Supporting Information

Unlocking High-Salinity Desalination with Cascading Osmotically Mediated Reverse Osmosis: Energy and Operating Pressure Analysis

Xi Chen[†] and Ngai Yin Yip[*,†,‡]

[†] Department of Earth and Environmental Engineering, Columbia University, New York, New York 10027-6623, United States

[‡] Columbia Water Center, Columbia University, New York, New York 10027-6623, United States

[*] Corresponding author: Email: n.y.yip@columbia.edu, Phone: +1 212 8542984

Number of Pages: 19
Number of Tables: 6
Number of Figures: 6

ENERGY REQUIREMENT AND OPERATING PRESSURES

Practical Specific Energy of Desalination with *N*-stage Configurations. Product of the applied hydraulic pressure, ΔP_i, and permeate volume, ΔV_i, gives the energy consumption in the i^{th} stage of a multistage desalination process. The practical specific energy, E, for an N-stage process is determined by summing the energy requirement along all the stages and dividing by the overall permeate volume (i.e., overall recovery yield, Y, multiplied by influent feed volume, V_f):

$$E = \frac{\sum(\Delta P_i \Delta V_i)}{YV_f} = \frac{\sum\left[\Delta P_i\left(\Delta \nu_i V_f\right)\right]}{YV_f} = \frac{k}{Y}\sum\left[\Delta\pi_i\left(\nu_i - \nu_{i\text{-}1}\right)\right] \quad \text{(S1)}$$

where $\Delta\nu_i$ is the permeate volume of the i^{th} stage normalized by the input feed volume ($\Delta\nu_i = \Delta V_i/V_f$) and can be considered as the "yield" in stage i. By corollary, ν_i is the cumulative permeate "yield rate" up to the i^{th} stage, $\nu_i = \nu_{i-1}+\Delta\nu_i$.

To maintain adequate water flux across the entire length of the membrane module for practical operation, the hydraulic pressure applied is in excess of the maximum osmotic pressure difference across the membrane within the stage, $\Delta\pi$. In this study, overpressurization applied in all stages and for all configurations is 1.15 and is denoted by k (i.e., $\Delta P = k\Delta\pi = 1.15\Delta\pi$, expressed in eq S1). The membranes in this analysis are assumed to be perfectly selective, i.e., rejecting 100% of the salt and only allowing water to permeate across. Osmotic pressures are determined using the van't Hoff equation with NaCl solutions at temperature of 298 K.

Specific Energy of Desalination with *N*-stage COMRO. Figure S1 shows an N-stage cascading osmotically mediated RO (COMRO), comprising N bilateral countercurrent (BCC) stages and terminated with a conventional RO stage. Stream parameter of osmotic pressure, π, and stage operating condition of applied hydraulic pressure, ΔP, are indicated in the schematic. In this analysis, the permeate volume across the terminal RO stage, i.e., product water, is set to be equivalent to the cumulative permeation attained across the N BCC stages. And because the membranes are assumed to be perfectly selective, the retentate exiting the terminal RO stage, thus, has the same osmotic pressure and flow rate as the input feed stream. Due to water permeation, the salinity and π along the membrane within each BCC stage are changing and, therefore, the osmotic pressure difference is also varying. For all BCC stages along the process, the highest osmotic pressure difference across the membrane invariably occurs on the left side of the stage (further elaborated in the later subsection on operating hydraulic pressure). Accordingly, the hydraulic pressure applied at the i^{th} BCC stage is $\Delta P_i = k\Delta\pi_i = k\left(\pi_i^{\text{H}} - \pi_i^{\text{L}}\right)$, where π_i is the osmotic pressure on the left side of stage i and superscripts H and L denote high and low concentration streams (bottom retentate and top dilute streams, respectively). Thus, π_i^{H} is the osmotic pressure of the high concentration retentate flow leaving the lower half of the i^{th} BCC stage, and π_i^{L} is the osmotic pressure of the low concentration dilute flow entering the upper half of the stage (Figure S1).

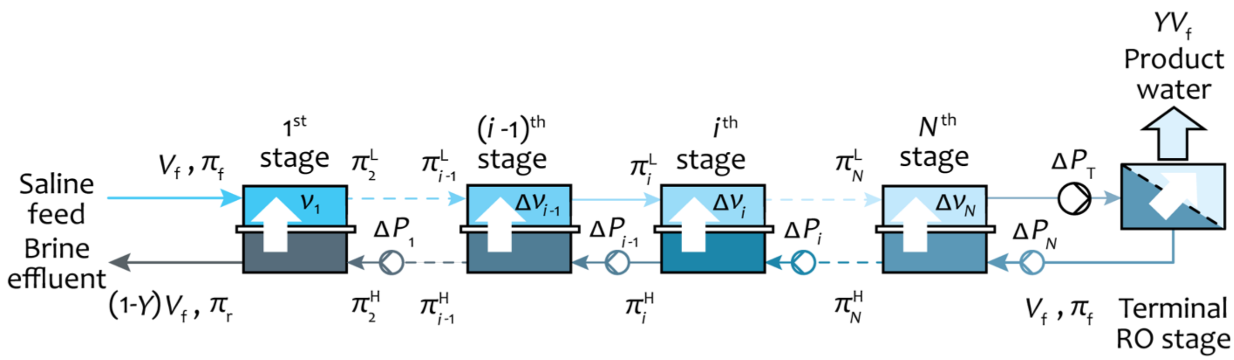

Figure S1. Schematic diagram of an N-stage COMRO. The osmotic pressure of the streams, π, and the applied hydraulic pressure of the stages, ΔP, are indicated. Color intensities of the streams and stages are representative of the salinities. Auxiliary components like energy recovery devices (ERDs) and circulation pumps are omitted for clarity of presentation.

To obtain a specific overall recovery, COMRO can be operated under different schemes, where the permeate volume of each BCC stage is altered but the sum across all the stages remains constant. An example of a stage operating scheme is to have equal permeate volume across all N BCC stages. Note that this stage operating scheme for COMRO is used for the results and discussions presented in the main manuscript, and is also employed for DPRO (working principles of CF/OARO dictate ΔV_i to be constant in all stages and equal to YV_f). Alternative schemes are presented and discussed in a later section. The operating scheme prescribes the permeate volume of each BCC stage to be constant, which means $\Delta v_i = v_i - v_{i-1} = Y/N$ in COMRO. The cumulative permeation up to the i^{th} stage is, hence, $v_i = iY/N$ (i.e., at the N^{th} BCC stage, $v_N = Y$). The osmotic pressure of high concentration retentate stream leaving and low concentration dilute stream entering the i^{th} BCC stage are expressed by eqs S2 and S3, respectively:

$$\pi_i^{\mathrm{H}} = \frac{\pi_{\mathrm{f}}}{1-\left(Y-\nu_{i-1}\right)} = \frac{N\pi_{\mathrm{f}}}{N-\left(N-i+1\right)Y} \tag{S2}$$

$$\pi_i^{\mathrm{L}} = \frac{\pi_{\mathrm{f}}}{1+\nu_{i-1}} = \frac{N\pi_{\mathrm{f}}}{N+\left(i-1\right)Y} \tag{S3}$$

The specific energy for COMRO is the sum of the energy requirement along all the BCC stages and the terminal RO stage. Using $\Delta P_i = k\Delta\pi_i = k\left(\pi_i^{\mathrm{H}} - \pi_i^{\mathrm{L}}\right)$ and substituting eqs S2 and S3 into eq S1 yields:

$$E_{\mathrm{COMRO}} = \frac{\Delta P_{\mathrm{T}}Y + \sum_{i=1}^{N}\left(\Delta P_i \Delta\nu_i\right)}{Y} = k\pi_{\mathrm{f}}\left(1 + NY\sum_{i=1}^{N}\left\{[N+Y(i-1)][N-(N-i+1)Y]\right\}^{-1}\right) \tag{S4}$$

where ΔP_{T} is the applied pressure in the terminal RO stage ($\Delta P_{\mathrm{T}} = k\pi_{\mathrm{f}}$, because retentate exiting the terminal RO stage has same osmotic pressure as the input feed stream). In eq S4, the BCC stages are numbered from the influent feed end (i.e., from left side). However, the stages can also be numbered in the opposite direction, starting from the terminal RO end (i.e., from right side). This enables the substitution of $i = N - i + 1$ into eq S4, to give eq 2 of the main manuscript:

$$E_{\mathrm{COMRO}} = k\pi_{\mathrm{f}}\left(1 + NY\sum_{i=1}^{N}\left\{\left[N + Y\left(N-i\right)\right]\left(N-iY\right)\right\}^{-1}\right) \tag{2}$$

Specific Energy of Desalination with *N*-stage DPRO. Figure S2 shows an N-stage direct pass RO (DPRO), made up of N conventional RO stages connected in series. Stream parameter of osmotic pressure, π, and stage operating condition of applied hydraulic pressure, ΔP, along the process are indicated in the schematic. For consistency of assessment, the permeate volume across each stage is set to be $V_{\mathrm{f}}Y/N$ (i.e., similar to the stage operating scheme of COMRO). Hence $y_i = iY/N$, where y_i is the cumulative recovery rate up to the i^{th} stage and is defined as the ratio of the cumulative permeate volume to the input feed volume (i.e., $y_N = Y$). Note that y_i is differentiated from ν_i of COMRO (and also CF/OARO) because permeate from all DPRO stages directly contribute to the overall product water. Because the membranes are assumed to have 100% salt rejection, the osmotic pressure of the retentate leaving the i^{th} stage is, thus, $\pi_i = \pi_{\mathrm{f}}/(1-y_i)$ and can be further expressed as:

$$\pi_i = \frac{\pi_f N}{N - iY} \tag{S5}$$

Substituting eq S1 with eq S5 and $\Delta y = y_i - y_{i-1} = Y/N$ yields:

$$E_{\mathrm{DPRO}} = \frac{\sum_{i=1}^{N}\left(\Delta P_i \Delta V_i\right)}{YV_f} = \frac{\sum_{i=1}^{N}\left[k\pi_i V_f\left(y_i - y_{i-1}\right)\right]}{YV_f} \tag{S6}$$

which further simplifies to give eq 3 of the main manuscript:

$$E_{\mathrm{DPRO}} = k\pi_f\left[\sum_{i=1}^{N}\left(N - iY\right)^{-1}\right] \tag{3}$$

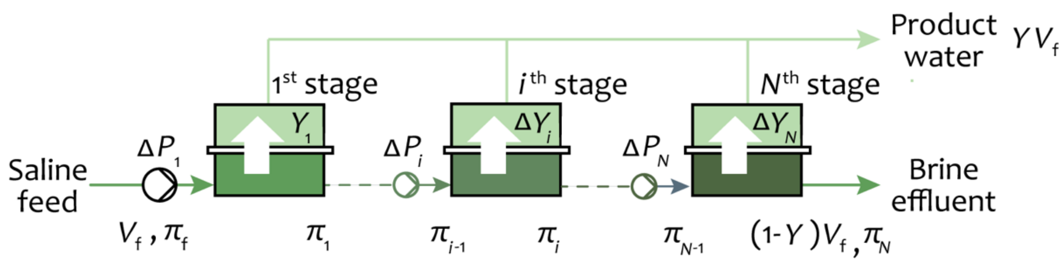

Figure S2. Schematic diagram of an N-stage DPRO. The osmotic pressure of the stream, π, and the applied hydraulic pressure of the stage, ΔP, are indicated. Color intensities of the streams and stages are representative of the salinities. Auxiliary components like ERDs and circulation pumps are omitted for clarity of presentation.

Specific Energy of Desalination with *N*-stage CF/OARO. Figure S3 shows an N-stage counter flow/osmotically assisted RO (CF/OARO), consisting of N bilateral stages terminated with a conventional RO stage. Note that this analysis considered countercurrent flow for the bilateral stages, although the system can also be configured to have co-current flow (i.e., flow on both sides are in the same direction). Stream parameter of osmotic pressure, π, and stage operating condition of applied hydraulic pressure, ΔP, along the process are similarly indicated in the schematic. Osmotic pressure of the stream leaving stage i and entering the preceding $(i-1)^{\text{th}}$ stage is denoted by π_i^{i-1} (i.e., subscript signify the stage exited from and superscript indicates the stage entered into). Based on the working principles of CF/OARO,[1, 2] the permeate volume at each stage along the process is constant, thus $v_1 = \Delta v_1 = \Delta v_2 = \ldots = \Delta v_i = \ldots = \Delta v_N = Y$.

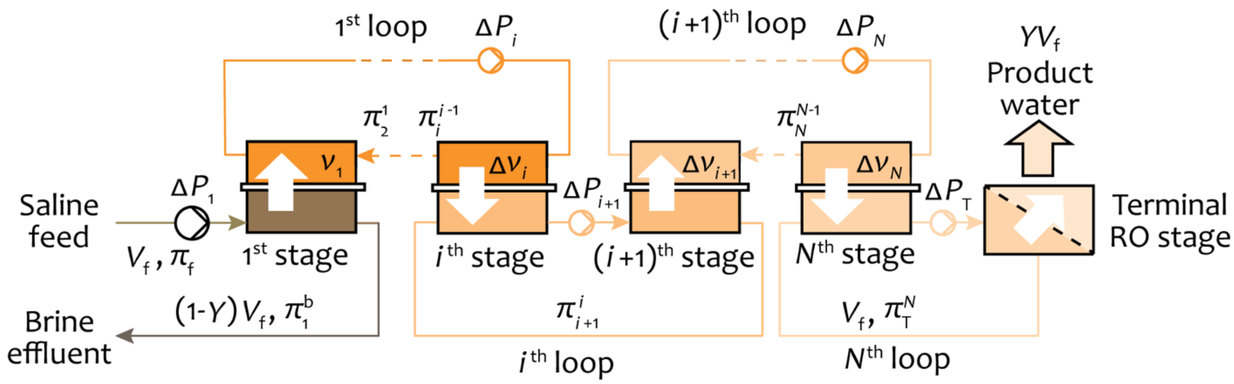

Figure S3. Schematic diagram of an *N*-stage CF/OARO. The osmotic pressure of the stream, π, and the applied hydraulic pressure of the stage, ΔP, are indicated. Color intensities of the streams and stages are representative of the salinities. Auxiliary components like ERDs and circulation pumps are omitted for clarity of presentation.

For all bilateral stages, the highest osmotic pressure difference across the membrane occurs on the side where the stream of the $(i-1)^{\text{th}}$ loop leaves the i^{th} stage and is circulated back to the preceding $(i-1)^{\text{th}}$ stage (or, alternatively, the side on the i^{th} stage where the i^{th} loop sweep stream enters). For instance, the highest osmotic pressure across the i^{th} stage is $\Delta\pi_i = \pi_i^{i-1} - \pi_{i+1}^{i}$ and the osmotic pressure across the terminal RO stage is $\Delta\pi_{\text{T}} = \pi_{\text{T}}^{N}$ (exiting retentate osmotic pressure), while $\Delta\pi_1 = \pi_1^{\text{b}} - \pi_2^{1}$ (π_1^{b} is the osmotic pressure of the brine effluent). Substituting the osmotic pressures of the streams into eq S1 yields:

$$E_{\text{CF/OARO}} = \frac{k}{Y}\left[Y\left(\pi_1^{\text{b}} - \pi_2^{1} + \pi_2^{1} - \pi_3^{2} + \cdots + \pi_i^{i-1} - \pi_{i+1}^{i} + \cdots + \pi_N^{N-1} - \pi_{\text{T}}^{N}\right) + Y\pi_{\text{T}}^{N}\right] \qquad \text{(S7)}$$

which simplifies to eq 4 of the main manuscript:

$$E_{\text{CF/OARO}} = \frac{k\pi_{\text{f}}}{1-Y} \qquad (4)$$

Eq 4 indicates that the specific energy requirement of desalination with CF/OARO is independent of the number of bilateral stages, *N*, and is only determined by the initial feed salinity and the overall recovery rate.

Operating Hydraulic Pressures in *N*-stage COMRO, DPRO, and CF/OARO. The maximum pressurization required for COMRO and CF/OARO operation is the greater of the

ΔP between the bilateral stages and the terminal conventional RO stage. The applied hydraulic pressure in the i^{th} bilateral stage, ΔP_i, is the highest osmotic pressure difference within the stage, $\Delta\pi_i$, multiplied by the operational overpressurization factor, k ($\Delta P_i = k\Delta\pi_i$). While for the terminal RO stage, the applied hydraulic pressure, ΔP_{T}, is determined by the osmotic pressure of the exiting retentate, which is set to be identical to the influent feed stream in this analysis, i.e., $\Delta P_{\text{T}} = k\pi_{\text{f}}$.

The largest osmotic pressure difference within the i^{th} BCC stage of COMRO, as discussed in the earlier subsection, is $\Delta\pi_i = \pi_i^{\text{H}} - \pi_i^{\text{L}}$. Substituting in eqs S2 and S3 and incorporating the overpressurization factor, k, gives the applied hydraulic pressure of the i^{th} BCC stage:

$$\Delta P_{i,\text{BCC-COMRO}} = k\Delta\pi_i = k(\pi_i^{\text{H}} - \pi_i^{\text{L}}) = k\pi_{\text{f}} N^2 Y \left\{[N + Y(i-1)][N-(N-i+1)Y]\right\}^{-1} \tag{S8}$$

Note that the stages are numbered from the influent feed end, i.e., as depicted in Figure S1 (unlike for eq 2). Eq S8 indicates $\Delta P_{i,\text{BCC-COMRO}}$ is lower when i increases. Thus, the applied hydraulic pressure among the BCC stages is greatest at the 1st stage, i.e., i = 1, and is $\Delta P_{1,\text{BCC-COMRO}} = k\pi_{\text{f}}Y/(1-Y)$. The applied hydraulic pressure of the terminal RO stage is $\Delta P_{\text{T-COMRO}} = k\pi_{\text{f}}$. The highest applied hydraulic pressure of an N-stage COMRO, $\Delta P_{\text{max,COMRO}}$, is the higher of the two:

$$\Delta P_{\text{max,COMRO}} = \begin{cases} \Delta P_{1,\text{BCC-COMRO}} = \dfrac{k\pi_{\text{f}}Y}{1-Y} & Y \in [0.5, 1) \\ \Delta P_{\text{T-COMRO}} = k\pi_{\text{f}} & Y \in (0, 0.5) \end{cases} \tag{S9}$$

For DPRO, the expression for $\Delta P_{i,\text{DPRO}}$, eq S10, is derived using $y_{\text{i}} = iY/N$ and $\pi_i = \pi_{\text{f}}/(1-y_i)$:

$$\Delta P_{i,\text{DPRO}} = \frac{k\pi_{\text{f}} N}{N - iY} \tag{S10}$$

The applied hydraulic pressure increases with i and is greatest at the N^{th} stage:

$$\Delta P_{\text{max,DPRO}} = \frac{k\pi_{\text{f}}}{1-Y} \tag{S11}$$

In the CF/OARO analysis, the sweep stream of all the recirculating loops entering a bilateral stage to be concentrated is set to have the same volumetric flow rate as the input feed stream (or, alternatively, the sweep stream leaving a bilateral stage after dilution has the same flow rate as the input feed). Because of water flux across the membrane in the bilateral stage, the sweep stream is concentrated and the volumetric flow rate shrinks by the factor $1-Y$, while on the other side the volumetric flow rate of the diluted sweep stream swells by the factor $1/(1-Y)$. Like COMRO, the osmotic pressure of the retentate exiting the terminal RO stage is also π_f. Thus, the difference in osmotic pressure between the feed influent and the retentate of the terminal RO stage is $\pi_f/(1-Y)-\pi_f = \pi_f Y/(1-Y)$. This overall difference is set to evenly distribute among the N bilateral stages. Hence the highest osmotic pressure difference experienced with each bilateral stage is identical for all stages and, consequently, the applied hydraulic pressure, $\Delta P_{i,\mathrm{CF/OARO}}$, is constant:

$$\Delta P_{i,\mathrm{CF/OARO}} = \frac{k\pi_f Y}{N(1-Y)} \tag{S12}$$

The maximum applied hydraulic pressure in CF/OARO is the higher of $\Delta P_{i,\mathrm{CF/OARO}}$ and the applied hydraulic pressure in the terminal RO stage, $\Delta P_{\mathrm{T\text{-}CF/OARO}} = k\pi_f$:

$$\Delta P_{\max,\mathrm{CF/OARO}} = \begin{cases} \Delta P_{i,\mathrm{CF/OARO}} = \dfrac{k\pi_f Y}{N(1-Y)} & N \in \left[1, \dfrac{Y}{1-Y}\right] \\ \Delta P_{\mathrm{T\text{-}CF/OARO}} = k\pi_f & N \in \left(\dfrac{Y}{1-Y}, \infty\right) \end{cases} \tag{S13}$$

ANALYSIS RESULTS OF OPERATING HYDRAULIC PRESSURES AND SPECIFIC ENERGY CONSUMPTION

Desalination of Hypersaline Brine. The specific energy consumption, E_i, and operating hydraulic pressure, ΔP_i, of the i^{th} individual stages to desalinate hypersaline feed of c_f = 70 000 ppm TDS to Y = 50% for COMRO, DPRO, and CF/OARO are presented in Tables S1, S2, and S3, respectively. Total specific energy requirement, E, and overall highest hydraulic pressure, $\Delta P_{\max}$, are also shown. For COMRO and CF/OARO, N = 1–4 bilateral stages are examined, while "Terminal" denotes the terminal conventional RO stage. I.e., N = 4 indicates 4 bilateral stages and one terminal RO stage, yielding 5 stages in total. For DPRO, N = 2–5 stages are considered (note that N = 1 is equivalent to conventional single-stage RO). The values presented in Tables S1–3 correspond to Figure 4A in the main manuscript.

Table S1. Specific energy consumption, E_i, and operating hydraulic pressure, ΔP_i, of COMRO to desalinate c_f = 70 000 ppm TDS hypersaline feed to Y = 50%.

Hypersaline desalination: c_f = 70 000 ppm TDS, Y = 50%
COMRO

N	Individual stage	Stage					Overall	
		i = 1	i = 2	i = 3	i = 4	Terminal		
1	E_i (kWh/m^3)	1.89	—	—	—	1.89	E (kWh/m^3)	3.79
	ΔP_i (bar)	68.2	—	—	—	68.2	ΔP_{max} (bar)	68.2
2	E_i (kWh/m^3)	0.95	0.51	—	—	1.89	E (kWh/m^3)	3.35
	ΔP_i (bar)	68.2	36.4	—	—	68.2	ΔP_{max} (bar)	68.2
3	E_i (kWh/m^3)	0.63	0.41	0.28	—	1.89	E (kWh/m^3)	3.22
	ΔP_i (bar)	68.2	43.8	30.7	—	68.2	ΔP_{max} (bar)	68.2
4	E_i (kWh/m^3)	0.47	0.34	0.25	0.20	1.89	E (kWh/m^3)	3.15
	ΔP_i (bar)	68.2	51.7	40.6	32.8	27.1	ΔP_{max} (bar)	68.2

Table S2. Specific energy consumption, E_i, and operating hydraulic pressure, ΔP_i, of DPRO to desalinate c_f = 70 000 ppm TDS hypersaline feed to Y = 50%.

Hypersaline desalination: c_f = 70 000 ppm TDS, Y = 50%
DPRO

N	Individual stage	Stage					Overall	
		i = 1	i = 2	i = 3	i = 4	i = 5		
2	E_i (kWh/m^3)	1.26	1.89	—	—	—	E (kWh/m^3)	3.16
	ΔP_i (bar)	90.9	136.4	—	—	—	ΔP_{max} (bar)	136.4
3	E_i (kWh/m^3)	0.76	0.95	1.26	—	—	E (kWh/m^3)	2.97
	ΔP_i (bar)	81.8	102.3	136.4	—	—	ΔP_{max} (bar)	136.4
4	E_i (kWh/m^3)	0.54	0.63	0.76	0.95	—	E (kWh/m^3)	2.88
	ΔP_i (bar)	77.9	90.9	109.1	136.4	—	ΔP_{max} (bar)	136.4
5	E_i (kWh/m^3)	0.42	0.47	0.54	0.63	0.76	E (kWh/m^3)	2.82
	ΔP_i (bar)	75.8	85.2	97.4	113.6	136.4	ΔP_{max} (bar)	136.4

Table S3. Specific energy consumption, E_i, and operating hydraulic pressure, ΔP_i, of CF/OARO to desalinate c_f = 70 000 ppm TDS hypersaline feed to Y = 50%.

Hypersaline desalination: c_f = 70 000 ppm TDS, Y = 50%

CF/OARO

N	Individual stage	Stage					Overall	
		i = 1	i = 2	i = 3	i = 4	Terminal		
1	E_i (kWh/m^3)	1.89	—	—	—	1.89	E (kWh/m^3)	3.79
	ΔP_i (bar)	68.2	—	—	—	68.2	ΔP_{max} (bar)	68.2
2	E_i (kWh/m^3)	0.95	0.95	—	—	1.89	E (kWh/m^3)	3.79
	ΔP_i (bar)	34.1	34.1	—	—	68.2	ΔP_{max} (bar)	68.2
3	E_i (kWh/m^3)	0.63	0.63	0.63	—	1.89	E (kWh/m^3)	3.79
	ΔP_i (bar)	22.7	22.7	22.7	—	68.2	ΔP_{max} (bar)	68.2
4	E_i (kWh/m^3)	0.47	0.47	0.47	0.47	1.89	E (kWh/m^3)	3.79
	ΔP_i (bar)	17.0	17.0	17.0	17.0	68.2	ΔP_{max} (bar)	68.2

High Recovery Seawater Desalination. The specific energy consumption, E_i, and operating hydraulic pressure, ΔP_i, of the i^{th} individual stages to desalinate seawater feed of 35 000 ppm TDS to enhanced recovery rate of Y = 70% for COMRO, DPRO, and CF/OARO are presented in Tables S4, S5, and S6, respectively. Total specific energy requirement, E, and overall highest hydraulic pressure, ΔP_{max}, are also shown. The number of stages assessed are the same as the earlier hypersaline desalination scenario. The values presented in Tables S4–6 correspond to Figure 4B in the main manuscript.

Table S4. Specific energy consumption, E_i, and operating hydraulic pressure, ΔP_i, of COMRO to desalinate c_f = 35 000 ppm TDS seawater feed to Y = 70%.

High recovery seawater desalination: c_f = 35 000 ppm TDS, Y = 70%
COMRO

N	Individual stage	Stage					Overall	
		i = 1	i = 2	i = 3	i = 4	Terminal		
1	E_i (kWh/m^3)	2.21	—	—	—	0.95	E (kWh/m^3)	3.16
	ΔP_i (bar)	79.6	—	—	—	34.1	ΔP_{max} (bar)	79.6
2	E_i (kWh/m^3)	1.10	0.38	—	—	0.95	E (kWh/m^3)	2.43
	ΔP_i (bar)	79.6	27.2	—	—	34.1	ΔP_{max} (bar)	79.6
3	E_i (kWh/m^3)	0.74	0.34	0.20	—	0.95	E (kWh/m^3)	2.22
	ΔP_i (bar)	79.6	36.3	21.2	—	34.1	ΔP_{max} (bar)	79.6
4	E_i (kWh/m^3)	0.55	0.30	0.19	0.13	0.95	E (kWh/m^3)	2.12
	ΔP_i (bar)	79.6	42.8	27.2	19.0	34.1	ΔP_{max} (bar)	79.6

Table S5. Specific energy consumption, E_i, and operating hydraulic pressure, ΔP_i, of DPRO to desalinate c_f = 35 000 ppm TDS seawater feed to Y = 70%.

High recovery seawater desalination: c_f = 35 000 ppm TDS, Y = 70%
DPRO

N	Individual stage	Stage					Overall	
		i = 1	i = 2	i = 3	i = 4	i = 5		
2	E_i (kWh/m^3)	0.73	1.58	—	—	—	E (kWh/m^3)	2.31
	ΔP_i (bar)	52.5	113.6	—	—	—	ΔP_{max} (bar)	113.6
3	E_i (kWh/m^3)	0.41	0.59	1.05	—	—	E (kWh/m^3)	2.06
	ΔP_i (bar)	44.5	63.9	113.6	—	—	ΔP_{max} (bar)	113.6
4	E_i (kWh/m^3)	0.29	0.36	0.50	0.79	—	E (kWh/m^3)	1.94
	ΔP_i (bar)	41.3	52.5	71.8	113.6	—	ΔP_{max} (bar)	113.6
5	E_i (kWh/m^3)	0.22	0.26	0.33	0.43	0.63	E (kWh/m^3)	1.87
	ΔP_i (bar)	39.6	47.4	58.8	77.5	113.6	ΔP_{max} (bar)	113.6

Table S6. Specific energy consumption, E_i, and operating hydraulic pressure, ΔP_i, of CF/OARO to desalinate c_f = 35 000 ppm TDS seawater feed to Y = 70%.

High recovery seawater desalination: c_f = 35 000 ppm TDS, Y = 70%
CF/OARO

N	Individual stage	Stage					Overall	
		i = 1	i = 2	i = 3	i = 4	Terminal		
1	E_i (kWh/m^3)	2.21	—	—	—	0.95	E (kWh/m^3)	3.16
	ΔP_i (bar)	79.6	—	—	—	34.1	ΔP_{max} (bar)	79.6
2	E_i (kWh/m^3)	1.10	1.10	—	—	0.95	E (kWh/m^3)	3.16
	ΔP_i (bar)	39.8	39.8	—	—	34.1	ΔP_{max} (bar)	39.8
3	E_i (kWh/m^3)	0.74	0.74	0.74	—	0.95	E (kWh/m^3)	3.16
	ΔP_i (bar)	26.5	26.5	26.5	—	34.1	ΔP_{max} (bar)	34.1
4	E_i (kWh/m^3)	0.55	0.55	0.55	0.55	0.95	E (kWh/m^3)	3.16
	ΔP_i (bar)	19.9	19.9	19.9	19.9	34.1	ΔP_{max} (bar)	34.1

STAGE OPERATING SCHEMES OF COMRO AND DPRO

Stage Operating Schemes of Multistage Desalination. To achieve the same overall recovery rate, Y, in a multistage desalination process, different operating parameters and objectives of ΔP_i and ΔV_i, respectively, for the stages can be selected. We term these different approaches “stage operating schemes”. The analysis presented in the main manuscript adopted a stage operating scheme of constant permeate volume at each bilateral stage in COMRO and CF/OARO, and at each conventional RO stage in DPRO. This operating scheme yields $\Delta v = v_i - v_{i\text{-}1} = Y/N$ and $v_i = iY/N$ for COMRO and CF/OARO (equivalently $\Delta y = y_i - y_{i\text{-}1} = Y/N$ and $y_i = iY/N$ for DPRO).

An alternative stage operating scheme is to obtain a constant increment in the applied hydraulic pressure along the stages, i.e., the difference in the applied hydraulic pressure between the i^{th} stage and the preceding $(i-1)^{\text{th}}$ stage is constant, i.e., $\Delta\pi_i - \Delta\pi_{i\text{-}1} = (\Delta\pi_N - \Delta\pi_1)/N$. Since CF/OARO is strictly confined by the requirement of constant permeation in all stages along the process, this operating scheme is only applicable to COMRO and DPRO. The stage operating scheme adopted in the main manuscript and discussed in the earlier section is termed “constant permeate increment” and the operation mode elaborated here in this section is termed “constant pressure increment”. The specific energy requirements under these two stage operating schemes are $E^{\Delta v}$ and $E^{\Delta P}$, respectively.

Specific Energy of *N*-stage COMRO with Alternative Stage Operating Scheme. For the constant pressure increment scheme, the largest pressurization among the BCC stages still occurs in the 1st stage (as with the constant permeate increment scheme). The osmotic pressure difference across the membrane in the N^{th} BCC stage is set as $\Delta\pi_1/N$. Thus, the constant stagewise increment in applied hydraulic pressure is $-\Delta\pi_1/N$. According to this stepped change, the osmotic pressure difference in the $(i+1)^{\mathrm{th}}$ stage is:

$$\Delta\pi_{i+1} = \frac{\Delta\pi_1}{N}\left[N-(i+1)+1\right] = \frac{\pi_{\mathrm{f}} Y (N-i)}{N(1-Y)} \tag{S14}$$

Meanwhile, $\Delta\pi_{i+1}$ is also defined as (eqs S2 and S3):

$$\Delta\pi_{i+1} = \pi_{i+1}^{\mathrm{H}} - \pi_{i+1}^{\mathrm{L}} = \frac{\pi_{\mathrm{f}} Y}{(1-Y+\nu_{\mathrm{i}})(1+\nu_{\mathrm{i}})} \tag{S15}$$

Equating eqs S14 and S15, and solving for ν_{i} yields:

$$\nu_i = \sqrt{\frac{(N-i)Y^2 + 4N(1-Y)}{4(N-i)}} - \frac{2-Y}{2} \quad i \in [1, N) \tag{S16}$$

Note that the water flux can be negative for certain N, i, and Y, i.e., water permeates from the more dilute feed stream in the top half to the more concentrated stream in the lower half of the BCC stage (further discussed in a later section). This is an artifact of the externally imposed constant pressure increment scheme and not an inherent operating regime of COMRO. Such modes of operation are counterproductive to the overall desalination objective and, hence, are not included for further assessment. The specific energy under the constant pressure increment operating scheme, $E_{\mathrm{COMRO}}^{\Delta P}$, is:

$$E_{\mathrm{COMRO}}^{\Delta P} = \frac{\Delta P_{\mathrm{T}} Y + \sum_{i=1}^{N}\left[\Delta P_i (\nu_i - \nu_{i\text{-}1})\right]}{Y} = k\pi_{\mathrm{f}}\left\{1 + \left[N(1-Y)\right]^{-1}\sum_{i=1}^{N}\left[(N-i+1)(\nu_i - \nu_{i\text{-}1})\right]\right\} \tag{S17}$$

Specific Energy of *N*-stage DPRO with Alternative Stage Operating Scheme. *N*-stage DPRO can similarly be operated under constant pressure increment scheme.[3] The constant osmotic pressure increment between stages is $\pi_i - \pi_{i-1} = \pi_f Y/[(1-Y)N]$. Therefore, the retentate osmotic pressure of the i^{th} stage, π_i, is:

$$\pi_i = \pi_f + i(\pi_i - \pi_{i-1}) = \frac{\pi_f \left[iY + N(1-Y) \right]}{N(1-Y)} \tag{S18}$$

As discussed in the previous section, $y_i = 1-\pi_f/\pi_i$. Substituting π_i with eq S18 and incorporating into $\Delta y_i = y_i - y_{i-1}$ yields the recovery rate of the i^{th} stage, Δy_i:

$$\Delta y_i = y_i - y_{i\text{-}1} = \frac{\pi_f (\pi_i - \pi_{i-1})}{\pi_i \pi_{i-1}} \tag{S19}$$

Further substituting eqs S18 and S19 into eq S1 gives the specific energy consumption under constant pressure increment scheme:

$$E_{\text{DPRO}}^{\Delta P} = \frac{\sum_{i=1}^{N} (\Delta P \Delta V_i)}{YV_f} = \frac{k\pi_f}{Y} \sum_{i=1}^{N} \frac{(\pi_i - \pi_{i-1})}{\pi_{i-1}} = k\pi_f \sum_{i=1}^{N} \left[(i-1)Y + N(1-Y) \right]^{-1} \tag{S20}$$

IMPACTS OF STAGE OPERATING SCHEMES ON SPECIFIC ENERGY REQUIREMENT

Impacts of Stage Operating Schemes on *N*-stage COMRO. An inspection of eq S16 reveals that the constant pressure increment operating scheme may not always result in positive water flux throughout all stages. For instance, while desalinating a 70 000 ppm TDS feed stream to 50% recovery (i.e., scenario discussed in the main manuscript), v_2 turns out to be equal to Y at $N = 3$, which means there is no net permeation in the 3rd stage, $\Delta v_3 = 0$. Adding more BCC stages such that $N > 3$ results in $v_i > Y$ in some stages for certain range of recovery rates, indicating negative water flux in some BCC stages, i.e., permeation from the low concentration half to the high concentration half ($\Delta v < 0$). Negative water flux in any of the BCC stages is counterproductive to the intended objective of diluting the feed stream and is, hence, not desirable. Therefore, the constant pressure increment scheme is only valid when $N = 1$ and 2 at $Y = 50\%$. The applicability of the constant pressure increment operating scheme is, hence, confined by this water flux limitation.

Figure S4 presents a comparison of specific energy requirement between the two stage operating schemes of constant permeate increment and constant pressure increment, $E_{\mathrm{COMRO}}^{\Delta v}$ and $E_{\mathrm{COMRO}}^{\Delta P}$, respectively. To desalinate an influent feed at 70 000 ppm TDS to 50% recovery (i.e., first scenario discussed in the main manuscript), $E_{\mathrm{COMRO}}^{\Delta P}$ is exactly identical to $E_{\mathrm{COMRO}}^{\Delta v}$ for $N = 1$, but is 0.8% higher for $N = 2$ (Figure S4A). In the second scenario discussed in the main manuscript, where an input feed stream at 35 000 ppm TDS is desalinated to a high recovery of 70%, the specific energy requirement difference between the two schemes is within ±3% for $N = 1$–5 (Figure S4B). This comparison reveals the specific energy requirements of the two stage operating schemes for COMRO desalination are only marginally different. Because the constant permeate increment operating scheme is not complicated by possible negative water flux under some conditions, it is used for the main analyses in this study.

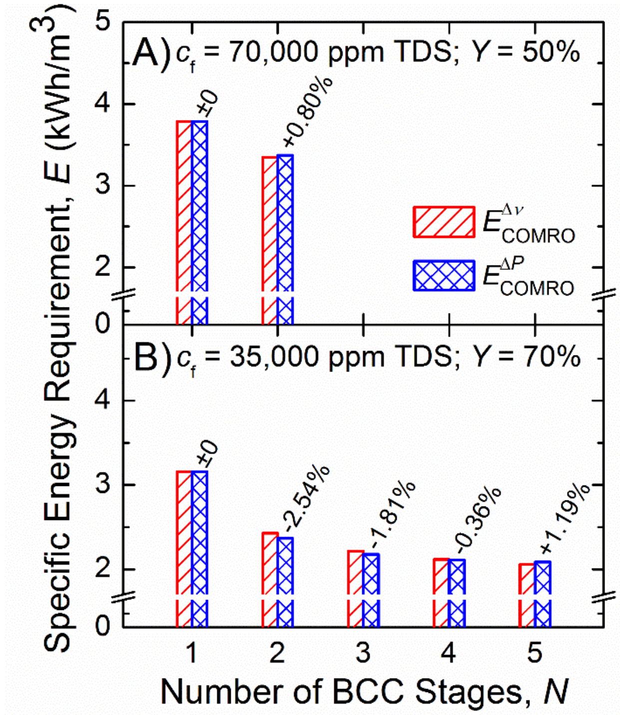

Figure S4. Specific energy requirement, E, with constant permeate increment and constant pressure increment stage operating schemes (red and blue column, respectively) for (A) desalinating a hypersaline feed at 70 000 ppm TDS to Y = 50% and (B) desalinating a seawater feed at 35,000 ppm TDS to Y = 70%. COMRO configurations with 1–5 BCC stages are evaluated. Osmotic pressures were determined using the van't Hoff equation with NaCl solutions at temperature of 298 K, and the operational overpressurization factor, k, is 1.15.

Impacts of Stage Operating Schemes on *N*-stage DPRO. Substituting $i = N-i+1$ into the expression for specific energy requirement with constant pressure increment stage operating scheme in DPRO, eq S20, recovers eq 3 of the main manuscript. (The substitution is equivalent to numbering the stages from the opposite direction.) This signifies that the two stage operating schemes of constant permeate increment and constant pressure increment result in exactly the same specific energy of desalination in N-stage DPRO. However, neither of these two schemes attains the lowest possible practical specific energy in DPRO. This is illustrated in Figure S5, which shows the specific energy requirement profiles of a 2-stage DPRO process as a function of the 1st stage recovery rate, y_1. In this example, input feed at 70 000 ppm TDS is desalinated to overall recoveries of 40%, 50%, and 60%.

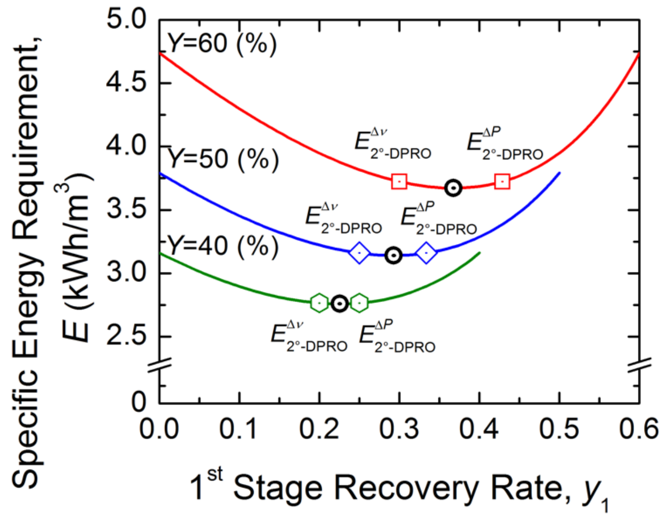

Figure S5. Specific energy requirement, E, of the 2-stage DPRO for desalination of hypersaline feed at 70 000 ppm TDS to recovery, Y, of 40%, 50%, and 60% (green, blue and red lines, respectively) as a function of 1st stage recovery rate, y_1. Black circle symbols denote the minimum E of each profile. Green, blue, and red symbols represent E attained by the constant permeate increment scheme and the constant pressure increment scheme, labelled $E^{\Delta\nu}_{2°\text{–DPRO}}$ and $E^{\Delta\nu}_{2°\text{–DPRO}}$, respectively. Osmotic pressures were determined using the van't Hoff equation with NaCl solutions at temperature of 298 K, and the operational overpressurization factor, k, is 1.15.

To attain overall recovery of Y in 2-stage DPRO, the recovery rate of the individual stages can be adjusted while maintaining $Y = y_1+y_2$. Note that the process is not operating in either constant permeate increment and constant pressure increment scheme, except at the points denoted by the symbols. Figure S5 shows that as y_1 increases, the overall specific energy reduces to a minimum and then increases. This minimum occurs when the derivative of E with respect to y_1 is equal to zero:

$$\frac{dE_{2°\text{–DPRO}}}{dy_1} = \frac{k\pi_{\mathrm{f}}}{Y}\left[\frac{1}{(1-y_1)^2} - \frac{1}{1-Y}\right] \tag{S21}$$

For each E profile with a certain Y, the specific energy requirements with the two stage operating schemes investigated here are always on either side of the minimum E (Figure S5). Therefore, neither the constant permeate increment nor the constant pressure increment operating scheme minimizes the desalination energy requirement. Nonetheless, these two stage operating schemes are able to achieve E that are relatively close to the minimum, as visually represented in Figure S5. Note that for $N \geq 3$, it is less straightforward to optimize for minimum E.

APPLICATION OF COMRO TO TREAT ULTRAHIGH SALINITY BRINE

COMRO offers great flexibility in stage configurations and operating schemes, and can be employed to treat hypersaline brines >>70 000 ppm TDS without exceeding the ≈85 bar hydraulic pressure upper limit in any of the BCC stages or the terminal RO stage. Figure S6 shows a 2-stage COMRO operation designed to desalinate hypersaline feed at 150 000 ppm TDS to 20% recovery (or, equivalently, dewater the stream by 25%). In this operation, the applied hydraulic pressure of the terminal RO stage is set to the RO operating upper limit of 85 bar (previously discussed operating schemes had the concentration and volumetric flow rate of the terminal RO stage retentate equivalent to the input feed stream), and the two BCC stages are operated under the constant permeate increment scheme ($\Delta v_1 = \Delta v_2$). Volumetric flow rate, TDS concentration, and osmotic pressure of the streams are indicated in the Figure. The applied hydraulic pressure along the BCC stages are ΔP_1 = 36.5 bar and ΔP_2 = 15.9 bar, far below the ≈85 bar limit.

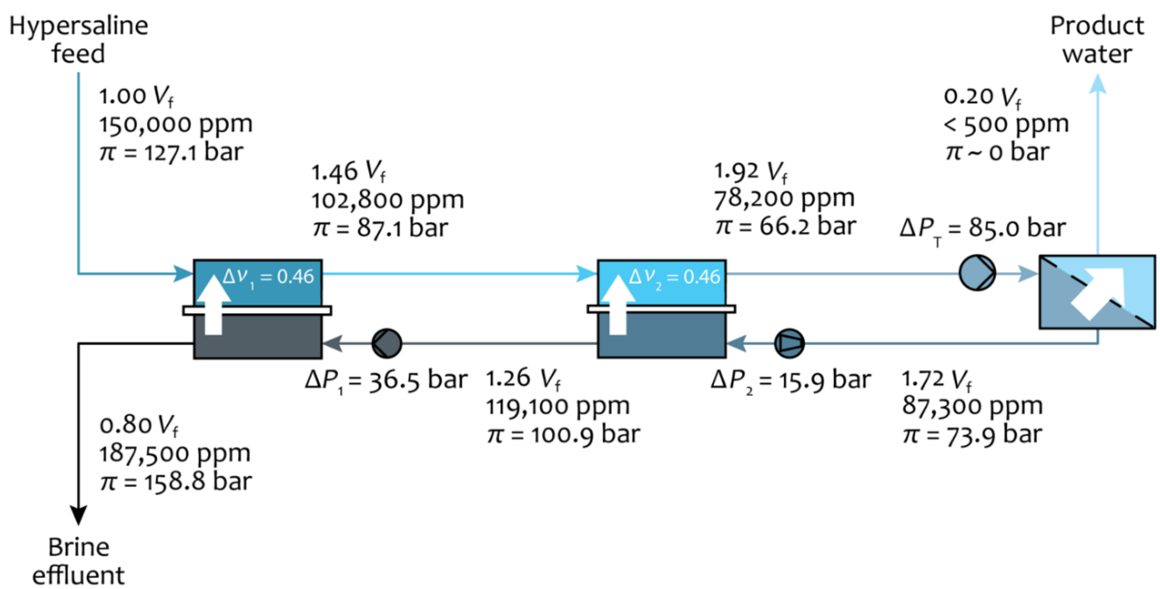

Figure S6. Desalination of hypersaline feed at 150 000 ppm to 20% recovery with 2-stage COMRO. Volumetric flow rate, TDS concentration, and osmotic pressure, π, of each stream are indicated. Operating pressures of booster pumps and high-pressure pumps are also labeled. Osmotic pressures are determined using the van't Hoff equation with NaCl solutions at temperature of 298 K, and the operational overpressurization factor, k, is 1.15.

REFERENCES

1. Bartholomew, T. V.; Mey, L.; Arena, J. T.; Siefert, N. S.; Mauter, M. S., Osmotically assisted reverse osmosis for high salinity brine treatment. *Desalination* **2017,** *421*, (Supplement C), 3-11.

2. Choong, L. T.; Govindan, P. N.; St. John, M. G.; Lam, S.; Andrews, J.-R.; Chehayeb, K. M. Osmotic desalination methods and associated systems. WO/2017/019944, 2017.

3. Lin, S. H.; Elimelech, M., Staged reverse osmosis operation: Configurations, energy efficiency, and application potential. *Desalination* **2015,** *366*, 9-14.