Electronic Supplementary Material (ESI) for Energy & Environmental Science.

Supplementary Information

The relative insignificance of advanced materials in enhancing the energy efficiency of desalination technologies

Sohum K. Patel,[a,#] Cody L. Ritt,[a,#] Akshay Deshmukh,[a] Zhangxin Wang,[a] Mohan Qin,[a] Razi Epsztein,[a] and Menachem Elimelech[a*]

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[a]*Department of Chemical and Environmental Engineering, Yale University, New Haven, Connecticut 06520, United States*

[b]*Nanosystems Engineering Research Center for Nanotechnology-Enabled Water Treatment (NEWT), Yale University, New Haven, Connecticut 06520, United States*

*Corresponding Author; Address: P.O. Box 208268, Yale University, New Haven, CT 06520; Email: menachem.elimelech@yale.edu; Phone: +1 (203) 432-2789; Fax: +1 (203) 432-2881

[#] These authors contributed equally to this work.

Permselectivity of pressure-driven membranes

Membrane permselectivity (P_W/P_S), defined as the ratio of the permeability of the more permeable penetrant (water) to that of the less permeable penetrant (salt), is a material property that describes a membrane's intrinsic selectivity. This property is inextricably linked with a membrane's real (intrinsic) rejection, R_{int}, which can be related to the observed rejection, R_o, by accounting for concentration polarization (CP) in the diffusive boundary layer at the feed channel-membrane interface:[1, 2]

$$\frac{P_W}{P_S} = \frac{R_g T}{\hat{V}_W (\Delta P - \Delta\pi_m)(1 - R_{int})} = \frac{R_g T}{\hat{V}_W (1 - R_o)\left[\Delta P - \Delta\pi_b \, exp\left(\frac{J_w}{k_f}\right)\right]} \quad \text{(S1)}$$

where R_g, T, and $\hat{V}_W$ are the ideal gas constant (83.145 cm^3 bar K^{-1} mol^{-1}), absolute temperature (298 K), and specific volume of water (18 cm^3 mol^{-1}), respectively. The driving force is described by the difference between the applied hydraulic pressure (ΔP) and transmembrane osmotic pressure ($\Delta\pi_m$). Using film theory, we can estimate the effect of CP by accounting for the measured water flux (J_W), the overall feed-side mass transfer coefficient (k_f), and the feed-side mass transfer coefficient for the solute of interest (k_{sol}). Considering a single-species feed, both mass transfer coefficients can be estimated as 100 L m^{-2} h^{-1},[2] allowing us to utilize the measured bulk osmotic pressure difference ($\Delta\pi_b$).

Using eq. S1, we calculated the permselectivities of novel material-based membranes and channels from previously published reports.[3-25] Table S1 details these calculations.

Table S1. Parameters used for membrane permselectivity calculation

Reference	Material	Observed Rejection, R_o (%)	Applied Pressure, ΔP (bar)	Osmotic Pressure, $\Delta\pi_b$ (bar)	Water Flux, J_W (L m^{-2} h^{-1})	Membrane Permselectivity, P_W/P_S
3	Nanotube	45.2	30	3.83	63	92
4		44.1	10	1.56	72	325

5		20.0	4	0.16	20	109
6		59.0	10	1.56	10	261
7[a,*]		-	-	-	-	1.9×10^4
8[b,i]	Laminate	-	-	-	-	300
9		68.0	15	0.077	8.6	211
10		35.0	10	0.78	42	126
11		18.0	3.4	0.91	17	152
12		39.2	15	1.56	73	155
13[ii]		-	-	-	-	1
14		25.0	4	0.39	13	145
15[a,i]		-	-	-	-	7.9×10^5
16	Biomimetic	32.9	5	0.156	171	889
17		88.1	5	0.156	19	2531
18		25.0	1	0.0456	10.5	531
19		39.0	5	0.156	114.5	606
20		45.1	5	0.156	41	353
21		70.0	1	0.156	3.1	1100
22[a,i]		-	-	-	-	10^{10}
23[ii]	Nanoporous sheet	-	-	-	-	1
24[ii]		-	-	-	-	1
25[a,i]		-	-	-	-	2×10^5

[a]Reference presenting the selectivity of isolated channels. [b]Simulation of large-scale laminate membrane. The permselectivity selected is based on a packing density of 75% with a permeability of 1 L m^{-2} h^1 bar^{-1}, values that are representative of typical laminate membranes. [d]Reference reported water and salt permeances, which can be directly related to membrane permselectivity. Note: input parameters were not provided for references that (i) reported permselectivity or values which can be directly related to permselectivity, such as water and salt permance; (ii) reported ≤ 0% rejection. In this case, a membrane permselectivity of 1 was listed as it represents the inability of the separation layer to discriminate between salt and water. [*]Salt permeance (P_S), needed to calculate permselectivity, was derived from conductance (*G,* 67 nS) measured across the channel.

P_S can be related to G by accounting for the applied voltage (V, 75 mV), channel area (A, 0.363 nm^2), and elementary charge (e, 1.6022×10^{-19} C) as follows: $P_S = \frac{GV}{eA}$.

Conventional- and Surface-heating Direct-Contact Membrane Distillation (DCMD) Modeling

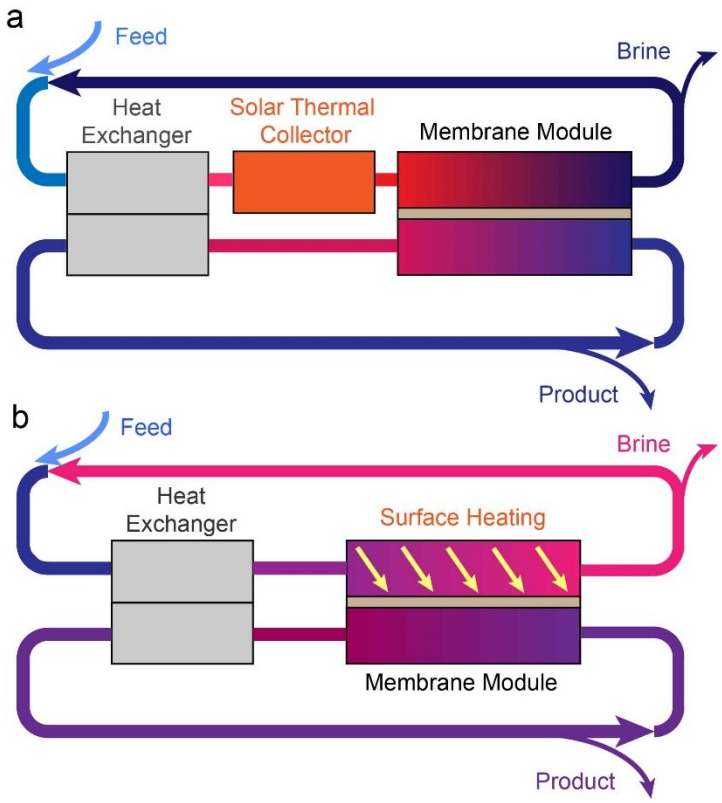

Figure S1. Schematic of DCMD with feed circulation. (a) conventional DCMD with an external solar thermal collector and (b) surface heating DCMD. The solution temperature is qualitatively described by color: red and blue colors denote hot and cold streams, respectively.

The detailed mass and heat transfer equations can be found in literature.[26-29] All the parameters in DCMD simulations are summarized in **Table S2**.

Table S2. Parameters in DCMD simulations

Membrane module length (cm)	9
Membrane module width (cm)	4
Membrane module thickness (mm)	2

Feed temperature (K)	293.15
Coolant temperature (K)	293.15
Ambient temperature (K)	293.15
Feed salinity (g L^{-1})	0
Feed/permeate flow rate ($\times 10^{-5}$ kg s^{-1})	8~17
Membrane mean pore size (μm)	0.2
PVDF membrane thickness (μm)	100
PVDF membrane tortuosity	2
PVDF membrane porosity (%)	75
Solar irradiance (W m^{-2})	1000

Calculating Energy Efficiency of CDI from Literature Data

The SE_{min} is the specific energy required for a thermodynamically reversible process determined by

$$SE_{min} = 2R_g T\left[\frac{c_F}{WR}\ln\left(\frac{c_B}{c_F}\right) - c_P\ln\left(\frac{c_B}{c_P}\right)\right] \quad \text{(S2)}$$

in which R_g is the ideal gas constant, T is the absolute temperature, c_F is the feed salinity concentration, c_B is the salinity of the brine, c_P is the salinity of the product water, and WR is the water recovery. The thermodynamic energy efficiency, η, is calculated as

$$\eta = \frac{SE_{min}}{SE} \quad \text{(S3)}$$

where SE is the actual specific energy required for the separation.

The SE_{min} of each separation was calculated according to eq. S2 and the reported values for c_F, c_B, c_P, and WR in the literature, which are provided in Table S3. The SE is calculated by taking the product of the operating voltage and the total transferred charge (determined by integrating current over time) in the duration of the charging step. We do not consider energy recovery during the discharging step. Further details regarding the applied methodology for calculating SE are described by Wang et al.[30] The collected and synthesized values from the literature are summarized in Table S3.

Table S3. Summary of the literature data used in Fig. 4A.

Electrode Material	Feed Concentration (g L^{-1})	Salt Removal	Water Recovery	Thermodynamic Minimum Specific Energy of Separation, SE_{min} (kWh m^{-3})	Energy Efficiency, η	Reference
Activated Carbon	1.2	54.5%	50%	8.7E-03	4.2%	[31]
	0.3	14.0%	50%	1.5E-04	0.6%	[32]
	0.3	24.0%	50%	4.1E-04	0.9%	[33]
	0.3	30.0%	50%	6.6E-04	1.1%	[33]
	1.2	28.0%	50%	2.2E-03	1.4%	[34]
	1.2	28.0%	50%	2.2E-03	1.3%	[34]
	0.2	73.7%	60%	3.2E-03	1.7%	[35]
	1.2	35.5%	50%	3.6E-03	5.2%	[31]
	1.2	49.6%	50%	7.1E-03	6.5%	[36]
	1.2	51.0%	50%	7.5E-03	2.4%	[31]
	1.2	54.0%	50%	8.5E-03	4.9%	[36]
	1.2	54.7%	50%	8.7E-03	8.9%	[36]
	1.2	55.5%	50%	9.0E-03	6.4%	[36]
	1.2	56.8%	50%	9.4E-03	5.7%	[36]

	1.2	57.3%	50%	9.6E-03	6.8%	[36]
	1.2	59.5%	50%	1.0E-02	4.1%	[36]
	1.2	60.9%	50%	1.1E-02	3.7%	[36]
	2.3	44.3%	50%	1.1E-02	2.1%	[37]
	1.2	61.8%	50%	1.1E-02	7.7%	[36]
	1.2	62.6%	50%	1.2E-02	2.4%	[36]
	1.2	63.1%	50%	1.2E-02	6.5%	[36]
	1.2	66.7%	50%	1.3E-02	3.9%	[36]
	1.2	69.0%	50%	1.4E-02	3.9%	[36]
	1.2	70.2%	50%	1.5E-02	1.3%	[36]
	1.2	73.0%	50%	1.6E-02	4.2%	[36]
	1.2	73.5%	50%	1.7E-02	4.4%	[36]
	1.2	74.5%	50%	1.7E-02	4.8%	[36]
	1.2	75.3%	50%	1.8E-02	2.0%	[36]
	1.2	75.9%	50%	1.8E-02	2.9%	[36]
	1.2	77.9%	50%	1.9E-02	3.3%	[36]
	1.2	77.9%	50%	1.9E-02	2.4%	[36]
	1.2	79.7%	50%	2.0E-02	3.6%	[36]
	1.2	80.1%	50%	2.0E-02	2.9%	[36]
	1.2	81.5%	50%	2.1E-02	3.2%	[36]
	1.2	85.7%	50%	2.4E-02	1.7%	[36]
	1.2	86.3%	50%	2.4E-02	2.3%	[36]
	1.2	86.3%	50%	2.4E-02	2.0%	[36]
	0.2	46.9%	67%	1.4E-03	0.67%	[38]
	0.2	40.0%	67%	8.8E-04	0.82%	[38]
	0.2	31.4%	67%	7.3E-04	1.0%	[38]
	0.3	30.0%	50%	6.6E-04	0.98%	[39]
	0.3	24.0%	50%	4.1E-04	0.86%	[39]
	0.6	7.0%	50%	6.2E-05	0.24%	[40]
	0.3	14.0%	50%	1.5E-04	0.62%	[41]
	0.3	5.2%	50%	1.9E-05	0.22%	[41]
	0.3	2.0%	50%	1.8E-06	0.22%	[41]
	0.3	6.0%	50%	2.5E-05	0.24%	[42]
	0.1	79.6%	50%	1.2E-05	0.14%	[43]
	0.1	69.9%	50%	1.2E-05	0.03%	[43]
	0.2	1.7%	50%	1.5E-05	0.05%	[44]
	0.2	2.3%	50%	1.6E-05	0.03%	[44]
	0.2	3.2%	50%	1.7E-05	0.01%	[44]
	0.5	18.7%	50%	1.8E-05	0.03%	[45]
	0.4	4.1%	50%	2.3E-05	0.04%	[45]
	0.4	4.9%	50%	2.5E-05	0.25%	[46]
	0.03	64.5%	50%	3.9E-05	0.05%	[47]
	0.5	2.5%	75%	4.4E-05	0.04%	[48]
Advanced	0.5	5.0%	63%	5.7E-05	1.24%	[48]
Carbon	0.5	5.5%	60%	6.0E-05	0.04%	[48]
	0.5	7.5%	44%	1.5E-04	0.62%	[48]
	0.5	2.3%	75%	2.0E-04	0.42%	[48]
	0.5	2.8%	63%	4.1E-04	4.53%	[48]
	0.3	24.0%	50%	4.1E-04	0.86%	[33]
	0.5	3.5%	60%	6.3E-04	1.58%	[48]
	0.5	4.0%	44%	7.7E-04	0.93%	[48]
	35	12.6%	50%	1.2E-03	1.62%	[49]
	1.1	61.0%	50%	1.4E-03	1.72%	[49]
	0.3	18.6%	50%	1.0E-02	2.30%	[50]
	0.3	9.3%	50%	1.3E-02	2.92%	[50]
Pseudo-	0.5	25.2%	50%	1.6E-06	0.5%	[51]

capacitive	0.3	25.1%	50%	2.0E-05	0.1%	[52]
	0.6	15.0%	50%	2.0E-05	0.0%	[53]
	0.3	16.0%	50%	2.5E-05	0.2%	[54]
	1.0	12.3%	50%	1.5E-04	0.4%	[55]
	0.4	12.5%	50%	1.5E-04	0.3%	[56]
	0.5	20.2%	50%	1.6E-04	0.2%	[57]
	0.5	56.6%	50%	1.8E-04	0.3%	[57]
	0.5	92.6%	50%	2.5E-04	0.3%	[57]
	1.2	7.4%	50%	2.7E-04	0.1%	[58]
	1.2	2.5%	50%	3.1E-04	0.1%	[58]
	0.3	40.9%	67%	3.2E-04	0.5%	[59]
	0.6	38.0%	50%	4.4E-04	0.6%	[60]
	0.2	10.3%	67%	4.8E-04	0.1%	[60]
	0.2	88.2%	60%	5.5E-04	0.1%	[60]
	1.5	30.8%	50%	6.3E-04	1.6%	[61]
	0.1	8.2%	50%	1.2E-03	1.6%	[62]
	0.5	11.2%	50%	1.4E-03	1.7%	[63]
	0.5	11.6%	50%	1.6E-03	1.4%	[63]
	0.5	14.5%	50%	2.0E-03	1.1%	[63]
	5.8	3.4%	50%	3.3E-03	6.5%	[64]
	0.8	3.3%	50%	4.6E-03	3.0%	[65]
	2.9	29.3%	50%	4.9E-03	3.6%	[66]
	2.9	25.6%	50%	6.0E-03	5.2%	[66]

MCDI Model

All MCDI simulations were performed using a 0-D dynamic ion transport model, as described by several previous works.[67-72] The model uses the extended Nernst-Planck equation to describe ion transport in the spacer channel, membranes, and electrode while utilizing modified Donnan theory for modeling of the EDL structure. The parameters used throughout the modeling are shown in Table S4.

Table S4. Specified parameters for MCDI simulations

Parameter	Value
Spacer thickness (mm)	0.25
Membrane thickness (mm)	0.15
Membrane charge density (mol L^{-1})	5
Spacer porosity	0.71
Electrode macroporosity	0.43
Electrode microporosity	0.30
Stern layer capacitance (F mL^{-1})	120
Specific external resistance (Ω cm^2)	40
Specific electrode resistance (Ω mmol cm^{-1})	2.2
Diffusion coefficient of NaCl in solution (m^2 s^{-1})	1.68×10^{-9}
Diffusion coefficient of NaCl in the membrane (m^2 s^{-1})	1.12×10^{-9}

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