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Integrated Processes for Desalination and Salt Production: A Mini-Review

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Abstract. The scarcity of fresh water due to the rapid growth of population and industrial activities has increased attention on desalination process as an alternative freshwater supply. In desalination process, a large volume of saline water is treated to produce freshwater while a concentrated brine is discharged back into the environment. The concentrated brine contains a high concentration of salt and also chemicals used during desalination operations. Due to environmental impacts arising from improper treatment of the brine and more rigorous regulations of the pollution control, many efforts have been devoted to minimize, treat, or reuse the rejected brine. One of the most promising alternatives for brine handling is reusing the brine which can reduce pollution, minimize waste volume, and recover valuable salt. Integration of desalination and salt production can be implemented to reuse the brine by recovering water and the valuable salts. The integrated processes can achieve zero liquid discharge, increase water recovery, and produce the profitable salt which can reduce the overall desalination cost. This paper gives an overview of desalination processes and the brine impacts. The integrated processes, including their progress and advantages in dual-purpose desalination and salt production are discussed.

INTRODUCTION

The need for freshwater during the current rapid growth of population and industries is urgent due to the depletion of freshwater resources and water pollution.[1] The increasing demand for the fresh water has resulted in an increased global interest in desalination technology as an attractive solution to the growing fresh water crisis due to a large amount of saline water resources such as brackish water and seawater. It was reported that more than 18,000 desalination plants have been operated worldwide with an average production rate of 86.8 million m^3/day.[2] Indeed, the gained interest of desalination technologies is also associated with the improvement of desalination technology such as seawater reverse osmosis (SWRO) that enables fresh water production with lower cost and energy requirement compared to thermal-based processes.[3] The development includes membrane materials and modules, system design, energy recovery devices, strategies in fouling mitigations, renewable energy sources, etc.[4-9]

Although desalination technology represents the most attractive supplementary non-conventional water source, management of the concentrate effluent or rejected brine is still the main problem.[10-14] The brine has strong negative impact on the sustainability of the environments due to their high concentration of salts and other pollutant contents.[15-19] The type of the pollutants may vary, depend on the type of feed water. The rejected brine from desalination concentrates contains higher salt concentration than that of seawater (used as feed for the desalination plant), and also contaminated with several chemicals utilized during the desalination operation. Therefore, the rejected brine from desalination processes must be properly managed to avoid environmental contamination.

Although the brine effluents will bring negative issues to the environment, but they are commonly discharged without any further treatment. Due to the environmental impacts arising as a consequence of brine discharge, many efforts have been devoted for waste brine minimization or seek for possible options to reuse the brine. In order to be

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reused, the brine should be treated with suitable technologies to obtain the desired quality. Moreover, the brine is also considered as a potential source of valuable compounds that may bring cost minimization of the process. The brine could be treated by performing simultaneous water recovery and salt production purpose that creates a dual-purpose plant of both desalination and salt production.[20-23] The salt products and the increased water recovery from the integrated process could reduce the cost of the overall process. Several integrated desalination processes (which are dominated by the application of advanced membrane-based processes) have been proposed to achieve those promising and interesting dual-purpose scheme of simultaneous desalination and salt production processes. In general, this paper is organized to cover several integrated processes for rejected brine management.

OVERVIEW OF DESALINATION PROCESS

Desalination Techniques

Desalination process refers to the removal of salts from saline water, such as brackish water and seawater, in order to obtain fresh water for domestic or industrial purposes. In this regard, the saline water is treated into two different streams. The first stream is fresh water containing a low concentration of salt while the second one is brine with concentrated salt. Generally, there are two classes of desalination techniques, namely thermal-based and membrane-based processes. The main thermal desalination technologies for seawater desalination are multi-stage flash (MSF), multi-effect distillation (MED) and vapor compression (VC).[24] MSF is operated based on the principle of flash evaporation in which the saline water is evaporated under vacuum condition and then followed by condensation of freshwater.[25] MSF consists series of stages (ranging from 4 to 40) with successively lower temperature and pressure.[24] Desalination process in MED occurs in evaporators arranged in series and uses the principle of reducing ambient pressure in the evaporators.[25] In this regards, saline water undergoes multiple boiling without supplying additional heat after the first evaporator. Meanwhile, VC utilizes heat from compression of vapor to evaporate saline water.[24, 25] Two methods are employed in this process, i.e. mechanical vapor compression (electrically driven) and thermo vapor compression. The thermal-based technologies are primarily used for high salinity solution and where inexpensive energy is available.[26]

In the membrane-based desalination technologies, desalination occurs based on permeation of components through a selective permeable membrane. Due to selectivity of the membrane, one component is preferentially permeable to others.[27] In the membrane-based process, freshwater could be obtained by permeating water or salt.[7] Reverse osmosis (RO), nanofiltration (NF), forward osmosis (FO), and membrane distillation (MD), are the typical example of desalination wherein water is permeated across the membrane. Meanwhile, in electrodialysis (ED), electrodeionization (EDI), and microbial desalination cell (MDC), freshwater is obtained by permeating salts or ion across the membrane.

In RO system, dissolved solutes including charged ion from water are separated via a semipermeable membrane that allows passing water in preference to the solute. The driving force is a pressure different between the two membrane sides, i.e. feed side and permeate side. If a saline solution is pumped into RO unit and a pressure gradient is applied (pressure required to exceed the osmotic pressure of solution) the water migrates across the membrane toward the permeate side while the salts and other components are retained by the membrane on the feed side. The overall result is two streams with different concentration of salt. The first stream is brine with increased concentration in concentrate side while the other is depleted of salts in the permeate side. RO has been recognized as the leading technology of desalination process and is now being used in various applications including water treatment, wastewater treatment, food and beverage processing, etc.[28-34]

NF is another type of pressure driven membrane process. The membrane used in NF has characteristics between RO and the ultrafiltration (UF) membrane. NF is able to remove turbidity, microorganisms, hardness, and a fraction of dissolved solid (especially for multivalent ions).[35] Due to its ability on removing multivalent ions and some portion of monovalent ion, NF is then introduced as pretreatment for RO system.[36, 37] As the pretreatment, NF reduces the osmotic pressure of RO feed water by removing a fraction of dissolved solid or acts as partial demineralization step thus the overall water recovery can be increased.[38, 39] By introducing NF as the pretreatment, it is possible to reduce energy consumption, chemical consumption, and the overall water cost of SWRO plant.[38]

Unlike the two previous membrane processes, MD is a thermally driven process that uses a hydrophobic porous membrane for separating a vapor phase from feed stream.[40] The driving force is vapor pressure difference generated by temperature difference across the membrane.[41] Vapor phase is transported through the membrane while the liquid phase is prevented due to the hydrophobic nature of the porous membrane.[1] The advantages of MD are relatively low operating temperature and pressure, high permeate quality, the possibility to treat highly concentrated solution, and not limited by the osmotic pressure.[42] Despite its advantages, MD technologies have not been applied in industrial scale due to several barriers, such as: low permeate flux, flux decline due to concentration and temperature polarization effects, membrane fouling and total or partial pore wetting, membrane and module design for MD, and relatively high thermal energy consumption compared to pressure-driven membrane.[43]

Desalination by removing salts or ion from a feed solution can be conducted by ED and related process. ED uses ion-exchange membrane as the selective separator and utilizes an electrochemical potential as the driving force for the mass transfer process.[44] Ion-exchange membrane is widely used in several processes and applications, such as desalination, wastewater treatment, chlor-alkali production, ultrapure water production, organic acid production, demineralization or purification of a product, and energy generation.[44-49] The success of those processes is strongly affected by the characteristic of the ion-exchange membrane, namely selectivity, electrical conductivity, chemical and mechanical stability, and production cost. Therefore, many efforts have been devoted to fabricate membrane with better characteristics.[50-57]

In ED unit, several cation-selective membranes and anion-selective membranes are arranged in alternating pattern inside a pair of electrode (anode and cathode) and are separated by spacers. When electrolyte solution is pumped into an ED, and electrical potential is applied to the electrode, cation and anion are transported across membrane toward electrode (anion to anode; cation to cathode). Since the membrane only selective for a specific charge, cation membrane allows passing cation while retains the anion, and vice versa. As the result, the electrolyte solution is concentrated with ionic components in one compartment (chamber) while the ions concentration is of the electrolyte is reduced in the adjacent compartment. The first solution is called as concentrate while the latter is known as diluate. ED has been used for brackish water desalination and the basic process has been significantly modified into several related processes such as EDI, electrometathesis, electrodialysis with bipolar membranes (EDBM), electrodialysis reversal (EDR), etc.[58] The advantages of using ED for desalination are high water recovery, long useful life of membrane, and less membrane fouling due to process reversal.[44] Unlike RO, there is no osmotic limitation thus higher water recovery and higher brine concentration can be achieved. Therefore, ED has been applied in table salt production as a pre-concentration step prior to evaporation.[44] However, ED also has some drawbacks such as the relatively low product purity and relatively high energy consumption when treating a dilute solution due to high electrical resistance and concentration polarization phenomena. To solve this problem, ED unit has been developed and modified by introducing ion-exchange resin inside ED cells. This modified ED unit is known as EDI or continuous deionization process. The use of ion-exchange resins as the bridge over current, conductivity of the overall process is enhanced. By adopting this strategy, it is then possible to produce high purity water and to treat a dilute solution with relatively low energy consumption. Furthermore, EDI eliminates chemical regeneration and the associated hazardous chemical that makes EDI as chemical free operation and environmentally friendly. With those advantages, EDI has continued to be an attractive deionization process with significant advantages over the conventional ion exchange deionization in the production of ultrapure water.[59]

Among the desalination techniques, RO is the most widely used desalination technology due to remarkable advances of this technology. The advances lead to significant cost reduction of RO process. Sorek desalination plant is an example of the largest SWRO plant which is located in Sorek, Israel, with a production capacity of 624,000 m^3/day.[60] This plant is expected to produce fresh water with a maximum energy consumption of 4 kWh/m^3 and contains 0.3 mg/L of boron.

Brine Discharge

Typically, desalination process extracts a large volume of water and discharges the rejected brine back into the environment. The brine contains a high concentration of salt (which is usually higher than its original feed water) and other pollutants. The pollutant components in the brine can be classified as corrosion products, anti-scaling additives, anti-fouling additives, halogenated organic compounds, anti-foaming additives, oxygen scavengers, acid, and concentrate.[61] The brine could potentially bring negative impact on the environment due to its salt concentration and chemical content. The impact involves physicochemical and ecological of receiving environment wherein the brine is being discharged.[12] The physicochemical impact is attributed to their salinity, temperature, and constituent which could alter the physicochemical properties of receiving water and may have adverse effects on the marine life. Other impacts of brine discharge are reported in literature.[15, 62, 63]

The impact of brine discharge into receiving water and its severity depends on volume, characteristics, dilution rate prior to dispose, and characteristics of receiving water.[61] Several options have been proposed to manage brine rejected from desalination plant. For example, Bukley et al. suggested several methods to treat the waste brine, such as reducing the dissolved solid in the brine, converting brine into useful products, indirect discharge, deactivating brine by transforming it into unreactive or insoluble compounds, immobilization of brine or storage, and direct discharge of brine.[64] According to Morillo et al., brine management options can be categorized into four different groups based on the final purpose:[65]

1. Reducing and eliminating brine disposal: solar evaporation, phytodesalination, evaporation and crystallization system, MD, two-stage reverse osmosis, closed-circuit desalination, forward osmosis, and ED.
2. Commercial salt recovery: SAL-PROC process, zero discharge desalination, and integrated processes.
3. Brine adaption for industrial uses: brine-adaptation for chlor-alkali industry and acid-basic production using bipolar membrane ED.
4. Metal recovery: sequential chemical extraction process using chemical and physical processes, adsorption.

Technologies oriented to commercial salt recovery is an attractive option for managing waste brine. Valuable recovered salt can be used to reduce the overall desalination cost. This option provides the possibility to obtain valuable components, both water and salts, that results in increased water recovery and reduced the overall water production cost. By doing so, a dual-purpose plant for desalination and salt production is achieved.

INTEGRATED PROCESSES

SAL-PROC is an integrated process for extracting dissolved solid from saline water by using multiple evaporations and/or cooling combined with mineral and chemical processing.[66] A case study of applying SAL-PROC process for producing salt from brine has been conducted.[66] Three different treatment options were used in the study. For example, one option involves solar evaporation, crystallizer pond, and reactive vessel for chemical mixing. The chemicals used for the study was lime and sodium carbonate. Various types of salts including gypsum, sodium chloride, magnesium hydroxide, calcium chloride, calcium carbonate, and sodium sulfate can be produced from 450 ML/year brine discharge with an approximate value of 895,000 $/year.

Dow Chemical process is another type of chemical process which can be used to produce magnesium metal from brine. Investigation on the use of this process for magnesium production from Arabian Gulf desalination brine has been reported.[67] In the Dow Chemical process, calcination, slaking, precipitation, and hydrochlorination reactions are involved. About 9 Mton/year of desalination brine is required to produce 2000 ton/year of magnesium with total cost of 2357 $/ton magnesium produced.

Integrated processes for the dual-purpose plant combining membrane and thermal process has been investigated by Turek and Coworker.[68-70] The integrated process includes ED, NF, RO, MSF, and crystallization (Fig. 1 (a)-(c)). In ED-MSF-crystallization system, seawater is firstly desalinated in ED system where in ED and ED reversal (EDR) are employed. Monovalent ion permeable membrane is used in ED. Consequently, the diluate solution contains multivalent ions, e.g. calcium, magnesium, and sulfate ions. This makes further processing by ED is difficult which is related to precipitation of such salts during the next desalination process. To overcome this shortcoming, EDR is employed. Results indicated that EDR can achieve 90% water recovery without gypsum crystallization in EDR concentrate. Seawater with TDS (Total dissolved solid) 35 g/L is concentrated up to 105 g/L. The ED-EDR system could produce potable water with 0.5 g/L of TDS. According to the economic analysis, the potable water cost is estimated to be only 0.44 $\$/m^3$ when using low resistance ion-exchange membrane (potable water cost= total desalination cost – salts sale). Other integrated processes that have been investigated are UF-NF-MSF-crystallization and UF-NF-RO-MSF-Crystallization. The cost of potable water produced from the first system is 0.71 $\$/m^3$ while the second system is 0.43 $\$/m^3$. Benefiting from the ability of NF in reducing the multivalent ion in pre-treatment step, the desalination system can be improved including higher water recovery and lower energy consumption for both RO and MSF.

Another interesting integrated process which has been developed for desalination and recovery of salt from the brine is integrated membrane desalination system.[22, 38, 71-73] The integrated membrane system combines UF, NF, RO, MD, and membrane crystallization (MCr) (Fig. 2 (a) and (b)). NF is used to remove bivalent ions from the feed stream. The NF permeate with reduced salt is delivered to RO unit. Due to lower osmotic pressure as the result of NF softening step, RO can reach higher water recovery. To recover more water from NF and RO, MD and MCr are introduced. MCr increases solute concentration of solution above its saturation limit in which crystal nucleate and growth. Similar to MD, MCr has some interesting features, such as low operating temperature and pressure, high permeate quality independent to feed quality, and the possibility to treat solution with a high concentration.[22]

Therefore, MCr is an attractive method to treat brine from RO and NF concentrates and to recover salts. By using this integrated system, various type of salts, including NaCl, $CaCO_3$, and $MgSO_4.7H_2O$ can be produced. Furthermore, by the integrated membrane system provides the possibility to improve the performance of desalination system. From the economic analysis, the potable water cost can be reduced due to high water recovery and the salts sale from the recovered salts (Table 1). The integrated membrane system will lead to seawater desalination system with improved performance and reduced environmental impacts.

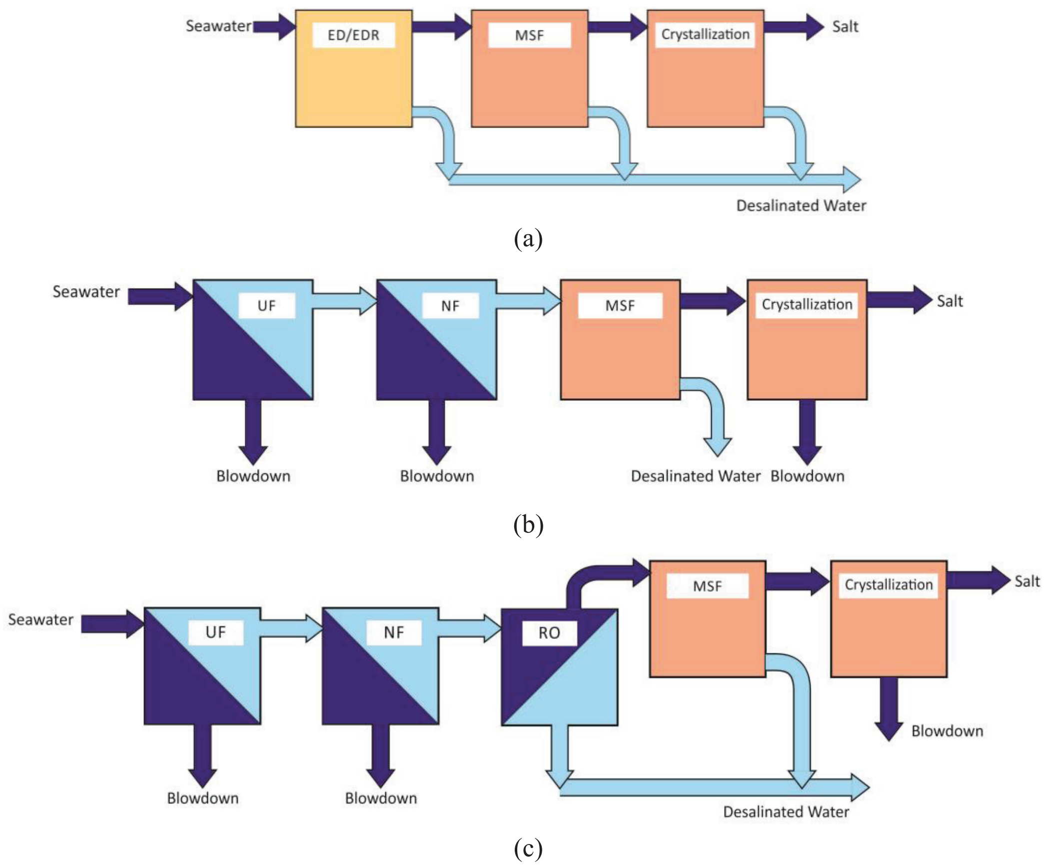
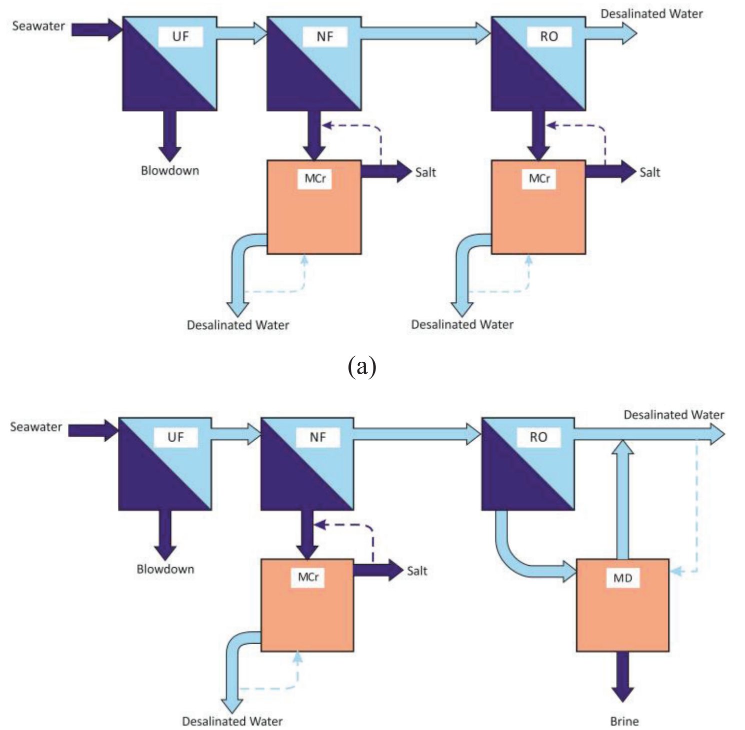


FIGURE 1. Membrane/thermal integrated processes. Integrated process scheme: (a) ED-MSF-Crystallization; (b) UF-NF-MSF-Crystallization; and (c) UF-NF-RO-MSF-Crystallization (adapted from references[68-70]).

Sorour et al. reported a preliminary techno-economic assessment of integrated process called as zero desalination discharge (ZDD) facility with 20,000 m^3/day of capacity.[74] The ZDD facility is based on membrane and thermal techniques (Fig. 3). The ZDD facility chemical precipitation, microfiltration (MF), NF, RO, ED, drying, MED, and evaporation crystallization. Seawater stream is subjected to chemical precipitation using sodium carbonate at pH 9.2 to precipitate calcium and magnesium ions as well as other water contaminants such as barium, strontium, silica, etc. The treated water is directed to MF unit. The permeate from MF is transferred to NF unit after pH adjustment, while a portion the MF reject stream is directed to a drying bed equipped with a clarifier and the other portion is recycled to the precipitator. Afterwards, the brine stream from NF is directed towards MED process. The MED concentrate stream is processed in "evaporator/crystallizer" unit wherein magnesium-rich salts are produced. The permeate stream from NF unit is directed to RO. The reject brine from RO unit is further desalinated using ED. NaCl concentration up to 20% is obtained in ED concentrate stream. This concentrated stream is further processed to the evaporator/crystallizer unit. Meanwhile, the diluate stream of ED is processed using chemical precipitation with sodium hydroxide at pH 11 and the filtrate is directed to the "evaporator/crystallizer". Result of economic analysis showed that about 0.98 \$/m^3 can be realized by selling the recovered salts ($CaCO_3$; $MgSO_4.7H_2O$; $Mg(OH)_2$; $MgCl_2$; NaCl).



(b)

FIGURE 2. Integrated membrane processes. Integrated process sheme: (a) MF-NF/MCr-RO/MCr and (b) MF-NF/MCr-RO/MD (adapted from ref. [72]).

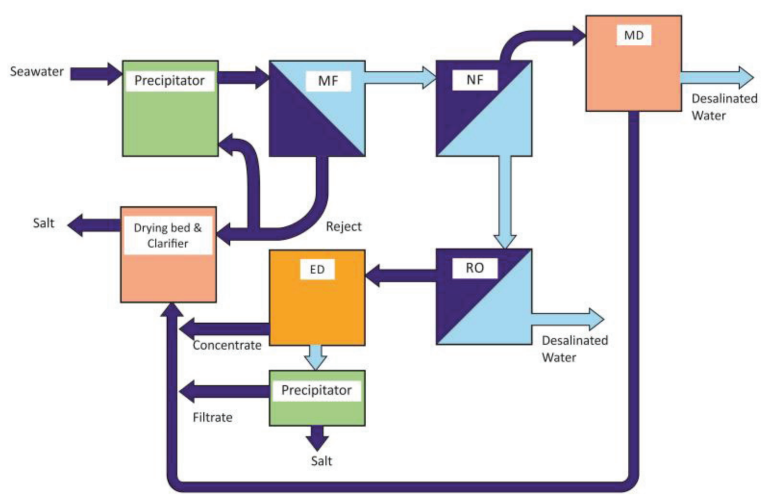


FIGURE 3. Integrated process of zero discharge desalination (adapted from ref. [74]).

The integrated process can be used not only for improving water recovery and producing salt from a desalination process, but also for producing hydrogen, chlor, and alkali. This type of integrated process includes RO, ED, evaporation, and electrolysis processes.[76] ED is used after RO unit to improve water recovery and to increase concentration factor of RO brine prior to evaporation. In evaporation unit, edible salt and bittern are produced. After purification, the solution from evaporation unit is processed in electrolysis in which hydrogen, chlor, and alkali are produced.

Typical example of integrated processes are listed in Table 1 while desalination cost of integrated process and the value of recovered salt is shown in Fig. 4. It is shown from the figure that integrated process for desalination and salt production can improve cost-effectiveness of the desalination process.

Besides salt, the brine also contains valuable metals which can be recovered for some profitable uses. However, the processes to extract the valuable metals are still in progress due to difficulties on targeting the selected components. In addition, the process is very complex and requires multiple sequential steps.[77, 78] The brine from desalination plant may also contain iodine.[79] Brine water consists of iodine in the form of an iodide salt. Therefore, oxidation of iodide ion is required to obtain the iodine. The oxidation of iodide to iodine using ozone as an oxidation agent in membrane contactor as contacting device exhibited an interesting result for iodine recovery.[80] Membrane contactor has some advantages, such as better contacting performance, larger interfacial area, the ease of direct scale up, and no catalyst is needed.[81, 82] The performance of membrane contactor could be more pronounced with development of superhydrophobic membrane.[83, 84] By using this kind of membrane, wetting phenomenon that is usually considered as one of the limitations of gas-liquid membrane contactor, could be tackled. With those advantages, this method could be potentially used for recovering iodine from the brine efficiently.

Another potential use of brine is for electrical energy generation by utilizing salinity gradient between brine and lower salinity water by pressure-retarded osmosis (PRO) and reverse electrodialysis (RED).[85, 86] The osmotic pressure difference induced by two solution with different salinity is utilized as a driving force. PRO uses semi-permeable membrane that allows to pass water across the membrane from a low salinity into high salinity solution.[87] Electrical energy is generated by a hydroturbine in which kinetic energy of the flowing water is converted to electrical energy. Meanwhile, RED is very similar to conventional ED. In RED, a number of anion and cation exchange membranes are stacked together in an alternating pattern between a pair of electrode (anode and cathode). The driving force in RED process is chemical potential owned by solutions. Due to the gradient salinity of two different solutions fed into RED cells, the chemical potential is converted to electrical energy by transporting ions from high salinity into low salinity solution called concentrate and diluate, respectively.[88]

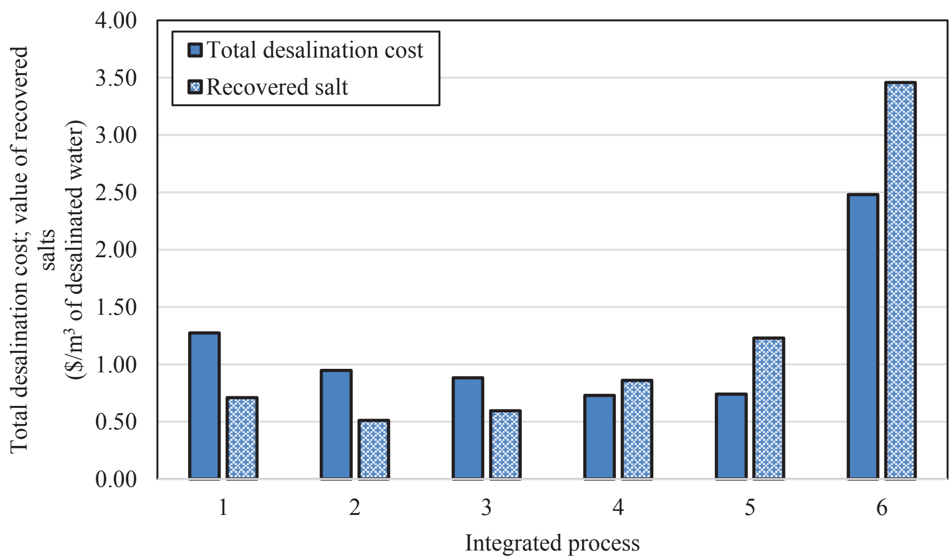


FIGURE 4. Total desalination cost and the value of recovered salts from integrated processes. Integrated process scheme: (1) ED-MSF-Crystallization; (2) UF-NF-MSF-Crystallization; (3) UF-NF-RO-MSF-Crystallization; (4) MF-NF/MCr-RO/MCr; (5) MF-NF/MCr-RO/MD; (6) Precipitator/MF/drying bed/NF/RO/ED/MED/Precipitator/Evaporator-crystallizer (data from references [68-70, 72, 74]).

TABLE 1. Typical example of integrated processes for dual-purpose desalination and salt production

No.	Process	Recovered salts	Remark	Ref.
1	SAL-PROC process	Gypsum; $Mg(OH)_2$; NaCl; $Na_2(SO_4)$; $MgCl_2$; $CaCl_2$;	Salts sale: 2.21 $/m^3 of brine	66
2	Dow Chemical process	Magnesium metal	Total production cost: 2357 $/ton magnesium.	67
3	ED-MSF-Crystallization	NaCl	Total desalination cost: 1.274 $/m^3. Salt sale: 0.710 $/m^3 of desalinated water. The recovery of ED system is 66.4%.	68
4	UF-NF-MSF-Crystallization	NaCl	Total desalination cost: 0.946 $/m^3. Salt sale: 0.51 $/m^3 of desalinated water.	69
5	UF-NF-RO-MSF-Crystallization	NaCl	Total desalination cost: 0.883 $/m^3. alt sale: 0.596 $/m^3 of desalinated water. Water recovery is 77.2%.	70
6	MF-NF/MCr-RO/MCr	$CaCO_3$; NaCl; $MgSO_4.7H_2O$	Total desalination cost: 0.73 $/m^3. Salt sale: 0.86 $/m^3. The overall water recovery up to 93%.	72
7	MF-NF/MCr-RO/MD	$CaCO_3$; NaCl; $MgSO_4.7H_2O$	Total desalination cost: 0.74 $/m^3. Salt sale: 1.23 $/m^3. The overall fresh water recovery: up to 87%.	
8	Precipitator-MF-NF-RO-ED-MED-Evaporator/crystallizer	$CaCO_3$; $MgSO_4.7H_2O$ $Mg(OH)_2$; $MgCl_2$; NaCl	Total desalination cost: 2.48 $/m^3. Salt sale: 3.46 $/m^3.	74
9	Precipitator-RO-evaporation pond	$CaCO_3$; $MgCO_3$; $Mg_3(PO4)_2$; Struvit; NaCl	-	75
10	RO-precipitator-ion exchange-evaporation pond	$CaCO_3$; $MgCO_3$; $Mg_3(PO4)_2$; Struvit; NaCl	-	
11	RO-ED-evaporation-electrolysis	Edible salt; Bittern; Cl_2, H_2, and NaOH are produced by electrolysis.	-	76

Integrated SWRO-PRO and SWRO-RED are promising process to alleviate water and energy demands.[89, 90] It is found from the study that the brine from SWRO desalination provides a better high salinity source for energy recovery. Moreover, the discharged brine can be controlled for minimizing the impact on the environment. In addition, the specific energy consumption of the RO system can be reduced. Comparison of PRO and RED performance during energy recovery from mixing of different type of saline water, i.e. seawater and brine, with river water has been reported.[91] From the study, RED could produce 2-4 W/m^2 of maximum power density while PRO is 1.2-1.5 W/m^2 when processing seawater and river water. In processing brine water and river water, PRO seems to be more attractive with higher power density and energy recovery compared to RED. By using these methods, not just electricity can be generated but the salt concentration of the brine can also be reduced.

CONCLUSION

In desalination, a large volume of rejected brine is generated during the production of freshwater from saline water (such as brackish water and seawater). The concentrated or rejected brine contains a high concentration of salt and also chemicals imparted during the desalination operations. Due to environmental impacts arising from improper treatment of the brine and more rigorous regulations of the pollution control, many attempts have been conducted for minimization, treatment, or reuse the brine. One of the most promising alternatives for brine handling is reusing the brine which results in pollution reduction, minimizing waste volume, and salt recovery. Integration of desalination and salt production can be applied to reuse the brine by recovering water and valuable salts. The integrated processes can achieve zero liquid discharge and a higher water recovery while the profitable salt is produced which can be used to reduce the overall water production cost. The brine may also contain metals on other valuable components which can be recovered. It is believed that the integrated membrane-based desalination process will be the preferred method in the future for the dual-purpose plant due to their promising properties of mild operation temperature thus saving vast amount of energy, high quality product, and combined with high productivity. However, there is still a lot effort to be placed on the research for development of the prospective integrated process that may produce water and salt in desalination system that is low energy consumption, low chemical consumption, and low cost.

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