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

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ABSTRACT: Minimum and zero liquid discharge (MLD/ZLD) are emerging brine management strategies that attract heightened attention. Although conventional reverse osmosis (RO) can improve the energy efficiency of MLD/ZLD processes, its application is limited by the maximum hydraulic pressure ($\Delta P_{\text{max}}$) that can be applied in current membrane modules. To overcome such limitation, novel RO-based technologies, including osmotically assisted RO (OARO) and low-salt-rejection RO (LSRRO), have been proposed. Herein, we utilize process modeling to systematically compare the energy consumption of OARO and LSRRO for MLD/ZLD applications. Our modeling results show that the specific energy consumption (SEC) of LSRRO is lower (by up to $\sim$30%) than that of OARO for concentrating moderately saline feed waters ($\sim$35,000 mg/L TDS) to meet MLD/ZLD goals, whereas the SEC of OARO is lower (by up to $\sim$40%) than that of LSSRO for concentrating higher salinity feed waters ($\sim$70,000 mg/L TDS). However, by implementing more stages and/or an elevated $\Delta P_{\text{max}}$, LSRRO has the potential to outperform OARO energetically for treating high-salinity feed waters. Notably, the SEC of both OARO and LSRRO could be 50% lower than that of mechanical vapor compressor, the commonly used brine concentrator in MLD/ZLD applications. We conclude with a discussion on the practicability of OARO and LSRRO based on membrane module availability and capital cost, suggesting that LSRRO could potentially be more feasible than OARO.

KEYWORDS: brine management, reverse osmosis, brine concentration, energy efficiency, practicability, minimal liquid discharge, zero liquid discharge

INTRODUCTION

Freshwater scarcity has become a critical global challenge.1–3 A substantial amount of freshwater is turned to wastewater due to the ever-growing public and industrial activities.4 Discharging wastewaters without adequate treatment could cause severe pollution of freshwater resources, exacerbating freshwater scarcity.5,6 To address this global problem, a strict wastewater management strategy, zero liquid discharge (ZLD), has been proposed.7 By recovering all the water in wastewater streams and leaving only solid waste, ZLD can obviate the risk of wastewater discharge and maximize freshwater usage efficiency. Because ZLD is very costly to achieve,9 minimum liquid discharge (MLD) that targets a water recovery of up to 95% has been proposed.10,11 Unlike ZLD, MLD allows the balance of economic factors with environmental impacts.

Early ZLD systems were built based on thermal processes, where the saline wastewater was evaporated first in a brine concentrator and then in a brine crystallizer or an evaporation pond.7,12 The application of such systems was economically prohibitive because of the high capital and operational costs of thermal processes. To improve the efficiency of brine management, reverse osmosis (RO), a membrane-based technology widely used in desalination, has been incorporated in ZLD systems before the thermal brine concentrator.5,13 The use of RO reduces the volume of brine treated using a thermal brine concentrator, thereby improving the energy and cost efficiency of ZLD systems. Due to its high energy utilization efficiency,14 RO is also commonly used in MLD systems.10,15

However, to date, ZLD systems are still costly because conventional RO cannot completely replace the thermal brine concentrator due to the restriction on the operating pressure.16,17 Specifically, with the current operating pressure of conventional RO (<85 bar),16 its maximum brine concentration cannot exceed 100,000 mg/L, whereas the required brine concentration for a brine crystallizer is over 200,000 mg/L.7 Furthermore, the amount of freshwater that can be recovered using current MLD systems is also limited because of the limit on the operating pressures in RO.17

To develop MLD/ZLD systems with improved performance, novel staged RO technologies that can overcome the
applied pressure limit of conventional RO are needed. Specifically, osmotically assisted RO (OARO) and low-salt-rejection RO (LSRRO) have been proposed. Both OARO and LSRRO are able to highly concentrate wastewaters using moderate operating pressures, thereby advancing MLD/ZLD systems. However, the “sweet spots” for these two technologies (i.e., scenarios in which a specific technology is preferred) for MLD/ZLD applications have not been established. Comparison of the performance of these technologies is therefore critical for guiding the development of future MLD/ZLD systems and for the ultimate goal of displacing thermal evaporators in MLD/ZLD systems.

In this study, we use process modeling to systematically compare the two novel RO-based technologies, OARO and LSRRO, for brine management and MLD/ZLD applications. We start by introducing the working principles of OARO and LSRRO, demonstrating that both technologies can achieve higher water recovery rates (i.e., discharge brines with higher concentrations and smaller volumes) than conventional RO with moderate operating pressures. Using specific energy consumption (SEC) as a performance metric, we systematically compare OARO and LSRRO in MLD/ZLD applications. By analyzing the impact of operating parameters, including feed salt concentration, number of stages employed, and maximum operating pressure on SEC, we identify the sweet spots of feed water salinities for each technology. We conclude with a discussion of practical considerations of the two technologies, highlighting potential advantages of LSRRO over OARO for MLD/ZLD applications.

**Novel RO-based Processes for MLD and ZLD**

**Limitations of Conventional RO.** In order to recover freshwater from the saline feed, the applied hydraulic pressure ($\Delta P$) in RO needs to be larger than the transmembrane osmotic pressure difference ($\Delta \pi$). In conventional RO with near perfect salt rejection, $\Delta \pi$ is equal to the osmotic pressure of the brine ($\pi_B$) because the osmotic pressure of the product water is negligible (Figure 1A). According to van’t Hoff’s approximation, $\pi_B$ increases with the brine concentration ($c_B$). During RO operation, the feed becomes more concentrated as more freshwater is being recovered, leading to an increased $\pi_B$. Once $\pi_B$ reaches $\Delta P$, the water recovery ceases and the feed cannot be further concentrated. In other words, the maximum brine concentration ($c_{B,\text{max}}$) of conventional RO is achieved as $\pi_B = \Delta P$ (Figure 1B). Following simple mass balance equations, the maximum water recovery ($R_{w,\text{max}}$) can be calculated, knowing $c_{B,\text{max}}$ and the initial feed salt concentration ($c_0$) (detailed derivations in the Supporting Information)

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

For a given $\Delta P$, $c_{B,\text{max}}$ is fixed. According to eq 1, with a fixed $c_{B,\text{max}}$, $R_{w,\text{max}}$ decreases with increasing $c_0$. To avoid detrimental effects of high hydraulic pressure on membranes and modules, $\Delta P$ is typically <85 bar. In most RO operations, $\Delta P$ does not exceed 80 bar, resulting in a $c_{B,\text{max}}$ of ~94,000 mg/L TDS (Figure 1B). Such $c_{B,\text{max}}$ leads to $R_{w,\text{max}}$ of ~93, 63, and 26% for feed solutions with $c_0$ of 10,000, 35,000, and 70,000 mg/L TDS, respectively. As discussed previously, for MLD/ZLD applications, a higher $R_{w,\text{max}}$ is desired. However, to further enhance $R_{w,\text{max}}$ in conventional RO, the allowable $\Delta P$ must be increased, which is currently not feasible.

**Osmotically Assisted RO.** To boost $R_{w,\text{max}}$ without elevating $\Delta P$, a novel staged RO configuration named OARO has been developed (Figure 2A). An N-stage OARO system comprises one conventional RO module and $N - 1$ bilateral RO modules, with the conventional RO module placed at the last stage. In each bilateral stage, two saline streams flow countercurrently with one being pressurized and the other remaining unpressurized. The pressure-driven water permeation results in concentration of the pressurized stream and dilution of the unpressurized stream. To maintain steady concentrations of the saline streams, the saline streams loop between adjacent stages, with the water permeation volume at each stage being fixed. Specifically, after being concentrated in one stage, the pressurized stream is depressurized and cycled back to the previous stage for dilution; after being diluted in that stage, the unpressurized stream is pressurized and sent to the next stage for concentration. Freshwater is produced from the last conventional RO stage, and the final brine is discharged from the first bilateral stage after recovering the energy embedded in the brine stream.

With the use of bilateral stages in OARO, the osmotic pressure of the unpressurized saline stream ($\pi_B$) is able to reduce $\Delta \pi$ from $\pi_B$ to $\pi_B - \pi_s$. Therefore, following the working principle of RO (i.e., $\Delta P \geq \Delta \pi$), the maximum $\pi_B$ in OARO ($\pi_{B,\text{max}}$) can exceed $\Delta \pi$ (i.e., $\pi_{B,\text{max}} = \Delta P + \pi_s$). According to van’t Hoff’s relationship, a larger $\pi_{B,\text{max}}$ corresponds to a higher $c_{B,\text{max}}$ and $R_{w,\text{max}}$ increases with increasing $c_{B,\text{max}}$ (eq 1). Thus, OARO can achieve a higher $R_{w,\text{max}}$ than conventional RO with the same $\Delta P$.

**Low-Salt-Rejection RO.** Another staged RO technology—LSRRO—that can achieve a high $R_{w,\text{max}}$ with moderate $\Delta P$ has recently been proposed (Figure 2B). In an LSRRO system, a conventional RO module is used in the first stage, while LSRRO modules are used in the subsequent stages. During operation, in each low-salt-rejection stage, the pressurized concentrated brine is directly sent to the next stage as feed, while the diluted permeate is repressurized and cycled back to the previous stage as additional feed (i.e., $\Delta P$ at each stage is constant). Freshwater is produced from the first conventional RO stage, while the final brine is discharged from the last low-salt-rejection stage after energy recovery.

Like OARO, LSRRO employs saline solutions to mediate $\Delta \pi$, albeit through a different pathway. Specifically, in LSSRO, the use of LSSRO membranes results in saline permeates in the low-salt-rejection stages. The osmotic pressure of the saline permeate ($\pi_s$) can reduce $\Delta \pi$ from $\pi_B$ to $\pi_B - \pi_s$, thereby resulting in a $\pi_{B,\text{max}}$ larger than $\Delta \pi$ (i.e., $\pi_{B,\text{max}} = \Delta P + \pi_s$). In other words, LSRRO can achieve a larger $\pi_{B,\text{max}}$ than conventional RO with the same $\Delta P$. Because a larger $\pi_{B,\text{max}}$ leads to a higher $R_{w,\text{max}}$ for given $\Delta P$, LSRRO can attain a higher $R_{w,\text{max}}$ compared to conventional RO.

**Performance of OARO and LSRRO in MLD Applications**

**Water Recovery in Two-Stage OARO and LSRRO for MLD.** Based on the working principles of OARO and LSRRO, the theoretical maximum brine concentration ($c_{B,\text{max}}$) and maximum water recovery rate ($R_{w,\text{max}}$) in an N-stage OARO/LSRRO system can be calculated as a function of the stage number ($N$) and maximum applied pressure ($\Delta P_{\text{max}}$) (detailed derivations in the Supporting Information).
\[ c_{B,\text{max}} = \frac{N \Delta P_{\text{max}} c_0}{\pi_0} \]  

where \( \pi_0 \) is the osmotic pressure of the feed solution.

In practical applications, the more stages an OARO/LSRRO system uses, the more complicated the system construction is. Therefore, for the MLD applications discussed in this section, OARO and LSRRO systems with two stages were investigated. Comparison of \( R_{w,\text{max}} \) between conventional RO and two-stage OARO/LSRRO is shown in Figure 3A. For all three feed solutions shown, two-stage OARO/LSRRO can achieve higher \( R_{w,\text{max}} \) than conventional RO. The difference in \( R_{w,\text{max}} \) between two-stage OARO/LSRRO and conventional RO varies with the feed salt concentration \( (c_0) \). With a \( c_0 \) of 10,000 mg/L, conventional RO and two-stage OARO/LSRRO can achieve \( R_{w,\text{max}} \) of 89 and 95%, respectively, resulting in an \( R_{w,\text{max}} \) difference of only 6%. In contrast, with a \( c_0 \) of 70,000 mg/L, conventional RO and two-stage OARO/LSRRO can achieve \( R_{w,\text{max}} \) of 26 and 63%, respectively, leading to an \( R_{w,\text{max}} \) difference of 37%.

We note that, despite the dependence of \( R_{w,\text{max}} \) on \( c_0 \) \((1 - R_{w,\text{max}}) \) in OARO/LSRRO is always half of that in conventional RO, suggesting that two-stage OARO/LSRRO can reduce the minimum brine flow rate \( (Q_{B,\text{min}}) \) from conventional RO by half. Reducing \( Q_{B,\text{min}} \) by a factor of two in two-stage OARO/LSRRO is directly related to \( c_{B,\text{max}} \) being two times of that in conventional RO. Based on mass balance of salts in RO operation \((i.e., Q_{B} c_0 = Q_{P} c_{B,\text{max}})\), a 100% increase in \( c_{B,\text{max}} \) results in a 50% reduction of \( Q_{B,\text{min}} \). Therefore, two-stage OARO/LSRRO can signifi-
cantly facilitate MLD applications because it can further minimize the volume of brines compared to conventional RO.

Energy Consumption in Two-Stage OARO and LSRRO for MLD. Because both OARO and LSRRO can achieve high $R_{w,max}$ with moderate $\Delta P$, the SEC for water recovery becomes a critical performance metric for the comparison between the two technologies in MLD applications. By definition, SEC in an $N$-stage OARO/LSRRO system is the total energy consumption normalized by the volume of the recovered or produced freshwater. Neglecting the inefficiencies of the ERDs, SEC can be calculated as

$$SEC = \frac{\sum_{i=1}^{N} Q_{P,i} \Delta P_i}{Q_P}$$

(3)

where $Q_P$ is the flow rate of the produced fresh water, $Q_{P,i}$ is the permeate flow rate in the $i$th stage, and $\Delta P_i$ is the operating pressure in the $i$th stage.

To maintain adequate water flux along the membrane module in practical RO operation, $\Delta P$ is in excess of $\Delta \pi$ everywhere inside the module (i.e., $\Delta \pi < \Delta P$). The ratio of $\Delta P$ to $\Delta \pi$ at the brine outlet is the over-pressurization factor denoted by $k$ (i.e., $\Delta P = k \Delta \pi$). Because $c_{B,max}$ and $R_{w,max}$ in eqs 2a and 2b and Figure 3A are obtained at the limit of $\Delta \pi = \Delta P$, they are theoretical performance limits that can hardly be achieved in practical applications because in practical applications, an over-pressure is applied to compensate for concentration polarization and the pressure drop along the module. Therefore, for the comparison of SEC in two-stage OARO and LSRRO for practical MLD applications, with a $\Delta P_{max}$ of 80 bar, a practical $c_B$ of 140,000 mg/L was set as the target brine concentration and $k$ was assumed to be 1.1. For a comprehensive comparison, three feed solutions with varying $c_0$ (i.e., 10,000, 35,000, and 70,000 mg/L) were investigated. With a target $c_B$ of 140,000 mg/L, the water recoveries ($R_w$) for the feed solutions with $c_0$ of 10,000, 35,000, and 70,000 mg/L were 93, 75, and 50%, respectively. The

Figure 3. Comparison between OARO and LSRRO in MLD applications. (A) Maximum water recovery ($R_{w,max}$) from conventional RO (gray columns) and two-stage OARO/LSRRO (blue columns) with varying feed salt concentrations ($c_0$). (B) SEC of two-stage OARO (orange columns) and LSRRO (green columns) for treating feed waters of varying salinities in MLD applications. The targeted brine concentration ($c_B$) was set at 140,000 mg/L, corresponding to water recoveries ($R_w$) of 93, 75, and 50% for $c_0$ of 10,000, 35,000, and 70,000 mg/L, respectively. SEC in each stage (SEC$_i$) of two-stage OARO for concentrating feed solutions of (C) 10,000 and (D) 70,000–140,000 mg/L. SEC of two-stage LSRRO for concentrating feed solutions of (E) 10,000 and (F) 70,000–140,000 mg/L. In panels (C–F), the $x$-axis refers to the permeate flow rate in each stage ($Q_{P,i}$) normalized by the flow rate of the produced fresh water ($Q_P$) and the dashed blue lines denote the osmotic pressure of the brine. In all calculations, the maximum hydraulic pressure ($\Delta P$) was assumed to be 80 bar, the over-pressurization factor ($k$) was set to 1.1, and the temperature was kept at 298 K. Detailed operating conditions ($\Delta P$) and SEC in each stage of OARO are presented in Table S1 of the Supporting Information. The SEC of LSRRO is the minimum SEC after optimizing the observed salt rejection rates of the stages and $\Delta P$; detailed information is shown in Table S4 of the Supporting Information.
calculated SEC for the different feed solutions are presented in Figure 3B (detailed calculation in the Supporting Information).

As shown in Figure 3B, the SEC of two-stage LSRRO increases with increasing $c_{0}$, whereas the SEC of two-stage OARO remains constant. To elucidate the difference in SEC of two-stage OARO and LSRRO with different $c_{0}$, the total SEC is broken into the SEC in each stage. By definition, the SEC in the $i$th stage (SEC$_{i}$) can be calculated as

$$\text{SEC}_i = \frac{Q_{f,i} \Delta P_i}{Q_{f}}$$  \hspace{1cm} (4)

Following the working principle of OARO (Figure 2A), the permeate flow rate from each stage is constant (i.e., $Q_{f,i} = Q_{f}$). Thus, SEC$_i$ is equal to $\Delta P_i$ (eq 4), and the total SEC is equal to the sum of $\Delta P_i$ (Figure 3C,D). As $\Delta P_i = k \Delta \pi$, the total SEC is equivalent to the sum of $k \Delta \pi$. In the bilateral stages, $\Delta \pi$ is the osmotic pressure difference between the concentrated streams at the outlets of the $i$th stage and the $i+1$ stage (i.e., $\Delta \pi_i = \pi_{B,i} - \pi_{B,i+1}$) in the last conventional RO stage, $\Delta \pi_{B,N} = \pi_{B,N}$. Thus, the total SEC can be calculated as

$$\text{SEC} = \sum_{i=1}^{N} k \Delta \pi_i = \sum_{i=1}^{N-1} k(\pi_{B,i} - \pi_{B,i+1}) + k \pi_{B,N} = k \pi_{B,1}$$  \hspace{1cm} (5)

For OARO, because the final concentrated brine is discharged from the first stage, $\pi_{B,1}$ is indeed the osmotic pressure of the concentrated brine from OARO, $\pi_{B}$. Therefore, the SEC of OARO is equivalent to $k \pi_{B}$ (Figure 3C,D). In other words, for a given $c_{B}$ in OARO, the SEC is determined. Therefore, the SEC of two-stage OARO is independent of $c_{0}$.

Following the working principle of LSRRO, $\Delta P_i$ in each stage is constant (i.e., $\Delta P_i = \Delta P$). For two-stage LSRRO with different $c_{0}$, SEC$_{1}$ is approximately the same (Figure 3E,F). The similar SEC$_{1}$ is because SEC$_{1}$ is equal to $\Delta P$, and to achieve the minimum SEC in two-stage LSRRO, $\Delta P$ is always close to $\Delta P_{\text{max}}$ (Table S4 in the Supporting Information). The difference in the overall SEC in two-stage LSRRO is attributed to SEC$_{2}$. For a given $\Delta P$, the flow rate of the brine from the 1st conventional RO stage ($Q_{B,1}$) increases with $c_{0}$. With a larger $Q_{B,1}$ sent to the 2nd stage, the permeate flow rate from the 2nd stage ($Q_{B,2}$) becomes larger. A larger $Q_{B,2}$ requires a larger SEC$_{2}$ for repressurization and recirculation, thereby leading to a larger SEC, as seen when comparing SEC$_{2}$ in Figure 3E,F.

As shown in Figure 3B, the SEC of two-stage LSRRO is ~70% of that of two-stage OARO for a $c_{0}$ of 10,000 mg/L, while LSRRO and OARO have comparable SEC for a $c_{0}$ of 35,000 mg/L. As $c_{0}$ is increased to 70,000 mg/L, the SEC of two-stage LSRRO increases and is ~65% greater than that of two-stage OARO. This comparison suggests that in MLD applications, two-stage LSRRO is more energy efficient than two-stage OARO for treating low-salinity feeds, while two-stage OARO is more energy efficient than two-stage LSRRO for treating hypersaline feeds.

### PERFORMANCE OF OARO AND LSRRO IN ZLD APPLICATIONS

**Energy Consumption in Three-Stage OARO and LSRRO for ZLD.** The goal of OARO/LSRRO in ZLD applications is to produce a highly concentrated brine that can be directly sent to the thermal brine crystallizer, thus eliminating the use of thermal brine concentrator. Typically, the required brine concentration ($c_{B}$) at the inlet of a thermal brine crystallizer is over 200,000 mg/L. Based on eq 2a, to achieve such concentrated brine in OARO or LSRRO, at least three stages are needed. Throughout this study, a target $c_{B}$ of 234,000 mg/L was set for ZLD applications. With a $\Delta P_{\text{max}}$ of 80 bar and over-pressurization factor $k$ of 1.1, the SEC of three-stage OARO and LSRRO in ZLD applications with different $c_{0}$ has been calculated, as shown in Figure 4A (detailed calculation in the Supporting Information).

Like MLD applications (Figure 3B), for ZLD applications with increasing $c_{0}$, the SEC of OARO remains constant, whereas the SEC of LSRRO increases (Figure 4A). As discussed previously, the SEC of OARO is only determined from $c_{0}$, which is independent of $c_{B}$. In contrast, the SEC of LSRRO increases with $c_{0}$ because a higher $c_{0}$ results in larger permeate flow rates in the low-salt-rejection stages, requiring more energy for repressurization and recirculation. Based on the SEC comparison (Figure 4A), with three stages, LSRRO is more competitive than OARO only when treating feed water with a relatively low $c_{0}$ (i.e., 10,000 mg/L), while for treating feed waters with higher $c_{0}$ (i.e., 35,000 and 70,000 mg/L), OARO is substantially more energy efficient than LSRRO (Figure 4A). We note that mechanical vapor compression (MVC), the most widely used thermal brine concentrator in ZLD applications, has an SEC of 20–25 kW h m$^{-3}$. Because the SEC in three-stage OARO is less than 30% of that in MVC, three-stage OARO is energetically superior to MVC. In contrast, three-stage LSRRO is more energy efficient than MVC for treating moderately saline feeds, while treating hypersaline feeds, the two technologies are energetically comparable.

**Energy Consumption in Four-Stage OARO and LSRRO for ZLD.** The SEC of four-stage OARO and LSRRO in ZLD applications with different $c_{0}$ has also been calculated (Figure 4B), assuming the same operating conditions as three-stage OARO and LSRRO (i.e., $\Delta P_{\text{max}}$ of 80 bar and $k$ of 1.1). Detailed calculations are presented in the Supporting Information. For OARO, the SEC remains constant with the stage number. To elucidate the independence of SEC on the stage number, the SEC of each stage in three-stage and four-stage OARO (SEC$_{i}$) for a $c_{0}$ of 35,000 mg/L was calculated and compared. As shown in Figure 4C,D, for a given $c_{B}$ in OARO, although the required hydraulic pressure in each stage ($\Delta P_{i}$) can be reduced with more stages being used, the sum of $\Delta P_{i}$ remains the same based on our previous discussion (i.e., $\sum_{i=1}^{N} k \pi_{B} = \sum_{i=1}^{N} k \pi_{B}$). Because the SEC of OARO is equal to the sum of $\Delta P_{i}$ (eq 3), it is independent of the stage number (i.e., SEC $= k \pi_{B}$).

Likewise, to understand the different SEC of LSRRO with different stage numbers, the SEC, in three-stage and four-stage LSRRO for a $c_{0}$ of 35,000 mg/L was calculated and shown in Figure 4E,F. The SEC of the first conventional RO stage (SEC$_{1}$) is similar in three-stage and four-stage LSRRO. As previously discussed, SEC$_{1}$ in LSRRO is always equal to $\Delta P$. In both three-stage and four-stage LSRRO, to achieve the minimum SEC, $\Delta P$ should be close to $\Delta P_{\text{max}}$ (Tables S2 in the Supporting Information), thereby resulting in similar SEC$_{1}$. The difference in SEC of three-stage and four-stage LSRRO is attributed to the different SEC in the low-salt-rejection stages. For a given $c_{B}$, the total feed volume reduction in LSRRO is fixed. When four stages are used, the permeate flow rates ($Q_{B,i}$) in the low-salt-rejection stages substantially decrease (Figure 4E,F, Table S5 in the Supporting Information). As smaller $Q_{B,i}$...
demands smaller energy consumption for repressurization and recirculation, the SEC of four-stage LSRRO becomes notably smaller than that of three-stage LSRRO.

With four stages, the SEC advantage of LSRRO toward OARO in treating feed waters with relatively low $c_0$ (i.e., 10,000 mg/L) is amplified, and for treating feed waters with moderate $c_0$ (i.e., 35,000 mg/L), LSRRO becomes more energy efficient than OARO. Furthermore, when treating feed waters with high $c_0$ (i.e., 70,000 mg/L), the SEC advantage of four-stage OARO over four-stage LSRRO decreases substantially compared to the case with three-stage OARO and LSRRO. Following such a trend, LSRRO could be energetically more favorable than OARO with more stages, even for treating hypersaline feed waters. Notably, four-stage LSRRO is much more energy efficient than MVC, with SEC of LSRRO being 2.5−10 kW h m$^{-3}$ compared to 20−25 kW h m$^{-3}$ of MVC.

Performance of OARO and LSRRO with Elevated Operating Pressure for ZLD. Although current $\Delta P_{\text{max}}$ of conventional RO modules is limited to $\sim$85 bar, it could be increased in the future if a demand for such modules arises. Several RO membrane manufacturers (e.g., Dow) have recently produced high-pressure RO elements that can withstand an operating pressure of up to 120 bar. With this technical development, it is necessary to compare the future performances of OARO and LSRRO at elevated $\Delta P_{\text{max}}$.

Assuming a $\Delta P_{\text{max}}$ of 120 bar (i.e., the maximum operating pressure of current high-pressure RO elements), the SEC of three-stage OARO and LSRRO in ZLD application with different feed solutions was calculated and shown in Figure 5A (detailed calculations in the Supporting Information). Comparing Figures 4A and 5A, the SEC of three-stage OARO with $\Delta P_{\text{max}}$ of 80 and 120 bar is the same, because $\Delta P_{\text{max}}$ is not a limiting factor in three-stage OARO for ZLD applications. To achieve a $c_B$ of 234,000 mg/L (feed for crystallizer in ZLD), three-stage OARO only requires a $\Delta P$ of $\sim$73 bar. Once $\Delta P_{\text{max}}$ exceeds 73 bar, three-stage OARO can be used in ZLD applications. A larger $\Delta P_{\text{max}}$ does not benefit SEC because SEC is only determined from $c_B$. However, for three-stage LSRRO, the SEC with a $\Delta P_{\text{max}}$ of 120 bar (Figure 5A) is substantially smaller than that with a $\Delta P_{\text{max}}$ of 80 bar (Figure 4A). Such difference can be explained by SEC (Figure 5B). With a larger $\Delta P_{\text{max}}$ despite an increased SEC, the brine volume from the first conventional stage RO decreases. A decreased brine
volume substantially reduces the permeate flow rates in the low-salt-rejection stages ($Q_p$) for repressurization and recirculation, thereby leading to a notably smaller SEC.

At an elevated $\Delta P_{\max}$ of 120 bar, three-stage LSRRO is more energy efficient than three-stage OARO for treating feed waters with relatively low and moderate $c_0$ (i.e., 10,000 and 35,000 mg/L). For treating hypersaline feed waters (i.e., $c_0 = 70,000$ mg/L), three-stage LSRRO is comparable to three-stage OARO with a slightly larger SEC. Based on the obtained SEC with a $\Delta P_{\max}$ of 120 bar, we conclude that LSRRO will be energetically more advantageous than OARO with the availability of high pressure RO modules. Notably, with a $\Delta P_{\max}$ of 120 bar, the SEC in three-stage LSRRO would be substantially lower than that in MVC (i.e., 3–6 kW h m$^{-3}$ in LSRRO versus 20–25 kW h m$^{-3}$ in MVC).

## IMPLICATIONS AND OUTLOOK

To overcome the operating pressure limitation of conventional RO in MLD/ZLD applications, novel staged RO technologies including OARO and LSRRO have been proposed. In this study, we performed the first direct comparison of the energy consumption in OARO and LSRRO in MLD/ZLD applications using process modeling. For a thorough comparison of these two technologies, the practicability and potential feasibility of the two technologies should be considered in addition to energy efficiency. However, because both OARO and LSRRO are emerging technologies, data regarding membrane module availability and capital costs are limited. Thus, we qualitatively compare OARO and LSRRO using a radar chart approach (Figure 6), followed by a discussion of the various components depicted in the radar chart.

Based on our analysis, both OARO and LSRRO can highly concentrate saline feed waters using moderate operating pressure. Hence, OARO and LSRRO are equally rated in this category of MLD/ZLD capability in Figure 6. In MLD/ZLD applications, LSRRO is more energy efficient than OARO for concentrating low-salinity feed waters, whereas OARO is energetically more competitive than LSRRO while treating hypersaline feed waters. Therefore, as shown in Figure 6, in terms of energy efficiency, LSRRO outperforms OARO for concentrating low-salinity feed waters, while OARO is better than LSRRO for treating high-salinity feed waters. However, we have shown that LSRRO could potentially outperform OARO for treating high-salinity feed waters by implementing more stages and/or elevating the operating pressure, which is illustrated by the dashed lines in Figure 6. Based on our analysis, the SEC of OARO and LSRRO could be less than 50% of that of MVC, suggesting that both technologies have the potential to replace MVC in MLD/ZLD applications.

The practicability of OARO and LSRRO was further compared using two performance metrics: membrane module availability and capital cost. Because membrane modules are essential to the practical implementation of OARO/LSRRO systems, their availability becomes a critical metric for evaluating the practicability of OARO/LSRRO. In addition, like other desalination systems, the practical implementation of OARO/LSRRO system is heavily determined by their overall costs. Thus, capital cost is another important performance metric for evaluating the practicability of OARO/LSRRO.

Distinctive membrane modules are used in OARO and LSRRO systems. Specifically, OARO uses bilateral RO membrane modules, in which spacers are needed on both sides of the membrane to simultaneously maintain channel structures and enhance mass transfer coefficients. For mitigating the parasitic energy consumption resulting from the
cross-flow streams in the module, loose spacers are required. However, loose spacers would cause severe membrane deformation under the applied hydraulic pressure that significantly compromises membrane performance.\textsuperscript{33,34} Because of the absence of robust bilateral spiral wound RO membrane modules, hollow fiber membrane modules have been used in pilot studies.\textsuperscript{35,36} However, these cellulose triacetate (CTA) hollow fiber membranes have very low water flux because of the inherent low permeability of the CTA active layer and the large structural parameter of the membrane which induces significant internal concentration polarization.\textsuperscript{37} The low water flux of such modules will require a very large membrane area, which will significantly increase the capital cost. Furthermore, with such membrane modules, there is a significant pressure drop of the stream in the hollow fibers, which would compromise the energy efficiency of the OARO system.

In contrast, LSRO uses conventional RO modules with low-salt-rejection membranes. Such membrane modules can be commercial nanofiltration modules (albeit their operating pressure will be limited to less than $\sim40$ bar) or commercial RO modules after chemically treating the RO membranes with an oxidant such as chlorine.\textsuperscript{38} The latter is a very appealing option as RO modules at their end-of-life can be chemically treated to be used in LSRO, instead of being discarded and creating solid waste. Therefore, regarding the availability of membrane modules, LSRO is advantageous over OARO (Figure 6), suggesting that LSRO is more ready to be implemented in practice than OARO.

In fact, even with commercially available bilateral RO modules, to avoid rupture of current thin-film composite membranes supported by a spacer, the applied $\Delta P$ cannot exceed $55$ bar,\textsuperscript{52} suggesting that the transmembrane water flux in OARO is limited. Additionally, the water permeation in the bilateral RO modules could cause severe concentration polarization on both sides of the membrane, further reducing the transmembrane water flux.\textsuperscript{53,40–42} For practical applications, a smaller transmembrane water flux requires a larger membrane area, corresponding to an increased capital cost of membrane modules. In comparison, the LSRO membrane modules can withstand high $\Delta P$ because densely woven mesh spacers can be used on the permeate side to support the thin-film composite membranes. Additionally, with the use of low-salt-rejection membranes, a large portion of salts permeates through the membranes, consequently mitigating concentration polarization effects. Thus, LSRO can maintain relatively high transmembrane water fluxes and therefore requires a smaller membrane area than OARO. In other words, the expense for membrane modules in LSRO is potentially lower than that in OARO.

The capital cost of an OARO/LSRO system includes not only the expense for membrane modules but also the expenses for high-pressure pumps and ERDs. Based on the working principles of OARO/LSRO (Figure 2), an $N$-stage OARO system comprises $N$ high-pressure pumps, $N$ ERDs, and $N$ membrane modules/stages, whereas an $N$-stage LSRO system contains $N - 1$ high-pressure pumps, only one ERD, and $N$ membrane modules/stages. Obviously, with a similar number of stages, an LSRO system requires fewer high-pressure pumps and ERDs than an OARO system. Thus, in terms of capital cost, LSRO should be more competitive than OARO as fewer membrane modules, high-pressure pumps, and ERDs are needed (i.e., LSRO outperforms OARO in Figure 6). According to the comparison of membrane module availability and capital cost, LSRO should be a more practical technology than OARO in ZLD/MLD applications.

We emphasize that this study is a theoretical investigation for providing guidelines for future technology development. The primary goal of this study is to compare the energy efficiency of OARO and LSRO, and the whole analysis is based on process modeling. For a more detailed and industrially relevant comparison of these two technologies, the levelized cost of water (LCOW) needs to be assessed.\textsuperscript{16,43} LCOW of a water treatment technology is defined as the cost per unit volume of product water. To estimate LCOW, a techno-economic analysis should be carried out.\textsuperscript{44,45} We further note that although the process modeling approach is able to evaluate the energy efficiency of the OARO/LSRO systems, it cannot provide the specific system design parameters (e.g., membrane properties, membrane module sizes, and feed flow rates) that are critical for techno-economic analysis. To acquire these design parameters, future work should carry out detailed module-scale analysis of OARO/LSRO systems.\textsuperscript{41,46}

In addition, several practical issues during system operation, such as membrane fouling and scaling, have not been considered. For practical MLD/ZLD applications, with the feed solutions being highly concentrated, fouling and scaling of the membranes could be significant challenges.\textsuperscript{37,48} Thus, to practically implement OARO/LSRO, effective pretreatment of feed waters (e.g., the addition of antiscalants, coagulation and softening, and nanofiltration) and/or other fouling and scaling mitigation strategies (e.g., development of novel membrane materials and operational modes) should be implemented.\textsuperscript{49–52}

\section*{ASSOCIATED CONTENT}

\section*{Supporting Information}

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.1c01638.

Derivation of eqs 1, 2a, and 2b; calculation of SEC in N-stage OARO; calculation of SEC in two-stage LSRO; calculation of SEC in three-stage LSRO; calculation of SEC in four-stage LSRO; operating conditions and SEC of OARO; and optimization results of SEC in LSRO (PDF)

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