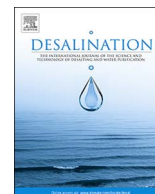




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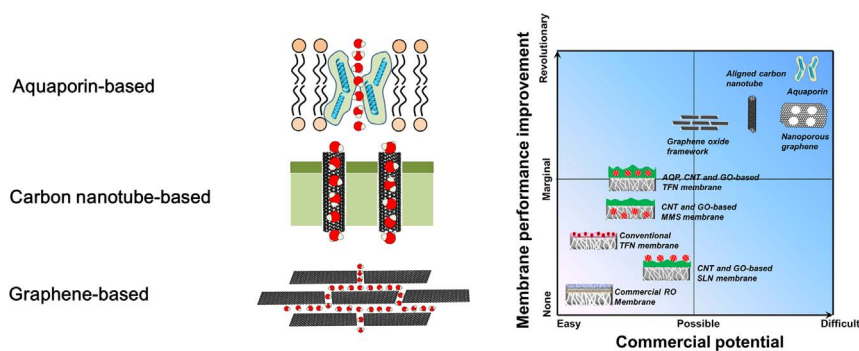
Desalination

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Recent development of novel membranes for desalination

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GRAPHICAL ABSTRACT



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ABSTRACT

In the past decades, novel materials (e.g., aquaporin proteins, carbon nanotubes, nanoporous graphene and graphene oxide) have emerged as promising candidates for synthesizing high performance desalination membranes. These materials can potentially achieve water fluxes of several orders of magnitude higher compared to the state-of-the-art thin-film composite polyamide reverse osmosis membranes. This paper provides a comprehensive summary of the current progresses and challenges in synthesizing aquaporin-based and carbon-based membranes. After a detailed review of the material properties of aquaporin proteins, carbon nanotubes, nanoporous graphene and graphene oxide, a general framework of membrane design and material incorporation is established. The fabrication methods and separation performance for each type of membrane are summarized. Future perspectives of aquaporin-based and carbon-based membranes are discussed in lieu with their ultimate separation performance and commercial scalability.

1. Introduction

With the increasingly growing world population, water crisis has become one of the grand challenges in the 21st century [1]. To date, reverse osmosis (RO) is the key technology for desalting water to alleviate the water scarcity [2]. Membrane is one of the most critical components in an RO desalination plant, and it largely determines the separation performance of the overall plant [3]. A recent analysis [4]

suggests that ultra-permeable membranes (UPMs) with tripling water permeability could save up to 15% energy consumption and use 44% fewer pressure vessels for seawater desalination. In the context of wastewater reclamation, even greater savings (e.g., 45% less energy input and 63% fewer pressure vessels [4]) can be achieved. Furthermore, increasing membrane selectivity results in improved quality of the product water. It can also potentially eliminate the second pass that is commonly adopted for boron removal, which significantly reduces

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the cost of desalination [5]. At the same time, membranes with enhanced antifouling properties reduce the frequency of membrane cleaning and prolong membrane lifespan [6].

Commercial RO membranes are dominated by thin-film composite (TFC) polyamide and its derivatives. These membranes face critical challenges of relatively low water permeability, low selectivity and high fouling tendency [6]. For example, typical water permeability of commercial TFC RO membranes range from $\sim 1\text{--}2 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$ for seawater reverse osmosis (SWRO) membranes and $\sim 2\text{--}8 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$ for brackish water reverse osmosis (BWRO) [7]. Synthesizing novel RO membranes with improved separation properties and better antifouling performance is therefore a key research focus in the field of desalination [6].

Tremendous advancements have been achieved in making ultra-permeable and antifouling membranes. Much of the exciting progresses are fueled by the recent emergence of promising novel materials for desalination. Among them, the most notable examples include aquaporin (AQP) proteins [8,9] and some carbon-based materials such as carbon nanotubes (CNTs) [10] and graphene-based materials [11,12]. These novel materials provide new dimensions for designing next-generation RO membranes. Fig. 1 shows the increasing number of publications in the recent decade on these topics.

This paper provides a comprehensive review of the recent progresses of novel desalination membranes prepared using AQPs, CNTs, and/or graphene-based materials. Following a brief presentation of conventional TFC polyamide membranes and its derivative thin film nanocomposite (TFN) membranes (Section 2), we provide a detailed summary of the material properties of AQPs, CNTs, nanoporous graphene and graphene oxide as well as a general framework for classifying novel RO membranes on the basis on how and where the materials are loaded into the membranes (Section 3). The fabrication methods using each type of materials and the properties of the corresponding membranes are then reviewed in Section 4–6, and the future perspectives and limitations of these novel membranes are also highlighted in Section 7.

2. Conventional and novel membrane materials

Although early RO membranes were of asymmetric cellulose acetate membranes, they have been largely replaced by TFC polyamide membranes [13]. Compared to the former, TFC polyamide membranes show better water permeability and salt rejection (e.g., $> 99.9\%$ NaCl rejection for some SWRO membranes), wider operating temperature range ($0\text{--}45^\circ\text{C}$) and better pH tolerance ($1\text{--}11$) [7]. A typical TFC membrane consists of three layers (Fig. 2a): a dense polyamide rejection layer, a porous substrate commonly made of polysulfone or polyethersulfone, and a non-woven fabric layer as a mechanical support. The polyamide rejection layer, which determines the water permeability and salt rejection of the membrane, can be prepared by the interfacial polymerization (IP) of an amine monomer in aqueous phase and acyl chloride monomer in organic phase (Fig. 2b). Researchers have applied various attempts to optimize the structure and chemistry of TFC polyamide membranes in order to enhance their separation performance and/or antifouling properties [7]. Typical approaches include changing monomer types and concentrations [14–18], membrane surface modification [19–22] and post-treatment [23–25]. Despite that many of these attempts lead to marginal improvements in membrane separation properties, the water permeability of the state-of-the-art TFC polyamide membranes remain relatively low.

A major improvement to TFC membranes was introduced by Hoek and co-workers in 2007 [26]. These researchers incorporated porous zeolite nanoparticles (NaA, entrance pore size $\sim 4 \text{ \AA}$) into the polyamide rejection layer during the interfacial polymerization process. In this membrane structure that is now termed as thin film nanocomposite (TFN, see Fig. 2a), the porous zeolite nanoparticles provide preferential water pathways with reduced hydraulic resistance while excluding

dissolved salts by size exclusion. The water permeability of the TFN membrane was nearly doubled compared to the TFC membrane that has no zeolite added. At the same time, the NaCl rejection was not compromised ($93.9 \pm 0.3\%$ for TFN vs. $93.4 \pm 1.1\%$ for TFC). Other porous nanoparticles, such as mesoporous silica [27,28] and metal organic framework (MOF) [29–31], also show similar permeability enhancement effect. Deng and co-workers [27] reported that, for mesoporous silica particles with pore size up to 3 nm, the resulting TFN membranes can maintain their rejection while improving their water permeability.

Researchers have also investigated the use of non-porous nanoparticles for the synthesis of TFN membranes. The loading of hydrophilic non-porous nanoparticles such as silica [32], silver [33,34], and TiO_2 [35–37] often show enhancement of water permeability compared to that of the control counterparts. In several studies, researchers found reduced salt rejection with the introduction of nanoparticles into the polyamide layer, which can often be attributed to the agglomeration of nanoparticles [38,39].

To date, TFN membranes have successfully reached commercialization (e.g., the zeolite-based LG NanoH₂O[®] membranes), partly because the relative easiness to scale up their production. In this kind of membrane structure, polyamide is indeed used as a salt-rejecting matrix that helps to minimize defects in the rejection layer and thus maintain high salt rejection. The addition of nanoparticles can also be easily adapted into the existing membrane fabrication lines that are originally designed for TFC polyamide membrane production. However, the improvements in membrane separation performance for the TFN membranes are incremental, since their performance is largely limited by the polyamide matrix. The use of the polyamide matrix further makes these TFN membranes vulnerable to chlorine attack [24]. Therefore, better membranes and materials for desalination are needed.

3. Novel materials and methods for synthesizing desalination membranes

3.1. Aquaporin

Nature provides a perfect solution for desalination – water molecules are transported across biological cells through a group of transmembrane proteins known as aquaporins (AQPs). AQPs, whose water transport channels resemble the shape of an hour glass, are highly efficient in delivering water molecules with high selectivity [40]. Readers interested in the detailed structures and properties of AQPs are referred to several existing reviews on this topic [41–43]. A brief description of the structure of AQP1 (the earliest water channel proteins discovered by Peter Agre in 1991 [40]) is presented in this section. The hour-glass-shaped channel of AQP1 (Fig. 3) has an internal vestibule of approximately 20 \AA in length, and the narrowest constriction of the vestibule is approximately 2.8 \AA . The asparagine groups near the middle of the vestibule give rise to positively charged sites, and the remaining of the vestibule is considerably hydrophobic. This particular structure forces water molecules to line up in the vestibule in a one-dimensional “water wire” with little frictional force from the hydrophobic wall. The tiny constriction only allows a single water molecule jump through the channel one at a time. The positive charged sites inside the vestibule ensures the proper orientation of the water molecules, as they undergo a “transient dipole reorientation” through the charge interaction and hydrogen bonding with the asparagine groups [44]. The combined site restriction and charge repulsion lead to nearly perfect rejection of solutes, including protons [40].

For membrane synthesis, Aquaporin Z (AqpZ, an AQP found in *Escherichia coli* cells) is more commonly used due to its relative simplicity for harvesting and extraction [45]. The key properties of AqpZ are summarized in Table 1. Based on stopped-flow measurements [42], AqpZ has a water permeability of approximately $2\text{--}10 \times 10^{-14} \text{ cm}^3/\text{s}$ [46,47]. According to Kumar and co-workers [8], polymeric vesicles

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