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TEMPORALLY MULTI-STAGED BATCH COUNTERFLOW REVERSE OSMOSIS FOR HIGH RECOVERY DESALINATION

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ABSTRACT

Osmotically assisted reverse osmosis (OARO) or counterflow reverse osmosis (CFRO) are recent RO configurations that uses saline streams on both sides of the membrane in counterflow. This reduces the osmotic pressure difference that needs to be overcome for permeation and allows water recovery from high salinity feeds at regular RO pressure. Batch RO is a new, transient RO configuration that closely follows the osmotic pressure profile of the feed and is marked by high energy efficiency. In this work we extend a transient version of CFRO, Batch CFRO for high recovery (~74%) desalination of seawater using a temporally multi-staged version of the process for the first time. In doing so, we introduce the first configuration to achieve Batch CFRO using entirely available components, including a pressure exchanger rather than high pressure tanks. Using a reduced order model, the terminal salinity of the brine leaving the system is calculated to be 183 g/kg. The key feature of this new configuration is that it is multi-staged in time rather than space. As such it can use the same hollow fiber membrane module for the different stages and hence reduce the component (pumps and pressure exchangers) count of the process. The brine produced in each stage is stored in inexpensive atmospheric pressure tanks. This is in contrast with other multi-stage processes where the number of flow devices usually scale with the number of stages needed for higher recovery and usually leads to high cost. Notably, the choice of membrane type can make a significant difference, as common narrow hollow fibers can experience large pressure drops that become significant. This leads to the conclusion that module design must be key to achieve the top-energy numbers of other batch CFRO configurations by the team, such as spiral wound membranes, turbulence-inducing spacers, or using feed on the shell side of the fibers.

Keywords: Recovery, salinity, multi-stage, batch, counterflow

I. INTRODUCTION

Water is an increasingly scarce natural resource in the world. The demand for freshwater is growing at a rate that can barely be met by its natural supply leading to acute shortages around the world. The World Resources Institute reports that by 2025, around 3.5 billion people could be affected by water scarcity[1]. Reverse osmosis (RO) is a membrane separation process that can help mitigate this problem given its high thermodynamic efficiency, compact footprint, and economic scalability[2]. However, RO today, is limited to seawater salinity and recovery ratios of around 50%, since operating with higher salinities or recoveries would most likely exceed the burst pressure of the membrane (~70 bar). Operation at higher pressures can also cause compaction of the active layer of the membrane with increased propensity for irreversible fouling[3]. Membranes rated for operation at higher pressures have thick support layers, necessary for mechanical strength, but lead to significant concentration polarization and reduced energy efficiency[4]. Treating high salinity feeds, as such, has been the forte of thermally driven processes which tend to be thermodynamically inefficient but economically feasible given that thermal energy is usually much cheaper than electricity.

Nonetheless, efforts at membrane-based desalination of high salinity feeds have been made with some reported in Table 1. Of these, osmotically assisted reverse osmosis (OARO)[3] or counterflow reverse osmosis (CFRO)[4] is a recent technology that can desalinate high salinity feeds and reach high recoveries while operating at regular RO pressures. In osmotically assisted processes, both sides of the membrane are saline, and the streams are in counterflow to maintain a relatively constant osmotic pressure difference across the membrane. The stream entering the feed side or concentrate side gets concentrated due to permeation. Likewise, the stream on the other side, which is called the diluate side, as the name suggests, gets diluted[4].

Batch RO processes have been shown to be highly energy efficient as they can closely follow the osmotic pressure profile of the feed while maintaining a relatively flat flux profile along the membrane[5]. Based on a similar idea, single stage Batch CFRO was introduced in[6] that could concentrate seawater to a recovery of around 62% with a terminal pressure of 70 bar. The working principle of the process has been shown in Fig. 1, where recirculation of the concentrate and diluate streams in their individual tanks allows for a uniform and gradual increase in pressure over time.

In this paper, we extend Batch CFRO to an even higher overall recovery (~74% of seawater) and introduce the first multi-stage batch process. In doing so, the same membrane module is used across different stages, requiring only one set of flow devices like pumps and pressure exchangers. The concentrate produced in one stage is used as the feed for the next stage, while the diluate produced in one stage is used in the next cycle for the previous stage. The system needs a well-designed valving system and an atmospheric pressure storage tank for each stage, which should be relatively less expensive than spatially multi-staged processes requiring flow devices for each stage.

Table 1: Comparison between high salinity and high recovery reverse osmosis desalination methods (adapted and modified from [6])

Configuration	Feed salinity (g/kg)	Recovery (%)	SEC (kWh/m^3)	Number of CFRO stages | pumps | ERDs
OARO[3]	60	50	3.70	3 | 3 | 3
COMRO[7]	70	50	3.16	4 | 4 | 1
Split feed CFRO[4]	35	81	3.90	5 | 5 | 4
BCFRO-BRO[6]	35	62	1.95	1 | 4 | 0
Multi-stage batch CFRO (this work)	35	74	4-21.4	1* | 4 | 1

*The single stages was run at 6 cycles to achieve this recovery

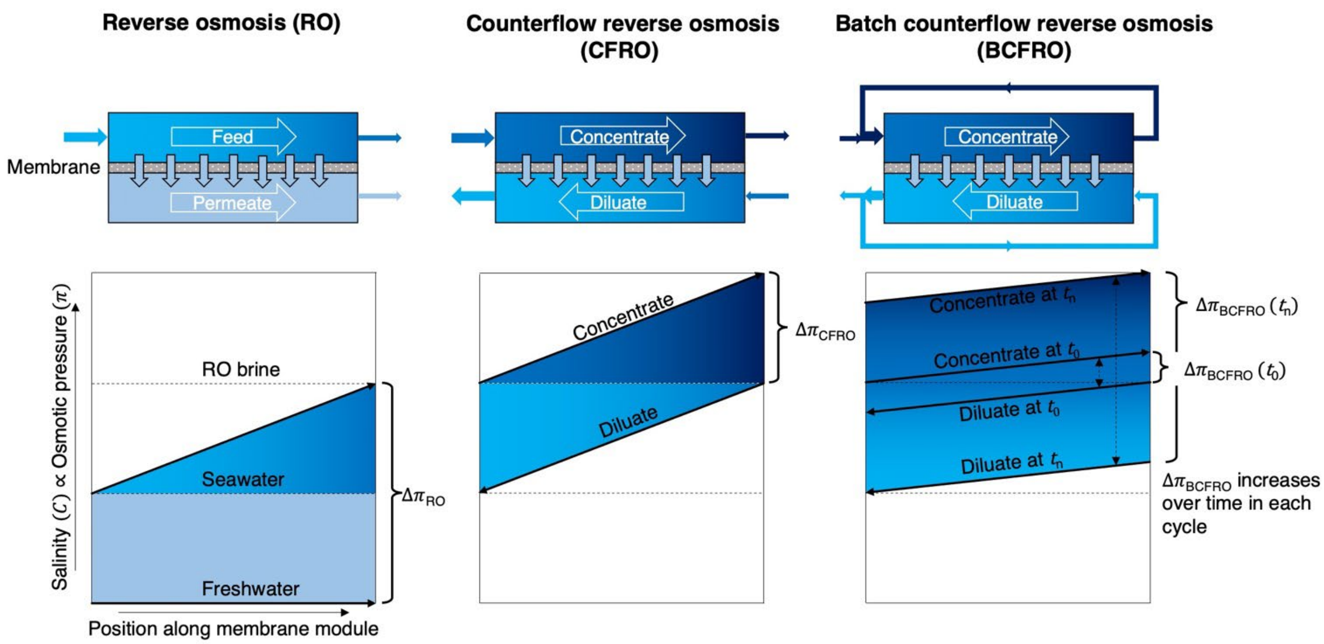

Fig. 1 Working principles of reverse osmosis (left), counterflow reverse osmosis (middle) and batch counterflow reverse osmosis (right). BCFRO allows for uniform and gradual increase in pressure overtime while achieving higher recoveries than CFRO and RO. Darker colors indicate higher salinity.

The recirculation of the concentrate and diluate streams leads to a gradual change in salinity over time and consequently the osmotic pressure difference that needs to be overcome to cause permeation (from [6]).

II. PROCESS DESIGN

The main idea behind multi-stage Batch CFRO is to gradually concentrate the feed over time using the same membrane module (Fig. 2, top right). The construction of a such a system involves a pressure vessel that can house multiple membrane elements, a high-pressure pump for setting the flux, circulation pumps for maintaining cross flow over the membrane surface and an energy recovery device (ERD) or pressure exchanger (PX) to recover energy from the concentrated feed (concentrate) exiting the membrane module. In regular Batch CFRO, two tanks are additionally needed, one for each side of the membrane for recirculation of the streams. The size of the tanks dictates the rate at which salinity of the tanks change over time and hence the overall process dynamics. The tanks can be either high-pressure tanks [6], for which a pressure exchanger is not needed, or atmospheric pressure tanks that require the use of a pressure exchanger (as in this work). In multi-stage Batch CFRO, the concentrate produced in one stage is used as the feed for the next stage. While the same concentrate tank can be used for the different stages, the diluate produced in each stage needs to be stored so that it can be used in the next cycle, but for the previous stage. As such, a tank is needed for each stage. These tanks (Fig. 2, bottom left) are atmospheric pressure tanks and as such only need appropriate valving to connect to the diluate side of the membrane module.

Some of the salient features of the process design are listed below:

1. The system is fed by seawater RO brine which has a salinity of ~70 g/kg (50% recovery). (Fig. 2, middle-right)
2. As permeation and recirculation happen in each stage, the salinity of the concentrate increases while that of the diluate decreases. This implies an increase in osmotic pressure difference across the membrane and consequently the applied pressure to maintain a certain flux. Increasing concentration for a stage is terminated when the applied pressure reaches a threshold value, usually a value close to the burst pressure of the membrane.
3. The diluate produced in one stage cannot be used immediately. The initial volume of diluate in each stage is chosen so that the salinity of the diluate at the end of the stage is equal to the salinity of the feed in the previous stage. Hence, the diluate needs to be stored so that it can be used in the next cycle. Therefore, small atmospheric tanks are needed for each stage, at each of these salinities (Fig 2., bottom left).
4. Only one of the diluate tanks (bottom left two rows) is used in a stage while the other tanks store diluate from other stages. Likewise, only one of the two feed tanks (top left) is used in a cycle while the other tank that has the terminal concentrate (brine) from the last stage of the previous cycle is drained and filled with fresh feed.
5. Additional recovery is driven by the diluate produced in the first stage. As mentioned above, the initial diluate volume is set so that the salinity of the diluate is close to that of seawater. Consequently, this reduces the amount sweater brine that needs to be fed to the system and in turn increases overall recovery.
6. The salinity of the diluate stored in each tank will change over the cycles in a cyclic manner. This is because each diluate tank will be eventually diluted to seawater salinity after which it will be emptied and fed back to the seawater RO plant.

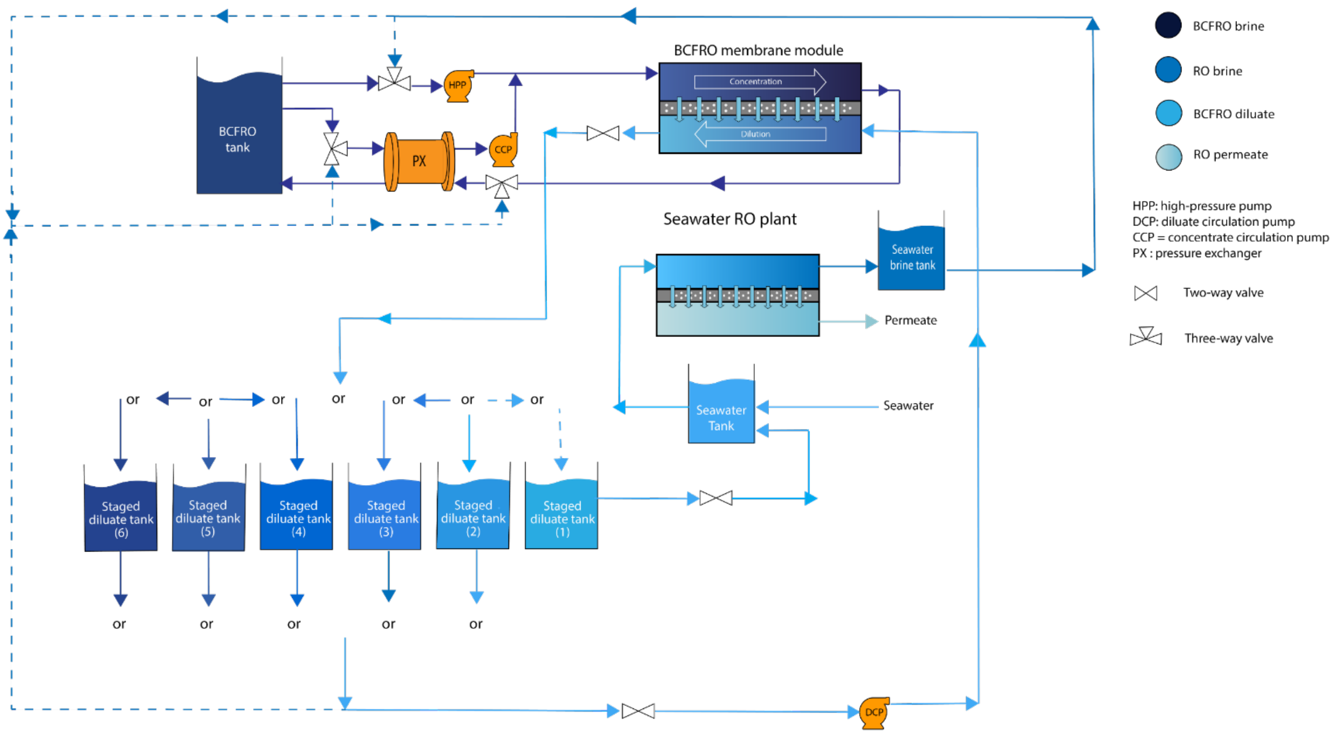

Fig. 2 Flow paths for concentrate and diluate streams in multi-stage Batch CFRO. HPP, CCP and PX are universal components in actual RO systems. While the circulation of the brine desalinates, the concentrate salinity increases as the diluate salinity decreases. Each step results in accumulating the diluate in tanks and matching the diluate salinity from the end of the step to be the same as the salinity of the feed from the previous step. This process helps keep the terminal pressure at 70 bar and ensure of achieving overall high recovery rates.

III. METHODOLOGY

We use a reduced order model for Batch CFRO developed in Simulink to capture the dynamics of the process. Variables of interest include time profiles of salinity on either side of the membrane both in bulk flow and at the membrane surface. The water flux across the membrane is set for each stage to maximize permeation with a relatively slow rate of pressure change. However, the circulation pump flow rates on the concentrate and diluate sides are held constant to maintain their rated flow rates. The osmotic pressure difference associated with the salinity profiles and the flux can then be used to calculate the required pressure that needs to be applied, which in turn can be used to calculate power requirements. It should be noted here that flux, salinity and pressure are coupled quantities and need to be solved for simultaneously.

Optimal values of flux and circulation pump flow rates were obtained using a pareto search algorithm. Since the volume of diluate produced in a stage is used in the next cycle for the previous stage, the difference between the volume of diluate required and produced was minimized using the optimizer. At the same time, the terminal salinity of the brine leaving the system was maximized. The minimum number of stages required to reach the target brine salinity was calculated to be 6 from initial runs. This gave a total of 8 parameters to be optimized: flux for the 6 stages, and the concentrate and diluate circulation pump flow rates. The key modeling equations have been listed below.

Ignoring the variation in density of saline streams and freshwater, mass conservation of water in the membrane module on the concentrate side can be written as

$$\dot{V}_p = J_w A_{mem} = \dot{V}_{c,in} - \dot{V}_{c,out} \qquad (1)$$

where $\dot{V}_p$ is the water permeation rate and can be calculated from flux, J_w and membrane area, A_{mem}. $\dot{V}_{c,in}$ and $\dot{V}_{c,out}$ are respectively, the concentrate flow rate into and out of the membrane module.

Mass conservation of salt on concentrate side (ignoring salt transport) can then be written as

$$\dot{C}_{c,in}(t) = \frac{\dot{V}_{c,out}C_{c,out}(t) - \dot{V}_{c,in}C_{c,in}(t) - \dot{V}_{tank,c}C_{c,in}(t)}{V_{tank,c}(t)} \qquad (2)$$

$$\dot{C}_{c,out}(t) = \frac{2(\dot{V}_{c,in}C_{c,in}(t) - \dot{V}_{c,out}C_{c,out}(t))}{V_{mem,c}} - \dot{C}_{c,in}(t) \qquad (3)$$

Here, $C_{c,in}$ and $C_{c,out}$ are the salinity of the of the concentrate stream entering and leaving the membrane module. It should be noted here that $C_{c,in}$ is also the salinity of the stream leaving the concentrate recirculation tank. $V_{tank,c}$ is the instantaneous volume of concentrate in the recirculation tank with $\dot{V}_{tank,c} = -\dot{V}_p$. $V_{mem,c}$ is the concentrate side membrane volume and can be calculated from the dimensions of the follow fiber membrane module; shell side for concentrate and bore side for diluate.

Similar equations can also be derived for the diluate side of the membrane and the recirculation tank, with a few sign changes to account for the flow of permeate into the control volume.

With the salinities calculated, osmotic pressure difference across the membrane can be calculated as[8]

$$\Delta\pi = \frac{\pi_c e^{\frac{J_w}{k}} - \pi_d e^{-\frac{J_w S}{D}}}{1 + \frac{B}{J_w}\left(e^{\frac{J_w}{k}} - e^{-\frac{J_w S}{D}}\right)} \qquad (4)$$

where π_c and π_d are respectively the osmotic pressure corresponding to bulk salinity on the concentrate and diluate side, J_w is the water flux, k is the mass transfer coefficient on the concentrate side, S is the support layer thickness, D is diffusion coefficient of salt in water and B is the salt permeability coefficient of the membrane.

The instantaneous pressure difference, ΔP required to cause permeation across a membran with water permeability coefficient, A_w can then be calculated using the equation

$$J_w = A_w(\Delta P - \Delta\pi) \qquad (5)$$

With the pressure difference calculated, the energy consumption of the process can be calculated and normalized by the total volume of permeate produced to obtain the specific energy consumption or SEC.

IV. RESULTS

For a single membrane module and specified process parameters, after optimizing the difference between the volume of diluate produced in different stages while trying to maximize the terminal concentrate salinity, the following performance numbers were obtained. The salinity profiles across the different stages of a cycle are shown in Fig. 3.

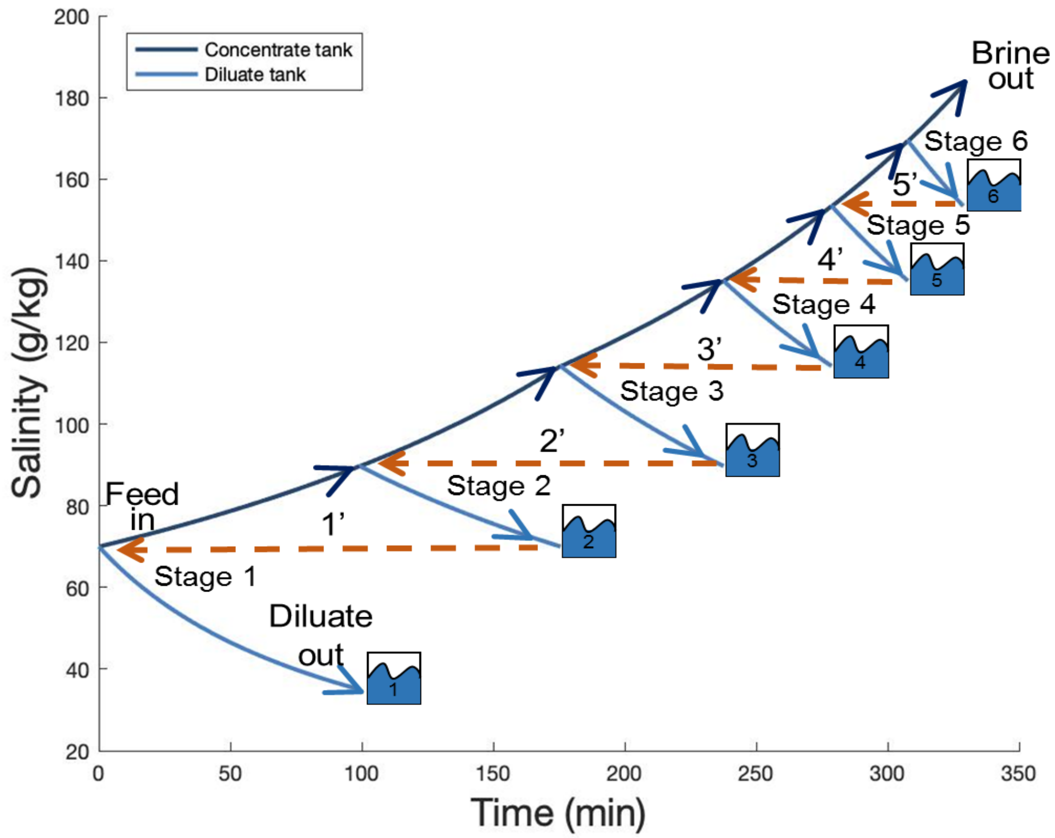

Fig. 3 Salinity profile for concentrate and diluate streams in multi-stage Batch CFRO. Stages are numbered, and prime symbols ' indicate a future cycle's stage. Feed enters at Time = 0 and progressively becomes more concentrated (proceeding to the top right). At each stage, the diluate exits and is stored in tanks. The next step the feed from the first stage will have the same salinity as the diluate from the second stage (see dashed arrows), thus causing the increase in the concentrate salinity. The process repeats until the system all diluate tanks becomes as saline as seawater. This achieves high recovery rates while the same constant terminal pressure since the pressure difference is held the same. (case 1 membrane)

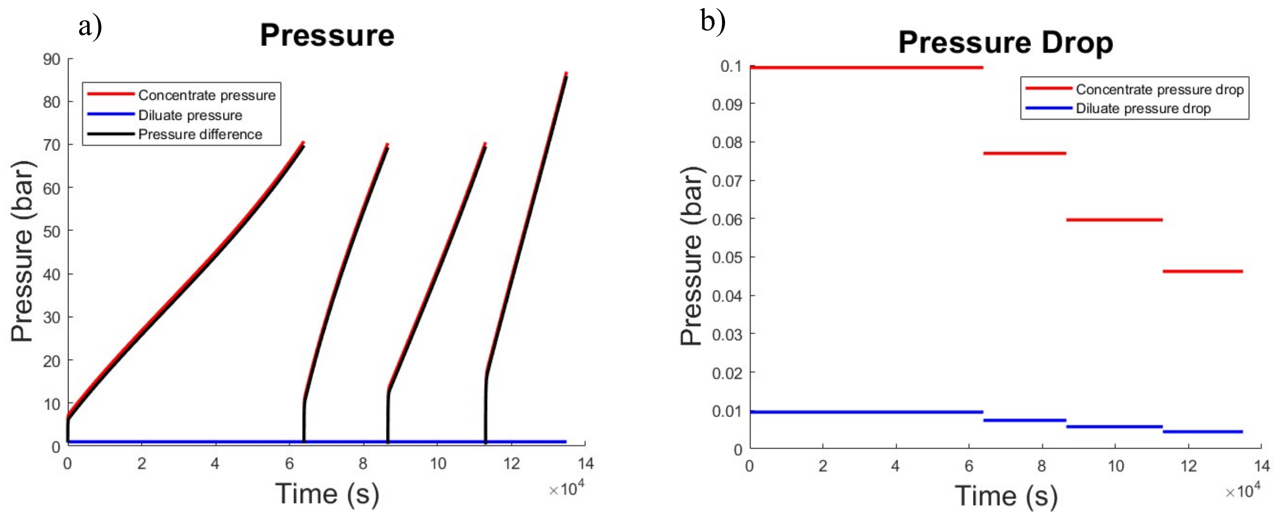

Fig. 4 Pressure in Temporally multi-staged batch counterflow RO. a) Pressure over the course of cycles concentrating to higher salinity with the same equipment. b) Pressure drop in each membrane module stream during said multistaging run (case 2 membrane)

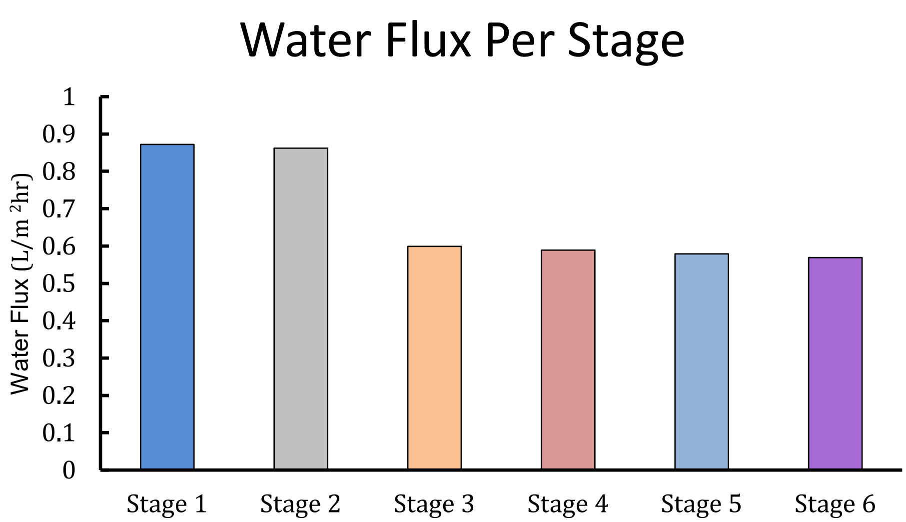

Fig. 5 Water Flux per stage, with seawater inlet and 74% recovery. (case 1 membrane)

<u>Membrane properties</u>[8]

Case 1)

Area based on hollow fiber outer diameter, A_{mem}: 76.8 m^2

Water permeability coefficient, A_w: 0.27 LMH/bar

Salt permeability coefficient, B: 0.035 LMH

Structural parameter S: 1000 μm

Case 2)

Area based on hollow fiber diameter: A_{mem}: 4.79 m^2

hollow fiber outer diameter [m] 1.06 mm
fiber module length 0.56 m
Water permeability coefficient 4430 LMH/bar

Salt permeability coefficient 1.88 LMH/bar

Structural parameter, S: 650 μm

Process parameters

Water flux

Stage 1: 0.872 LMH	Stage 2: 0.862 LMH	Stage 3: 0.599 LMH
Stage 4: 0.589 LMH	Stage 5: 0.579 LMH	Stage 6: 0.569 LMH

Concentrate circulation pump flow rate, $\dot{V}_{c,out}$: 18 l/min

Diluate circulation pump flow rate: $\dot{V}_{d,in}$: 36 l/min

High-pressure pump overall efficiency: η_{HPP}: 85%

Circulation pump overall efficiency: η_{CP}: 65%

Pressure exchanger overall efficiency: η_{PX}: 92%

Performance metrics

Overall water recovery, $RR_{overall}$: 74%

Terminal concentrate salinity, C_{term}: 183 g/kg

Overall specific energy consumption, SEC: 3.64 kWh/m^3 (Case 1)

Overall specific energy consumption SEC: 21.2 kWh/m^3 (Case 2)

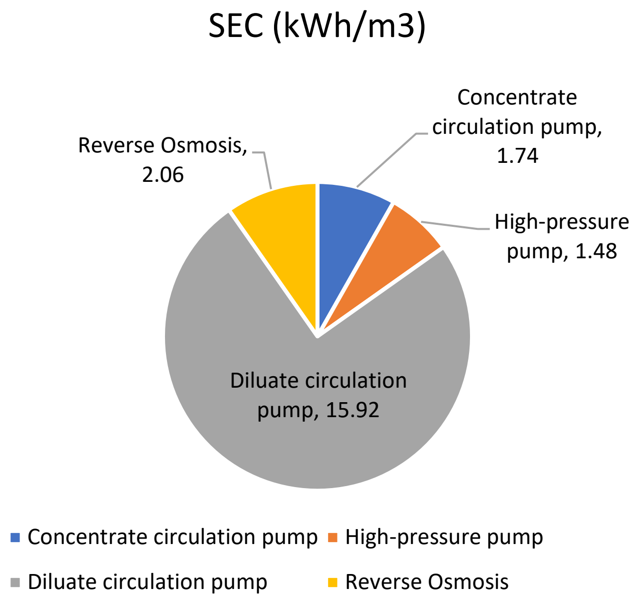

Fig. 4 Specific energy consumption (SEC) of different pumps in multi-stage Batch CFRO in kWh/m^3 (Case 1)

V. CONCLUSIONS

We report the first temporally multi-staged batch process that is capable of handling high salinity feeds and attaining high recoveries while using the same membrane module, pumps and pressure exchanger. The specific energy consumption of was 21.4 kWh/m^3 for the case 1 membranes, and ~4 kWh/m^3 for the case 2 membranes, for seawater at 74% recovery. Notably, currently available membrane modules may experience a fairly large pressure drop (case 1), as was analyzed here with Toyoba hollow fiber membranes. For these, fiber diameters and module lengths may not be the best fit for multi-stage Batch CFRO. The permeability coefficients are insufficient as well. Theoretically, the performance could get much closer to the team's piston-tank Batch CFRO design, whose models predicted an SEC of 1.95 kWh/m^3 for 62% recovery of seawater. Also, there are some losses in the diluate recirculation stream and concentration polarization that make it difficult to achieve the exceptional efficiencies of other batch reverse osmosis configurations [9-11]. Substantially more optimization is needed, including for module length, fiber diameter, membrane structural parameter, and any enhanced-mixing methods. Additionally, the process might benefit from using specially designed spiral wound membranes that offer a large flow area without significant confinement to reduce the pressure drop. Future work in this direction will assess the suitability of such membranes for the process and reducing pressure drop in the recirculation loops.

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