Manuscript Details

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Title	Thermodynamic Optimization of Multistage Pressure Retarded Osmosis (MPRO) with Variable Feed Pressures for Hypersaline Solutions
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Abstract

Salinity gradient processes, such as Forward Osmosis and Pressure Retarded Osmosis, have been proven to be promising technologies for reducing the energy consumption in water treatment processes, for energy production, and for energy recovery. In such processes higher power densities can be achieved by applying higher hydraulic pressures on the draw solution, this requires greater mechanical stability of the membrane to be able to withstand these higher hydraulic pressures. Therefore, there is a limitation to the salinity of the draw solution which can be used in the PRO processes. This being dependent on the concentration of the hypersaline solution and hence overall hydraulic pressure, necessitating the use of an ultra-thick support layer for maximum energy production and/or recovery. In this theoretical and simulative optimization of the PRO process, we achieved the optimum energy recovery from a hypersaline solution (TDS ~ 300,000 mg/L) by using a multistage PRO (MPRO) system which included implementing variable applied feed pressures to each stage. The results showed that the volumetric flow rate of the hypersaline draw solution increased by up to a factor of 10 during the MPRO process in single pass, and the concentration of the hypersaline draw solution diluted up to 10x accordingly.

Keywords	Pressure Retarded Osmosis; Hypersaline Solution; Energy Recovery; Osmotic Pressure; Gibbs' Free Energy of Mixing; Salinity Gradient
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Dear Editor

Please find enclosed the article entitled ‘Thermodynamic Optimization of Multistage Pressure Retarded Osmosis (MPRO) with Variable Feed Pressures for Hypersaline Solutions’ which I am submitting for consideration for publication in the Desalination Journal.

This paper describes theoretical optimization of pressure retarded osmosis (PRO) processes to be used to utilize osmotic pressure in hypersaline solutions. When hypersaline solutions are used, the draw solution can be diluted as much as 10 times based on the theoretical osmotic potential calculations. However, such dilution would be impossible with single pass PRO process. In our paper we implemented multistage PRO process in order to overcome the drawing limitations from a single PRO process. In further optimization attempt we implemented a variable feed pressure set-up, which was proven to be effective to increase the volumetric flow rate of the diluted draw solution.

Therefore, we believe our work, as summarized above, can be considered as a breakthrough on utilizing the osmotic potential of hypersaline soltuions. We believe the manuscript that I am submitting will receive worldwide attention from various research and private organisations.

Thank you for your consideration of our work for publication.

Sincerely,
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HIGHLIGHTS

- The optimum osmotic energy recovery from a hypersaline solution (TDS ~ 300,000 mg/L) was theoretically achieved by using a multistage PRO (MPRO) system.

- Using variable feed pressures to MPRO process further increased the theoretical energy recovery from a hypersaline solution.

- The simulation results showed that the volumetric flow rate of the hypersaline draw solution can increase by up to a factor of 10 during the MPRO process in single pass.

Thermodynamic Optimization of Multistage Pressure Retarded Osmosis (MPRO) with Variable Feed Pressures for Hypersaline Solutions

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Abstract:
Salinity gradient processes, such as Forward Osmosis and Pressure Retarded Osmosis, have been proven to be promising technologies for reducing the energy consumption in water treatment processes, for energy production, and for energy recovery. Based on the thermodynamic concepts, specifically Gibbs' Free Energy of Mixing, the concentration of the draw solution plays an important role in determining whether the selected salinity gradient process is economically feasible or not. An increase in the salinity of a draw solution does not only increase the osmotic pressure difference between the draw and feed solutions, , but also allows a higher hydraulic pressure to be applied on the draw solution, which together greatly increases the volumetric flux of the draw solution per single pass when PRO is used. Even though higher power densities can be achieved by applying higher hydraulic pressures on the draw solution, this requires greater mechanical stability of the membrane to be able to withstand these higher hydraulic pressures. In order to increase the mechanical stability of the membranes, generally, thicker support layers can be applied, which have a direct negative impact on membrane permeability. Therefore, there is a limitation to the salinity of the draw solution which can be used in the PRO processes. This being dependent on the concentration of the hypersaline solution and hence overall hydraulic pressure, necessitating the use of an ultra-thick support layer for maximum energy production and/or recovery. , In this theoretical and simulative optimization of the PRO process, we achieved the optimum energy recovery from a hypersaline solution (TDS ~ 300,000 mg/L) by using a multistage PRO (MPRO) system which included implementing variable applied feed pressures to each stage. The results showed that the volumetric flow rate of the hypersaline draw solution increased by up to a factor of 10 during the MPRO process in single pass, and the concentration of the hypersaline draw solution diluted up to 10x accordingly.

Keywords: Pressure Retarded Osmosis; Hypersaline Solution; Energy Recovery; Osmotic Pressure; Gibbs' Free Energy of Mixing; Salinity Gradient

1. **Introduction and Background**

The treatment of hypersaline solutions (TDS ~ 300,000 mg/L) has been considered challenging because of the high energy input required to achieve the necessary desired water quality for human consumption and/or irrigation. Traditionally, thermal processes have been used to treat the hypersaline solutions, generally only when there is a requirement for treatment, such as environmental regulations or forced reuse because of the lack of fresh water sources [1].

Thermal distillation and vapor compression are two of the most common technologies to treat high salinity brines (usually only up to 125,000 mg/L TDS), but such treatments come with a levelized cost of up to \$39/m^3 for complete desalination [2–4]. As a result of this high energy and cost requirement, the main purpose of hypersaline solution treatment has been generally limited to lowering the salinity level enough to a degree that is acceptable for discharge [2].

Electro-membrane processes have also been used to reduce the salinity of hypersaline solutions, such as electrodialysis and electrodialysis reversal (EDR) [5–7], shock electrodialysis (SED) [8] and ion concentration polarization (ICP) [1], and/or produce energy by utilizing the salinity gradient potential, such as reverse electrodialysis (RED) [9]. Salinity, permselectivity of the ion exchange membranes, fouling related ionic resistance, ionic current, and scalability are some of the important limitations of the electro-membrane processes when used for the desalination of hypersaline solutions [5,7,9,10].

Membrane salinity gradient processes, where the salinity gradient is utilized as the driving force to draw water from the target feed solution to the higher salinity draw solution, can offer an economically feasible solution to the treatment of hypersaline solutions, either for a complete desalination or energy production via osmotic conversion [11–13]. Forward osmosis (FO) and pressure retarded osmosis (PRO) do not have upper limits for salinity but where in fact, having higher salinities increases the driving force through the membrane, hence resulting in higher flux values [14,15]. The main fundamental difference between a FO and a PRO process is that the FO often requires a secondary treatment stage in order to separate the draw solutes from the produced (drawn) water [16,17]. Whereas, PRO does not require such a secondary process stage to further treat the diluted draw solution, as the main product of the PRO is the hydraulic energy converted from the osmotic energy [18,19]. Therefore, application of PRO process to desalination of hypersaline solutions would be, theoretically, more favorable than that of FO processes.

PRO is a near isobaric process (Fig. 1), where the pressure of the draw solution does not change, considerably, during the process [11]. PRO converts osmotic pressure to hydraulic pressure, via a volume change, by drawing water from the feed solution at a near-constant pressure. Based on one of the most basic energy descriptions, if the pressure is constant and the volume is increased the extractable work from the system increases.

$$\textit{Pressure x Volume = Energy} \qquad (1)$$

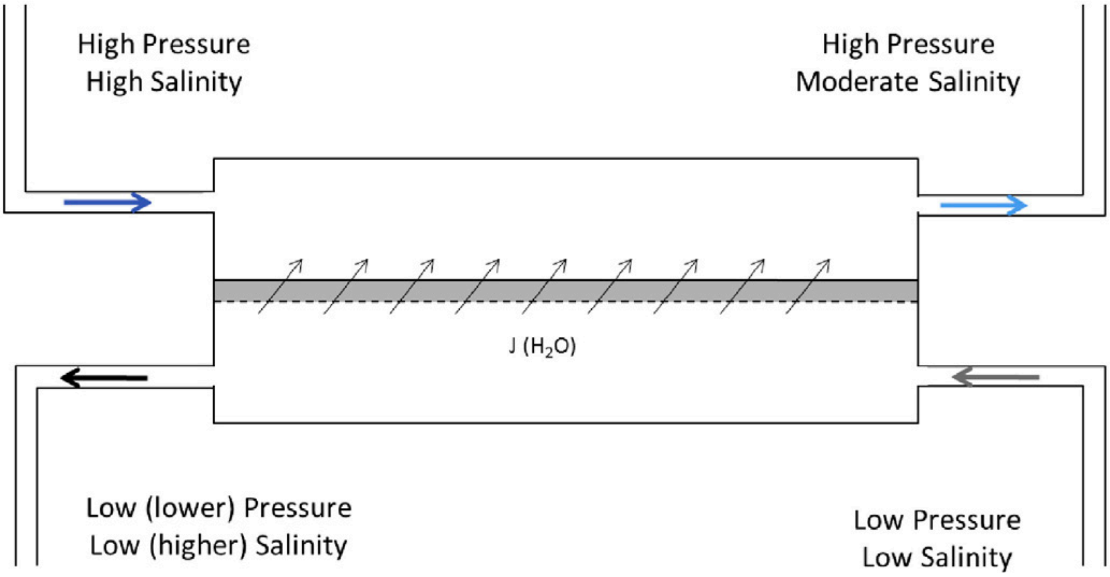

Fig. 1. Simplified representation of a near-isobaric PRO process [11].

This increase in the energy during PRO processes can be explained theoretically by Gibbs Free energy of mixing [15].

$$-\frac{\Delta G_{mix,V^0_{LC}}}{\nu RT} \approx \frac{c_M}{\emptyset}\ln c_M - c_{LC}\ln c_{LC} - \frac{1-\emptyset}{\emptyset} c_{HC}\ln c_{HC} \tag{2}$$

where ν is the number of dissociated ions for each electrolyte molecule, c is the molar salt concentration of the solutions, v^0 represents the specific volume at the reference state, LC stands for low concentration, HC stands for high concentration, M is for the mixed solution, and ϕ is the ratio of the total moles in given solution.

The produced energy in a PRO process is directly related to the amount of volume drawn from the feed solution and the applied hydraulic pressure on the draw solution. The optimum applied hydraulic pressure (ΔP), which gives the maximum extractable work production, is theorized to be the half of the osmotic pressure (π) of the draw solution [20].

$$\Delta P = \frac{\Delta\pi}{2} \tag{3}$$

If a hypersaline solution (TDS ≅ 300,000 mg/L) is used as a draw solution, with an osmotic pressure value of (approximately) 230 bar, the optimum applied hydraulic pressure should be around 115 bar to produce the maximum extractable work. However, this hydraulic pressure level will require a membrane with a substantial mechanical strength, which may require a very thick support layer and special spacers. Such a thick support layer will increase the *S* value (structural parameter) of the membrane, which will also increase the internal concentration polarization (Fig. 2) and subsequently restrict the water flow pathways[21–23]. With today's current technology, it is not possible to produce PRO membranes which can withstand ultrahigh hydraulic pressures (ex. 115 bars), and still give a positive water flux. The flux, therefore, in PRO processes can be given based on the solution-diffusion thermodynamic model:

$$J_w = A_{PRO}[(\pi_{net}) - (P_{net})] \tag{4}$$

Where A is the Pure water permeability constant, and hydraulic and osmotic pressures were given as “net” values.

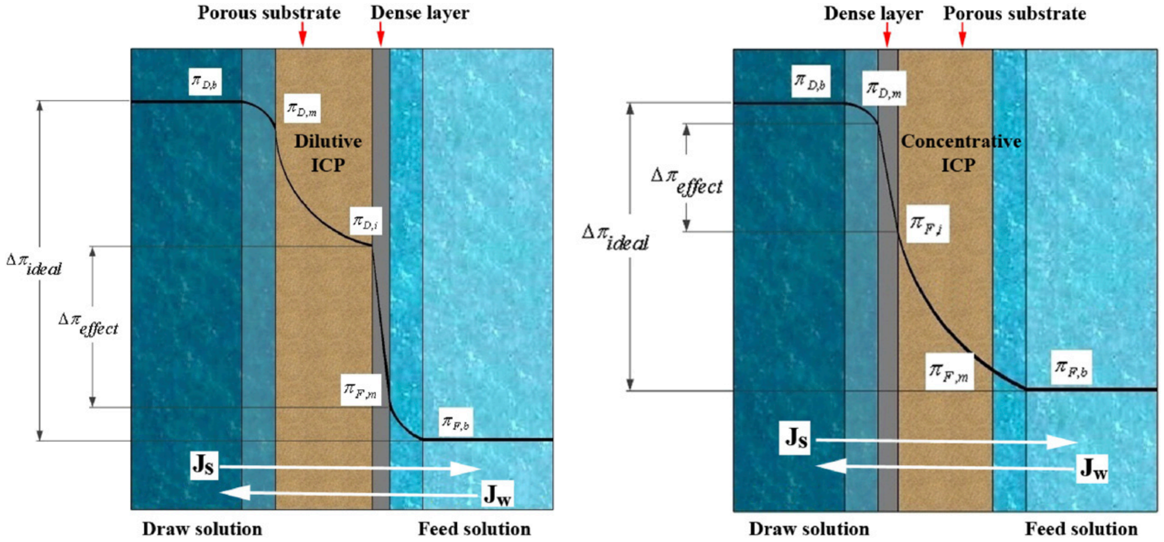

Fig. 2. Dilutive and concentrative ICP in salinity gradient processes [24]

In addition to the maximum applied hydraulic pressure limitations, there are mechanical limitations which determine the maximum volume of water which can be drawn by PRO in a single pass. In the above scenario (hypersaline solution), if the feed water salinity is around 1.5 bar (2,000 mg/L TDS), the draw solution can be diluted up to a maximum of 8-9 times, depending on the hydraulic and mechanical properties of the PRO module. However, it is not possible to dilute the draw solution 8-9 times in a single pass in a PRO system, due to the mechanical and hydraulic design limitations of the PRO membrane modules. Therefore, to overcome this limitation a robust and optimized multistage PRO process design is required.

In this work, we have designed and theoretically optimized a multistage PRO process with variable feed pressures. The pressure of the feed solution was increased in the later stages of the PRO in order to enhance the water flux, and hence optimize energy production.

2. Design Parameters and Operating Conditions

Theoretical optimization of a process requires a high level of accuracy in order to be accepted as a trustworthy reference and to be used in further process development. Therefore, pre-determined design parameters and operating conditions from various literatures and past experimental results were used in this work. We used 2 different designs for the optimization of MPRO, with and without variable feed pressures, in order to determine the effect of variable feed pressure on the performance of the MPRO process. Operating conditions and design parameters of these selected processes are given in Table 1.

Table 1. Operating conditions and design parameters for hypersaline MPRO with and without feed pressure manipulation.

Condition/Parameter	MPRO without Variable Feed Pressure	MPRO with Variable Feed Pressure
Draw solution TDS (mg/L)	330,000	330,000
Draw solution flowrate (m^3/h)	100	100
Draw solution pressure (bar)[1]	30	30
Draw solution pressure drop (%)[2]	1	1
Feed solution TDS (mg/L)	2,000	2,000
Feed solution flowrate (m^3/h)	1,000	Calculated [3]
Feed solution pressure (bar)	1	1-10
Feed solution pressure drop (bar)[2]	0.5	0.5
Number of PRO stages	4	4
Flowrate ratio in each PRO stage (Draw/Feed)	1	1
Draw solution flowrate increase in each stage (%)[4]	80	80
Flow pattern	Counter-current	Counter-current
Minimum absolute pressure difference in the membrane cell (bar) (hydraulic and osmotic)[5]	~ 5	~ 4

[1] The draw solution pressure was selected as 30 bar as recent reports on the membrane performances at this pressure, in both laboratory and pilot scale, are confirmed to be mechanically and economically feasible [25–29].

[2] The pressure drop in the draw solution, during the PRO process, was selected as 1%, based on the near isobaric nature of the process and previous experimental results [11, 25].

[3] Feed solution flowrate for the variable feed pressure MPRO was back calculated using the design parameters and operating conditions.

[4] Draw solution' volumetric flow rate increase was capped at 80% in order to satisfy the thermodynamic limitations. Where more than an 80% volumetric increase would result in a very low-pressure difference to draw substantial amount of water (Equation 3 and [5]).

3. Mass and Energy Calculations for the MPRO Process Designs

3.1. MPRO without Variable Feed Pressure Figure 3 shows the process flow diagram of the hypersaline MPRO without feed pressure manipulation. Hypersaline solution is fed into a pressure loop, between booster pump, PRO module, and isobaric pressure exchangers, where the pressure drop during the PRO process is compensated for by the booster pump. In order to operate the isobaric pressure exchanger at maximum efficiency, similar volumetric flow rates were supplied on both ends of the pressure exchanger. Isobaric pressure exchangers have more than 95% pressure transfer efficiencies (Energy Recovery Inc.) and therefore the pressure recovery of the isobaric pressure exchangers was capped at 95% efficiency for this theoretical simulation. The MPRO stages and mass balance calculations are shown in Figure 4, and all mass balance calculations were done according to the assumptions shown in Table 1. The calculations showed that 4 PRO stages (Figure 4) will give the optimum osmotic pressure recovery from the given hypersaline solution (TDS 330,000 mg/l). Calculations showed that after the 4th PRO stage the osmotic pressure and hydraulic pressure difference between the two sides of the membrane where to narrow and the efficiency of the process would be substantially reduced, even if a counter-current flow pattern was chosen (Figure 5).

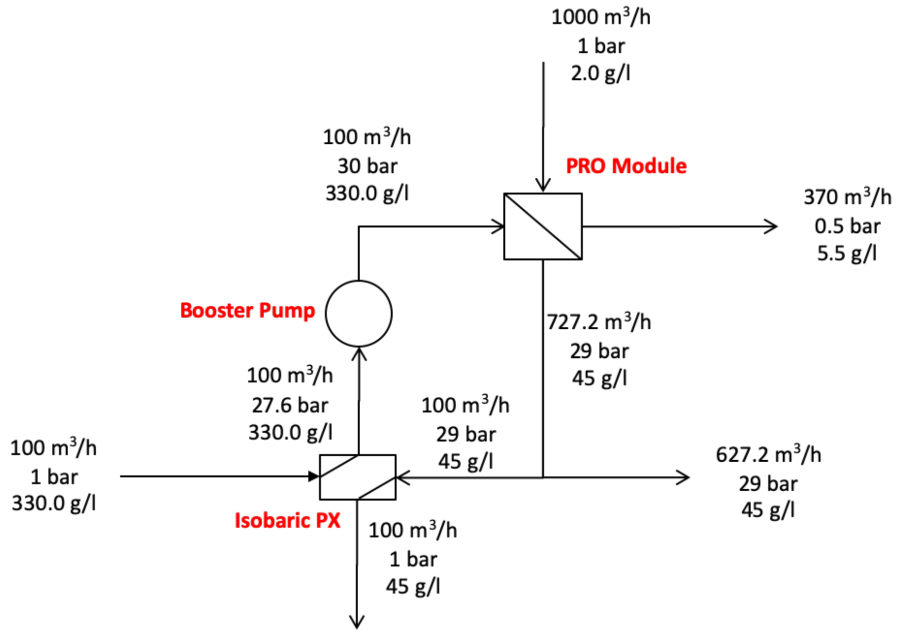

Figure 3 Overall process flow diagram for hypersaline MPRO without feed pressure modification.

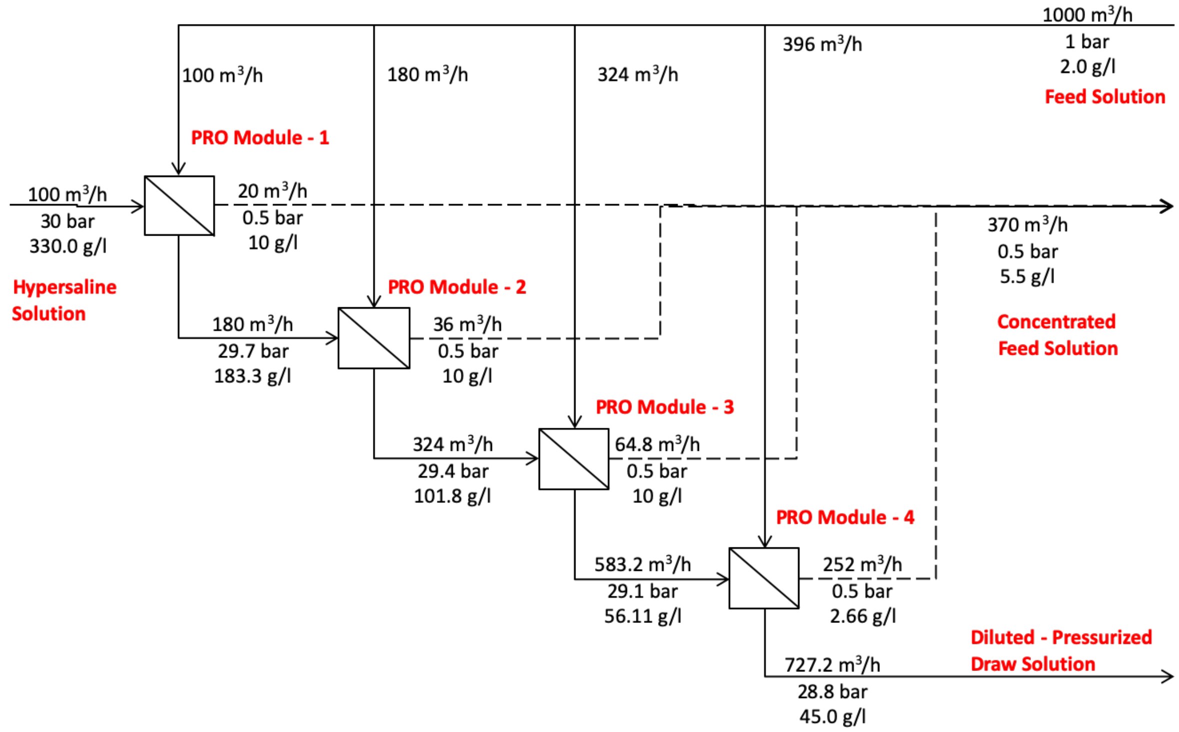

Figure 4 Flow diagram and mass balance calculations for Multistage PRO for hypersaline solutions without feed pressure manipulation.

Figure 5 shows the net pressure differences between the draw and feed solution sides of the PRO membrane. Because the MPRO process chosen here is without feed pressure manipulations, the MPOR process could be assumed as a single process (Figure 3) in order to make calculations regarding the thermodynamic limits for net pressure and water flux.

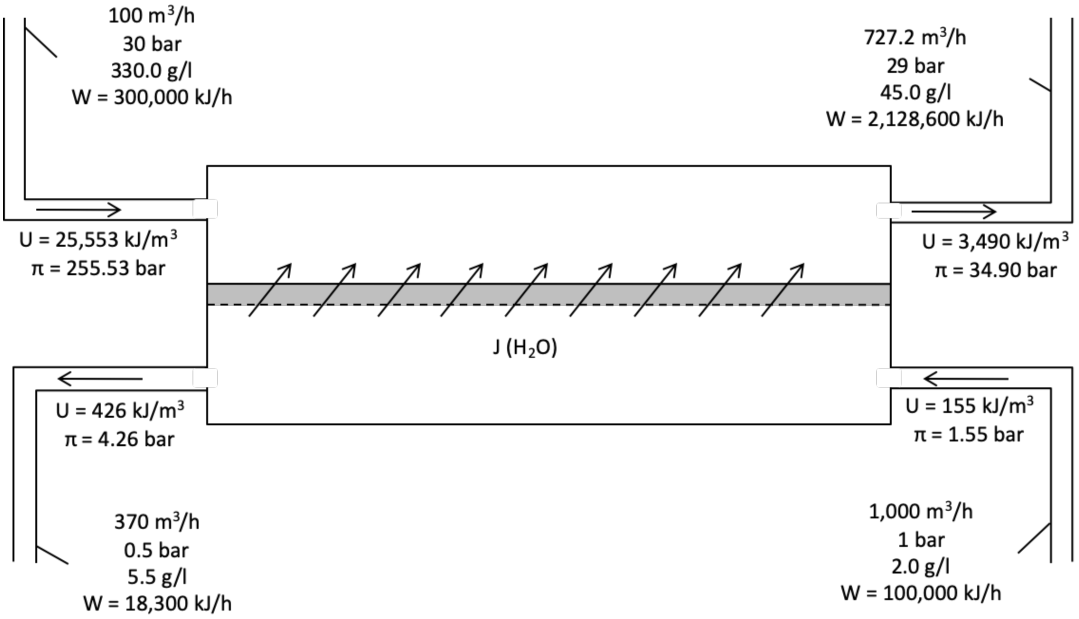

Figure 5 Mass and energy balance for the MPRO membranes, assumed as a single PRO module for calculation purposes.

At any point of the process, the net pressure difference (Osmotic – Hydraulic) between draw (D) and feed (F) sides of the PRO membrane can be calculated from:

$$(\Pi_D - P_D) - (\Pi_F - P_F) \tag{5}$$

If the net pressure difference is greater than zero, the water flux will theoretically be positive. However, below certain pressure differences, flux values will become too small to be considered as feasible for process applications. Therefore, the net pressure difference at the end of the module (Figure 5 right hand side) was kept higher than 3 bar for the design and calculations from Equation 5:

(34.90 bar – 29.00 bar) – (1.55 bar – 1.00 bar) = 5.35 bar net pressure difference.

MPRO without the variable feed pressure can reduce the salinity of the hypersaline solutions from 330,000 mg/l to around 49,000 mg/l, which will then be suitable for reverse osmosis (RO) processes. If an RO process was used to produce drinking water from the diluted PRO draw solution (Figure 3-5), the hydraulic pressure of the solution (29 bars) will result in low energy consumption for the RO process. Alternatively, the diluted draw solution can be fed to a hydro turbine and the hydraulic pressure of the solution (29 bars) can be converted directly to electricity, the depressurized diluted draw solution could then be safely discharged.

3.2. **MPRO with Variable Feed Pressure** In order to further decrease the salinity of the diluted draw solution and increase the amount of pressurized solution ready for RO process, the hydraulic pressure of the feed was modified, and a variable feed pressure design was developed (Figure 6).

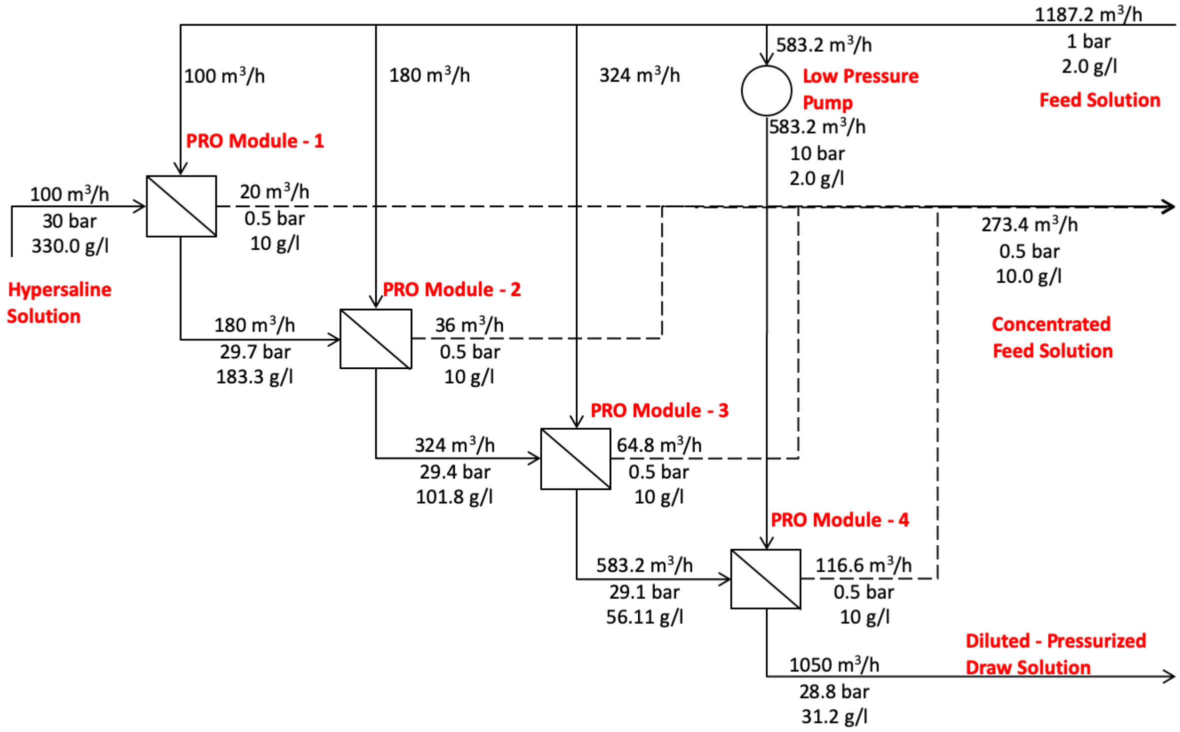

Figure 6 Flow diagram and mass balance calculations for Multistage PRO for hypersaline solutions with feed pressure manipulation of stage 4.

The feed solution to the 4th PRO stage was pressurized to 10 bar in order to increase the water flux through the PRO membrane (Equation 3) and therefore reduce the salinity of the diluted draw solution still further. A pressure of 10 bar was selected based on the thermodynamic limitations and the calculations made using Equation 5. Since the feed pressure is variable, two sets of mass and energy calculations were made (Figures 7 and 8).

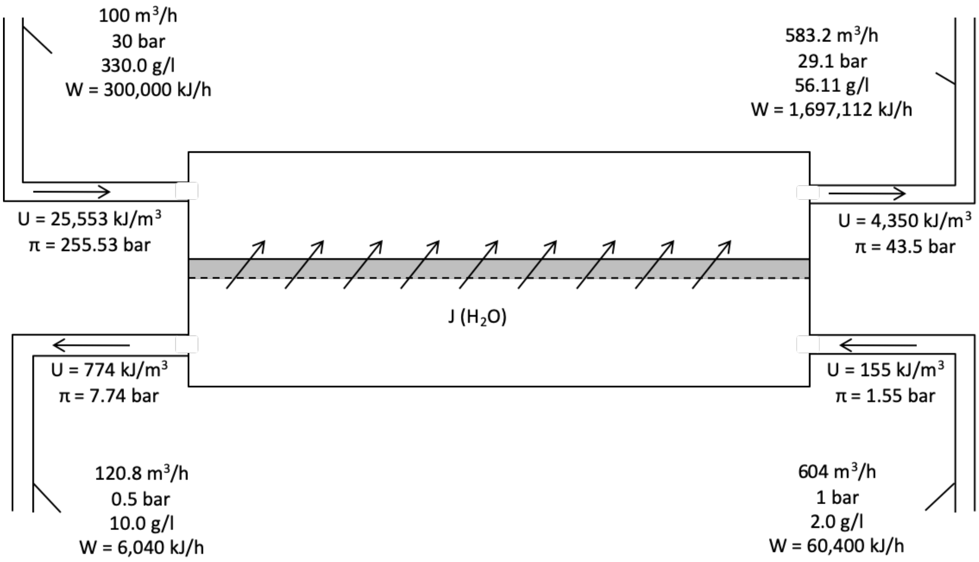

Figure 7 Mass and energy balance for the first 3 stages of PRO processes, where the feed pressure is constant (1 bar)

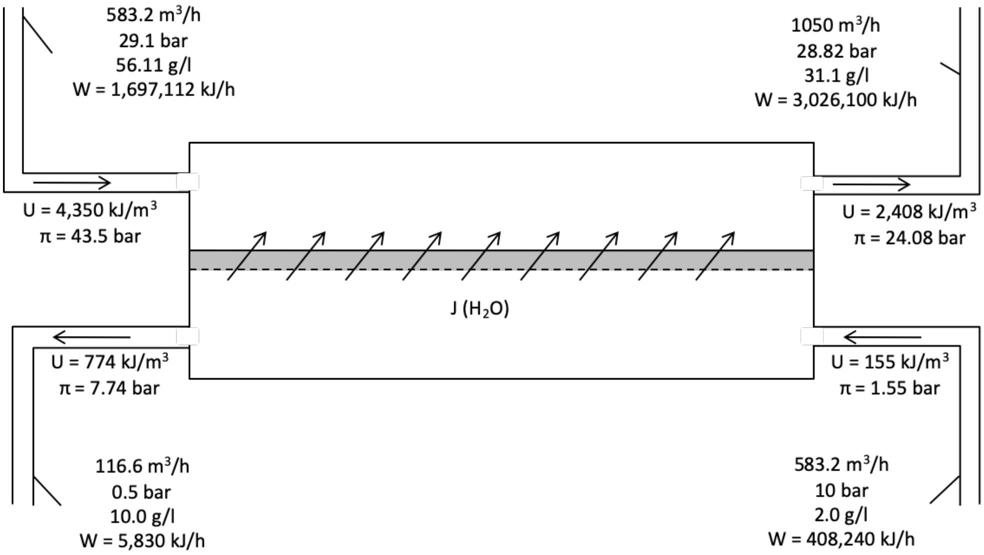

Figure 8 Mass and energy balance for the 4th PRO stage, where the feed pressure is increased to 10 bar.

The end of the 4th PRO module (Figure 8, right hand side) was the limiting zone for the net pressure difference between draw and feed side of the PRO membrane. The pressure difference here was also kept above 3 bar based on the previous discussed assumptions. The net pressure difference was (Equation 5)

$(24.08 - 28.82) - (1.55 - 10.00) = 3.71$ bars

The overall MPRO process with variable feed pressure is shown in Figure 9. MPRO with variable feed pressure can further reduce the hypersaline solution's salinity from 330,000 mg/l to 31,100 mg/l, which is lower than most seawater salinity levels. Additionally, by increasing the feed solution's hydraulic pressure to 10 bar for the 4th PRO stage the volumetric flow rate of the diluted draw solution was increased to 1050 m^3/h (compared to 727.2 m^3/h without stage pressurization), while the pressure of this draw solution was kept constant (28.82 bars) to the previous design without the variable feed pressure (29 bars)

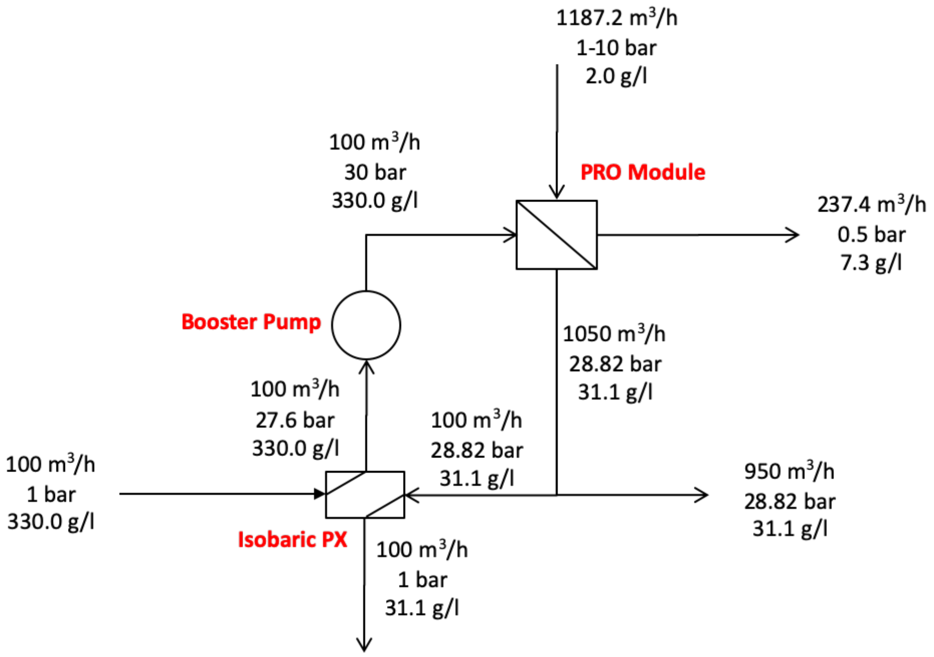

Figure 9 Overall process flow diagram for MPRO with variable feed pressure.

4. Conclusion

Hypersaline solutions have very high osmotic pressures (up to 250 bars) which could be harnessed as a sustainable energy source as long as the osmotic pressure could be converted to hydraulic pressures with little loss of efficiency. However, the current available salinity gradient process applications (single pass PRO process) cannot increase the draw solutions volumetric flow rate to greater than 100%, due to the limitations of the thermodynamics (absolute difference between osmotic and hydraulic pressure in membrane module) as well as the mechanical limitations(the restricted volume of the membrane modules) of the current systems. The new proposed multi-stage PRO process (MPRO) design can overcome such limitations, whereby the draw solution can continuously be diluted over the number of stages. This being achieved by pressurizing the feed to each separate stage to maintain the absolute pressure difference high enough to maintain an effective water flux through the PRO membranes.

MPRO process is theoretically shown to increase the draw solutions volumetric flow rate (via PRO process) by up to 7.2 times with relation to the initial inlet draw solution. The pressure drop over the MPRO was calculated to be around 4%, which was based on previous studies [11]. Further improvements on the MPRO process design were done by applying hydraulic pressure (10 bar) to the feed solution on the 4th PRO stage. This additional hydraulic pressure on the feed solution effectively increased the overall draw solution volumetric flow rate to approximately 10.5 times its original inlet volume. The proposed theoretical MPRO with variable stage feed solution pressures is shown to be a thermodynamically effective process which greatly increases the efficiency of converting the osmotic potential of a hypersaline water sources, such as hypersaline lakes and/or oil and gas co-produced waters to sustainable energy (hydraulic energy). Further improvements in the process can be made by following on from this work with experimental and pilot tests.

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Declaration of interests

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: