Closed-Loop Pressure Retarded Osmosis Draw Solutions and Their Regeneration Processes: A Review

Ali Etemad Zadeh[a], Khaled Touati[a], Catherine N. Mulligan[a]*, Jeffrey R. McCutcheon[b], Md. Saifur Rahaman[c]

[a]Department of Building, Civil, and Environmental Engineering
Concordia University,
EV-6.139, 1455 Boul de Maisonneuve Ouest
Montreal, QC, Canada, H3G 1M8
[b]Department of Chemical & Biomolecular Engineering, Center for Environmental Sciences and Engineering, University of Connecticut
191 Auditorium Road, Unit 3222, Storrs, CT 06269-3222

[C]Department of Civil, Geological and Mining Engineering, Polytechnique Montreal, QC H3C 3A7,

Canada

*Corresponding author: Catherine Mulligan, Email: catherine.mulligan@concordia.ca

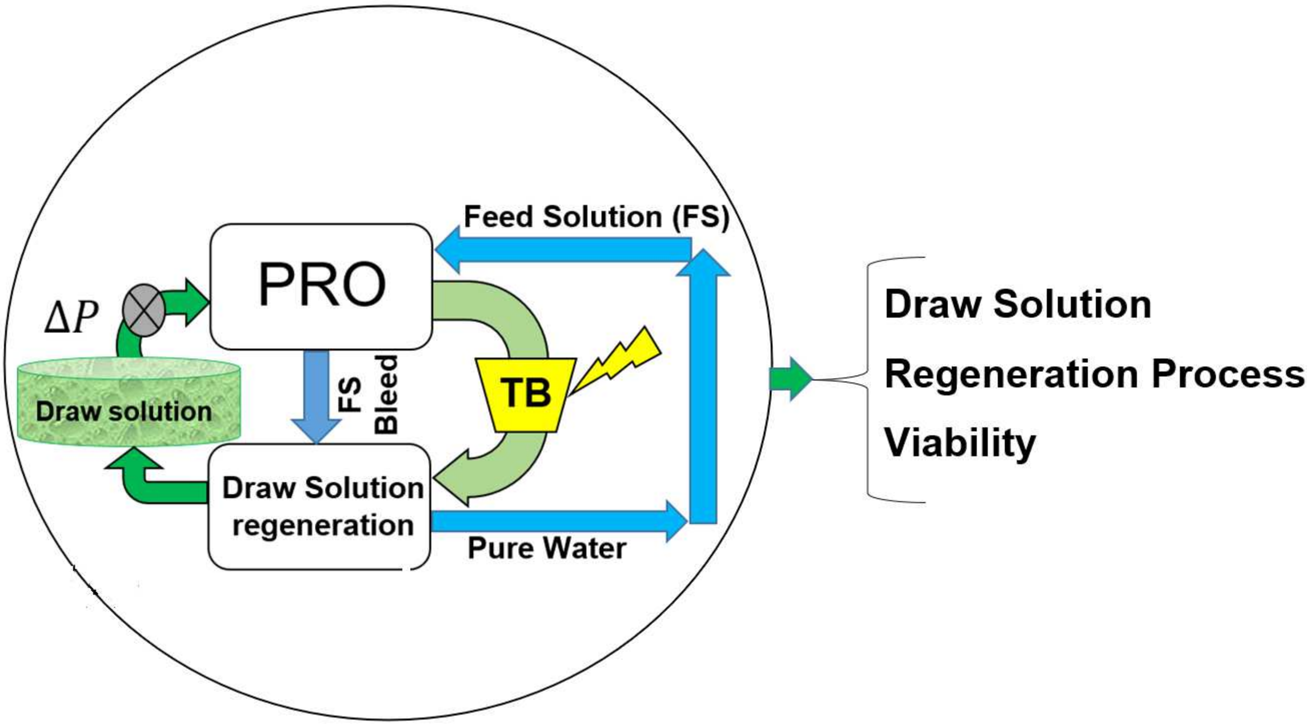
Phone: +1 5148482424, Ext. 7925

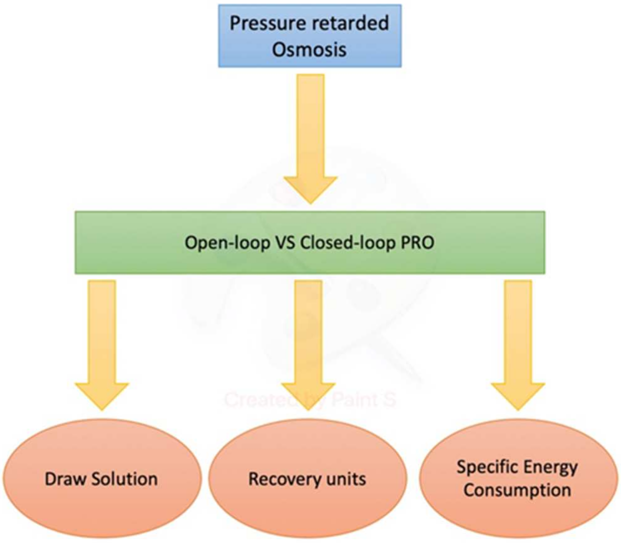
Abstract

Pressure-Retarded Osmosis (PRO) is an osmotic process that has been used to harvest energy from salinity gradients using a semi permeable membrane. A comparison between open-loop PRO (OLPRO) and closed-loop PRO (CLPRO) was made regarding their performance and costs. In CLPRO, where the diluted draw solution is re-concentrated in the regeneration system to be reutilized in the process, has recently received an intensive focus as the most viable configuration for a standalone power plant. The choice of the PRO draw solution in CLPRO is crucial to garner a high osmotic pressure as the key for the feasibility of the process. In this review, the draw solutions are critically evaluated in the literature in terms of energy output as well as the method of regeneration used to recirculate them. A set of practical criteria has been suggested to appraise the adequacy of the solution for CLPRO application. It was concluded that $\mathrm{NH_3 - CO_2}$ theoretically can produce 170 $\mathrm{W/m^2}$ of power density. Inorganic draw solutes such as NaCl can generate high power density up to 87 $\mathrm{W/m^2}$. Organic draw solutes with their remarkably low reverse salt flux (RSF) have promising potential for future application in PRO. Similarly, the regeneration systems of the diluted draw solutions have also been reviewed and discussed. How the energy consumption of the regeneration process affects the feasibility of CLPRO is explained. For the specific case of osmotic heat engines (OHEs), when the energy of the regeneration process is supplied by heat waste, the range of applicability of the heat waste in CLPRO in terms of efficiency is defined and compared to Organic Rankine Cycle (ORC). The results showed that CLPRO has better efficiency than ORC for temperatures T < 80°C, which makes it a promising process or low-grade heat energy recovery. In addition, a PRO-RO hybrid system coupled with solar power can reduce the net specific energy consumption (SEC) to 0.39 $\mathrm{kWh/m^3}$. The conditions that regeneration processes should operate under to make PRO viable are discussed in the last section. Overall, the study indicates the key factors for optimizing the performance of CLPRO process.

Keywords: Closed-loop PRO; Draw solution; Regeneration process; Specific energy; Cost effectiveness.

Graphical Abstract


Feed Solution (FS)
PRO
ΔP
Draw solution
FS Bleed
TB
Draw Solution regeneration
Pure Water
Draw Solution
Regeneration Process
Viability


Pressure retarded Osmosis
Open-loop VS Closed-loop PRO
Draw Solution
Recovery units
Specific Energy Consumption

Nomenclature

Abbreviation

CLPRO	Closed-loop Pressure retarded osmosis
CAPEX	Capital expenditure
CTA	Cellulose triacetate
ECP	External concentration polarization
ERD	Energy recovery device
FO	Forward osmosis
HMIS	Hazardous material identification system
ICP	Internal concentration polarization
LCOE	Levelized cost of electricity
LCST	Least critical solution temperature
LGH	Low-grade heat
MED	Multi-effect distillation
MD	Membrane distillation
MNP	Magnetic nanoparticles
MSF	Multi-stage flash
OHE	Osmotic heat engine
OLPRO	Open-loop pressure retarded osmosis
ORC	Organic Rankin cycle
PRO	Pressure retarded osmosis
RED	Reversible electrodialysis
RO	Reversible Osmosis
RSF	Reverse salt flux
SEC	Specific energy consumption
SG	Salinity gradient
SPS	Switchable polarity solvents
TB	Turbine
TFC	Thin-film composite

Symbols

S	Membrane structural parameter
t	Membrane tortuosity
ε	Membrane porosity
D	Diffusion coefficient
K	Solute resistivity

Subscript

C	Cold reservoir
D	Draw solution
F	Feed solution
H	Hot reservoir
s	Salt
RP	Regeneration process
w	Water

1. Introduction

Due to strong economic growth and rapid population growth, providing energy for human consumption will become a great challenge. Currently, conventional fossil fuels are the most prevalent source which are being used for providing energy in industrial and domestic sectors [1]. However, industrial emissions such as power plants, as well as transportation vehicles have grave harmful effects on health. Burning fossil fuels leads to emitting dangerous substances such as carbon monoxide, nitrogen oxides, sulfur oxide, methane, and volatile organic compound [2]. Moreover, these emissions are not only harmful to people but also to the environment. Aforementioned issues created unprecedented challenges for humankind to provide energy. To reduce the reliance on such limited sources, preventing climate change and global warming, and reducing health risks, researchers are investigating alternative sources of energy and processes, which are renewable and sustainable with minimal impact on the environment.

Renewable energy has a promising role in providing energy in the future. Solar, wind, biomass, geothermal, and hydroelectricity are the most “mature” sources of renewables [1]. However, these technologies have some challenges. In solar energy (1) low efficiency (2) high levelized cost of electricity (LCOE) (3) large footprint (4) storage challenges (5) availability [3], are some drawbacks of this technology. Also, wind energy is facing some problems such as (1) high material cost like rotor blades (2) noise and aesthetic pollutions (3) harmful impacts on local wildlife (4) wind forecasting models [4]. Application of geothermal and hydroelectric energies is limited to certain areas and is not universal. Among renewable energy sources, biomass has the largest contribution (72.3%) for providing energy worldwide with a potential capacity of 33,000 EJ [5]. However, the technologies for harvesting this energy are still not productive and cost-effective. In fact, the production cost that ranged between 0.13 USD to 0.99 USD/L, is still high compared to the cost of electricity production (0.03 USD – 0.24 USD/kWh) [5].

Another potential source of clean energy is generated from salinity gradients (SG) [6]. SG energy can be harvested as electricity utilizing promising technologies such as pressure retarded osmosis (PRO) and reversible electrodialysis (RED). PRO has been shown to be more efficient compared to RED for extracting power from SG [7]. In terms of energy conversion efficiency (the ratio between the generated energy and the energy input), SG shows better efficiency compared to other renewable energy sources [6,7] as shown in Table 1, despite the various energy losses that occur during the energy conversion process. In PRO, there are two types of standalone systems based on their configuration: open-loop (OLPRO) and closed-loop (CLPRO). An open-loop (OLPRO) system is based on the mixing of relatively fresh water with saline water, where both water streams are discharged. On the other hand, in the CLPRO configuration, both streams exiting the PRO membrane modules, go to the regeneration process and then, are circulated back into the process.

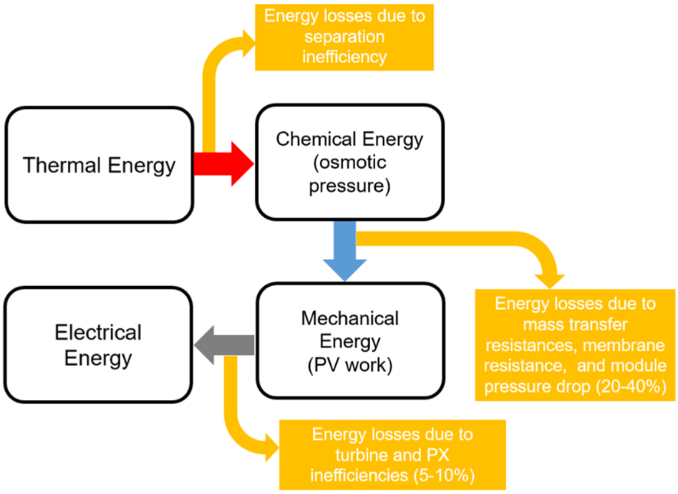


Figure 1: Energy losses during the conversion of SG to electrical energy.

Table 1: Energy conversion efficiency of salinity gradient in comparison to other renewable energy sources

Renewable energy sources	Energy conversion efficiency (%)	References
Solar PV	22 - 50	[8]
Geothermal	10 - 17	[9]
Wind	35 - 45	[10]
Tidal	70 -80	[11]
Hydro Power	60 – 90	[12]
Salinity Gradients	20 - 55	[16]

Several studies have investigated CLPRO systems. Straub et al. [13] investigated the power density, which can be obtained by using the hypersaline draw solution. They used 3 M NaCl as the draw solution under applied hydraulic pressure of 48 bars and demonstrated an unprecedented power density up to 60 W/m^2. However, the used commercial membranes cannot withstand an applied pressure of more than 48 bars. So, the full power density cannot be obtained. Altaee et al. [14] modeled a dual-stage CLPRO and compared it with conventional (single-stage) PRO using multi-effect distillation (MED) and waste heat as the regeneration process. It is stated that the overall efficiency of the process increases by 18% by adding the second stage, but the capital cost of the system, especially membrane costs, is high. Lin et al. [15] conducted a systematic analysis of an osmotic heat engine (OHE) consisting PRO for generating electricity and membrane distillation (MD) for the diluted draw solution regeneration. The thermodynamic limit of energy conversion efficiency and significant factors that control system performance is examined. According to the results, the MD process's efficiency, especially in applying low grade heat (LGH), is not high enough and needs further improvements to be economically viable and practical for the regeneration section of PRO. However, these tests were performed using NaCl-based draw solutions. Other studies were conducted with engineered draw solutions that showed better performance and energy output than the NaCl draw solution in small-scale applications [22,26].

In this review, the choice of draw solution for CLPRO process is examined. First, a brief comparison between CLPRO and OLPRO is presented, followed by a summary of draw solution chemistry and regeneration schemes. The review covers studies published between 2007 and 2020 that are related to closed-loop PRO, where experimental and simulating findings are thoroughly discussed along with the methodology of each study. Based on these studies, insights for the future of CLPRO and new opportunities for research are provided. Criteria for the choice of draw solutes and recovery processes are suggested and discussed referring to recent findings, as well as the energetic and economic evaluation of the CLPRO processes. Then, as a real case discussion, the efficiency of OHEs to harvest energy from LGH is studied in comparison to the organic Rankine cycle (ORC). Finally, limitations and future work are discussed

2. Closed-loop versus Open-loop Pressure Retarded Osmosis

CLPRO and OLPRO, both illustrated in Figure 2, are the two main configurations for hydraulic pressure-based salinity gradient power. Despite some common elements and overall operating principles, the two designs present substantial differences in terms of operation, draw solution types and sources, power production potential, economics, and environmental impact. In OLPRO, natural saline sources are generally the primary origin of the draw solutions, such as seawater and hypersaline lakes [17,18]. For the feed solution source, the majority of studies have considered freshwater [16,13], wastewater [16,20], or even seawater [16,17] as a low concentration feed. OLPRO can be located where these feeds and draw solutions are both abundant and within close proximity to each other. In CLPRO, the feed and draw solution are continuously regenerated using heat or another power source. CLPRO requires this energy to re-concentrate the draw solution and remove water, which can then both be remixed in a conventional PRO process. Illustrated in Figure 2, this separation produces pristine feed and draw solutions. Unlike with OLPRO, the system designer can choose precisely which draw solution they want to use, giving flexibility in design based on available energy sources at a particular location.

In OLPRO, not only pre-treating both feed and draw solutions, but also the energy consumption due to transporting streams from natural or industrial sources to the PRO plant highly affects cost. Also, the risk of membrane fouling is higher in an open-loop system which leads to more membrane cost. The OLPRO plant location is limited to places close to the inlet streams and thus cannot be applied far from these sources. However, CLPRO can operate with more flexibility and with minimal footprint, except for the which must be close to the low-grade heat source to recover the diluted draw solution. In CLPRO, there is no discharged streams and technically, all the outlet streams (diluted draw and concentrated feed solutions) are sent to the recovery unit and circulated back to the process. Thus, compared to the open system it is more environmentally friendly. However, in OLPRO, the outlet streams are discharged, especially when natural sources are used as the streams. Overall, CLPRO seems to be more practical and the best option for the applicability of PRO. Table 2 briefly describes the general performance of the two PRO configurations. To make it feasible, two parameters should be well-controlled and minutely selected in CLPRO: the draw solution and the regeneration process.

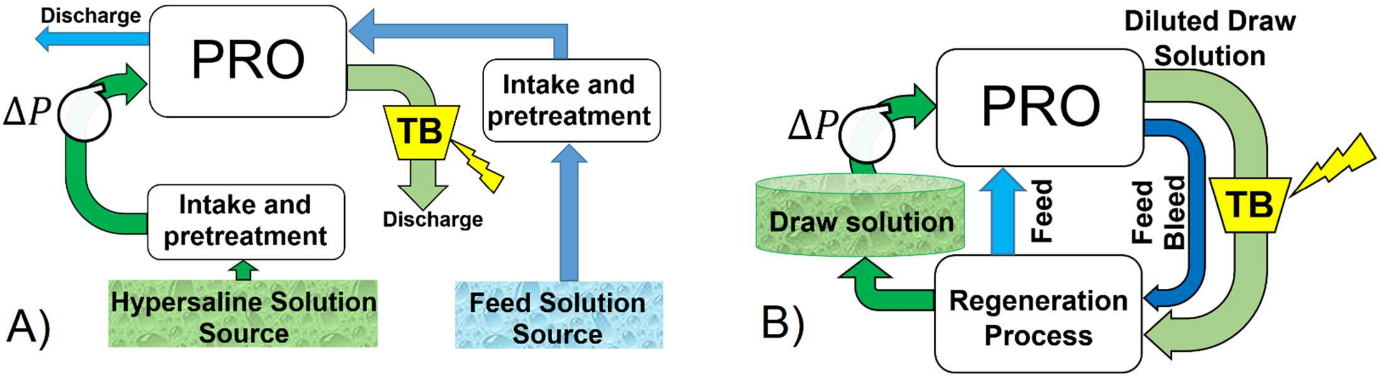


Figure 2: Schematic of (A) open –loop PRO (OLPRO) and (B) closed-loop PRO (CLPRO) for energy harvesting from solutions with high osmotic pressure. In CLPRO both feed and draw solutions are regenerated and recirculated to the process using a downstream separator. OLPRO generally requires intake and pre-treatment steps for both feed and draw solutions. TB refers to the turbine.

Table 2: Advantages, disadvantages and challenges for CLPRO and OLPRO

	Energy production	Operation	Cost	Environmental impact
CLPRO	Advantages • Operates at higher osmotic and hydraulic pressures for higher power density • Can generate more power per module and in a smaller system footprint than other PRO systems Disadvantages • Draw solution regeneration requires thermal or equivalent energy source • Draw recovery adds an energy efficiency loss • Overall low Carnot efficiency	Advantages: • Can operate wherever waste heat is available • Has no liquid waste or intake Challenges: • Relies on effective capture of waste heat for draw solution regeneration • Must have components that can handle operating pressures to maximize power production and justify installation • System components must be tolerant to the draw solution chemistry • Draw solute leakage to the feed must be recovered to ensure closed loop operation	Advantages: • High power density can lead to smaller systems with smaller footprint • Draw solution is 100% recycled. The solution does not need to be replaced, unless it ages or spoils • Better revenue from higher power density system using heat that would otherwise be discarded Challenges: • Specialty, high pressure and chemically tolerant components may be needed to accommodate higher pressures that make CLPRO relevant • Exotic draw solutes may constitute an expensive up-front cost • Draw solution regeneration process adds CAPEX	Benefits: • Capture of waste heat that would otherwise be discarded • CO_2 life-cycle offsets from conventional electricity generation • No waste streams Challenges: • Membrane and draw solution life-cycle must be considered based on lifetime of the plant
OLPRO	Advantages • Draw solution does not require energy • Disposal, rather than recycling, of solution mixture Disadvantages • Generates limited power because of osmotic and hydraulic pressure limitations to all but the most naturally occurring brine solutions (and these natural brines often occur in arid places with little freshwater) • Net power production is limited by power consumption for extensive pretreatment and pumping costs for the intake and outfall	Advantages: • Naturally occurring draw solution • Extensive experience with intakes and outfalls of saline and freshwater sources Challenges: • Natural water requires extensive pre-treatment to prevent fouling, leading to complex systems with more maintenance and failure points • Geographically limited to locations where freshwater and saline water mix naturally • Permitting challenges for intakes and outfalls	Advantages: • Naturally occurring salinity gradients offer basic energy source for “free” • Lower operating pressures enable use of off-the-shelf components for systems Challenges: • Pretreatment system incurs significant CAPEX and OPEX costs and power losses • OLPRO systems have yet to yield net positive power production • Low energy generation potential makes ROI targets challenging to meet.	Benefits: • Capture of energy of mixing that would otherwise be lost • CO_2 life-cycle offsets from conventional electricity generation • May offer a means of reconstituting inland saline seas Challenges: • Intake and outfall environmental impacts • Disruption of delicate estuary environments • Requires use of freshwater or lower salinity waters in arid regions • Pretreatment chemical use and disposal life cycle • Membrane life cycle costs

3. Draw solutes

3.1 Criteria for draw solute selection

The draw solution is a crucial parameter in the PRO process since it controls several factors, such as the driving force and salt leakage which affects the process performance directly, and consequently, the generated power density. Achilli et al. [21] proposed a procedure for the selection of suitable inorganic draw solutions, including a desktop screening process, followed by a laboratory and modeling analysis. According to his work, a suitable draw solute must have the following criteria: (1) soluble in water, (2) be solid at ambient temperature and pressure, (3) not toxic, nor a code below 2 within the Hazardous Materials Identification System (HMIS), with 0 denoting minimal danger and 4 representing a severe or lethal hazard, (4) produces an osmotic pressure more than 1 MPa (145 psi) at saturation concentration, and (5) has a specific cost, i.e., the cost of solutes to produce one liter of draw solution which can generate 2.6 MPa (406 psi) of osmotic pressure, and at a cost of less than 10 USD/L. However, requiring a non-toxic draw solution, especially for a CLPRO, is not a requirement as it is not used for human or animal consumption. As for the cost, this value was not suggested based on existing processes or developed cost estimation. Also, even if the draw solution price is relatively high, it might have a slight impact on the overall energy cost especially in the case of CLPRO when the solute is recoverable.

Moreover, there are some other important criteria that should be taken into consideration such as (a) solutes solubility, (b) solute flux tolerance, (c) viscosity, (d) recoverability of the draw solution, (e) solute diffusivity, (f), and (g) compatibility with the membrane. High solubility has a direct relationship with the generated osmotic pressure. The trans-membrane osmotic pressure difference is the driving force in PRO. Therefore, higher solubility leads to better osmotic performance. Also, the solute must have minimal reverse salt flux (RSF). As not only diffusing solutes from the draw side into the feed side leads to reduction in effective driving force across the membrane, the trace of the draw solute in the feed side also may exacerbate fouling. Moreover, the draw solution's viscosity at high concentrations is an important factor. High viscosity may affect the hydrodynamics at the surface of the membrane (low mass transfer) due to the laminar flow, lead to more energy consumption for pumping, severe external concentration polarization (ECP) in the draw side, and reduction in particle dissociation that lowers water flux. In terms of cost effectiveness, the diluted draw solution must be re-concentrated at a competitive cost and energy-efficient way. The regeneration stage is the most crucial part of the system's total energy consumption. The extracted energy from the process must outbalance the required energy in the regeneration system, and it is the main factor in indicating whether the process is applicable or not.

3.2. Categorization of draw solutes described in the literature

Draw solutes can be categorized by their physio-chemical properties, and many research groups have worked on optimizing draw solutions in PRO. Table 3 demonstrates a list of the main types of draw solutions investigated in the literature for CLPRO application, categorized by their generated osmotic pressure and power density, the method of their recovery, and their advantages and drawbacks as the draw solution in PRO. In the following sections, these draw solutes are defined, and their advantages and drawbacks are discussed. Also, their performance under the closed-loop system is critically discussed. The draw solutes in the literature are categorized into gaseous and volatile compounds, inorganic, organic, and functionalized nanoparticles.

Table 3: Summary of potential draw solutes described in the literature in stand-alone closed-loop PRO systems

Draw solutes	Osmotic pressure /Concentration	Water flux /Power density	Regeneration	Advantages	Drawbacks	Ref
KCit	42 bar/0.62 M	11.12 W/m^2	MD	* High osmotic pressure, water flux, and power density * Low RSF	* high viscosity at high concentrations	[22]
CaAc	42 bar/1.22 M	12.03 W/m^2	MD			
KOxa	42 bar/0.55 M	12.45 W/m^2	MD			
KAc	42 bar/0.97 M	12.70 W/m^2	MD			
NH_4Ac	42 bar/1.32 M	13.06 W/m^2	Thermolytic			
NH_4C	42 bar/0.58 M	13.55 W/m^2	Thermolytic			
NH_4F	42 bar/0.91 M	13.70 W/m^2	Thermolytic			
KF	42 bar/0.99 M	13.85 W/m^2	MD			
NaGly	42 bar/1.07 M	14.36 W/m^2	MD			
NaP	42 bar/1.01 M	14.56 W/m^2	MD			
CaP	42 bar/0.87 M	14.67 W/m^2	MD			
NaCl	42 bar/0.93 M	10.21 W/m^2				
NH_4HCO_3	42 bar/1.03 M	9.57 W/m^2				
$Na_5[Fe(C_6H_4O_7)_2]$	58 bar/1 M	48.6 LMH/16.2 W/m^2 (applied pressure: 12 bar)	Precipitation	High osmotic pressure and power density at low concentrations	High viscosity at high concentrations	[23]
NaCl	1.5 M (20 °C) 1.5 M (40 °C)	8.8 ± 1.0 W/m^2 18.0 ± 2.3 W/m^2	Thermal	Higher water flux and power density due to temperature increase	Increase in RSF	[24]
LiCl − methanol	2 M 3 M	37.8 LMH/31.3 W/m^2 47.1 LMH/72.1 W/m^2 (theoretical)	MD	More efficient regeneration, better performance	* Difficult to incorporate with OHE Due to high volatility of methanol * Methanol is more expensive than water	[25]
$NH_3 - CO_2$	191.6 bar/4.6 M (applied pressure: 101.3 bar)	170 W/m^2 (theoretical)	Distillation	High solubility, low molecular weight, high diffusivity, completely removeable (60 °C), produces high osmotic pressure	High RSF, volatile	[26]
$CaCl_2$	174 bar/1.6 M		MD	High solubility, High pressure osmosis	High RSF for inorganic draw solutes	[27]
HCOONa	174 bar/4.1 M	30 LMH/17.6 W/m^2				
KBr	174 bar/3.2 M					
LiBr	174 bar/2.2 M					
LiCl	174 bar/2.6 M					
$MgCl_2$	174 bar/1.5 M					
$Na(C_2H_5COO)$	174 bar/4.1 M	9.5 LMH/11.2 W/m^2				
NaCl	174 bar/3.0 M					
NaCl	1 M	14.88 W/m^2	MD	High solubility, High pressure osmosis	High RSF	[28]
$MgCl_2$	0.67 M	10.2 W/m^2				
$MgSO_4$						
NaCl	4 mol /kg		MD	-	-	[15]
NaCl	3 M	87 W/m^2		High power density		[29]

3.2.1. Gases and volatile compounds

McGinnis et al. [26] assessed theoretically OHEs and their potential for power generation and introduced ammonium-carbon dioxide as a novel draw solute for PRO. $\mathrm{NH_3 - CO_2}$ is highly soluble in water, thus producing high osmotic pressure. However, it has a high diffusion constant, causing high RSF and severe water flux decline. Ammonia is a nutrient and may promote biofouling on the feed side of the membrane [30]. McGinnis et al. [26] stated that this draw solute at 4.6 molar which generates 199.6 bars of osmotic pressure, can theoretically produce 170 $\mathrm{W/m^2}$ under 101.3 bar applied pressure. Furthermore, by increasing the crossflow velocity which leads to reduction in ECP, or using a higher applied pressure, higher power densities are achievable. Based on modelling of OHE with increased crossflow velocities (5 m/s in a 0.05 cm high flow channel), the author concluded that the power density would be increased by almost 61% (from 170 $\mathrm{W/m^2}$ to 274 $\mathrm{W/m^2}$) compared to the system used in their study (0.46 m/s in a 0.3 cm high channel). In the matter of higher applied pressure, increasing the applied pressure from 101.3 bars (100 atm) to 202.6 (200 atm) could result in 47% more power density. However, higher crossflow velocities and applied pressures will result in higher energy consumption and high-performance equipment. Therefore, capital and operation costs should be optimized in the process design. Although, increasing the draw solution concentration can result in higher PRO performance, it increases the RSF which leads to lowering the effective driving force in the system and increasing energy consumption in the regeneration process. This draw solution can be separated efficiently using waste heat. The unavailability of commercial membranes that are sturdy enough to handle high applied pressure (more than 50 atm), is still a critical issue. The availability of membranes with higher mechanical strength and salt rejection plays a vital role in process viability.

3.2.2. Inorganic draw solutes

The most common inorganic draw solutes are monovalent salts, and more precisely, NaCl. These salts have some advantages such as: (a) they can produce a relatively high osmotic pressure with high water flux, (b) at high concentrations they have low viscosity, (c) they have high diffusion coefficients due to their small size, reducing the effect of ICP, (d) they can be separated easily through thermal processes, and (e) they are inexpensive and available in large quantities. However, their small size leads to high RSF, which causes flux reduction and lowers PRO performance. Also, their reverse flux can promote organic fouling on the feed side [30]. Moreover, multivalent salts outperform monovalent ones. These ions have less RSF due to their larger hydrated radius and produce higher osmotic pressures with the same molar concentrations [21].

Straup et al. [13] investigated 3 M NaCl as the draw solute and claimed an unprecedent power density of 59.7 $\mathrm{W/m^2}$, with an impressive water flux of 44.5 LMH using an HTI TFC membrane, which withstood 48.3 bars of hydraulic pressure. Based on osmotic pressures simulated by the OLI software, they predicted higher power densities than can be achieved using this draw solution. It is illustrated that with the availability of more robust commercial membranes, under 100 bars of applied pressure, the power density could increase to 75 bars. This study showed flow channel and spacer design, and suitable membrane availability allow higher applied pressure which results in higher power densities. Anastasio et al. [24] investigated the impact of draw solution (NaCl) temperature on power density under conditions similar to OHE. As the temperature increases, the water flux increased due to enhanced mass transfer, which results in a higher power density. At 1.5 M NaCl, by increasing temperature from 20 ℃ to 40 ℃, the resulting power density increases from $8.8 \pm 1.0\ \mathrm{W/m^2}$ to $18 \pm 2.3\ \mathrm{W/m^2}$ at 20.7 bars. However, elevated temperature will lead to increased salt flux which is caused by increased salt diffusivity and reduced dilutive ECP, resulting in water flux decline and ICP promotion [7]. Although, higher temperature causes better PRO performance, increased RSF will result in higher required energy in the re-concentration of the diluted draw solution which decreases process energy efficiency. This required energy is provided by available waste heat. This heat might be in large amounts but harnessing and using it in the regeneration unit comes with costs. Therefore, a trade-off should be made in this case.

Hickenbottom et al. [31] also used a NaCl solution as the draw solution. They studied different FO membranes which were commercially available and evaluated their performance in PRO, and assessed the effects of feed and draw streams spacers and cross-flow velocities on generated power density. Three polyamide-based thin-film composites (TFC), and one HTI cellulose triacetate (CTA) were used in the experiments. According to the results, HTI TFC showed the highest mechanical strength, power density (15.5 $\mathrm{W/m^2}$ under 35 bars), selectivity, and lowest salt diffusion among the tested membranes. This membrane was tested under different applied pressures. Although, it can withstand pressures up to 48 bars (~ 700 psi), under pressures more than 35 bars (~ 500 psi), the support layer becomes thinner which causes porosity reduction and tortuosity increase. This phenomenon leads to higher membrane parameter structure which promotes ICP and power density reduction. Three types of spacers were investigated in this study: 20-channel tricot, 35-channel tricot, and extruder mesh spacer, with different orientations. The highest power density was achieved when the feed spacer was a two 20-channel tricot positioned at 45° to the feed flow. The reason behind it is due to increased mass transfer at the membrane interface, and consequently, decreasing the concentration polarization. For the draw side, an increase of 76% in power density was observed when a one 20-channel tricot spacer and one extruded mesh spacer were used compared to the two extruded mesh spacers. It is concluded that by using these two configurations for the feed and draw sides, the power density of 22.6 $\mathrm{W/m^2}$ can be achievable in comparison with when there are no spacers under 35 bars applied pressure. Moreover, they concluded that feed cross-flow velocity has more impact on power density than the draw side. Mass transfer in the feed side affects process performance more. By increasing the feed and draw solutions cross-flow velocity from 2.6 cm/s to 13 cm/s, the power density increased 48% and 36%, respectively.

Shaulsky et al. [25] investigated an organic solvent, methanol, compared its performance to water for LiCl draw solute. LiCl is highly soluble in polar solvents due to the existence of a strong ion-dipole force between Li^+ and polar solvents. It is depicted that the membrane solvent permeability for water (37.78 LMH) is ~55% more (26.25 LMH for LiCl-methanol) when the draw solution concentration is 1 M. Higher polarity of water molecules cause higher permeability due to more sorption of these molecules on the membrane polyamide layer. On the other hand, the solute permeability in methanol is almost half of the water solution. It can be deduced that Li^+ and Cl^- ions form a larger solvated ion size in methanol, so, the solute flux is less. Moreover, the membrane structural parameter (*S*) in the LiCl-methanol case is lower. The reason for this phenomenon is that the methanol surface tension is higher than water. Therefore, the support layer of the membrane will be wetted better resulting in lower *S,* and less severe ICP. Although LiCl-methanol has a lower water flux than LiCl-water, the ratio of J_w/J_s is higher for methanol solutions, thus, it performed more efficiently than water in terms of reverse diffusion. The predicted power density for LiCl-methanol solution at 3 M is 72.1 $\mathrm{W/m^2}$ which makes it a potent potential draw solution for PRO. To achieve this power density, an applied pressure of 114 bars is needed which is not possible with the currently available membranes. Methanol has lower enthalpy of evaporation than water. In terms of thermal separation and recovery, methanol is more efficient. However, methanol is highly volatile, and it is hard to incorporate with OHEs, and it is more expensive than water.

Hickenbottom et al. [27] assessed alternative inorganic and organic ionic salts for an optimized closed-loop OHE using MD as the regeneration process. The experimented draw solutions are listed in Table 3. Solutions with different concentrations, which produce 174 bars of osmotic pressure were used as the draw solution. Among the selected draw solutes, HCOONa had the highest (30 LMH and 17.6

W/m^2), and $Na(C_2H_5COO)$ had the lowest (9.5 LMH and 11.2 W/m^2) water flux and generated power density. The difference in water fluxes is caused by ICP (the same membrane was used in all experiments) which is related to different solute diffusivity and salt permeability (B). Analyzing RSF, HCOONa had the lowest solute flux (11.8 gMH) due to its large hydration diameter affecting salt permeability. Thermal efficiency (the ratio of water flux to total heat flux), is another criterion investigated by the author. It was determined that KBr had the highest thermal efficiency (94%), and $Na(C_2H_5COO)$ had the lowest one, due to its high heat capacity and low MD water flux. Working fluids with high thermal efficiency are the most suitable ones for OHE. The total electricity generation costs were calculated for each draw solution. The total electricity costs for $CaCl_2$ was the lowest, 1.65 USD per kWh. The MD membrane cost has the biggest contribution to the system cost. Since, $CaCl_2$ had low RSF, the MD membrane cost was low. Also, it had the highest net energy production, the lowest system costs, the second highest power density, and the second highest MD flux. Considering specific cost, RSF, power density, MD water flux and thermal efficiency, net power generation, and electricity generation cost, the author suggested that $CaCl_2$ is the best draw solute. Still, compared to other sources of renewable energies, energy generation cost for OHE is high. The introduction of more robust and selective membranes for PRO could be helpful in harnessing the full potential of this draw solution and resulting in reasonable electricity generation costs.

Gong et al. [28] investigated the transport of ions during PRO in order to find better draw solutes for an OHE. Three inorganic salts were selected for this study: NaCl, $MgCl_2$, and $MgSO_4$. Apparently, ions with a larger hydrated ion diameter have lower solute permeability because more water molecules surround ions with higher charge density, which leads to higher polarization. Smaller ions with higher valence have higher charge density, and consequently, higher larger hydration diameter. According to their results, $MgSO_4$ had the lowest power density due to its lower osmotic coefficient (φ) compared to the other two. The φ for $MgSO_4$ at 1 M is 0.525 which is much lower than 1.004 ($MgCl_2$) and 0.926 (NaCl) at 0.67 M [40]. Moreover, this draw solute showed poor mass transfer due to its higher viscosity and lower diffusion coefficient, resulting in more ECP on the draw side. $\mathrm{MgCl_2}$ generated more power density compared to NaCl because of its larger hydration diameter, which increases the membrane selectivity and reduces the effect of ICP. Overall, draw solutes with smaller hydrated ion sizes have higher water fluxes and diffusion coefficients, leading to an increased RSF, which promotes ICP. On the other hand, larger ions have lower RSF, but higher viscosity at the same concentration, promotes ECP. Therefore, trade-offs of these parameters should be considered to select the optimized draw solution with the best performance.

Moon et al. [29] investigated a new type of membrane in OHE for comparing its performance with other renewable technologies based on LCOE. Direct fluorination (converting C-H bonds to C-F bonds within a polymer) is used in a one-step gas-phase reaction to enhance the surface characteristics of the support layer. Fluorination of PBO-TFC increased the membrane hydrophilicity, resulting in a better rejection (R value) for the selective layer (increased from 90.7% to 95.9 ± 1.7 $\%$). Although the fluorination resulted in increased A and B values compared to other membranes, the significantly low structural parameter (S = 142 μm) is promising for better PRO performance. Using 3 M $\mathrm{NaCl_{aq}}$, PBO-TFC-F5 showed way more transmembrane water flux (116.2 – 147.2 LMH) than the unfluorinated PBO-TFC (33.9 – 86.0 LMH). Obviously, the reduced ICP is the reason behind this result due to super hydrophilicity (high R) nature and a crumpled selective layer (high A) of PBO-TFC-F5. Also, considering that the mechanical strength of PBO-TFC cannot be operated at pressures above 21 bars, implementing 1 M and 3 M NaCl draw solutions, PBO-TFC-F5 generated 17.6 $\mathrm{W/m^{-2}}$ and 87.2 $\mathrm{W/m^{-2}}$ power density under hydraulic pressure of 18 and 27 bars, respectively. For the first time, a research group claimed this high power density in CLPRO, which outperforms HTI-TFC membranes (6-8 times better performance) under the same experimental conditions. Furthermore, using feed solutions with higher concentrations (industrial applications) from DI water to 0.5 M NaCl (seawater), increases the power density ratio of

PBO-TFC-F5 to HTI-TFC even more. This modified membrane has a much lower S value of the support layer compared to commercially available HTI-TFC, and consequently, much less severe ICP happens in its support layer. This study shows promising results for feasibility and practical implementation of PRO at industrial scale.

3.2.3. Organic draw solutes

Organic draw solutes have two main advantages compared to inorganic salts: (1) the former's hydrated ion sizes are larger than the inorganic salts, resulting in better membrane selectivity. Therefore, their RSF is much lower, and less water flux decline happens during the operation. Additionally, due to lower RSF the required energy for feed solution regeneration, and replenishment costs are lower as well. (2) Organic draw solutes can be engineered and tailored to change their properties and increase their PRO-related performance. However, their larger size leads to lower diffusion coefficients (D), hindering solute flow within the membrane. This phenomenon, increases the solute resistivity (K) parameter of the membrane and promotes ICP ($K = \frac{\tau t_s}{\varepsilon D}$). They also have high viscosity at high concentrations which increases required pumping energy and reduces process efficiency. In later sections, different types of organic draw solutes are explained, and scientific work related to the use of these solutes in CLPRO is reviewed.

a. Simple organic ionic salts

Islam et al. [22] explored organic ionic salts (Table 3) in closed-loop PRO and compared their performance with commonly used inorganic salts such as NaCl and $\mathrm{NH_4HCO_3}$. They concluded that at concentrations that generate equal osmotic pressure (42 bars), these salts produce more water flux and power density compared to inorganic salts due to their lower RSF which was caused by the larger sizes of their hydrated ions. The salt fluxes of organic draw solutes were between 0.0325 $\mathrm{mol\,m^{-2}\,h^{-1}}$ and 0.0854 $\mathrm{mol\,m^{-2}\,h^{-1}}$ which are remarkably lower than NaCl (0.854 $\mathrm{mol\,m^{-2}\,h^{-1}}$) and $\mathrm{NH_4HCO_3}$ (0.952).

Having a C-O double bond in their structures which makes the polarization of the anion better in their aqueous solutions, could be the reasonable explanation. Thus, more hydration of the solute ions happens due to the polarized nature. Also, it was observed that by increasing the draw solution concentration, the RSF remains almost constant for these solutes. In terms of water fluxes, obtained water fluxes for organic salts ranged between 19.02-25.09 LMH, while for NaCl and $\mathrm{NH_4HCO_3}$, permeate fluxes were 17.50 and 16.40 LMH, respectively. Peak power density values (11.10-14.64 $\mathrm{W/m^2}$) for these salts were 8.72% to 43.39% and 15.99% to 52.98% higher compared to NaCl (10.21 $\mathrm{W/m^2}$) and $\mathrm{NH_4HCO_3}$ (9.57 $\mathrm{W/m^2}$) at 21 bars of applied pressure. Higher RSF for inorganic salts is the most critical factor causing the inferior performance of these salts. In conclusion, organic salts showed better performance regarding their minimal RSFs, higher water fluxes, and generated power densities than their inorganic counterparts. In terms of power density, while considering thermal distillation as the regeneration process, authors concluded that $\mathrm{NH_4C}$ holds the highest potential for PRO application. However, the unavailability of membranes with enough mechanical strength, still remains a critical issue for harnessing the full potential of these draw solutes. At their solubility, these solutes produce osmotic pressures ranging from 70-1300 bars. Excluding CaAc, the maximum generated power density can be achieved when the applied pressure is half of the transmembrane osmotic pressure difference. The most robust commercially available membrane can tolerate up to 48 bars [13]. Furthermore, this paper did not investigate the comparison of the performance of these draw solutes with inorganic ones at high concentrations such as 3 M. While at high concentrations, inorganic draw solutes result in higher salt flux. Organic solutions will have much more viscosity which increases required pumping energy and mass transport resistance. Thus, choosing the draw solute with better performance at high concentrations remains unresolved.

Switchable polarity solvents (SPS) are another category of simple organic draw solute. The SPSs can be separated from water through a reversible transition from water soluble to water insoluble by adding

CO_2 [32,33]. The draw solute can dissolve into the miscible state using heat from low grade sources and N_2 gas to strip off the CO_2. Due to these properties, SPSs can be considered as great draw solutes in PRO.

b. Hydro-acid complexes

Hydro-acid complexes consist of metal(s) and ligand(s) parts that, when the metal center bonds to the ligand, the configuration of it expands. According to the required function, they can be manipulated by changing either metals or ligands. These compounds are highly soluble in water, large enough to have negligible salt diffusion, easily regenerated, and provide a high osmotic pressure.

Han et al. [23] demonstrated a novel closed-loop PRO for sustainable osmotic energy production using $Na_5[Fe(C_6H_4O_7)_2]$ (Na-Fe-CA) hydro-acid complex as the draw solution. In all experiments, the TFC PRO hollow fiber membrane with a polyamide selective skin was used. In this study, the draw solute consists of a ferric complex with hydroxyl acids of citric acid (CA) as ligands. It was observed that this draw solute has a better performance compared to conventional inorganic draw solutes, such as NaCl, at the same molar concentration. Higher osmotic pressure of 58 bars at 1 M was achieved due to its hydrophilic groups with multi-charged anions, while for NaCl it is only 47 bars at the same concentration. The difference in osmotic pressure becomes even more at higher concentrations because CA ligands improve water solubility, generating higher water fluxes. At 1 M and 12 bars applied pressure, the obtained water flux was 48.6 LMH which is slightly more than NaCl (41 LMH). Based on the water flux and 12 bars applied pressure, the produced power density was 16.2 W/m^2 and 13.7 W/m^2 for Na-Fe-CA and NaCl, respectively. Also, the hydro-acid showed negligible salt flux due to expanded octahedral structure. For NaCl, the RSF increased significantly by increasingapplied pressure from 10.5 gMH at 0 bar to 32.7 gMH at 12 bars. On the contrary, Na-Fe-CA showed remarkably low salt diffusion ranging between 0.44 gMH at 0 bar to almost 3.0 gMH at 12 bars. Consequently, solute loss for the proposed system is minimal which reduces the replenishment cost compared to conventional draw solutes like NaCl. The diluted draw solution was re-concentrated by adding ethanol for the precipitation of Na-Fe-CA. So, it can easily re-concentrated and it is not energy consuming. The regenerated draw solution used again in the process and in terms of J_w, J_s, and power density, the results were almost the same as the primary draw solution proving that the diluted raw solute was recovered almost completely. On the other hand, Na-Fe-CA has a much larger molecular weight and a bulkier structure than NaCl. Therefore, at concentrations more than 1 M, for example 1.5 M, its viscosity is much higher (almost 5.5 times more) compared to NaCl, which increases the energy consumption in pumping and promotes ECP.

3.2.4. Functionalized nanoparticles

Recently, nanoparticles functionalized with hydrophilic groups have received significant attention, especially magnetic nanoparticles (MNP) due to their high surface-area-to-volume ratio and relatively easy separation from water by utilizing magnetic separation. MNPs are materials consisting of a magnetic core and a surrounding polymer shell. The magnetic separation can be achieved by using an external magnetic field due to the existence of nanoparticles in the magnetic core. The polymer shell not only decreases particle aggregation in the solution, but also enables surface modification for better osmotic performance. It seems that the particle agglomeration during the magnetic recovery remains the main problem in using MNPs, making the process economically not viable for long-term operation. Ling and Chung [34] investigated the use of ultrasonication and its effect on particle agglomeration. Although utilizing ultrasonication reduced nanoparticle agglomeration effectively, the magnetic properties were also decreased over time, limiting the total recovery by magnetic separation methods over several cycles.

4. Draw solution regeneration

In order to ensure sustainable production of electricity, diluted draw solutions must be re-concentrated periodically. The regeneration process has a crucial role in the energy efficiency of the whole system and the net energy generated in CLPRO systems. The recovery method must be energy efficient, cost-effective, and practical. It must not only re-concentrate the diluted draw solution completely (i.e. up to initial concentration of the draw solution), but also not allow salt leakage while producing permeate water with high quality. If none of these conditions are satisfied during the regeneration, the effective driving force will decline, lowering the efficiency of the whole process. A summary of relevant regeneration processes for recovery of diluted draw solutions investigated in literature is shown in Table 4.

4.1. Processes for draw solution recovery

4.1.1. Thermal separation

Research has indicated that thermal separation could be good a choice for draw solution recovery [35,36]. During this process, heat can be utilized to turn volatile solutes into gaseous form, which can then be separated from the solution. Water evaporates through the distillation process, and for thermo-responsive polymers, draw solutes precipitate at a temperature above the least critical solution temperature (LCST), allowing the separation via filtration to happen. Moreover, in switchable polarity solvents, heating removes the dissolved $\mathrm{CO_2}$ which causes draw solutes to shrink and release water [37].

McGinnis and Elimelech [38] investigated FO as a method of seawater desalination process and its required energy compared with other desalination processes such as multi-stage flash (MSF), multi-effect distillation (MED), and RO. They simulated thermal recovery of the diluted ammonia-carbon dioxide draw solution using single vacuum column and multi-stage distillation (MSD) for very low temperature (40 ℃) and high temperature sources, respectively. In a low pressure single column system, the diluted draw solution is exposed to LGH counter-currently and volatile $\mathrm{NH_3} - \mathrm{CO_2}$ mixture separates from water. Then, all gases were condensed under vacuumand removed with steam thermojet. In multi-stage distillation the same configuration as MSF and MED is used. At the top stage, a portion of feed water was vaporized by exposing to heat and then, the produced vapor was condensed in contact with second stage (lower temperature and pressure) on a heat transfer surface. This exposure to heat, vaporizes more feed water, and so on. In MSF and MED unlike MSD, both energy and material streams move in series, while in MSD, partitioned parallel streams of the diluted solution enter each column. Potable water is produced out of 0.5 M NaCl as the simulated seawater, with the recovery rate of 75%, and concentration of less than 1 ppm. The FO draw solution inlet and outlet streams are 5 M and 1 M, respectively, at 25 ℃. For measuring the thermal desalination efficiency, a term of gained output ratio (GOR) was used which is defined as the ratio of each kg of steam condensed to each kg of water produced. Typically, the estimated GOR values for MSF and MED are between 8-15 at temperature of 70-120 ℃ [39], while the calculated range of GOR for FO is between 4.4-20.2 for temperatures between 40-250 ℃. They concluded that using low temperature (40-44 ℃) for FO shows significant energy saving. Compared to MSF, low temperature multi effect desalination (LT-MED), MED using thermal vapor compression (MED-TVC), and RO, the saved energy is 85.1%, 73.8%, 79.2%, and 72.1%, respectively. In addition, the electrical energy consumption for current desalination process lies between 1.6-3.02 $\mathrm{kWh/m^3}$, while, for FO, due to benefitting from large recoveries and unpressurized fluid pumping, the electrical consumption is about 0.25 $\mathrm{kWh/m^3}$.

McGinnis et al. [26] investigated the separation of $\mathrm{NH_3-CO_2}$ from a diluted draw solution in PRO using a distillation column. This draw solute can decompose at temperature and pressure of 60 ℃ and 1 atm, respectively, and be removed from water up to less than 1 ppm. Then, this water was circulated back to the feed side as the working fluid. In modeling of OHE some assumptions were made regarding the efficiencies of the pressure exchanger and turbine, 95% and 90%, respectively. The efficiency of the OHE calculated considering thermal and Carnot efficiencies. Thermal efficiency is the ratio of the amount of power produced by the engine over the heat used for recovering and separation draw solution and working fluid. Carnot efficiency is defined as the ratio of the engine over Carnot engine which is the maximum theoretical efficiency. Thermal and Carnot (between 25 and 50 ℃) efficiencies of the OHE were calculated with the HYSIS® chemical simulator model and Aspen HYSIS®, respectively, considering the draw solution at concentrations ranging from 1 to 6 M (CO_2 basis). A distillation column, containing 30 theoretical stages, supplied heat at 50 ℃, bottom pressure and temperature of 10.62 kPa and 46.96 ℃, and top pressure and temperature of 10.54 kPa and 35.55 ℃ was considered as the recovery unit. When the effective driving force ($\Delta\pi - \Delta P$) approaches zero the maximum engine efficiency is achieved which was almost 16% of Carnot engine (probable operation efficiency lies between 5-10%). With increasing the draw solution concentration, and consequently the increase of osmotic pressure, water flux and power density will increase. However, using a more concentrated draw solution will increase the heat duty in the recovery unit as well.

It was concluded that the separation of NH_3 and CO_2 in the distillation column is inefficient. Some water vapor is also removed which consumes heat, and therefore, not all the supplied heat is converted to power generation. As the draw solution becomes more concentrated, the amount of vaporized water in distillation column also increases, and leads the separation to the equivalent state, which lowers the efficiency of separation and results in decreasing the overall efficiency of OHE. On the other hand, higher draw solution concentrations will lead to higher power densities, requiring less membrane area and therefore, less membrane cost. Thus, a trade-off should be made between membrane capital cost and engine efficiency. Moreover, in thermal recovery, when using waste heat, one of the most important factors that should be considered is the efficiency of the heat exchanger, which affects its area and the capital cost of the system for suppling heat. The system efficiency has a direct relationship with the temperature difference between supply and rejecting reservoirs. The lower the difference, the lower the thermal efficiency of the heat engine is, which results in an increased heat exchanger area and cost.

MD is a thermal membrane process, in which water transfers across the membrane due to the vapor pressure difference across a hydrophobic membrane. The feed solution is in direct contact with one side of the membrane and draw solutes are prevented from penetrating inside the dry pores of the membrane, allowing vapor molecules to pass through it. The feed solution is prevented from flowing into its pores by a hydrophobic membrane due to the surface tension forces. Due to the many advantages of MD, some research groups utilize it as the method for re-concentration of the diluted draw solution. MD can reject non-volatile feed draw solutes up to 100% theoretically [40]. In addition, MD efficiency is independent of feed water salinity, allowing the treatment of highly saline waters [40-42]. Membrane distillation also can use LGH, such as industrial waste heat, leading to a low energy requirement in the process [43,44]. However, this system has some drawbacks and limitations, such as: (1) low permeate flux compared to other separation techniques [45], (2) permeate flux decline due to temperature polarization, membrane fouling, scaling, and membrane wetting [45], (3) high heat loss by conduction, and (4) it is energy intensive because, although the pumping energy demand is similar to FO (0.25 $\mathrm{kWh/m^3}$) [38], the energy required to heat the water from room temperature to 50 ℃ is 29 $\mathrm{kWh/m^3}$. Many studies investigated OHE, trying to make it more feasible and practical using MD as the regeneration process for re-concentrating the diluted draw solution for PRO.

Hickenbottom et al. [46] performed a techno-economic analysis of OHE to recover low temperature waste heat. In this process MD is coupled with PRO to re-concentrate diluted draw solution utilizing LGH. They modeled an OHE system producing 2.5 MW of net power, the capacity of commercially available small-scale ORC (benchmark technology for this study) plants with twenty-year plant life, as the basis. In this modelling, 3 M NaCl was assumed as the working fluid in the PRO with operational pressure of 34 bars (~500 psi), and recovery rate of 15%. For MD, average water recovery of 6% based on the literature, and negligible membrane wetting were assumed. The temperature of feed and distillate streams were 70 and 30 ℃, respectively. LGH temperature (heat source) and cooling water system (heat sink) were assumed to be 80 and 20 ℃, respectively. Considering base-case (2.5 MW of net power), the capital and O & M costs were estimated to be 57.3 and 3.6 million USD/yr, respectively. It was found that electricity is generated at the cost of 0.48 USD per kWh, which compared with average electricity cost for wholesale U.S. grid energy (0.04 USD per kWh [48]) or the organic Rankine cycle (ORC) (0.08-0.13 USD per kWh [49,50]), is not competitive.

The reason for high levelized cost of electricity is due to the energy required for pumping, which accounts for 40% of the gross electrical power produced. According to this study, the system efficiency is less than 1% due to low operating temperature. However, by increasing the generated net power, PRO power density, and MD water recovery, the efficiency can be improved. Another study [15] demonstrated that the maximum theoretical efficiency is almost 9-10% when MD feed and distillate is 60 and 20 ℃, respectively, 4 M NaCl as the working fluid in PRO. Operating with a draw solution, generating high osmotic pressure at high applied pressure, increases the PRO power density significantly and reduces membrane area, leading to better efficiency. Also, increasing the partial vapor difference across the membrane (i.e. operating at higher feed and lower distillate temperatures) will increase MD water flux and system efficiency and decrease the required membrane area. In another study, different draw solutes were tested in OHE, trying to assess better draw solutions for better system performance [27]. It was concluded that a lower RSF can reduce the MD load on the feed side, lowering the required pumping energy and membrane area. Therefore, the draw solutes with high RSF, such as inorganic ones, are not appropriate for solute recovery using MD, unless PRO membranes with higher selectivity were developed. According to the sensitivity analysis that was done on the operating parameters in this study, increasing PRO water recovery from 15% to 40% can decrease the electricity cost by 20% (from 0.48 USD to 0.38 USD per kWh), and improve system efficiency by 32% (from 0.1% to 0.13%). Increasing MD water recovery from 6% to 30% has the same influence on electricity cost and system efficiency. In fact, electricity cost decreases 25% (from 0.48 USD to 0.36 USD per kWh), while efficiency will increase six-fold (0.1% to 0.6%). Increasing the operating pressure in PRO (up to 90 bars), significantly reduces electricity cost (0.48 USD to 0.21 USD per kWh), and triples system efficiency (0.1% to 0.3%). Although, increasing hydraulic pressure will increase RSF resulting in higher feed stream bleeding and more heat duty in MD, the benefit of increased power density outweighs its drawbacks. Surprisingly, although increasing MD feed temperature leads to reduction in electricity cost, system efficiency (ratio of output work to input heat) would increase because, more heat is needed to increase the feed's temperature to the desired level.

Finally, this analysis shows that by increasing system size from 2.5 MW to 25 MW, a 30% reduction in electricity cost would happen (from 0.48 USD to 0.34 USD per kWh). Considering the best case scenario resulted from the sensitivity analysis, while, the system size, MD and PRO recovery rate, and operation pressure in PRO are 25 MW, 30% and 40%, and 76 bars, respectively, electricity generation costs would decrease remarkably from 0.48 USD to 0.1 USD per kWh (system efficiency increases from 0.1% to 0.8%). In conclusion, with currently available membranes and advancements, continuous operating OHE is not comparable with other technologies especially, ORC. Availability of highly permeable and selective PRO membranes with high mechanical strength can be promising in the viability of OHE. Also, draw solutions which produce higher osmotic pressure and possessing minimal RSF, play crucial role in reducing PRO and MD membrane costs.

4.1.2. Physical separation (Reverse osmosis)

RO is a membrane separation process which operates under a hydraulic pressure which is greater than the difference in osmotic pressure across the membrane. In RO, water flows from the draw side into the permeate side. This process is commonly used as a desalination system for producing potable water.

Due to its high-water recovery and salt rejection rates, it can be utilized as a secondary process for recovering diluted draw solution as well. Several studies investigated the PRO-RO hybrid system [51-55]. The energy consumption of conventional SWRO using one pressure exchanger (PX) is 2.5 $\mathrm{kWh/m^3}$, while the thermodynamic minimum specific energy consumption (SEC) for this system under 50% recovery is ~1.1 $\mathrm{kWh/m^3}$[56]. This amount is higher than the energy produced by PRO. In fact, according to a previous study [51], closed-loop PRO can address only 50% of the SEC required in RO for seawater desalination when the water recovery is 50%. Furthermore, because of the membrane sensitivity and severe fouling, highly saline draw solutions cannot be used in this system. On the other hand, the hybrid RO-PRO system can be used for recovering required energy in RO via utilizing energy recovery device (ERD), as well as dilute the RO brine to sea water concentration to allow its discharge back to sea.

Wang et al. [52] investigated integration of RO-PRO system with double ERDs regarding specific energy reduction using temperature enhanced PRO. The concentration of feed and draw solutions are 0.01 M NaCl (municipal wastewater) and 1.2 M NaCl (RO brine), respectively. There are some parameters that affect the hybrid system SEC such as PRO permeate and operating pressure and temperature. The amount of PRO permeate has a direct relationship with power density and hence, with the whole system SEC. It is stated that in a situation that PRO and RO permeates are equal along with 50% recovery 30 bars of applied pressure in the PRO unit, the lowest SEC of RO-PRO system can reach 1.57 kWh/m^3. This amount deviates from the theoretical limit of SEC (1.14-1.20 kWh/m^3) mentioned in the literature [53,54], which is due to the SWRO capacity or low RO permeate flow (RO recovery assumed to be 50%). By increasing the capacity from 1500 to 150,000 m^3/d, SEC decreases 1.27 kWh/m^3. Likewise, increasing operating pressure from 30 to 35 bars leads to SEC reduction by 12.94% (from 1.27 to 1.11 kWh/m^3), resulting from the higher generated power density. However, increasing the operational pressure, will decrease the permeate flow significantly. On the other hand, higher temperature leads to higher water permeate due to improved membrane selectivity and transmembrane effective osmotic pressure. It was stated that when using 1.2 M NaCl (RO brine), increasing temperature from 25 to 50 ℃ while increasing applied pressure from 28.5 to 29 bars, the permeate flow will rise from 501.8 to 878.4 $\mathrm{m^3/d}$ and consequently, leads to power density increase (from 16.55 to 29.84 $\mathrm{W/m^2}$). This result implies that elevating temperature along with moderate pressure can improve water permeability and power density significantly. In terms of SEC, increasing temperature to 50 ℃ will result in SEC reduction by 17.93%.

Renewable energy sources such as wind and solar can be coupled with RO to compensate electricity demand in this system [57-60]. However, these sources are not always available (solar power at nights). Thus, another source of renewable energy should be integrated with this system. Some studies investigated PRO-RO hybrid system coupled with solar PV [61,62]. Tong et al. [62] proposed PRO-RO hybrid system coupled with solar PV in a large-scale plant with capacity of 5000 $\mathrm{m^3/d}$. In this system, solar energy is used to heat up the inlet streams in PRO to improve its performance by increasing the osmotic pressure. Tertiary wastewater (0.02 M) and seawater (0.61 M) are the feed and draw solutions in this system. In order to calculate the maximum amount of recoverable energy, PRO membrane or module inefficiencies were neglected. According to the presented results, when the PRO operating temperature (T_H) increases from 293.15 K to 353.15 K, the maximum generated energy (when the applied pressure is half of the osmotic pressure difference) that can increase from 0.183 to 0.221 kWh/m^3 because, higher temperature leads to higher osmotic pressure which results in higher power density. Increasing the operational pressure and RO recovery rate will increase the net SEC of the system, while elevating the temperature has the opposite effect. For instance, increasing temperature from 293.15 K to 353.15 K, considering the optimal value of applied pressure at each temperature, and when the recovery rate is 0.5, the net SEC reduces from 0.47 kWh/m^3 to 0.39 kWh/m^3. However, increasing temperature will reduce the second law efficiency (η); For the same range reduction in temperature, η will decrease from 9.16%

to 3.14%. This can be justified by the fact that increasing T_H, decreasing the implementation efficiency of the solar thermal energy due to more required heat duty to maintain the elevated temperature. Consequently, solar collectors with a larger area are needed which increases one-time infrastructure cost. Therefore, an optimization between net SEC and η should be made while choosing the operation temperature. The analytic hierarchy process (AHP) was made to choose the optimal T_H, considering net SEC, η and the area of the solar collector. It was concluded that higher T_H is beneficial to the system. Another system is also considered using the hybrid system, taking seawater as the RO feed and utilizing RO brine and treated wastewater as the draw and feed solutions while getting heated by solar energy prior to PRO. The achievable net SEC for this system, when T_H and RO recovery are 353.15 K and 50%, respectively, is 1.34 kWh/m^3 which is higher than the previous hybrid system (0.39 kWh/m^3). The reason behind this result is that the dilution effect in PRO was neglected. Although more energy could be generated in PRO due to higher osmotic pressure difference, higher energy consumption in RO makes the overall SEC more.

4.1.3. Chemical separation: Precipitation

Precipitation is one of the most energy-efficient methods for recovery of draw solutes. The draw agents precipitate by adding another compound, changing the solution pH or temperature, which allows water to be separated from draw solutes. Han et al. [16] utilized precipitation for the regeneration of a Na-Fe-CA hydro-acid complex. By adding ethanol to the solution of $Na_5[Fe(C_6H_4O_7)_2]$, draw solutes start to precipitate because they cannot dissolve in ethanol. The re-concentration process worked effectively (yield>99%) without requiring an energy input. However, a small amount of water/ethanol in the mixture can be lost due to evaporation. In other studies [63,64], the precipitation and regeneration of $MgSO_4$ and $CuSO_4$ were investigated by adding $Ba(OH)_2$. After the draw solutes precipitated, the soluble $Ba(OH)_2$ in water will precipitate through the addition of CO_2. This regeneration process suffers from some disadvantages, such as the product water contains residues of heavy metal ions. Moreover, the removal of $Ba(OH)_2$ cannot be completely achieved and a portion of it remain soluble. Thus, the recovered water still needs further purification [65]. In addition, a large amount of $Ba(OH)_2$ is consumed because of metathesis precipitation. Although this method is energy-efficient, this process suffers from low water recovery and the precipitate costs may be very high. Moreover, \in most cases, a downstream process is needed to separate draw solutes completely from produced water.

4.1.4. Magnetic recovery

Magnetic nanoparticles (MNPs) can be functionalized with hydrophilic species to create superparamagnetic nanoparticles to enable easy recovery by an external magnetic field in magnetic separators. In the matter of energy consumption, this method is effective. However, it has some drawbacks. The most important problem is particle agglomeration which reduces driving force drastically after a couple of cycles. In this process the separation efficiency can be low regarding the separation of the smaller particles because the magnetic force was not dominant compared to the diffusion force and gravity. Therefore, the product water may need to be further processed [66].

Ling et al. [67] investigated the use of novel surface-functionalized MNPs draw solutes such as 2-Pyrol-MNPs, TREG-MNPs and PAA-MNPs. The result indicated that after each re-concentration process, the water flux decreases due to agglomeration. PAA-MNP had the highest water flux. In addition, they concluded that by decreasing the PAA-MNP diameter, water flux increases, while the magnetic ability becomes weaker as a result of higher polyacrylic acid contentcovering each particle. Therefore, the recovery of smaller MNPs becomes impossible. Magnetic heating was utilized for regeneration of nanocomposite polymer hydrogels (SA-*co*-NIPAM+ $\gamma - Fe_2O_3$) by Razmjou et al. [68] in FO desalination. Unlike conventional heating, magnetic heating can result in reducing the case hardening phenomena due to a significantly lower temperature gradient. Moreover, higher water recovery rates can be achieved due to high uniformity of heating throughout the volume of hydrogels. In conventional heating, case hardening significantly reduces water recovery. In this process, most of the recovered water is in liquid form. The water recovery and liquid water portion increase with increasing MNP concentration, but not above a certain level. The problem with this system is that the water flux is low, and also, implementation is complicated and not efficient enough.

Table 4: Regeneration processes in the literature

Draw solutes	Regeneration process	Advantages	Drawbacks	References
NaCl	Thermal	Complete recovery of draw solution	Not energy efficient	[24]
$NH_3 - CO_2$	Distillation column		Not energy efficient and cost effective	[26]
KCit	MD	Independent of feed salinity, efficient rejection of non-volatile compound, use LGH	Low water flux, temperature polarization, membrane wetting	[22]
CaAc	MD			
KOxa	MD			
KAc	MD			
NH_4Ac	Thermolytic			
NH_4C	Thermolytic			
NH_4F	Thermolytic			
KF	MD			
NaGly	MD			
NaP	MD			
CaP	MD			
LiCl − methanol	MD	Minimal salt leakage	Very low efficiency, not economically practical	[25]
$CaCl_2$ HCOONa KBr LiBr LiCl $MgCl_2$ $Na(C_2H_5COO)$ NaCl	MD	-	-	[27]
NaCl $MgCl_2$ $MgSO_4$	MD	-	-	[28]
NaCl	MD			[15]
$Na_5[Fe(C_6H_4O_7)_2]$	Precipitation	Effective separation, low energy consumption	Permeated water needs further separation	[23]

5. Energy consumption of the regeneration process

The viability of CLPRO is strongly dependent on the energy consumption of the regeneration processes used to recycle the draw and the feed solutions back into the system. In fact, this dependency manifests in the fact that the overall energy produced should compete with other renewable energy sources. As presented in the previous section, many regeneration processes have been used in CLPRO. Several authors have not discussed the impact of the energy consumption of the used regeneration processes on the overall produced energy. Herein, the feasibility of some potential regeneration processes based on the net energy production is discussed. For that, first, the maximum theoretical energy production is identified as the upper value of energy that can be produced by PRO under perfect conditions such as ideal membrane, ideal salt rejection, and perfect hydrodynamics. This energy can be expressed as a function of the osmotic pressure difference as follows [69]:

$$SE_{PRO}^{M} = \frac{1}{4}\left(\frac{(\pi_D - \pi_F)^2}{\pi_D + \pi_F}\right) \tag{1}$$

where π_D and π_F are the draw and feed osmotic pressures, respectively. In real conditions, this maximum is unachievable due to fouling, scaling, salt diffusion, and imperfect hydrodynamics. For that, it is assumed that optimized PRO systems can produce 70% - 80% of the maximum extractable energy [16]. Then, the first condition to discuss the feasibility of CLPRO is to satisfy the following criterion:

$$SE_{PRO}^{R} - SEC_{RP} > 0 \tag{2}$$

SE_{PRO}^{R} and SEC_{RP} are the PRO specific energy production and the specific energy consumption of the regeneration process, respectively. Figure 3 shows the energy consumptions of various regeneration processes used in the literature, as well as the PRO specific energy that can be theoretically and practically produced with potential draw solutions. To satisfy Equation 2, thermal processes and reverse osmosis are excluded. In fact, the SEC of RO process ranges between 2.4 and 3.4 kWh/m^3, which makes the net power production negative, thereby deeming the process energetically inefficient. Similarly, thermal processes such as MD or MED have a very high energy consumption (100 to 1000 times the energy produced by PRO), which is unsuitable for a viable CLPRO.

Several studies suggested that MD can operate with low grade waste heat released from industrial activities (i.e., nuclear plant, thermal power plant factories, etc.) and therefore can be used as a downstream separator for PRO. This seems too ambitious due to two reasons: 1) MD should be fully powered by waste heat, which is not practically achievable, and 2) if we assume that the previous condition is fulfilled, that waste heat should be unrecoverable by any other process because if so, around 100 kWh/m^3 of recoverable energy will be consumed to produce less than 2 kWh/m^3. For that, MD is likely unsuitable for PRO. Similarly, besides the high energy consumption, the range of RO applicability is limited (<100 bars). Also, like MD, the use of renewables is not reasonable. For the other possible regeneration processes like precipitation, waste heat to degrade thermolytic solution, and electromagnetic regeneration, the energy consumption is relatively attractive (< 0.2/0.4 kWh/m^3). However, these processes are still suffering from low recovery rate as well as various practical drawbacks (agglomeration, cost efficiency, low recovery ratio, etc.). Fig.3-A shows two main zones: the zone of inefficiency and the targeted zone. The zone of inefficiency is where the SEC of a system used for solution recovery does not satisfy criterion 1 as mentioned in Equation 2, and the targeted zone is characterized by the regeneration process handling solutions with high osmotic pressure with low energy requirements.

The second criterion that should be considered is the economic competitiveness of CLPRO. More precisely, the cost of the produced energy must be comparable to the renewable energies, such as wind, solar, and hydroelectricity. As a comparison tool, economists use the levelized cost of electricity (LCOE) to compare between energy harvesting processes. To determine the LCOE, an estimation of the cost is required. Several studies have performed a techno-economic evaluation of PRO process [46,69-72]. The analysis showed promising results with high solutions with osmotic pressure ($\Delta\pi$ > 150 bars) [71]. However, these studies have considered OLPRO and very few studies have been performed for CLPRO [46,72]. As shown in Table 2, CLPRO has some economic advantages when compared to OLPRO such as lower capital and operation costs, as well as a controllable produced energy (by controlling the input osmotic pressure). Therefore, the choice of the draw solution and the way to regenerate it is a determining factor on the cost effectiveness of CLPRO. Overall, higher osmotic pressure with a low SEC of regeneration is the key for a sustainable, viable, and competitive renewable source of energy. The van't Hoff relationship was used to calculate the osmotic pressure. For a highly concentrated solution, there is a gap between the real osmotic pressure and that provided by the van't Hoff relationship. Therefore, the energy produced must be greater than the energy calculated using Equation 1. Future studies should be performed to quantify the real extractable energy at high draw solution concentrations.

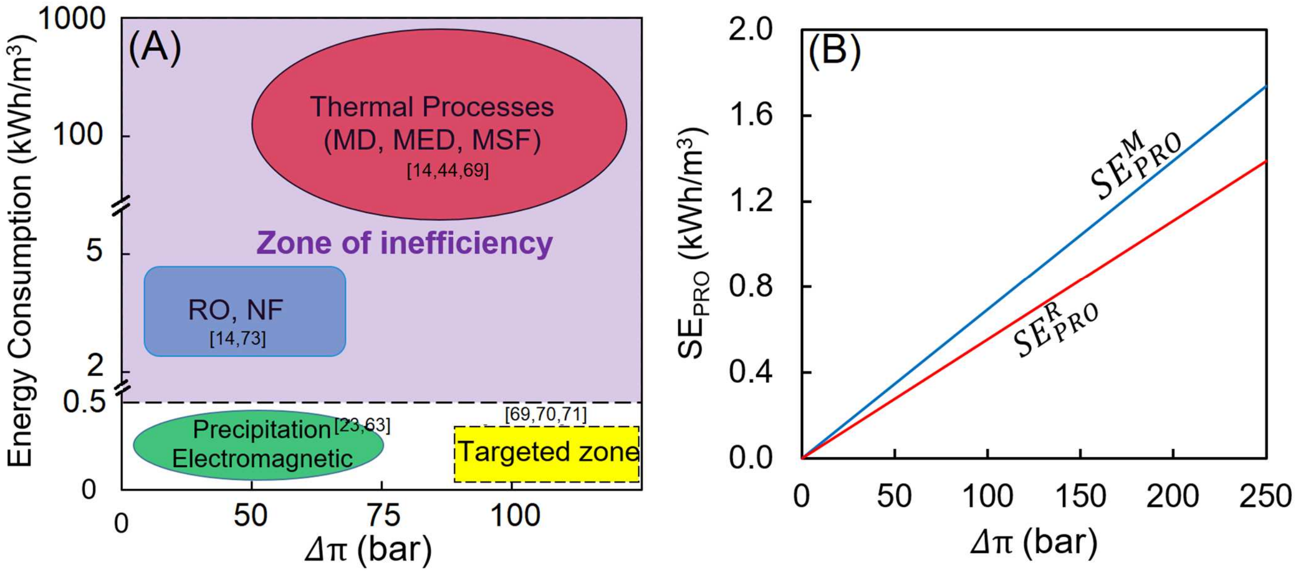


Figure 3: Specific energy consumption of draw solution regeneration processes, SEC, and the specific energy production of PRO, SE_{PRO}. (A) The SEC of potential recovery processes used in the literature. It shows also the zone of inefficiency and the targeted zone in term of energy consumption. (B) Shows the maximum and the real achievable energy by PRO. Here the real energy is considered 80% of the maximum theoretical extractable energy as shown in previous work [9]. The calculation that the PRO feed solution is pure water is assumed. The van't Hoff relationship was used to calculate the osmotic pressure for 25°C.

For the specific case of an OHE, presented in Figure 4-A, where the PRO is coupled to MD to generate electricity from LGH, the thermal efficiency, η_{OHE}, is defined as the ratio between the generated power through PRO- turbine and the heat duty for separating and recycling the diluted draw solution via MD:

$$\eta_{OHE} = P/q_h = Y\pi/c_p(T_H - T_C)\,\rho_W \qquad (3)$$

Here, P, q_h, Y, π, c_p, ρ, T_H, and T_C are respectively the power produced by the PRO-TB component, the amount of heat, the mass flow recovery of MD, the osmotic pressure of the working fluid (water in this case), the heat capacity of the working fluid, the working fluid density, the heat source temperature, and the cold sink temperature. Carnot efficiency is the relative comparison of an engine efficiency with maximum theoretical efficiency of the Carnot engine which operates with the same high and low temperatures heat streams ($\eta_{Carnot} = 1 - T_C/T_H$). Carnot efficiency is presented as a reference. For comparison, the energy conversion efficiency of Organic Rankine Cycle (ORC), operating in the same range of temperature was plotted. This efficiency was determined from the Entropy-temperature diagram for and ideal ORC process operating with isopentane as a working fluid. The Carnot efficiency has little practical value due to the fact that, even being very efficient, a Carnot engine has to operate at infinitesimally low velocities to allow the heat transfer to occur, then, no power will be generated (i.e., a thermodynamically reversible process). For that, the Carnot efficiency at maximum power output of Carnot engine (dashed red line) is defined as:

$$\eta_{Carnot}^{Pmax} = 1 - \sqrt{T_C/T_H}. \qquad (4)$$

This efficiency is more practical than the classic Carnot efficiency since it describes the performance of the engine when power is produced. Figure 4-B shows, as for any heat engine, the increase of the temperature difference leads to the increase of the OHE efficiency. This efficiency is still remarkably lower than η_{Carnot} due to losses occurring during the processes (heat losses: in MD, heat exchanger, hydraulic losses: in PRO due to constant applied pressure that leads to friction losses, unutilized energy, pressures losses in the PRO module). However, the OHE efficiency is still greater than η_{Carnot}^{Pmax}, which means that the OHE reaches higher efficiency for PRO energy production lower than its maximum. Therefore, pushing PRO for maximum energy production should not be the target for an OHE. However, tracking the optimal PRO energy generation for maximum efficiency is one of the important operational parameters that should be readily controlled. Consequently, the required applied pressure in the PRO side for maximum efficiency is lower than the half of osmotic pressure difference between the draw and the feed side, a condition widely utilized for PRO energy optimization.

The comparison between OHE and ORC in the same temperature range shows the main zones: below and beyond 70°C. In the first zone, OHE efficiency is slightly lower than that of ORC (1-3% of efficiency difference) which means that of OHE can be practical alternative for ORC to harvest energy from low temperatures specially when considering that ORC is unable to tolerate fluctuations in the heat source temperatures, the operational constraints imposed by the working fluids, as well as the economic unviability of ORC for low temperatures [74]. In the second zone, it is clearly seen that ORC has much better efficiency than OHE that makes it incapable of competing with binary cycle processes at relatively high temperatures. Note that the previous statements are valid for OHE operating with water as the working fluid characterized by an elevated heat of vaporization which causes losses in the membrane side as well as in the heat recovery device (heat exchanger). For that, losses may be mitigated by employing other working fluids with a low heat of vaporization. However, this working fluid should also provide high osmotic pressure to optimize the energy production in the PRO side as well as guarantee a certain tolerance to MD membrane (fouling and wetting). Undoubtedly, developing a PRO membrane withstanding high pressures is a vital condition toward the viability and the competitiveness of OHE. Overall, CLPRO, including the specific case of OHE, seems to have great potential to produce energy from salinity gradients as well as harvesting power from heat waste and has the potential to compete with other sources of renewable energy and energy conversion processes. This also is still relying on the economic viability of CLPRO and OHE that is not definitely proven.

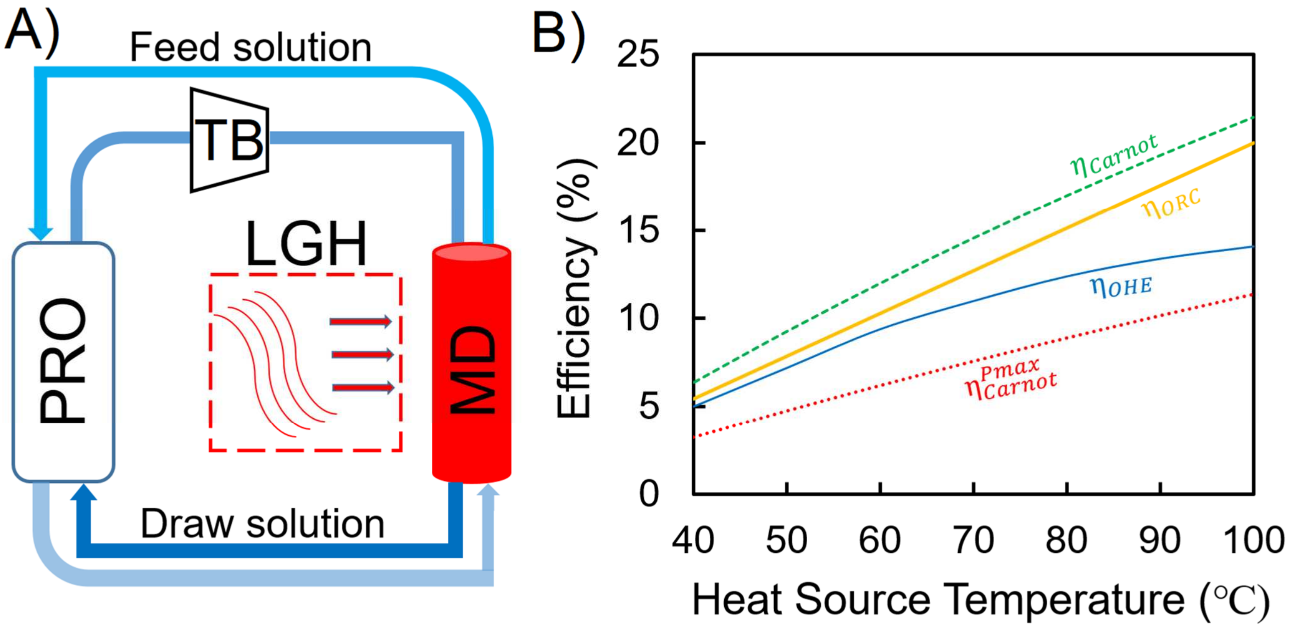


Figure 4: (A) Schematic of a MD-PRO OHE. The power generation process consists of units such as a PRO, a turbine (TB) and a membrane distillation (MD) including heat exchange connecting to a heat energy source (LGH). (B) Energy efficiency of the PRO-MD OHE at different heat source temperatures of the working solution (NaCl, Concentration = 4 M, osmotic pressure π = 200 bar), Carnot efficiency of the OHE, and the Carnot efficiency at maximum power produced. Also, the energy conversion efficiency of ideal Organic Rankine Cycle with isopentane as a working fluid. The temperature of the cold sink is 20 °C. The mass recovery rate of MD process is assumed, and the thermodynamic properties of the process were adopted from [14]. The relative flow rate, defined as the ratio between the permeate mas flow and the feed mass flow, was assumed to be 0.918.

6. Conclusions and Future Directions

In the last decade, SG has been widely studied as a potential source of energy that can be harvested using PRO which has shown promising results. In this study, the potential of CLPRO to harvesting energy from SG was reviewed. First, the opportunity to use CLPRO configuration instead of OLPRO was shown by presenting the advantages and challenges of each process. As well known, CLPRO operates with engineered draw solutions with controlled properties, which mostly do not require pre-treatment and reduces the risk of fouling in the membrane. Moreover, regarding the plant location and footprint, CLPRO has more flexibility compared to open-loop one. Second, essential criteria were set that should be respected in the selection of an adequate draw solution to optimize the energy production. Then, draw solutes used in the literature were reviewed regarding their qualitative and quantitative performance. Inorganic draw solutes, mostly NaCl, showed potential for generating high power density (87 $\mathrm{W/m^2}$) at high concentrations. However, their high RSF due to their small sizes reduces effective driving force and efficiency of the process. On the other hand, organic draw solutes, have the benefit of remarkably low RSF compared to inorganic salts due to their larger sizes, though, this advantage promotes ICP. Engineering these salts makes them more flexible for use in PRO. Future studies should focus on synthesizing novel draw solutes which produce high osmotic pressures at low concentrations with minimal RSF which have efficient regeneration.

Third, regeneration processes for recovering the draw and feed solutions, in the literature, were critically reviewed. The analysis showed that the regeneration step should operates with very low energy consumption and high recovery to guarantee an energy-efficient process. Physical separation methods such as RO are still energy consuming due to the required pumping energy. With current advances in PRO, the net SEC of the whole process is not below zero (minimum 0.39 kWh/m^3), and thus, reliance on electricity still exists. Regarding using waste and low-grade heat in thermal separation methods, future work should focus on designing the process for harvesting and transferring that heat into the process including all the necessary unit operations, and their energy and cost calculations. After that, as a specific case, the efficiency of PRO-MD based OHE (5%) to harvest energy from LGH was examined. The analysis showed that OHE is efficiently comparable to ORC for relatively low temperatures (< 70°C) when operating with water as the working fluid. Beyond 70°C, OHE requires optimization to compete with ORC such as better working fluid and mitigated energy losses in the MD membrane side, the heat exchanger, as well as optimized energy production by PRO. This will require the fabrication of better PRO membranes that withstand high hydraulic pressures, a working fluid that provides high osmotic pressure as well as a low heat of vaporization, optimizing the energy losses in the heat exchangers or developing a better heat recovery device. After that, as a final step toward implementation, an economic analysis should be performed to confirm the viability and the competitiveness of the process. Although precipitation is an energy-efficient way to regenerate the draw solution, its low water recovery is still not suitable for continuous electricity generation. Also, according to many cases, the product water needs more purification using a downstream process which increases the cost. Magnetic separation suffers from particle agglomeration which reduces water recovery and process efficiency.

The most crucial issue in CLPRO is the membrane. Improving membrane selectivity as well as its mechanical strength (up to 100 bars) should be focused on and studied by research groups. Developing membranes with thinner support layers and higher porosity and robustness at the same time, especially in the PRO-RO case, can make this process viable. Applying more pressures on the membrane makes it possible to harvest the full potential of draw solutions which leads to higher power density. Improved membrane's selectivity and porosity can lower ICP significantly. Therefore, effective driving force reduction and feed recovery load will be reduced due to minimal RSF. Furthermore, there are several methods to assess productivity, efficiency, envirosafety, and sustainability of an energy system such as life-cycle assessment (LCA), techno-economic analysis, emergy analysis, energy analysis, and exergy analysis. It was concluded that among the methods mentioned, exergy analysis is the most effective one [75]. Exergy analysis not only evaluates the quantity of the energy streams but also the quality of them. This method considers both first and second laws of thermodynamic, and, through assuming reversible processes, it determines the maximum achievable work a thermodynamic system can generate until it reaches the equilibrium state. Furthermore, this method can be integrated with environmental and economic concepts which are known as exergoenvironmental and exergoeconomic. By applying thermodynamic limits, this assessment can show precisely economic and energy losses assessing the sustainability of a system. Therefore, future work should investigate PRO under exergy analysis and to be specific exergoeconoenvironmental assessment which proposed by Aghbashlo and Rosen [76] to quantify the efficiency of the system, LCOE, envirosafety and sustainability.

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Declaration of Competing Interest

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.

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