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The Critical Need for Increased Selectivity, Not Increased Water Permeability, for Desalination Membranes

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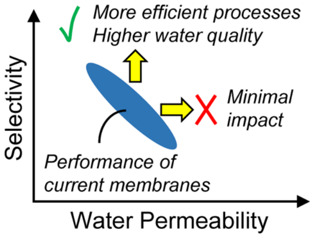
Supporting Information

ABSTRACT: Desalination membranes are essential for the treatment of unconventional water sources, such as seawater and wastewater, to alleviate water scarcity. Promising research efforts on novel membrane materials may yield significant performance gains over state-of-the-art thin-film composite (TFC) membranes, which are constrained by the permeability−selectivity trade-off. However, little guidance currently exists on the practical impact of such performance gains, namely enhanced water permeability or enhanced water−solute selectivity. In this critical review, we first discuss the performance of current TFC membranes. We then highlight and provide context for recent module-scale modeling studies that have found limited impact of increased water permeability on the efficiency of desalination processes. Next we cover several important examples of water treatment processes in which inadequate membrane selectivity hinders process efficacy. We conclude with a brief discussion of how the need for enhanced selectivity may influence the design strategies of future membranes.



INTRODUCTION

As the impact of water scarcity grows in regions around the globe, there is an ever-increasing need to augment municipal, industrial, and agricultural water supplies through the purification of unconventional water sources, such as seawater and municipal wastewater.[1,2] Due to the inextricable linkage between water and energy consumption, often called the water−energy nexus, the augmentation of water supplies must not come at the cost of large amounts of consumed energy. As such, with their high energy efficiency and often superior efficacy, membrane-based technologies have gained widespread implementation in various water treatment processes.[2,3]

Desalination membranes, membranes that allow passage of water but largely reject salt and most other solutes, play a critical role in many of these processes.[3−5] These membranes lie at the heart of traditional reverse osmosis (RO) processes, including (i) seawater reverse osmosis (SWRO), which is the dominant seawater desalination technology globally, and (ii) brackish water reverse osmosis (BWRO), which allows for desalination of low-salinity water, such as brackish groundwater that makes up 55% of global groundwater supplies.[4,6] In addition, RO is a key step in advanced municipal wastewater treatment schemes that allow for industrial and potable reuse.[2,7] Lastly, the emerging technology of forward osmosis (FO), which also relies on desalination membranes, has enabled the treatment of highly saline wastewaters, such as produced waters from shale gas processes, that cannot be treated by RO due to high required hydraulic pressures.[8]

The increased use of desalination membranes has come with a renewed focus on membrane materials research.[9] Because of recent advances in nanomaterial synthesis and assembly, potential step-change improvements in performance may be possible. However, the existing body of literature lacks guidance on the practical impact of improvements in the critical active-layer properties, namely, membrane water permeability and water−solute selectivity. In other words, in the design of novel desalination membranes, what active-layer properties are most desired?

In this critical review, we first cover the performance of state-of-the-art desalination membranes. We then review recent analyses and modeling studies that have found limited impact of improvements in membrane water permeability on the performance of RO and FO processes. Next, we highlight several important examples of processes that are adversely affected by inadequate solute retention, demonstrating the need for enhanced water−solute selectivity. Lastly, we discuss how current and potential future membranes fit into this landscape. Lessons gained from this critical review should influence the design strategies of novel desalination membranes.

SELECTIVE-LAYER PERFORMANCE OF CURRENT MEMBRANES

Aromatic thin-film composite (TFC) polyamide membranes, the current state-of-the-art membranes, serve as the benchmark for any novel desalination membrane.[5] The selective layer, also called the active layer, in TFC membranes is a dense, highly cross-linked polyamide film, formed via the interfacial polymer-

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ization of two aromatic monomers: *m*-phenylenediamine and trimesoyl chloride. The transport of water and solutes through the active layer is governed by the solution-diffusion model.[10,11] In this model, transport through the active layer, which is considered nonporous, is diffusive in nature. Water and solutes partition into the polymeric active layer, diffuse down their chemical potential gradient, and desorb into the permeate solution.

Water flux according to the solution-diffusion model is given by[10,11]

$$J_w = A(\Delta P - \Delta \pi_m) \tag{1}$$

where J_w is the volumetric water flux, A is the water permeability coefficient (also called permeance), ΔP is the applied hydraulic pressure, and $\Delta \pi_m$ is the osmotic-pressure difference across the membrane active layer between the feed and permeate sides. In RO, flow is driven by hydraulic overpressure, i.e., the difference between hydraulic and osmotic pressures. In FO, flow is driven by an osmotic-pressure difference created using a highly concentrated draw solution. For RO, eq 1 can be modified using film theory to account for concentration polarization in the diffusive boundary layer at the feed channel−membrane interface:[10]

$$J_w = A\left[\Delta P - \Delta \pi_b \exp\left(\frac{J_w}{k_f}\right)\right] \tag{2}$$

where $\Delta \pi_b$ is the osmotic-pressure difference between the bulk feed and permeate solutions and k_f is the overall feed-side mass transfer coefficient averaged for all feed solutes. In both RO and FO, solute flux is modeled as Fickian diffusion:

$$J_s = B\Delta c_m \tag{3}$$

where J_s is the solute flux, B is the solute permeability coefficient, and Δc_m is the concentration difference across the membrane active layer. As can be seen from eqs 1−3, the contribution of the membrane active layer to water and solute fluxes is entirely contained in the lumped coefficients A and B.

The impact of membrane properties on water flux differs between RO and FO. In RO, only active-layer properties (i.e., A coefficient) affect water flux, while in FO, support-layer properties are also important. During FO operation, permeating water molecules dilute the draw solution at the interface of the active layer and support layer. This dilution, combined with hindered diffusion within the support layer, results in a draw-solute concentration gradient, termed internal concentration polarization, that sharply decreases the osmotic-pressure driving force and the achievable water flux.[8] Support-layer properties that impact resistance to diffusion, including thickness (δ_s), tortuosity (τ), and effective porosity (ε_{eff}), are contained in the structural parameter (S):[8,12]

$$S = \frac{\delta_s \tau}{\varepsilon_{\text{eff}}} \tag{4}$$

Minimizing the structural parameter (i.e., achieving high porosity, low tortuosity, and low thickness) maximizes draw-solute diffusion and the resulting water flux in FO.

Solute retention in RO and FO is predominantly influenced from a materials perspective by active-layer properties, namely A and B.[8,11] For example, solute rejection ($R = 1 - c_{\text{permeate}}/c_{\text{feed}}$) in RO can be modeled as a function of A and B (see the Supporting Information for the derivation):

$$\frac{R}{1-R} = \frac{A\left[\Delta P - \Delta \pi_b \exp\left(\frac{J_w}{k_f}\right)\right]}{B \exp\left(\frac{J_w}{k_{\text{sol}}}\right)} = \frac{J_w}{B \exp\left(\frac{J_w}{k_{\text{sol}}}\right)} \tag{5}$$

where k_{sol} is the feed-side mass transfer coefficient for the solute of interest. It is useful to consider eq 5 at a fixed hydraulic pressure while neglecting changes in the bulk osmotic-pressure difference (i.e., $\Delta P - \Delta \pi_b$ is constant), which is permissible when salt rejection is high. At a very low A, water flux is low, concentration polarization is minimal ($J_w \ll k_f$, k_{sol}), and R approaches a function of A/B. For such membranes with very low water permeability, increasing A or decreasing B enhances rejection through increased water flux or decreased solute flux, respectively. Conversely, at a very high A, severe concentration polarization due to high water flux decreases the effective driving force, limiting further increases in water flux through increased A. At the constant-flux limit, R is purely a function of B.

The effect of A and B on rejection is shown in the red lines in Figure 1, wherein A and B values correspond to given salt rejection values under standard SWRO test conditions. Under these conditions, an increased A at a constant B yields substantially enhanced salt rejection when $A < 1$ L m^{-2} h^{-1} bar^{-1}, while the impact is greatly diminished when $A > 2$ L m^{-2} h^{-1} bar^{-1}. The key message is that decreasing B at a constant A always significantly increases salt rejection, whereas increasing A at a constant B impacts rejection only when A and the resulting water flux are low.

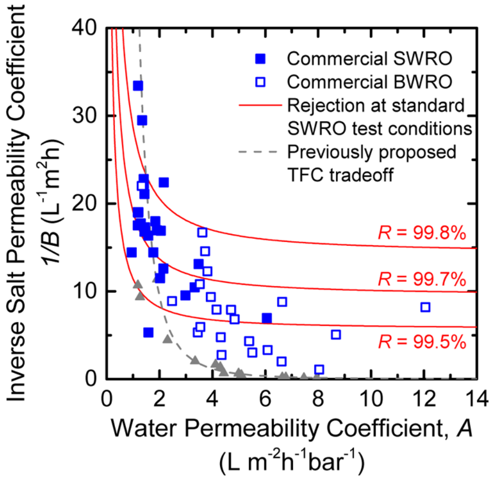

Figure 1. Water permeability−salt permeability trade-off for commercial thin-film composite (TFC) polyamide membranes. Data points for commercial seawater reverse osmosis (SWRO) and brackish water reverse osmosis (BWRO) membranes were calculated from manufacturer technical specifications.[15] Values depicted as gray triangles were measured for a chlorine/alkaline-treated commercial SWRO membrane and were used previously to establish the proposed TFC trade-off relationship (gray dashed line).[14] Red lines correspond to A and B values that result in the labeled RO rejections under standard SWRO test conditions (55.15 bar applied pressure and 32000 ppm sodium chloride feed solution) at coupon scale, calculated using eq 5 with a mass transfer coefficient, k_f, of 100 L m^{-2} h^{-1}.

The ideal desalination membrane has a high A coefficient paired with a near-zero B coefficient for all solutes, i.e., moving toward the top right corner in Figure 1. Achieving such a membrane is extremely difficult. Dense polymeric membranes are postulated to follow a permeability–selectivity trade-off, in which increases in water permeability result in even greater increases in solute (e.g., salt) permeability.[13] This trade-off has been extended to TFC membranes, for which $B \propto A^3$, shown as the gray dashed line in Figure 1.[14] For comparison with the trade-off relationship, Figure 1 also shows performance data for commercial SWRO and BWRO membranes, calculated from manufacturer technical specifications.[15] The data does indeed show that B for NaCl tends to increase as A increases, and several SWRO membranes have performance that falls along the previously proposed TFC trade-off relationship (gray dashed line). However, small improvements in TFC performance (e.g., slightly increased water permeability without a sharp drop in selectivity) have been achieved by membrane manufacturers.[15,16] As such, several SWRO membranes in Figure 1 have performance that exceeds (lies to the right of) the previously proposed TFC trade-off. BWRO membranes in Figure 1 also tend to exceed the trade-off relationship, but the comparison is less clear as these membranes are tested at lower salt concentrations and hydraulic pressures.

On the basis of the data depicted in Figure 1, state-of-the-art SWRO membranes can achieve a high rejection with water permeability coefficients of 2–3 L m^{-2} h^{-1} bar^{-1}, while BWRO membranes can achieve moderate rejection with permeability coefficients of 3–5 L m^{-2} h^{-1} bar^{-1}. The performance of TFC membranes will likely continue to incrementally improve through optimization of polyamide film structure. However, improvements are expected to be limited because of the trade-off relationship and the use of similar polyamide-based materials to effect water–solute separation.

For dramatic improvements in performance, fundamentally novel desalination membranes will likely be needed. In recent years, a rich variety of new membrane materials have been considered, including carbon nanotubes,[17,18] graphene,[19−21] aquaporin,[22] self-assembled surfactants,[23,24] and synthetic water channels.[25−27] While these materials have the potential to achieve breakthrough performance, the impact of such improved performance on full-scale desalination systems is often unclear. To help guide research efforts toward performance gains that are critically needed, we now discuss the practical impact that increased water permeability (increased A at constant B) and increased water–solute selectivity (decreased B at constant A) would have on desalination processes.

■ RELATIVE INSIGNIFICANCE OF HIGH WATER PERMEABILITY

Recent studies using novel materials for desalination membranes have emphasized the potential for extremely large increases in water permeability.[17,22,26,28,29] For example, densely packed carbon nanotubes and nanoporous graphene are projected to have permeabilities 2–3 orders of magnitude higher than those of current TFC membranes.[17,28] The implication of these and similar studies is that a dramatic increase in water permeability would lead to an equally dramatic increase in process efficiency (e.g., lower energy usage and/or lower capital and operating costs). However, this assumption has come under heavy scrutiny for both RO and FO desalination processes.[3,30−35]

Reverse Osmosis. Energy consumption of RO desalination, which for SWRO can be up to 50% of operation and maintenance costs,[4] depends on several factors, including membrane properties, process design and efficiency, and required pre- and post-treatment steps. As membrane water permeability affects only the desalination stage, the discussion here is restricted to exclude intake, pre- and post-treatment, and brine discharge, which combined can consume more than 1 kWh of energy/m^3 of product water.[3]

Over the past 40 years, the specific energy consumption — the energy required to produce a unit volume of permeate water — of the desalination stage has decreased from ~16 to 2 kWh m^{-3} for SWRO.[3] Much of this decrease stems from increases in pump efficiency and the incorporation of pressure exchangers, which recover up to 95% of the energy in the high-pressure brine.[4] Further reductions in energy consumption have been achieved through the development of TFC polyamide membranes, which have water permeability coefficients and selectivities higher than those of the cellulose acetate membranes that were initially used in SWRO.[3] This improved active-layer performance reduced the hydraulic overpressures (and the corresponding energy) required to achieve adequate water flux.

Although the advent of TFC membranes resulted in energy savings, further gains are constrained by single-stage operation of RO, in which the minimum hydraulic pressure and minimum specific energy are equal to the osmotic pressure of the exiting brine.[3,36] Figure 2A illustrates the impact of practical operation on energy requirements. For SWRO with a feed of 35000 ppm and 50% recovery, the thermodynamic minimum specific energy (gray shaded area) is 1.06 kWh m^{-3}.[3] Single-stage operation increases the practical minimum specific energy (gray and yellow shaded area combined) to 1.56 kWh m^{-3}.[3] Material enhancements, including water permeability, affect only the energy related to the hydraulic overpressure (cyan shaded area), which is large for membranes with low permeability (e.g., for cellulose acetate) and small for membranes with higher permeability (e.g., for current TFC membranes, as discussed later). As illustrated for BWRO, additional energy can be saved through use of multiple RO stages, a common practice in BWRO,[4] allowing for operation at multiple hydraulic pressures.[3]

A recent module-scale modeling study explicitly analyzed the effect of an increased A coefficient on specific energy requirements.[30] The study was modified and extended here (see the Supporting Information for details), with results shown in Figure 2B. For both SWRO and BWRO, as the A coefficient is increased, the specific energies decrease precipitously before eventually plateauing. In SWRO, increasing A from 0.2 to 1.0 L m^{-2} h^{-1} bar^{-1} facilitates a 45% reduction (1.55 kWh m^{-3}) in energy consumption, while increasing A from 2 to 10 L m^{-2} h^{-1} bar^{-1} results only in a 3.7% reduction (0.06 kWh m^{-3}). Even a 10-fold increase in A from 10 to 100 L m^{-2} h^{-1} bar^{-1} yields a mere 1.0% reduction in energy consumption. In single-stage BWRO with a 0.1 M NaCl feed, effects of enhanced water permeabilities are similarly limited: increasing A from 4 to 10 L m^{-2} h^{-1} bar^{-1} yields a 2.2% reduction. For two-stage BWRO, the amount of energy required at 4 L m^{-2} h^{-1} bar^{-1} is 22% lower (0.11 kWh m^{-3}) than for single-stage BWRO due to the effects of staging. In addition, the effect of increased water permeability is slightly larger, with a 12% reduction (0.05 kWh m^{-3}) as A increases from 4 to 10 L m^{-2} h^{-1} bar^{-1}. Nevertheless, the absolute decrease in the required energy is still quite small, similar in magnitude to the change for SWRO.

Using state-of-the-art TFC membranes (2–3 L m^{-2} h^{-1} bar^{-1} for SWRO and 3–5 L m^{-2} h^{-1} bar^{-1} for BWRO) as a reference, the modeling results demonstrate that increasing the

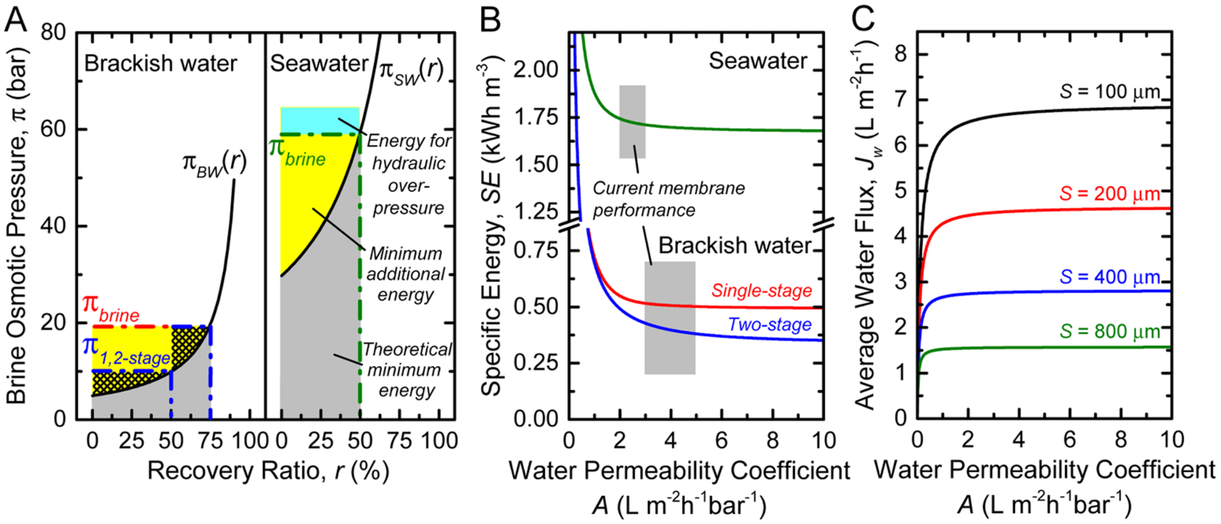

Figure 2. Energetics and the impact of increased water permeability for reverse osmosis (RO) and forward osmosis (FO) desalination. (A) Impact of system design on the energy required in RO. In an RO stage, the minimum hydraulic pressure is the osmotic pressure of the exiting brine, resulting in irreversible losses (yellow shaded area). Hydraulic overpressure (cyan area) is needed to achieve the desired water flux. Using two stages, shown for brackish water RO (cross-hatched area), can decrease irreversible losses. Brackish water (BW) and seawater (SW) are modeled as 0.1 and 0.6 M NaCl, respectively. (B) Specific energy requirements for RO desalination with an increasing water permeability coefficient, *A*, considering SWRO with 50% recovery and BWRO with 75% recovery. Performance was obtained via module-scale modeling (modifying and extending a previous study)[30] with an average water flux of 15 L m^{-2} h^{-1}. For two-stage BWRO, recovery and water flux in each stage were 50% and 15 L m^{-2} h^{-1}, respectively. To isolate membrane-related contributions from other process inefficiencies, the effects of fouling, frictional-pressure losses, and inefficiencies of pumps and pressure exchangers were neglected. More details are available in the Supporting Information. (C) Impact of the increased water permeability coefficient, *A*, and the decreased structural parameter, *S*, on average water flux in FO for high-salinity feed streams. Water flux was determined using module-scale modeling following methods in a previous study[31] for counter-current flow mode with a 1.5 M NaCl feed solution, a 4 M NaCl draw solution, and 60% recovery. For panels B and C, the salt permeability coefficient, *B*, and mass transfer coefficient, k_f, were set to 0.1 and 100 L m^{-2} h^{-1}, respectively.

water permeability coefficient beyond current values would facilitate only minor (<0.06 kWh m^{-3}) reductions in the energy consumption of RO. This limited impact is due to two factors. First, the hydraulic overpressures are already small when using TFC membranes, limiting the potential for further efficiency gains. For example, the modeled hydraulic pressure in single-stage SWRO for an *A* of 2 L m^{-2} h^{-1} bar^{-1} is only 7.6% above the brine osmotic pressure. Second, at high water permeabilities, concentration polarization limits further increases in flux. Concentration polarization can be reduced if k_f is increased (eq 2), but this generally requires greater frictional-pressure losses due to increased turbulence.[37]

We note that a higher water permeability may result in membrane-area savings.[30] This finding, however, is inconclusive as models did not account for fouling, which is exacerbated at higher water fluxes.[38,39] A small decrease in membrane area may be possible without an increased level of fouling by incorporating high-permeability elements only at the back end of an RO module, where the driving force is relatively low because of the increased osmotic pressure of the retained feed.[34,40]

Forward Osmosis. FO has enabled membrane-based desalination of difficult to treat feed-waters, particularly those with high total dissolved solids (TDS), which cannot be treated by RO.[8] For high-TDS feed-waters, water fluxes in FO are generally very low, typically around 3 L m^{-2} h^{-1}.[41] Increasing the average water flux, and thus reducing the membrane area, would be highly impactful because of decreased capital and operational costs.

As described above, the effects of FO membrane properties on water flux are mainly contained in the *A* coefficient and the structural parameter, *S*. A recent module-scale modeling study (extended here) has shown that the support layer, rather than the active layer, provides the main resistance to water permeation across FO membranes due to high levels of internal concentration polarization.[31] Figure 2C shows that reducing *S*, which typically ranges from 300 to 500 μm, leads to significant increases in the average water flux across a membrane module. For example, for a high-TDS feedwater (modeled as 1.5 M NaCl), halving *S* values from 400 to 200 μm produces a 63% increase in water flux. In contrast, doubling the *A* coefficient from 2 to 4 L m^{-2} h^{-1} bar^{-1} yields a mere 1.5% increase in average water flux. In other words, as for RO, increased water permeability above currently achieved levels has a minimal impact on FO performance. Thus, efforts to increase water flux for FO membranes should focus on decreasing the structural parameter.

■ CRITICAL NEED FOR HIGH SELECTIVITY

The water–solute selectivity of TFC membranes is by most considerations very high, with NaCl rejections of 99.7–99.8% possible for some commercial SWRO membranes under standard test conditions (Figure 1).[3] Even so, currently achieved selectivities are often inadequate for water treatment applications, generally because of one or both of the following: (i) inadequate ion rejection for applications in which very low TDS levels are required (Figure 3A) and (ii) incomplete rejection of small neutral solutes (Figure 3B), which can have hydrodynamic radii smaller than those of hydrated ions.

Although selectivity needs differ greatly between water treatment applications, the impact of improved selectivity on various processes is qualitatively similar, with two general outcomes. First, improved retention of inadequately retained

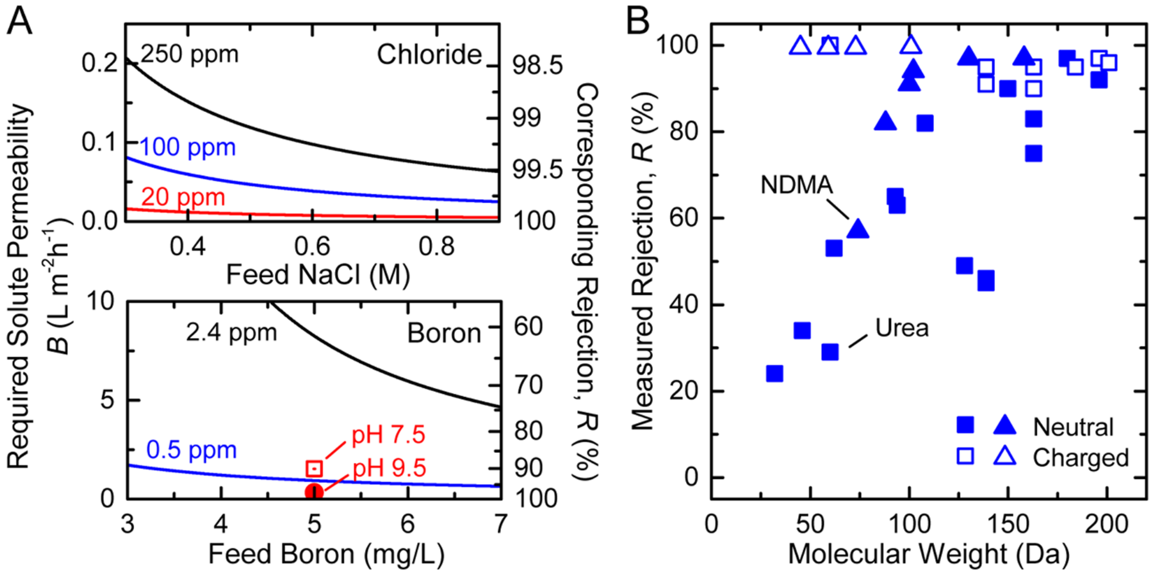

Figure 3. (A) Solute permeabilities needed at varying feed concentrations to achieve permeate water quality levels for chloride and boron using single-pass reverse osmosis (RO). A chloride concentration of 20 ppm is the target permeate quality in the Ashkelon seawater RO plant in Israel.[47] Boron levels of 2.4 and 0.5 ppm are standards for drinking water and irrigation water for sensitive crops, respectively.[43,44] Data points for boron permeability are from Hyung and Kim[42] and are averaged for six commercial seawater RO membranes. Curves are from module-scale modeling of seawater RO (details in the Supporting Information), assuming an average flux of 15 L m^{-2} h^{-1}, 50% recovery, and a mass transfer coefficient, k_f, of 100 L m^{-2} h^{-1}. The corresponding rejection axes refer to the coupon-scale rejection calculated for a given solute permeability coefficient using eq 5 when the water flux is 15 L m^{-2} h^{-1}. (B) Rejection data for neutral and charged organic solutes using RO membranes. Data were included only for solutes measured >1 pH unit from the solute pK_a, and only from studies that verified that the membrane salt rejection was near expected levels (>98%). NDMA refers to *N*-nitrosodimethylamine, a disinfection byproduct. Data depicted as squares are from ref 69. Data depicted as triangles are from ref 52.

species would lead to more effective processes, yielding higher quality and, in some cases, safer water. Second, pretreatment and post-treatment steps necessitated by inadequate solute removal in the desalination stage could be eliminated, leading to significant chemical, energy, and cost efficiency gains. These efficiency gains through eliminated process steps would typically outweigh the small energy savings obtained through enhanced water permeability (described above), which affects only the desalination stage.

The need for enhanced selectivity is best illustrated for specific water treatment processes. We now discuss four examples in which suboptimal membrane selectivity hinders process performance and efficiency.

Seawater Desalination for Irrigation. In SWRO, a contaminant of considerable interest is boron. Boron is ubiquitously present in seawater, typically in the form of boric acid, ranging from 4.5 to 6.0 ppm in concentration.[4] With a pK_a in seawater of 8.7 (increasing to 9.1 in fresh water),[42] boron predominantly exists during RO operation as a small neutral species, resulting in typical rejections of ~80%.[4] While the need for boron removal partially stems from its impact on human health (the World Health Organization has set a drinking water guideline for boron at <2.4 ppm),[43] restrictive standards for irrigation (<0.5 ppm due to leaf damage and premature ripening of fruit) pose the greatest technical challenge.[44] In addition, chloride and TDS levels in water for irrigation are recommended to be less than 105 and 450 ppm, respectively, due to sodium- and chloride-related toxicities and salt build-up in the soil.[44] Because desalinated water in some regions (e.g., Israel and Spain) is commonly used for irrigation, often indirectly through the reuse of wastewater effluent,[45] solute removal in SWRO to levels at or even below these restrictive standards can be critical.

Using current membranes, SWRO process schemes require extra processing to decrease the boron concentration to acceptable levels for irrigation, as the required boron permeability coefficients for single-pass operation are below currently achieved levels at typical operating pH values (Figure 3A).[42] To remove residual boron, processes often employ an extra RO step, in which at least a portion of the permeate from the first RO pass is adjusted to pH ~10 and then processed via a second RO pass.[46,47] Because boron is predominantly charged at pH 10, boron rejections of 99% are typical for SWRO membranes.[48] A noteworthy example of the multipass approach is the Ashkelon plant in Israel, the world's largest RO plant when built in 2005, which has highly restrictive standards of <0.4 ppm for boron and <20 ppm for chloride.[47] To meet these standards, the plant uses a complicated four-pass operation with intermediate acid/base additions.

For SWRO for irrigation, the critical need is thus a membrane with enhanced selectivity for water over salt and boron in its neutral form. Such a membrane would allow for single-pass operation at neutral pH, obviating the need for extra RO passes with pH adjustments. This would not only lead to significant efficiency gains but also decrease safety risks caused by storage of large quantities of caustic solutions.

Potable Reuse of Wastewater. Reclamation of municipal wastewater to augment potable and nonpotable water supplies will play a major role in alleviating water scarcity.[7,49] For example, treatment and reuse of coastal wastewater effluent, which is typically discharged into the ocean without opportunity for downstream reuse, would account for 27% of the public supply of drinking water in the United States.[7] For such potable reuse, advanced water treatment plants, such as the Groundwater Replenishment System (490000 m^3/day planned capacity) in Fountain Valley, CA, and NEWater

(460000 m^3/day total capacity) in Singapore, are essential. Although almost all potable reuse schemes are indirect, meaning there is an environmental buffer (i.e., water body or aquifer) between treatment and consumption, the National Research Council (NRC) has recommended that direct potable reuse be considered a viable option.[7]

A typical advanced treatment process for potable reuse has three steps: microfiltration or ultrafiltration for pretreatment, RO, and advanced oxidation processes (AOPs).[2,7] RO is critical; in addition to reducing TDS to appropriate levels, RO serves as a redundant barrier for pathogens and the main barrier for small organic and inorganic compounds. It is also relatively economical, as low TDS levels result in energy requirements that are lower than those of SWRO. The AOP step, which is present mainly due to inadequate selectivity in RO, degrades residual organic compounds, typically transforming them into less toxic products.[7]

RO is the most effective technology for the removal of micropollutants, which can have deleterious effects at trace levels and thus pose great concern in potable reuse.[7,50] For example, analysis of pilot- and full-scale plants showed near-complete removal by RO of a variety of pharmaceutical and personal care products (all with molar masses of >150 Da).[51] However, very small neutral micropollutants, such as the industrial solvent 1,4-dioxane (88 Da) and the disinfection byproduct *N*-nitrosodimethylamine (NDMA; 74 Da), are poorly rejected by TFC membranes.[7,52−54] Both compounds were found downstream of Water Factory 21, a now-defunct advanced water treatment plant in Fountain Valley, CA, that pioneered using RO for indirect potable reuse, resulting in suspended operations for the treatment plant and downstream drinking water production wells until levels could be decreased (e.g., by installation of high-energy ultraviolet treatment to degrade NDMA).[55,56] The NRC identified NDMA as particularly concerning because of its potent carcinogenic properties; incomplete removal of NDMA during RO necessitates the AOP step.[7,55]

Desalination membranes with increased selectivity would ideally yield near-complete rejections for all known micropollutants (including NDMA and its precursors), permitting more efficient processes wherein AOPs are replaced by significantly less costly and less energy-intensive treatment steps. Because AOPs in some cases produce undesired or toxic byproducts,[57] effective removal of micropollutants in RO is also preferred from a water quality standpoint. In addition, a strengthened RO barrier would prevent permeation of unidentified and emerging micropollutants, potentially mitigating future water quality issues.

Forward Osmosis with Thermolytic Draw Solutes. Critical to FO operation is the draw solution, for which the thermolytic ammonia−carbon dioxide (NH_3-CO_2) system is particularly promising.[8,58] In this system, the constituent solutes have a high mutual-diffusion coefficient to decrease the extent of internal concentration polarization. In addition, the diluted solutes can be thermally distilled to readily regenerate a highly concentrated draw solution. The NH_3-CO_2 system has been used at pilot scale to treat produced water from shale gas operations[41] and at commercial scale as part of a zero-liquid discharge (ZLD) scheme to treat flue gas desulfurization wastewater.[59]

The main drawback to the NH_3-CO_2 draw solution is its high rate of reverse draw-solute flux, the permeation of draw solutes into the feed/brine solution. Reverse draw-solute flux increases operating (material) costs, potentially increases rates of fouling and scaling, and impacts the environment because of the toxicity of ammonia.[8] NH_3-CO_2 specific reverse draw-solute fluxes, with deionized water as the feed, are approximately 3 times higher than for NaCl-based draw solutions through typical TFC membranes[60] and 10 times higher through cellulose triacetate membranes.[61] In addition, reverse draw-solute flux sharply increases when using more realistic feed streams. Recent studies found a 5-fold increase in reverse ammonium flux when the feed stream was changed from deionized water to a NaCl solution, with similarly high levels of forward permeation of sodium ions.[62,63] This sharp increase in cation flux was attributed to cation exchange, mediated by surface carboxyl groups on the TFC membrane.[64,65]

Increased solute flux directly impacts FO process performance. In a pilot-scale demonstration plant, high rates of reverse ammonium flux necessitated a brine stripper to recover draw solute from the brine, while the corresponding forward ion flux necessitated a polishing RO step to remove TDS remaining in the product water after regeneration of the draw solution.[41] Improved membrane selectivity would thus have a clear practical impact on FO. Decreased solute flux through enhanced membrane selectivity would not only improve product water quality via the retention of undesired feed solutes[66] but also eliminate the need for costly and energy-intensive process steps such as brine stripping.

High-Purity Water Production. High-purity water, sometimes called ultrapure water, has water quality standards far exceeding those of drinking water. High-purity water is used for a variety of industrial applications, including high-pressure boilers in thermoelectric power plants (to mitigate scale formation on turbine blades), pharmaceutical manufacturing, and semiconductor device fabrication.[67] In the semiconductor (i.e., microelectronics and photovoltaics) industry, ultrapure water is used to rinse wafers between fabrication steps. Ultrapure water requirements for these rinse steps are extensive. For example, Intel Corp. in 2014 withdrew an average of 87000 m^3/day of water from rivers and aquifers, with much of that going toward ultrapure water production.[68]

High-purity water production is usually conducted on site, using fresh water or reclaimed wastewater as the feed. Typically, RO is the central step in an extensive process that includes polishing steps such as electrodionization, ion exchange, and further RO passes to remove residual ions, and UV irradiation to remove residual organic species.[67] With the need to remove nearly all solutes, improved selectivity in the RO membrane would certainly enhance the performance and efficiency of high-purity water production, for example by eliminating some polishing steps. Considering the large volumes of water treated, such efficiency gains could be very impactful.

OUTLOOK FOR CURRENT AND FUTURE DESALINATION MEMBRANES

On the basis of the discussion presented above, it is clear that membranes with increased water−solute selectivity, not water permeability, are needed for desalination applications. As such, a reasonable target for materials research would be a membrane with a water permeability coefficient of 2−4 L m^{-2} h^{-1} bar^{-1}, near-perfect rejection of NaCl at any ionic strength, and very high rejection of small neutral species. A membrane with these properties would enhance a wide variety of important desalination applications, including SWRO and wastewater RO for potable reuse.

For TFC membranes and possibly all solution-diffusion-based polymeric membranes, the permeability−selectivity trade-off inhibits the achievement of this goal. Although TFC membranes will continue to be successfully applied for a wide variety of processes, they necessarily have drawbacks in operation due to the trade-off relationship. For example, TFC membranes with the highest boron rejection also tend to have the lowest water permeability (from specified water fluxes,[42] typical A coefficients can be estimated to be ∼1 L m^{-2} h^{-1} bar^{-1}), which is low enough to meaningfully hinder SWRO efficiency. The converse example is the application of low-pressure RO membranes to potable reuse of municipal wastewater. These highly permeable membranes offer minor energy and membrane-area savings, but at the cost of increased permeation of small neutral solutes.[52,54,69] Concern over micropollutant permeation may incentivize usage of high-rejection RO (e.g., SWRO) membranes with lower water permeabilities to yield a higher-quality permeate.[54]

Novel desalination membranes may provide a viable route for achieving extremely high selectivities. Promising approaches in this active area of research were recently reviewed.[9] Particularly notable are materials with uniform-diameter nanochannels that retain solutes via size exclusion and for which the size of the nanochannel is intrinsically determined by the starting material and the assembly process. Proper tuning of the nanochannel size and incorporation within a robust, relatively impermeable thin film could result in a high-performance membrane.

The potential influence of the need for high selectivity on novel desalination membranes is best illustrated for carbon nanotubes (CNTs), which have long held promise because of ultrafast water permeation through their atomically smooth interiors.[17,70] Because of the limited impact of very high water permeability, a densely packed carbon nanotube membrane is unnecessary. Instead, the critical research goals to yield a highly selective membrane are scalable synthesis of uniformly sized CNTs with diameters small enough to reject salt and small neutral solutes and stable alignment of CNTs within a thin polymeric film. The density of CNTs needs to be only high enough, approximately 2.4×10^{11} nanotubes per square centimeter or 0.25% areal density for (6,6) CNTs,[9] to achieve water permeabilities comparable to those of TFC membranes.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.estlett.6b00050.

> Detailed derivation of the apparent rejection equation in RO (eq 5), methodology for coupon-scale modeling of RO, and methodology for module-scale modeling of RO (PDF)

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Notes

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