Supporting Information

Comparison of Energy Consumption of Osmotically Assisted Reverse Osmosis and Low-Salt-Rejection Reverse Osmosis for Brine Management

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Derivation of Equations 1 and 2

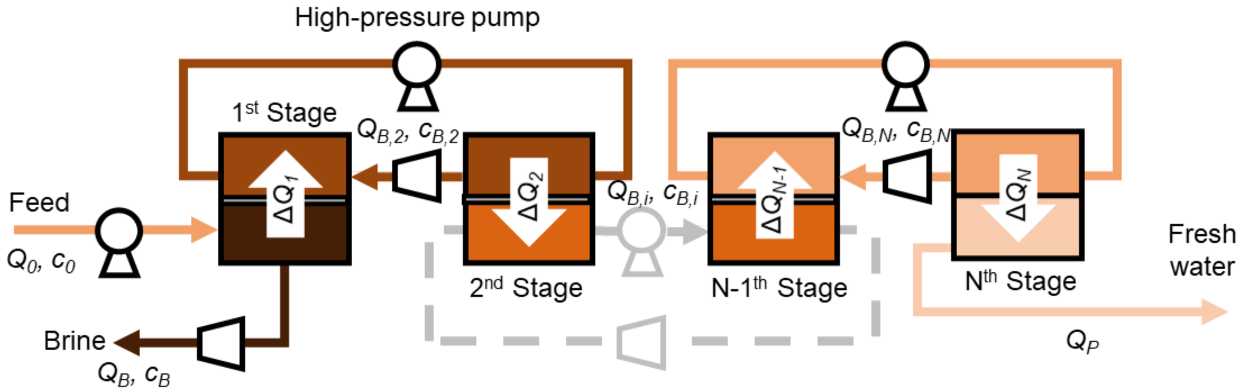

Figure S1. Schematic of an N-stage OARO.

In an N-stage OARO (Figure S1), following the working principle of RO (i.e., $\Delta\pi \leq \Delta P$), we can calculate the maximum osmotic pressure of the concentrated brine out of the last stage ($\pi_{B,N,max}$)

$$\pi_{B,N,max} = \Delta P_{max} \tag{S1}$$

where, ΔP_{max} is the maximum operating pressure in the system.

After going through an energy recovery device (ERD), the concentrated brine out of the Nth stage serves as the unpressurized stream in the N-1th stage. Then, the maximum osmotic pressure of the concentrated brine out of the N-1th stage ($\pi_{B,N-1,max}$) can be obtained:

$$\pi_{B,N-1,max} = \pi_{B,N,max} + \Delta P_{max} = 2\Delta P_{max} \tag{S2}$$

Likewise, we can obtain the osmotic pressure of the final concentrated brine out of the 1st stage ($\pi_{B,max}$)

$$\pi_{B,max} = N\Delta P_{max} \tag{S3}$$

According to van't Hoff's approximation,[1, 2] the osmotic pressure of a solution is proportional to the solution concentration. Given the feed concentration (c_0) and feed osmotic pressure (π_0), we can estimate $c_{B,max}$ from $\pi_{B,max}$:

$$c_{B,max} = \frac{\pi_{B,max} c_0}{\pi_0} \tag{S4}$$

Combing eqs S3 and S4 yields

$$c_{B,max} = \frac{N\Delta P_{max} c_0}{\pi_0} \tag{S5}$$

We note that eq S5 is eq 2a in the paper.

Based on mass balance of salts in the OARO system, the minimum flow rate of the final brine ($Q_{B,min}$) can be calculated:

$$Q_{B,min} = \frac{c_0 Q_0}{c_{B,max}} \tag{S6}$$

where Q_0 is the feed flow rate.

Then, we can calculate the maximum water recovery ($R_{w,max}$) as

$$R_{w,max} = \frac{Q_{P,max}}{Q_0} = \frac{Q_0 - Q_{B,min}}{Q_0} \tag{S7}$$

Substituting eq S6 into eq S7 yields

$$R_{w,max} = 1 - \frac{c_0}{c_{B,max}} \tag{S8}$$

Combing eqs S5 and S8 gives

$$R_{w,max} = 1 - \frac{\pi_0}{N\Delta P_{max}} \tag{S9}$$

We note that eqs S8 and S9 are eqs 1 and 2b in the paper, respectively.

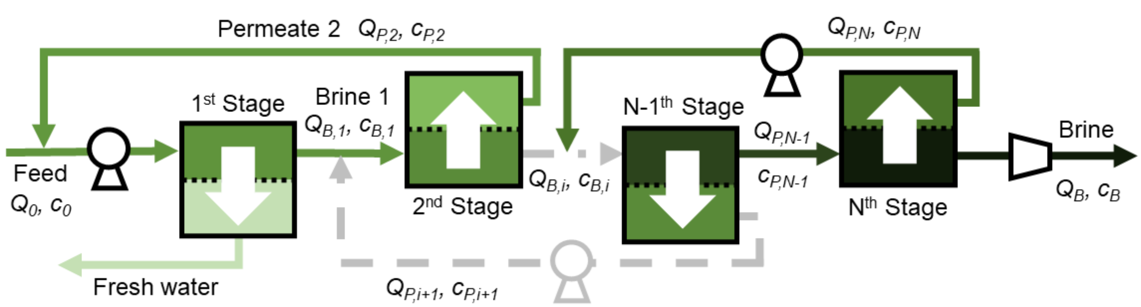

Figure S2. Schematic of an N-stage LSRRO

In an N-stage LSRRO, following the working principle of RO, the maximum osmotic pressure of the brine out of the 1st stage ($\pi_{B,1,max}$) can be obtained from the maximum operating pressure (ΔP_{max}):

$$\pi_{B,1,max} = \Delta P_{max} \tag{S10}$$

As the brine out of the 1st stage serves as the feed in the 2nd stage, the maximum osmotic pressure of the permeate stream out of the 2nd stage ($\pi_{P,2,max}$) is capped by $\pi_{B,1,max}$. Then, the maximum osmotic pressure of the brine out of the 2nd stage ($\pi_{B,2,max}$) can be obtained:

$$\pi_{B,2,max} = \pi_{P,2,max} + \Delta P_{max} = 2\Delta P_{max} \tag{S11}$$

Likewise, the maximum osmotic pressure of the final brine out of the Nth stage ($\pi_{B,max}$) can be calculated:

$$\pi_{B,max} = N\Delta P_{max} \tag{S12}$$

Note that eq S11 is the same as eq S4, and eqs S5 to S9 apply to both OARO and LSRRO. Therefore, in an N-stage LSRRO, eqs 1 and 2 in the paper still hold.

Calculation of *SEC* in N-stage OARO

Following the working principle of OARO (Figure S1), in steady-state operation, the permeate flux rate in each stage (ΔQ) is constant,[3]

$$\Delta Q_1 = \Delta Q_2 = \cdots = \Delta Q_i = \cdots = Q_P \tag{S13}$$

where Q_P is the permeate flow rate.

By definition, the specific energy consumptions of the ith stage (SEC_i) in an N-stage OARO is

$$SEC_i = \frac{Q_{P,i}\Delta P_i}{Q_P} = \Delta P_i \tag{S14}$$

where ΔP_i is the operating pressure in the ith stage. The SEC of an N-stage OARO is the sum of SEC_i:

$$SEC = \sum_{i=1}^{N} SEC_i = \sum_{i=1}^{N} \Delta P_i \tag{S15}$$

According to the working principle of RO, in the bilateral RO stages, ΔP_i is equal to the product of the transmembrane osmotic pressure difference at the outlet of the concentrated brine ($\Delta\pi_i = \pi_{B,i} - \pi_{B,i+1}$) and overpressurization factor (k):

$$\Delta P_i = k\Delta\pi_i = k(\pi_{B,i} - \pi_{B,i+1}) \tag{S16}$$

In the last conventional RO stage, ΔP_N is equal to the product of k and $\pi_{B,N}$:

$$\Delta P_N = k\pi_{B,N-1} \tag{S17}$$

Substituting eqs S16 and S17 into eq S14 yields

$$SEC = k\pi_B \tag{S18}$$

where π_B is the osmotic pressure of the final brine out of the 1st stage. Based on the van't Hoff approximation, SEC for an N-stage OARO can be calculated as

$$SEC = kvRTc_B \tag{S19}$$

where k is the overpressurization factor; v is a dimensionless van't Hoff index (e.g., v=2 for NaCl); R is the gas constant; T is the absolute temperature; and c_B is the molar concentration of the final brine out of the 1st stage.

Throughout this study, T was assumed to be 298 K because RO systems in the industry are usually rated at 25 °C (298 K). We note that the brine temperature in RO systems could vary during operation.[4] However, based on the van't Hoff approximation, even if the brine temperature varies by 10 K, the resulting variation of the calculated osmotic pressure would be only ~3 %. Such small variation would not influence the modeling results and conclusions of our study. In addition, since the impact of T on the osmotic pressure would be the same for OARO and LSRRO, the choice of T would not alter the results for the comparison between OARO and LSRRO.

Calculation of *SEC* in Two-stage LSRRO

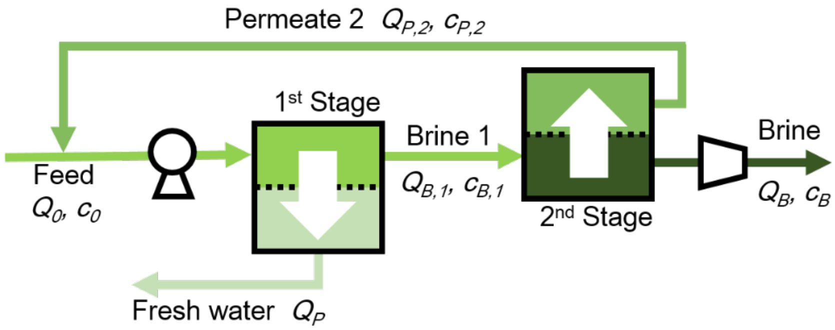

Figure S3. Schematic of a two-stage LSRRO.

In a two-stage LSRRO (Figure S3), a mass balance of salts yields

$$c_0 Q_0 = c_B Q_B \tag{S20}$$

$$c_0 Q_0 + c_{P,2} Q_{P,2} = c_{B,1} Q_{B,1} \tag{S21}$$

where c_0 and Q_0 are the molar concentration and volume flow rate of the feed, respectively; c_B and Q_B are the molar concentration and volume flow rate of the final brine, respectively; $c_{P,2}$ and $Q_{P,2}$ are the molar concentration and volume flow rate of permeate 2, respectively; and $c_{B,1}$ and $Q_{B,1}$ are the molar concentration and volume flow rate of brine 1, respectively.

Based on mass balance of water, we can obtain

$$Q_0 = Q_B + Q_P \tag{S22}$$

$$Q_{B,1} = Q_B + Q_{P,2} \tag{S23}$$

where Q_P is the flow rate of produced freshwater.

From the definition of observed salt rejection,[5, 6] we can get

$$\frac{c_{P,2}}{c_{B,1}} = 1 - r_2 \tag{S24}$$

where, r_2 is the observed salt rejection in the 2nd stage.

Following the working principle of RO (i.e., $\Delta\pi = \Delta P$) and van't Hoff's approximation, we can attain

$$c_{B,1} = \frac{\Delta P}{kvRT} \tag{S25}$$

$$c_B = c_{P,2} + \frac{\Delta P}{kvRT} \tag{S26}$$

where, k is the overpressurization factor; v is a dimensionless van't Hoff index (e.g., v=2 for NaCl); R is the gas constant; T is the absolute temperature.

Given ΔP, k, c_0, Q_0, and r_2, we can attain $Q_{P,2}$ and Q_P from solving eqs (S20) to (S26). Then, SEC can be calculated based on its definition:[7]

$$SEC = (1 + \frac{Q_{P,2}}{Q_P})\Delta P \tag{S27}$$

We note that the SEC of two-stage LSRRO reported in this study is the minimum SEC after optimization. The optimization is performed by varying ΔP and r_2 in the calculation for a target c_B (i.e., $c_B = 2.4$ M or 140,000 mg/L TDS for MLD in our study). The target c_B can be achieved by different combinations of ΔP and r_2, and each combination results in a different SEC. The minimum SEC was selected from all the resulting SEC, and the corresponding ΔP and r_2 are shown in Table S1.

Calculation of *SEC* in Three-stage LSRRO

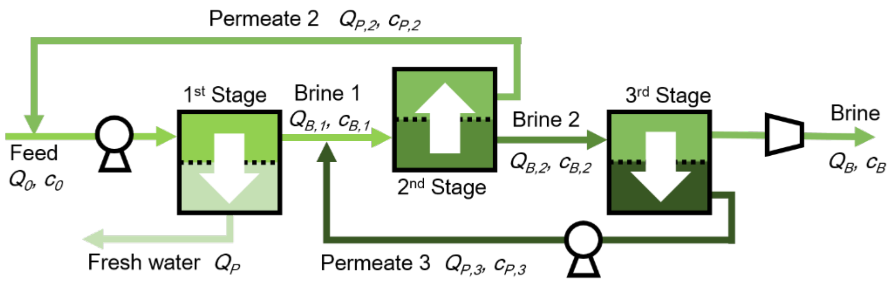

Figure S4. Schematic of a three-stage LSRRO

In a three-stage LSRRO (Figure S4), a mass balance of salts yields

$$c_0 Q_0 = c_B Q_B \tag{S28}$$

$$c_0 Q_0 + c_{P,2} Q_{P,2} = c_{B,1} Q_{B,1} \tag{S29}$$

$$c_{B,1} Q_{B,1} + c_{P,3} Q_{P,3} = c_{B,2} Q_{B,2} + c_{P,2} Q_{P,2} \tag{S30}$$

where c_0 and Q_0 are the molar concentration and volume flow rate of the feed, respectively; c_B and Q_B are the molar concentration and volume flow rate of the final brine, respectively; $c_{P,i}$ and $Q_{P,i}$ are the molar concentration and volume flow rate of permeate i, respectively; $c_{B,i}$ and $Q_{B,i}$ are the molar concentration and volume flow rate of brine i, respectively.

Based on the mass balance of water, we can get

$$Q_0 = Q_B + Q_P \tag{S31}$$

$$Q_{B,1} = Q_B + Q_{P,2} \tag{S32}$$

$$Q_{B,2} = Q_B + Q_{P,3} \tag{S33}$$

where Q_P is the flow rate of fresh water.

From the definition of observed salt rejection, we can attain

$$\frac{c_{P,2}}{(c_{B,1} Q_{B,1} + c_{P,3} Q_{P,3})/(Q_{B,1} + Q_{P,3})} = 1 - r_2 \tag{S34}$$

$$\frac{c_{P,3}}{c_{B,2}} = 1 - r_3 \tag{S35}$$

where, r_i is the observed salt rejection of the i$^{\text{th}}$ stage.

Following the working principle of RO and van't Hoff's approximation, we can obtain

$$c_{B,1} = \frac{\Delta P}{k v R T} \tag{S36}$$

$$c_{B,2} = c_{P,2} + \frac{\Delta P}{k v R T} \tag{S37}$$

$$c_B = c_{P,3} + \frac{\Delta P}{kvRT} \quad \text{(S38)}$$

where, k is the overpressurization factor; v is a dimensionless van't Hoff index (e.g., v=2 for NaCl); R is the gas constant; T is the absolute temperature.

Given ΔP, k, c_0, Q_0, r_2, and r_3, we can obtain $Q_{P,2}$, $Q_{P,3}$, and Q_P from solving eqs (S28) to (S38). Then, SEC can be calculated based on its definition:[7]

$$SEC = (1 + \frac{Q_{P,2}}{Q_P} + \frac{Q_{P,3}}{Q_P})\Delta P \quad \text{(S39)}$$

The SEC of three-stage LSRRO reported in this study is the minimum SEC after optimization. The optimization is performed by varying ΔP, r_2, and r_3 in the calculation for a target c_B (i.e., $c_B = 4$ M or 234,000 mg/L TDS for ZLD in our study). The target c_B can be achieved from different combinations of ΔP, r_2, and r_3, and each combination results in a different SEC. The minimum SEC was selected from all the resulting SEC, and the corresponding ΔP, r_2, and r_3 are shown in Tables S2 and S3.

Calculation of *SEC* in Four-stage LSRRO

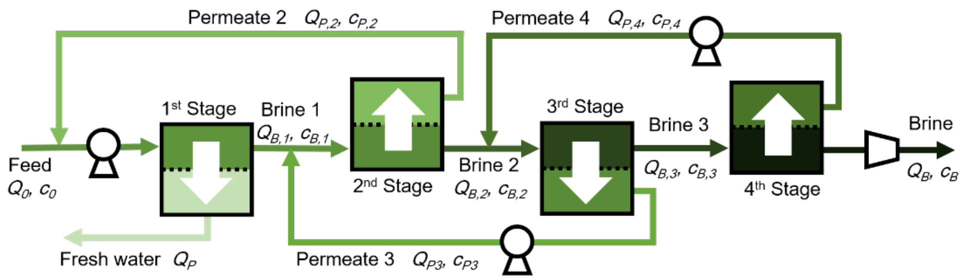

Figure S5. Schematic of a four-stage LSRRO

In a four-stage LSRRO (Figure S5), mass balance of salts yields

$$c_0Q_0 = c_BQ_B \tag{S40}$$
$$c_0Q_0 + c_{P,2}Q_{P,2} = c_{B,1}Q_{B,1} \tag{S41}$$
$$c_{B,1}Q_{B,1} + c_{P,3}Q_{P,3} = c_{B,2}Q_{B,2} + c_{P,2}Q_{P,2} \tag{S42}$$
$$c_{B,2}Q_{B,2} + c_{P,4}Q_{P,4} = c_{B,3}Q_{B,3} + c_{P,3}Q_{P,3} \tag{S43}$$

where c_0 and Q_0 are the molar concentration and volume flow rate of the feed, respectively, c_B and Q_B are the molar concentration and volume flow rate of the final brine, respectively, $c_{P,i}$ and $Q_{P,i}$ are the molar concentration and volume flow rate of the final brine, respectively, $c_{B,1}$ and $Q_{B,1}$ are the molar concentration and volume flow rate of brine i, respectively.

Based on mass balance of water, we can get

$$Q_0 = Q_B + Q_P \tag{S44}$$
$$Q_{B,1} = Q_B + Q_{P,2} \tag{S45}$$
$$Q_{B,2} = Q_B + Q_{P,3} \tag{S46}$$
$$Q_{B,3} = Q_B + Q_{P,4} \tag{S47}$$

where Q_P is the flow rate of fresh water.

According to the definition of observed salt rejection, we can attain

$$\frac{c_{P,2}}{(c_{B,1}Q_{B,1} + c_{P,3}Q_{P,3})/(Q_{B,1} + Q_{P,3})} = 1 - r_2 \tag{S48}$$
$$\frac{c_{P,3}}{(c_{B,2}Q_{B,2} + c_{P,4}Q_{P,4})/(Q_{B,2} + Q_{P,4})} = 1 - r_3 \tag{S49}$$
$$\frac{c_{P,4}}{c_{B,3}} = 1 - r_4 \tag{S50}$$

where, r_i is the observed salt rejection of the i$^{\text{th}}$ stage.

Following the working principle of RO and van't Hoff's law, we can obtain

$$c_{B,1} = \frac{\Delta P}{kvRT} \tag{S51}$$

$$c_{B,2} = c_{P,2} + \frac{\Delta P}{kvRT} \tag{S52}$$

$$c_{B,3} = c_{P,3} + \frac{\Delta P}{kvRT} \tag{S53}$$

$$c_B = c_{P,4} + \frac{\Delta P}{kvRT} \tag{S54}$$

where k is overpressurization factor$v$ is a dimensionless van't Hoff index (e.g., v=2 for NaCl); R is the gas constant; T is the absolute temperature.

Given ΔP, k, c_0, Q_0, r_2, r_3, and r_4, we can obtain $Q_{P,2}$, $Q_{P,3}$, $Q_{P,4}$, and Q_P from solving eqs (S40) to (S54). Then, SEC can be calculated based on its definition:[7]

$$SEC = (1 + \frac{Q_{P,2}}{Q_P} + \frac{Q_{P,3}}{Q_P} + \frac{Q_{P,4}}{Q_P})\Delta P \tag{S55}$$

The SEC of four-stage LSRRO reported in this study is the minimum SEC after optimization. The optimization is performed by varying ΔP, r_2, r_3, and r_4 in the calculation for a target c_B (i.e., $c_B = 4$ M or 234,000 mg/L TDS for ZLD in our study). The target c_B can be achieved from different combinations of ΔP, r_2, r_3, and r_4, and each combination results in a different SEC. The minimum SEC was selected from all the resulting SEC, and the corresponding ΔP, r_2, r_3, and r_4 are shown in Table S2.

Operating Conditions and *SEC* of OARO

Table S1. Operating pressures and *SEC* of two-stage OARO for MLD with a ΔP_{max} of 80 bar

Stage Number	2	2	2
c_0 (mg/L TDS)	10,000	35,000	70,000
ΔP_1 (bar)	65.4	65.4	65.4
ΔP_2 (bar)	65.4	65.4	65.4
SEC (kWh m^{-3})	3.63	3.63	3.63

Table S2. Operating pressures and *SEC* of three-stage and four-stage OARO for ZLD with a ΔP_{max} of 80 bar

Stage Number	3	4	3	4	3	4
c_0 (mg/L TDS)	10,000	10,000	35,000	35,000	70,000	70,000
ΔP_1 (bar)	72.7	54.5	72.7	54.5	72.7	54.5
ΔP_2 (bar)	72.7	54.5	72.7	54.5	72.7	54.5
ΔP_3 (bar)	72.7	54.5	72.7	54.5	72.7	54.5
ΔP_4 (bar)	-	54.5	-	54.5	-	54.5
SEC (kWh m^{-3})	6.06	6.06	6.06	6.06	6.06	6.06

Table S3. Operating pressures and *SEC* of three-stage OARO for ZLD with a ΔP_{max} of 120 bar

Stage Number	3	3	3
c_0 (mg/L TDS)	10,000	35,000	70,000
ΔP_1 (bar)	72.7	72.7	72.7
ΔP_2 (bar)	72.7	72.7	72.7
ΔP_3 (bar)	72.7	72.7	72.7
SEC (kWh m^{-3})	6.06	6.06	6.06

Optimization Results of *SEC* in LSRRO

Table S4. Detailed information about the salt rejections and applied pressures to achieve SEC_{min} in two-stage LSRRO for MLD with a ΔP_{max} of 80 bar

Stage Number	2	2	2
c_0 (mg/L TDS)	10,000	35,000	70,000
r_2 (%)	34	37	37
r_3 (%)	-	-	-
r_4 (%)	-	-	-
ΔP (bar)	78.5	80	80
SEC (kWh m^{-3})	2.51	3.49	6.02

Table S5. Detailed information about the salt rejections and applied pressures to achieve SEC_{min} in three-stage and four-stage LSRRO for ZLD with a ΔP_{max} of 80 bar

Stage Number	3	4	3	4	3	4
c_0 (mg/L TDS)	10,000	10,000	35,000	35,000	70,000	70,000
r_2 (%)	32	48	32	46	32	46
r_3 (%)	7	30	7	30	7	30
r_4 (%)	-	18	-	19	-	19
ΔP (bar)	80	79.4	80	70	70	80
SEC (kWh m^{-3})	4.04	3.02	9.38	5.38	19.61	9.90

Table S6. Detailed information about the salt rejections and applied pressures to achieve SEC_{min} in three-stage LSRRO for ZLD with a ΔP_{max} of 120 bar

Stage Number	3	3	3
c_0 (mg/L TDS)	10,000	35,000	70,000
r_2 (%)	43	55	53
r_3 (%)	20	43	44
ΔP (bar)	94.6	119.7	119.9
SEC (kWh m^{-3})	3.28	4.54	6.26

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