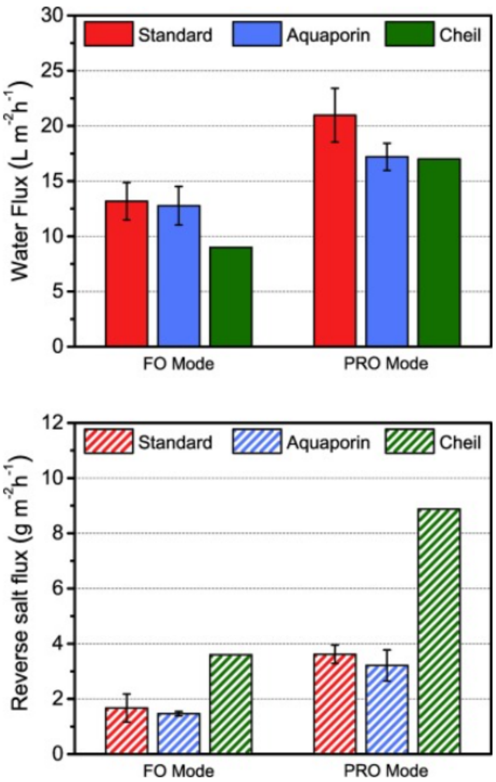


Fig. 15 Water flux and reverse salt flux of FO and PRO mode tests for hollow fiber membranes using Standard and Aquaporin A/S prescribed methods (red and blue bars respectively).[49]

6. Conclusion and future perspectives

Table. 2 Summary of different nanomaterials-based hollow fiber membrane.

Type of nanomaterials	*Applications*	*Interesting highlights*	*Limitations/drawbacks*	*Ref.*
CNTs	·MD	·High flux, salt rejection, and wetting resistance ·Improved antifouling with EA	·Energy consumed by EA ·Preparation of superhydrophobic CNT hollow fiber membrane	42
	·FO	·Low internal concentration polarization	·Fouling ·Wettability of RGO active layer	43

		·High water flux and low reverse salt flux		
GO	·Pervaporation	·Excellent water permeability and 99.8% salt rejection ·High stability	·Smaller tensile strength	45
	·Membrane filtration	·86% water permeability ·Shorter cross-linking time (30min)	·Salt rejection	46
Zeolites	·RO ·OARO	·98% salt rejection ·Higher permeability	·High energy cost	47
AQP	·FO ·RO	·Super-high water flux without an increase in salt flux ·Higher salt rejection	·Low-pressure RO	48
	·FO ·PRO	·Commercially available ·Good performance in FO	·Not better in PRO	49

Hollow fiber membranes are extremely applicable with industries. How to improve its performance in desalination is crucial to solve water scarcity. And the work of Liang et al. made hollow fiber membranes have ability to afford high pressure (100 bar) and achieve high water flux and low salt rejection. On the other hand, nanotechnology contributes to novel and creative medium for desalination. Some nanomaterials (e.g., CNTs, GO, zeolites and AQP) have been applied in many ways (e.g., MD, FO, RO and pervaporation) to increase the efficiency of desalination. And according to the papers I reviewed, it is found that these hollow fiber membranes were incorporated with nanomaterials, all had better performances than before. Therefore, it is reasonable to think that if adding these nanomaterials onto the surfaces of ultra-strong hollow fiber membranes of Liang et al., it is possible to additionally improve the performance of desalination.

As shown in Table. 2, although CNTs hollow fiber membranes could enhance fouling resistance with EA, it would consume more electricity. Also, the synthesis of zeolites hollow fiber membranes needed consume considerable energy. Thus, energy supply is a problem when these membranes need scale-up. Moreover, trade-off between water flux and salt rejection still existed in GO hollow fiber membranes. It is worth noting that AQP hollow fiber membranes showed exceptional performances of both water permeance and salt rejection in FO but were barely satisfactory in the results of RO, especially when the applied pressure was high. This was not surprising since AQP is a protein and cannot stand high pressure. Consequently, servals following suggestions can be minimized the above-mentioned drawbacks of different nanomaterials-based hollow fiber membranes for wide-range application in desalination.

1- The utilization of renewable energy sources for electrochemical treatments and the process of synthesis of different nanomaterials-based hollow fiber membranes, such as sunlight or by wind.

2- To find the most suitable method for different nanomaterials-based hollow fiber membranes. For example, AQP hollow fiber membranes are more suitable in FO compared to RO, then using FO to produce pure water in factories is enough.

In conclusion, the future for the applicability of different nanomaterials-based hollow fiber membrane on large-scale is bright because their applications for desalination is conceptually feasible.

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