

A Review on Recent Progress in Membrane Distillation Crystallization

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Abstract

Membrane distillation crystallization (MDC) is a promising hybrid separation technology that can play an important role in desalination, mineral recovery from liquid solution as well as in carbon dioxide fixation. MDC combines membrane distillation and crystallizer into one integrated unit that allows excellent recovery of clean water and high purity salt from highly concentrated salts solution (i.e., brine), which is otherwise detrimental when discharged to the environment. The process intensification addresses the limitation of standalone

membrane distillation and a standalone crystallizer (i.e., temperature and concentration polarization, membrane properties) when operated as individual technology. This review discusses the fundamental of MDC focused on how the process intensification addresses those standalone units' limitations. Later, MDC's potential applications in addressing some pressing issues such as water scarcity and climate change are also evaluated. Lastly, current trends in the MDC research are discussed to project the required future developments.

Keywords: Crystallization, Desalination, Membrane crystallization, Membrane distillation, Membrane distillation crystallizer

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1 Introduction

Over the past few decades, freshwater scarcity has become one of the major issues due to the rapid increment of the population as well as industrial activities. Even though Earth's surface area is covered with 71 % of water, the challenges to meet the demand of humans, animal and plants towards freshwater are still high [1]. It is reported that only 2.5 % of the total water quantity is freshwater, and only 0.008 % are accessible surface freshwater. Meanwhile, the rests are glaciers, ice caps, and groundwater [2]. As estimated by Seckler et al. [3], more than 1.4 billion people will face absolute water scarcity by 2025, and about 73 % of the affected are Asian. Water scarcity is defined as a condition when the water supply is below 1000 m³ per person per year [4]. Many technologies had been introduced to overcome this issue. In this context, the desalination of saline water has been immensely important in fulfilling the demand for high-quality fresh water as well as sustaining the water resources [5, 6].

Desalination is the process of producing potable water by removing dissolved salts and minerals from saline water. The permissible limit of salinity in potable water is 500–1000 ppm [7]. Desalination is an energy-intensive process as it requires around 10 000 tons of fossil fuel per year to produce 1000 m³ water per day [8]. It is estimated that 10 tons of carbon dioxide

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or 5000 m³ of greenhouse gases are produced with every 5 tons of fossil fuels consumed [9]. Substantial increment in total global desalination capacity is reported within the last few years, from 66.48 million m³ per day in 2011 to 86.6 million m³ per day in 2015. It is expected to rise to over 100 million m³ per day in 2020 [10, 11]. The substantial increment of desalination capacity may lead to high production costs as high energy is consumed. In contrast, fossil fuel use leads to greenhouse gas emissions promoting global warming and climate change.

Several well-known commercial desalination technologies are currently being used, namely solar still distillation (SD), multistage flashing (MSF), multi-effects distillation (MED), reverse osmosis (RO), and electrodialysis (ED). SD has advantages in terms of energy requirement, environmentally friendly, investment and maintenance cost. However, this system is relatively low in productivity per unit area [12]. MSF is easy to be managed and operated and can treat very high salt concentrations of up to 70 000 mg L⁻¹ [13]. However, it cannot be performed under 60 % capacity and consumes a lot of energy, about 3 to 5 kWh m⁻³ for electricity and 233 to 258 MJ m⁻³ for heating [13]. MED is highly flexible in operational capacity but required anti-scalants to avoid scaling [14]. RO is the most well-known system among others as the plant size can be adjusted to meet the demand, remove silica, and low cost in treating brackish groundwater [15]. However, RO is limited by a complex configuration, membrane fouling formation, and requires skilled personnel for operation and maintenance [15]. ED has the advantages of a high recovery rate, which can reach up to 94 %, longer membrane lifetime, reaching up to 15 years when appropriately operated and combined with RO to achieve higher water recovery of up to 98 %. However, it is still high in terms of capital cost compared to RO [16, 17].

Since most desalination technologies have limitations in energy consumption and environmental issues, many strategies have been developed to overcome these issues. One of the strategies involves process intensification (PIS). PIS has been used as a good practice of sustainability. Generally, it engages with several objectives: minimizing energy consumption, raw materials, environmental impacts, waste minimization, and maximization of product output [18]. Membrane distillation crystallization (MDC) is one form of PIS for greater sustainability [19]. MDC integrates membrane distillation (MD) and crystallization which offers high-quality freshwater and valuable resources (salts) from highly saline solution [20].

MD is a thermally driven process where the vapor of the feed solution's volatile components is transported across a porous hydrophobic membrane [21, 22]. It is favorable due to its high separation efficiency, ease of scale-up, suitable for the treatment of solution with high solute concentration, can be operated at low pressure, can work under moderate temperature and economically attractive when the heat can be obtained from waste stream [19, 23]. MD is particularly attractive when using low-grade heat as energy sources, as such heating energy can be omitted from the cost factor.

Like most other membrane processes, the MD process's main limitation is membrane fouling or (more precisely) membrane scaling [24, 25]. Scaling in MD can be described as precipitation and accumulation of solute on the membrane surface or inside the pores [26, 27]. After depositing on the membrane,

salt deposits grow into larger crystals, depleting the flux when treating feed with high solute concentrations. Besides reducing the flux, scaling promotes membrane wetting which complicates operation that eventually shortens the membrane life span [20, 24].

Meanwhile, crystallization is a solid-liquid separation process in which solid crystal precipitates from a liquid solution [28]. It is widely used mainly in chemicals, pharmaceuticals, food and the electronic industry [29, 30]. Despite its wide range of applications, crystallization has several drawbacks related to product quality. To date, since most of the full-scale crystallizers operate batch-wise, they are more likely to suffer from production inefficiency and overwhelmed workload. Hence, continuous operation is required to reduce the workload and increase efficiency [29].

Meanwhile, in terms of product quality issue, it is crucial to control the produced crystal properties. Other critical issues that affect the crystal quality include limited control of supersaturation, poor mixing, non-homogeneous distribution over the solvent removal, dosing of antisolvent, low possibility to modulate the rate of supersaturation and high energy requirement for heating in conventional evaporators or pumping in vacuum system [31, 32]. Important crystal properties include morphology, structure, and purity, significantly affecting the end products' quality [31].

By looking into MD and crystallization alone, one can see an intimate relationship. MD uptakes water as the main product in desalination when separating the solvent from a solution, leaving a more concentrated solution. When the solution reaches a certain degree of saturation, it creates a detrimental impact on MD, crystallizing the solute that eventually fouls the membrane. Conversely, crystallization aim for solute recovery by precipitating it to form pure crystals and most of the energy input is required to concentrate the solute to reach saturation. Based on the two processes' characteristics, the process integration between MD and crystallization is seen as a promising alternative to address their limitations.

In an MDC, the water concentration and the crystallization of salt occur simultaneously [33]. The MD brings about the feed solution to supersaturation by withdrawing the water to produce pure water as permeate. At the same time, the retentate is sent to the crystallizer column to create the crystal cake [23, 25]. It is claimed that operation as MDC can improve the limitations of the individual process (standalone MD and standalone crystallization) by achieving high water recovery, excellent controlled saturation ratio, high crystallization kinetic, low induction time, low fouling potential, and eventually reduce the environmental issue [19, 24]. Moreover, Quist-Jensen et al. [18] highlighted another advantage of MDC over the standalone crystallization of a high nucleation rate combined with a slow induction time. In addition, MDC can concentrate the feed solution up to a saturation point with minimum flux decline while managing membrane scaling properly [34, 35]. The most significant advantage of this integrated system is its ability to exploit both MD and crystallization processes' full benefits while reducing their limitations, as detailed later in this review.

In the past few years, Jiang et al. [23] reviewed the development of MDC process models, specifically crystallization con-

tol strategies. This review summarized the importance of MDC applications in crystal engineering and pharmaceutical manufacturing for the last five years. On the other hand, Salmon Ruiz et al. [36] discussed MDC regarding membrane performance and operating conditions. They also clarified the technical obstacles for the system and current technology related to MDC, including their applications and future development. In addition, Choi et al. [19] explained the basic concept of MDC as an emerging technology platform for brine mining and zero liquid discharge. They also addressed the technical challenges for MDC, such as scaling prevention and membrane wetting. Moreover, their review also includes different crystallization techniques that suitable for MDC operation.

Meanwhile, this review provides a thorough discussion of the fundamental, current status and perspectives on MDC's further developments. It also discusses the MDC's operational problems as an individual process that includes membrane fouling, poor crystal quality and limited supersaturation control, and how those issues interplay between MD and crystallization. This review also addresses how the process hybridization overcomes the limitations of individual MD and crystallizer standalone units. Besides, MDC's potential and feasibility as an alternative solution to the current pressing issues such as water scarcity and climate change are also addressed.

2 Limitations of Standalone Membrane Distillation and Standalone Crystallization

As mentioned earlier, MDC able to improve the limitations of the individual process (standalone MD and standalone crystallization) by achieving high water recovery, excellent controlled saturation ratio, high crystallization kinetic, low induction time, low fouling potential and eventually reduce the environmental issue [19, 24]. The limitation of standalone MD and standalone crystallization was further discussed in this section and was summarized in Fig. 1

2.1 Standalone Membrane Distillation

MD offers many advantages, namely low operating pressure (usually ambient pressure [37]), moderate operating temperature (30–70 °C [38], 20–80 °C [39]), low energy consumption (3–22 kWh m³, which can be discounted when waste heat is used [39]), environmentally friendly, and able to treat a feed with high solute concentrations with minor effect on the performance. In addition, high purity of permeate water can be produced when no volatile contaminant presents since only vapor can theoretically pass through the hydrophobic membrane. At the same time, it completely rejects the non-volatile compounds [19]. Many studies reported that for highly saline feed solutions (1–4 M NaCl), MD could achieve >90 % water recovery [40, 41].

Membrane fouling/scaling remains the most significant limitation that overwhelms its advantages, apart from heat source availability. It deteriorates the flux and permeates quality by promoting membrane wetting. When wetted, water occupies the available pore for vapor transport. When the whole pore is wetted, it allows diffusion of non-volatile components from the feed to the permeate side, which deteriorates rejection [42, 43]. Fouling and scaling also promote temperature and concentration polarization and, in the long run, may damage the membrane [44]. Among all of those drawbacks, membrane wetting is very critical because, when it happens, it fails the operation due to the need to dry the MD membrane to restore its performance [45].

Scaling on the membrane surface blocks the available pore for vapor permeation and contributes to heat transfer resistance from the bulk to the pore, which eventually lowers the flux [46]. On the other hand, membrane wetting allows penetration of the feed liquid either partially or fully into the membrane pore inflating the temperature polarization that depletes the vapor flux [47]. Those two significant problems remain the most challenging issues in MD [48]. Feed temperature is a dominant factor affecting the membrane fouling and scaling in MD. The risk of scaling increases at higher feed temperature due to the decrease in solubility of some solutes and its thermal

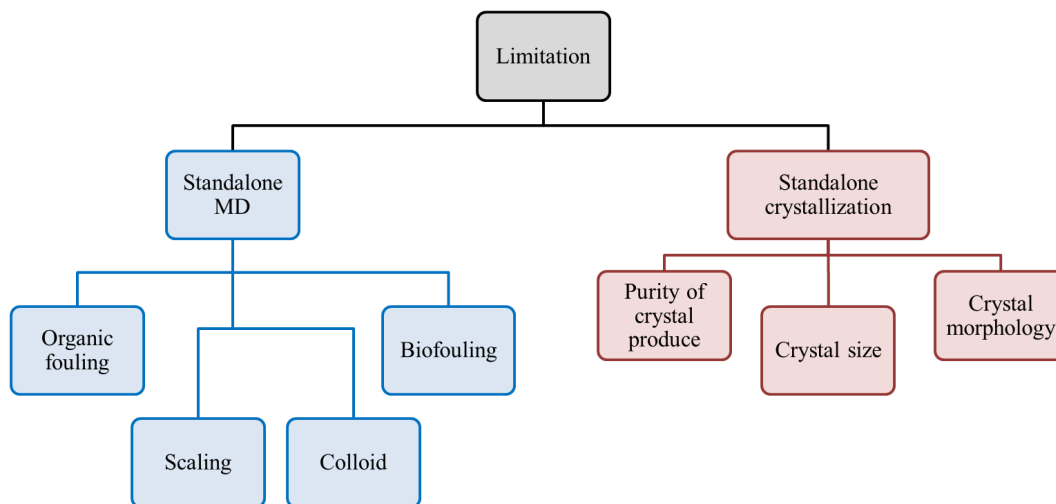


Figure 1. Limitations of standalone MD and standalone crystallization.

effects on organic compounds [44]. The type of fouling may vary with the feed properties.

In MDC, MD unit is required to perform optimally. MD's poor performance would slow down both the throughput and the rate of solute saturation in crystallization. A problem associated with membrane materials must be limited to avoid interruption of operation. For instance, membrane wetting would require stopping the operation from restoring the dry condition of membrane material strictly required by MD process.

There are four different types of foulant materials: organic, inorganic (scaling), colloid, and biofilm, which can reside solitary or simultaneously, as illustrated in Fig. 2. The nature of fouling is strongly affected by the composition of the feed. For instance, lake and river water typically contain high silica and calcium, a high concentration of biological compounds, and suspended solids compared to seawater [49]. Seawater, on the other hand, often shows a relatively high propensity of fouling for calcium sulfate, calcium carbonate, possible biological compounds and organisms compared to other surface waters, as well as a significant dry-out concern from very high levels of sodium chloride [44, 50]. The subsequent sub-sections discuss in more details the foulant materials how they affect MDC.

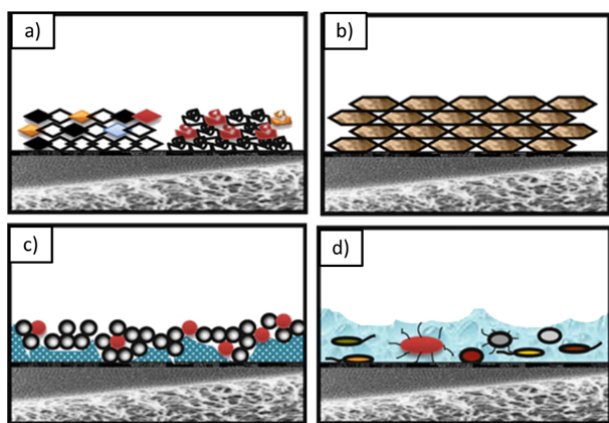


Figure 2. A schematic representative of membrane fouling; (a) organic fouling, (b) inorganic fouling, (c) colloidal fouling, and (d) biofouling [157].

2.1.1 Organic Fouling

Organic fouling is caused by organic matters in the feed solution that deposit on top of the membrane surfaces or within the membrane pores [51]. The presence of organics aids the formation of biofilm in the presence of microorganisms that survive the applied conditions (i.e., low-temperature vacuum MD) [52]. Organic fouling becomes a concern since its MD employs a hydrophobic membrane, a preferred surface for the foulant materials (typically hydrophobic) to adhere [53, 54]. In the context of MDC, organics in the feed solution may result in salts contamination. Therefore, pre-treatment for removal of organic is required. As the water is recovered from the solution for poorly soluble organics, increasing the mother liquor's organic concentration would promote precipitation. While for a highly soluble organic, its presence in the feed solution requires

purging to limit its concentration in the system. Purging lower both water and salts recovery.

2.1.2 Inorganic Fouling (Scaling)

Scaling is caused by solutes precipitation on the membrane surface or in the membrane pores. Precipitation occurs as the solute concentration exceeds saturation, at a point where the solute is more stable in the form of the crystal rather than ions. When dealing with a mixture of solutes, solutes of low solubility (i.e., MgSO_4 , CaSO_4) are likely to precipitate first to initiate the scaling. The primary factor affecting scaling is the availability of nucleation sites on the membrane surface. Minerals can quickly deposit on nucleation sites which eventually stimulate the growth of microcrystals. Hence, it may promote damage to the membrane's selective layer (i.e., by enlarging the pores) [51, 55]. The concentration gradient is created when MD and crystallization is integrated as one system, in which the nucleation sites are concentrated in the crystallization zone. Therefore, the segregation of zone between MD and crystallizer would indirectly enhance MD unit resistance from scaling.

2.1.3 Colloidal Fouling

Colloidal fouling is caused by the accumulation of colloidal particles on the membrane surface, worsening if particles smaller than the pore mouth entrapped inside the membrane pores. Organic colloids have large molecular weight, while inorganic colloids have compact and rigid particles [55]. Examples of the particles that would contribute to colloidal fouling are the clay, iron oxide and, silica particles, macromolecules, bacteria and viruses [44]. They typically have sizes in the range of 1 to 1000 nm [15, 55].

2.1.4 Biofouling

Biofouling is caused by biofilm formation, where microorganisms form a colony and attach to the membrane's surface. The microorganisms build up their gel-type diffusion barrier layer known extra-cellular polymeric substances [26, 56]. Due to the high feed temperature in MD, biofouling is less likely to happen. However, for the feed with low temperature like in the vacuum MD, it has biofouling potential. Biofilm reduces the MD performance as it becomes the additional transport resistance and induces temperature polarization or pressure drop [26, 57]. Biofouling is particularly challenging because it is difficult to control. The microorganisms increase rapidly as they can grow and multiply quickly in the presence of ample nutrients [55, 57]. When combined with crystallization, biofilm is detrimental for MD and can severely affect the crystal quality. Therefore, the application of high feed MD shall be preferred with feed temperature beyond the tolerance of microorganism to grow.

2.2 Standalone Crystallization

Crystallization is a fundamental and straightforward process for the separation and purification of solid products from a liquid solution; it is widely used in the chemicals, pharmaceuticals and food industries as well as for water and wastewater treatments [19,23]. It offers higher resources and water recovery ratio [19]. It is based on a solid-liquid separation process using control of solubility. A pure solid crystal is produced and extracted from the liquid solution [19,24]. In crystallization, the quality of the solid products is defined by the purity, polymorphic form, crystal shape, and crystal size distribution [58]. The driving force for crystallization is supersaturation. The generation of supersaturation can be achieved by controlling the saturation level of the process, lowering the solute's solubility or removing solvent from the solution [59].

Despite the advantages mentioned, a number of limitations faced by a standalone crystallization unit have been reported [60], including the low purity of the product due to impurities and additives, polymorph, solvate screening and small surface area for evaporation. Those limitations may affect the crystallization in terms of poor reproducibility of the final crystals' characteristics, limited control of supersaturation and less ability to modulate the supersaturation generation rate [20,24]. Discussion on each limitation is further elaborated in Sects. 2.2.1–2.2.3. Industrial crystallization techniques often used evaporative crystallization to overcome limitations of the standalone crystallization [61–63] to allow continuous and large scale production [64]. In evaporative crystallization, the solution is removed via evaporation of the solution to the point where the precipitation of solids exceeds its solubility limits. Like in an evaporator, only that the solution is boiled up to its solubility point until a solid slurry product is obtained [65]. Nevertheless, this process consumes high energy due to large latent heat [64]. In addition, the rate of supersaturation generation is often limited due to the limited surface area for evaporation in an evaporative crystallization [64]. Therefore, MDC could potentially overcome the issue of high energy consumption as well as a low rate of supersaturation generation when employing a low-grade heat at a relatively lower temperature that is more cost attractive than evaporative crystallizer. Detailed explanation on MDC was elaborated in Sect. 3.

2.2.1 Crystal Purity

The most significant limitation for crystallization is maintaining the purity of the crystal product [66]. Operating variables such as draw temperature, dasher speed, residence time, and throughput rate affected the size distributions of the produced crystal. Operations at low draw temperature, short residence time, and high dasher speed and high throughput result in smaller crystals' formation [66]. Hence, it is imperative to apply suitable operating conditions to obtain a high-quality product. The crystallizer needs to produce high-quality products as it is often used either for water purification, water discharge, or cooling purposes. An understanding phase diagram is the key to develop an appropriate crystallization process with a desirable product specification. Phase diagram maps the

thermodynamic properties of materials under certain range of parameters (solubility – temperature) [60,67]. However, the phase diagram of actual materials is rare. The phase diagram is complex, and experiments to compose the diagrams will be time-consuming and costly [67,68].

2.2.2 Crystal Size

As mentioned in the previous subsection, membrane fouling is the biggest limitation for standalone MD. Hence, it has been found out that many studies highlighted the use of non-membrane-based technology such as freeze crystallization (FC) to overcome that limitation [69]. FC is solid-liquid separation involving separation of ice and highly soluble dissolved salts concentrate [70,71]. However, the major drawback of FC is the formation of a huge amount of small-sized ice-crystals, which results in a high specific surface area (surfaces covered by a slew of salts due to solid adhesive force between ice and salt concentrate), which entails washing to remove those salts [71,72]. Many potential ways were suggested to minimize the surface area of ice formed. According to Chang et al. [71] the optimum amount of washing water to clean raw ice is about half of the raw ice produced under optimal operating conditions of FC for seawater. It is found out that minimizing washing can avoid dilution of concentrate and increase FC performance. Recently, a new membrane-based technology has been introduced to overcome the limitation of standalone MD and FC. Qu et al [73] designed a MDC system known as vacuum membrane distillation crystallization (VMDC). They compared an VMDC and an evaporation crystallization (mentioned in Sect. 2.2). They reported that the crystal product obtained from the VMDC has more uniform size of particle compared to the ones produced from the evaporation crystallization.

2.2.3 Crystal Morphology

On top of the crystal's size, the crystal morphology is also affected by impurities and additives that affect the nucleation rate, crystal growth and stability, and the metastable zone [74,75]. Moreover, polymorph and solvate are listed as the main limitations. Polymorph is a substance's ability to form more than two crystalline phases with different molecules arrangements in their crystal lattice while retaining its chemical composition. Meanwhile, the solvate is a condition of incorporate molecules with one or more solvents [76,77]. The polymorph and solvate condition may cause destructive impacts on the produced crystal's physical properties, such as its habit, solubility, density, hardness, color, melting point, and reactivity [78,79]. Another limitation of this process is the limited area available for the evaporation process, necessitating a continuous process [20,24]. It is reported by Diprofito et al. [80] the supersaturation condition created by the membrane may affect the crystal size and morphology. Hence, crystal with uniform size and controlled morphology may be produced by applying MDC due to the homogeneous nucleation of crystals at the membrane pores where solvent removal occurred.

2.2.4 Crystal Kinetic (Nucleation and Growth)

Crystallization kinetics via the nucleation and growth mechanism would strongly affect the quality of the crystal product [81]. A comprehensive investigation reported that crystal nucleation and growth mechanism in MDC could improve the quality of the crystal product [73]. To date, only a study by Qu et al. [73] focused on the crystal kinetics for the $\text{MgSO}_4\text{-H}_2\text{O}$ system in MDC. $\text{MgSO}_4\text{-H}_2\text{O}$ system is often used in crystallization studies [82, 83] since magnesium sulphate is a common chemical in industries. They are often found from brines and reclaimed as magnesium salt [84, 85]. Thus, the $\text{MgSO}_4\text{-H}_2\text{O}$ system is appropriate to investigate the performance of MDC in terms of crystal nucleation and growth. Qu et al. [73] evaluated the crystal nucleation and growth in MDC through the population balance equation and compared it with evaporation crystallization. Results showed that the size of the crystal in MDC was slightly smaller than the ones obtained from evaporation crystallization due to the reduction in collision frequency between the crystals in MDC [73]. Further investigation on the performance of MDC for crystal kinetic are required to improve the performance of MDC in terms of crystal nucleation and growth.

3 Membrane Distillation Crystallization

3.1 Principle of Membrane Distillation Crystallization

MDC principles involve simultaneously concentrating sub-saturated solute in the feed solution and imposing a high and a low-temperature zone. The high-temperature zone is required for MD, and the low-temperature zone is required for crystallization. The gradient of temperature is achieved through the heat transfer process, in which external heating is provided for MD and external cooling is provided for the crystallization [86]. In MD, the temperature difference between the feed and permeate side across a microporous hydrophobic membrane drove the vapor transport. In MDC, the vapor mass transport accompanied by solid rejection promotes the solute's concentration process, bringing it closer to the saturation point [87, 88]. Further increase in the solute concentration would reach the solute saturation from which further uptake of the solvent (via permeation in MD) triggers the precipitation/crystallization of the solute [87]. Thus, it is crucial to create a condition with significant concentration and temperature differences to produce high-quality permeate and solid crystal with maximum recovery [24].

In conventional standalone MD and standalone crystallization (Fig. 3), both processes are run simultaneously. In MD, the hot feed is pumped through the MD module, from which the clean water permeate is withdrawn via multiple mechanisms (direct-contact, vacuum, air gaps, etc.) [42]. The MD retentate is sent as the crystallization unit's feed where its temperature is lowered to reach supersaturation and induce crystallization [89, 90]. The crystallization process's mother liquor is recycled back, combined with the fresh feed and heated before fed back to MD [91]. This operation mode is applied when the solute

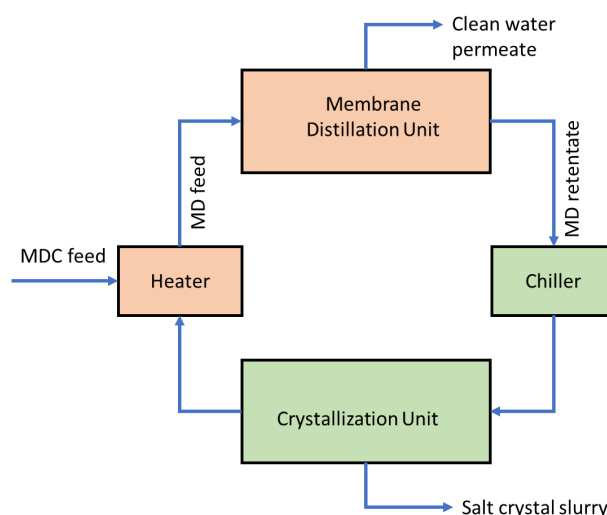


Figure 3. The process flow of the simultaneous operation of (simplified) continues MD and crystallization. The process for MD is simplified by not showing various option to draw permeate depending on the process configurations (vacuum, direct-contact, air gaps, etc.).

concentration in the feed solution is near the saturation concentration. This operation allows almost complete recovery of water and salts without leaving any waste stream. It is highly attractive when MDC treats a single solute but could result in low salt purity when MDC treats a solution with multiple types of solutes.

The combination of standalone MD and standalone crystallization can be further intensified in a hybrid MDC, as illustrated in Fig. 4. Such hybridization enables simultaneous pure

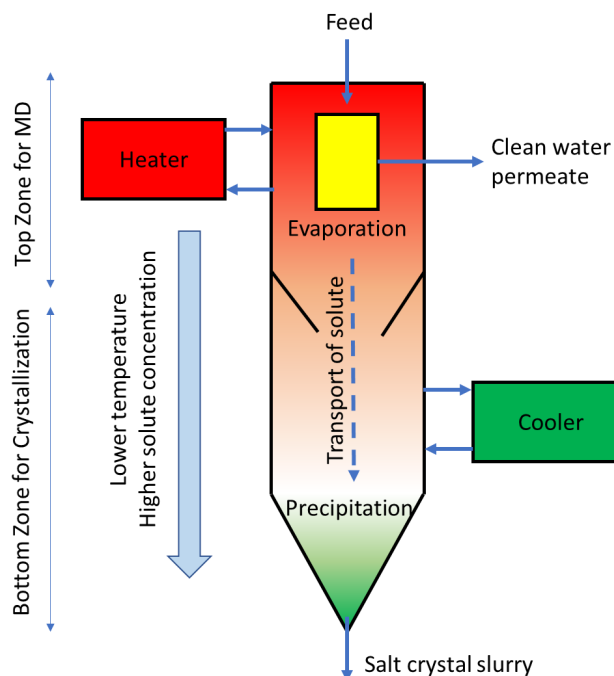


Figure 4. Configurations of an integrated MDC. The colors intensity shows the gradient of the solute concentration and temperature gradient in a combined column system.

water recovery and pure salt production in a single tank [24]. The tank is basically divided into two zones: the top and the bottom, to allow simultaneous operation of MD and crystallization in one shared tank/column. The top zone is used for the MD part. Meanwhile, the bottom zone is used for crystallization. The temperature of the fluid in the top zone is higher (to allow MD operation) than the one in the bottom zone (to allow crystallization) [20]. The temperature gradient is promoted by heating the feed on the top zone. Since part of the liquid evaporates, the sensible heat is transferred into the latent heat, turning the liquid to be colder. On the other hand, the liquid's temperature in the bottom zone is lowered to promote crystallization/precipitation of salts harvested from the stream at the bottom of the column. The difference in temperature (hot at the top and cold at the bottom) drives salts' mobility from top to bottom and promotes a continuous process.

By considering the MDC's operational principle, the supply of heat at the top column must be maintained for pure water recovery via MD and to withdraw heat at the bottom column for pure salt production via crystallization [24]. It is essential to maintain the gradient in salt concentration to achieve supersaturation salt condition [24]. The ability to maintain the temperature and the solute concentration gradient in the MDC process enhances the performances of the standalone methods, increases the efficiency of pure water and salt recovery, and strengthens the difference of the saturation level of the solution enhances the crystallization rate [24].

3.2 Operational Parameters of Membrane Distillation Crystallization

3.2.1 Volumetric Concentration Factor

The volume concentration factor (VCF) plays a vital role in MDC as it measures the level of water recovery in MDC [92]. VCF is a ratio of the initial volume of the feed to the final volume of the concentrate after recovery of water [93], as shown in Eq. (1).

$$\text{VCF} = \frac{\text{Initial volume of feed}}{\text{Final volume of concentrate}} = \frac{V_0}{V} \quad (1)$$

A variation of VCF with time and the relation with flux was reported by Chhaya et al. [93]. The flux measured showed decrement as time increase; meanwhile, the VCF showed increment as a function of time [93]. Hence, it can be concluded that the relationship between flux and VCF is inversely proportional. However, in the same study, Chhaya et al. [93] reported that the relation between VCF and pressure is linearly proportional. From the results obtained in their study, the amount of VCF increased when operated under high pressure as more permeate is being filtrated [93]. According to Lok et al. [94], VCF in membrane application is directly associated with fouling formation rate. This statement was inline with the report of Chhaya et al. [93] where higher VCF would contribute to flux decrement due to the high formation of fouling.

It is reported that for applying the fractional-submerged MDC (F-SMDC) process, low flux reduction can be achieved

at a higher VCF of 3.5 [20], [24]. A modelling approach using solubility software (PHREEQC) has been used to estimate MDC's capability in terms of the crystallization process for treating the brine of reverse osmosis [92]. The results reveal that halite, NaCl and brucite, Mg(OH)₂ start to precipitate at VCF of 4.4 and 10.99, respectively. Meanwhile, glauberite (Na₂Ca(SO₄)₂) begin to precipitate at a VCF of 7.3 [92]. The knowledge of the precipitation condition enables process design to produce multiple salts/minerals at different VCF. This topic is fascinating to explore to enhance MDC's competitiveness and enhance its chance for full-scale implementation.

3.2.2 Feed Solute Concentration

The solute concentration in the feed significantly affects MDC performance [24]. It is desirable to maintain the liquid in the top zone at a lower solute concentration to ease the MD process from scaling, especially when treating feed at a high solute concentration [95]. The scaling in MD can deteriorate the performance, which also detriment the overall MDC. According to Alsahy et al. [96], the permeate flux and rejection decreased when the feed concentration is high, as shown in Fig. 5. It is reported that the leading cause of this finding is the increment in osmotic pressure which encouraged the formation of concentration polarization [96]. Concentration polarization occurred due to a boundary layer of higher concentration on membrane surface than the concentration in bulk solution, which causes by convective transport between solute and solvent [96].

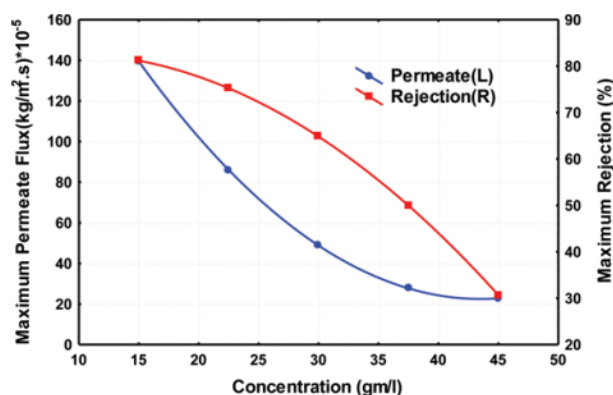


Figure 5. Effect of feed solute concentration on permeate flux and rejection [96].

On the other hand, the formation of salt crystallization at the bottom zone is promoted by a high solute concentration [24]. Depending on the liquid's thermodynamic condition in the bottom zone, the solute can reach a supersaturation concentration. For the MD process, the solute concentration affects the cation's electronegativity, which eventually affects the flux trend. An experimental study demonstrated that lower cation electronegativity allows sustained operation by maintaining the water flux over the prolonged process [24]. Conversely, at concentrations near-critical supersaturation, the flux shows gradual decrement [25]. This condition is due to the scaling, rapid

growth of crystal deposited on the membrane surface. Creusen et al. [33] stated that flux remained almost constant unaffected by the scaling effect during 4.5 days of MD operation when the system operates under 93–97% of saturated NaCl concentration in an MDC process with only pure water and dry salts as the products. To the best of our knowledge, none of the current research scope using feed solution with more than one type of salt.

3.2.3 Feed Temperature

The temperature difference generated between the MD side (top zone) and the crystallization side (bottom zone) in MDC significantly influence overall performance. The permeate flux is linearly proportional to feed temperature, while the rejection is inversely proportional to feed temperature, as shown in Fig. 6 [96]. Cheryan [97] reported the permeate flux is higher at higher feed temperature due to the low viscosity of feed solution; hence, the solution became easier to pass through the membrane.

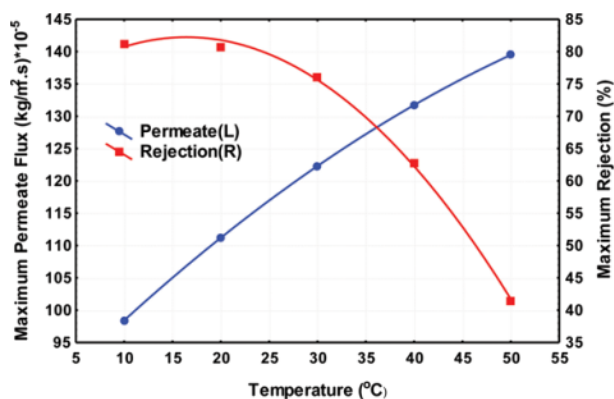


Figure 6. Effect of temperature difference on permeate flux and rejection [96].

Like the feed concentration, the temperature difference generated was also influenced by the cation's electronegativity [24]. Lower cation's electronegativity leads to lower flux decline. The bottom zone liquid temperature exhibits a vital role in controlling the salt solubility or saturation degree in the feed solution. Choi et al. [24] demonstrated that solute in the feed solution promotes heat transfer in a fractional submerged MDC. It shows that the temperature gradient created in the column favors the process performance. Effective crystallization at the bottom zone reduces the scaling by crystals at the top zone. Rapid crystallization draws solute mobility (driven by temperature difference) from the top zone and prevents it from precipitating/scaling on the MD membrane.

Creusen et al. [33] claimed that MDC could achieve high efficiency at high boiling point elevation around 6°C with electrical conductivity of below 10 $\mu\text{S cm}^{-1}$. Based on the Antoine equation, the vapor pressure exponentially increases with temperature, suggesting its role in affecting the MDC flux [98]. According to Tun et al. [25], flux increased alongside the vapor pressure. It is also reported by Quist-Jensen et al. [18] that the

highest flux was obtained when the most elevated vapor pressure was applied across the membrane in the MD system.

3.2.4 Membrane Properties

Membrane properties play a critical role in the MDC process as the membrane surface directly connects with the hot feed solution. The interaction of the membrane surface with the feed solution depends on the materials, morphology and fluid dynamics that occurred throughout the interaction [31]. The membranes used in MDC application should possess high chemical, thermal and mechanical strength [99], [100]. According to Edwie and Chung [90], it is recommended to use membranes with a more compact morphology substructure or smaller pore size with a macro void free structure for the MDC process involving a saturated feed solution.

Cui et al. [101] highlighted relationships between membrane properties and vapor flux and crystals growth. Higher surface porosity leads to a high crystal growth rate on the membrane surface [101]. The evaporation of the solvent near the pore mouth utilizes a large amount of heat that locally lower the film temperature and increases the likelihood of the total concentration of the solute to precipitate. As commonly well acknowledged in standalone MD, enhanced MD performance results in improved MDC performance as well.

As highlighted by Ko et al. [102] the membrane properties should exhibit high surface porosity, sharp pore size distribution and low thermal conductivity to enhance MDC performance. They reported applying ceramic membrane in MD for the MDC to cater to the polymeric membranes' limitation such as low thermal, mechanical and chemical resistance and has a shorter lifetime [102]. They evaluated two different hydrophobic ceramic membranes prepared using different methods [102]. One of the membranes was prepared by coating hydrophobic polymethylsilsesquioxane aerogels on alumina membrane supports via a sol-gel process. The membrane exhibited stable hydrophobic properties in MDC tests but deteriorated its flux performance. The other membrane was synthesized using combined phase-inversion and sintering methods by applying fluoroalkyl silanes (1H, 1H, 2H, 2H-perfluorooctyltriethoxysilane) hydrophobic agent at the relatively thin and more porous as-sintered alumina hollow fibers. It showed an improved average flux while maintaining its hydrophobic character throughout the performed tests.

3.2.5 Hydrodynamics

Another important parameter is the hydrodynamics near the membrane surface to provide adequate mixing on the liquid/membrane boundary layer. Poor mixing results in severe temperature gradient from the bulk to the film which eventually diminishes the MD flux [103,104]. When operated under a cross flow setting in an external MD module, the mixing comes from the feed flow across the module in the flow channel. When the module is submerged inside the top zone of the column, additional mixing is required. A recent approach for enhancing the local mixing near the top of the MD mem-

brane can be induced by applying patterned membranes [105,106].

4 Applications of Membrane Distillation Crystallization

4.1 Water and Salt Recovery from Highly Saline Water

Seawater desalination produces brine that impairs the aquatic ecology near the discharging point [107,108]. Brine is made from salt mining, RO plant for seawater treatment, and effluent treatment for industrial wastewater that contains high saline such as saline wastewater and hypersaline wastewater. The brine contains a high concentration of organic and inorganic matters as well as a high amount of chemicals such as coagulant, chemical washing agent and pH adjusting agent. Hence it needs to undergo concentration and purification before being disposed to the environment [20]. According to William [109], the main difference between brine and highly saline water is the total dissolved solids (TDS) in their properties. It is reported that for highly saline water, the amount of TDS is more than $10\,000\text{ mg L}^{-1}$; meanwhile, for brine, the amount of TDS is higher than $35\,000\text{ mg L}^{-1}$. Direct discharge of both solutions towards the environment had become a significant problem recently, as the environmental regulations become more stringent to protect the aquatic environment [33,92].

The typical treatment method used for seawater desalination is RO. However, the major drawback of the RO process is the disposal of brine (RO concentrate). Hence, the MDC process has been introduced to overcome this issue [19,20], particularly for post-treatment of the brine. This recent development prompts the implementation of zero liquid discharge (ZLD) in desalination [110,111], in which MDC can play an important role. MD work at ambient pressure and even better than conventional distillation processes in terms of rejection capacity of non-volatile compounds, operation temperature and footprint [112], most importantly only modestly affected by the solute concentration. For brine management, MDC can play crucial role. In MDC, the membrane acts as a tool to generate supersaturation condition prior crystallization [19]. This is possible due to solvent removal's simultaneous effect via evaporation and decrement of feed temperature [19].

In this regard, MDC shows promising potential in brine treatment. Tun and Growth [5] claimed that integrated nanofiltration with MDC could achieve flux of $15\text{--}20\text{ L m}^{-2}\text{ h}^{-1}$ with water recovery of 80 % and overall salt rejections of 95–97 %. The system achieved a flux of $4\text{ L m}^{-2}\text{ h}^{-1}$ at a higher water recovery of 95 % and increment of total dissolved salt from 19 200 to 212 899 mg L^{-1} when treating an RO brine [5]. Curcio et al. [113] stated that the rejection rate decreases from 99.6 to 99.1 %, and permeate flux decreases from 17.31 to $0.1541\text{ m}^2\text{ h}^{-1}$ when the solute concentration increases from 60 to 120 g L^{-1} for membrane crystallizer unit with the combination of nanofiltration and evaporative crystallization. It was also reported that the flux increased by 156 % when the feed temperature was increased from 30 to $40\text{ }^\circ\text{C}$ with an overall water recovery factor of 87 % [113], which aligned with a statement made by

Alsahly et al. [96] where the permeate flux increased as the feed temperature increased. These reports show that the rejection rate and permeate flux decrease at a higher solute concentration in the feed because of the need to overcome the osmotic pressure by the pressure-driven membrane process (nanofiltration).

It is also proven by Li et al. [114] that the mass transfer coefficient also dependent on the feed concentration. The mass transfer coefficient reached the highest value of $1.25 \times 10^{-10}\text{ m Pa}^{-1}\text{ s}^{-1}$ at 60 g L^{-1} concentration of Na_2SO_4 and 100 g L^{-1} concentration of NaCl. It reached the lowest value, $5.57 \times 10^{-11}\text{ m Pa}^{-1}\text{ s}^{-1}$ at 180 g L^{-1} concentration of Na_2SO_4 and 300 g L^{-1} concentration of NaCl [114]. When employing MD for solute concentration within sub-saturation, the feed solution's impact is modest [115].

4.2 Water and Mineral Recovery from Shale Gas Produced Water

Shale gas produced water is an aqueous waste produced from shale gas extraction, known as an emerging energy source, that can prolong fossil fuel usage by several decades [116]. Shale gas is a natural gas trapped within the shale formation and was first categorized as unconventional natural gas due to its low economic feasibility upon its extraction process [116]. As the technology began to emerge rapidly, the shale gas extraction process has been advancing, resulting in more recoverable global gas reserves than 40 % since 2010 [88]. Based on environmental impact assessment's prediction in 2016, shale gas production would make the most outstanding contribution as the amount to be produced is estimated to be tripled from 2015 ($97 \times 10^8\text{ m}^3$) to 2040 ($157 \times 10^8\text{ m}^3$) [117].

Unfortunately, in 2013, it is reported that approximately around 8000 to 27 000 m^3 wastewater beings generated from shale gas extractions [118]. The waste produced contains high concentrations of salts, metals, oil, grease, and organic compounds, which cause harm to human health and the environment [88,118]. Many technologies have been introduced as shale gas produced waste treatments, but the drawbacks of these conventional technologies are expensive, the high toxicity of chemicals used and the large footprint [119,120]. However, a recent study shows interest in the use of membrane process offering significant advantages and inexpensive operation and low usage of chemical additives [119]. The only challenge of using membrane is the severe membrane fouling. Regarding that challenge, an integrated system between MDC shows higher efficiency by mitigating the membrane fouling formation [121,122].

According to Kim et al. [30], operation in hybrid MDC offer lower membrane fouling propensity in the MD process with an increment in total recovery by 62.5 % from shale gas produced water. The system is effectively recovering water and minerals by 84 % and 2.72 kg m^{-2} , respectively [30]. Kim et al. [116] claimed that critical temperature and cross flow velocity would affect the water recovery results. Under the critical temperature of 30, 40, and $50\text{ }^\circ\text{C}$, the obtained water recovery rates were 84, 80, and 76 %, respectively. This finding was able to be supported by Alsahly et al. [96] where rejection decreased with the

feed temperature. Meanwhile, under cross flow velocity of 0.1, 0.2, and 0.3 m s⁻¹, the water recovery rate obtained were 78, 84, and 75 %, respectively. Hence, it was concluded that the optimal critical temperature and cross flow velocity in view of quantitative water production was found to be at 30 °C and 0.2 m s⁻¹, respectively.

MDC has also been explored for water and salt recovery from produced water with promising results. Integration of microfiltration [123] or ultrafiltration/RO [124] as the pre-treatment stage has proven beneficial in enhancing overall performance. Nonetheless, it is worth noting the significant contribution of energy for the MD process from the treatment of produced water. Techno-economic analysis of employing standalone MD for the concentration of produced water can be primarily lowered if the energy for feed heating is available freely. The implications of utilizing waste heat on the total cost of treating produced water using MD is \$5.70 m⁻³ feed, decreasing significantly to \$0.74 m⁻³ feed when MD is integrated with a source of waste heat [125].

4.3 Carbon Capture

Carbon dioxide is the most abundant and critical greenhouse gas as it contributes to the climate change. The tremendous rise of carbon dioxide concentration is reported to be 280 ppm from the pre-industrial era, reached the value of 379 ppm, in 1999, and recently reached 408 ppm in 2005 [126, 127]. About three-quarters of the anthropogenic emissions of carbon dioxide during the past 20 years, 30 % of them come from fossil fuel, and around 40 % of them come from power generation [126, 127].

An increment of 2.2 to 3.0 °C from 1961 to 1990 is due to the anthropogenic contribution towards natural climate change [126]. As reported by Flis [128], global carbon dioxide levels continue to rise from 2019 (4119 ppm) to 2020 (4145 ppm) despite the Covid-19 pandemic, which causes a slowdown of the industry and traffic all around the world. According to Vega et al. [129] the current carbon dioxide levels increase about 2.5 ppm every year. By 2025, the values are expected to exceed any significance in the last 3.3 million years. Carbon dioxide concentrations increase rapidly unless less carbon-intensive energy alternatives and energy-saving infrastructure using new materials and technologies are realized soon. Therefore, a method to capture and recover the carbon dioxide for further reuse or storage is currently being researched intensively to overcome this issue.

Membrane-based alkaline absorption has recently been proposed for the direct capture of carbon dioxide from the ambient air [130]. Alkaline absorption is an environmentally friendly process as it used alkaline solutions (inorganic reagents) [131]. However, this technology's main challenge is to obtain dried carbonate to recover the reagent, either reused directly as a reagent in the industry or converted back to its initial state [130]. MDC has been introduced as a potential alternative to produce sodium carbonate with high purity from salts solutions with various concentrations to overcome this challenge.

Ruiz Salmon et al. [126] designed MDC for carbon dioxide capture to obtain sodium carbonate decahydrate (Na₂CO₃·10H₂O) as the solid product. The system's performance has been evaluated based on the mass and heat transfer coefficients of about 4.34 × 10⁻¹² to 6.53 × 10⁻¹¹ m Pa⁻¹s⁻¹ and 255.75 to 262.46 W m⁻², respectively [126]. Ye et al. [131] mentioned that the transmembrane flux decreases when the concentration of other compounds increases, indicating higher solute or impurities in the feed, thus reducing the driving force for the transport. The decrements of transmembrane flux for sodium nitrate are 1, 6.3, and 13 % for the concentration of 0.2, 0.4, and 0.6 mol L⁻¹ for sodium chloride are 1.7, 8, and 14.8 % while for sodium sulphate are 3.1, 10.4, and 16.4 %, respectively [131]. It is also reported that the purity of sodium carbonate crystals can reach up to 99.5 % by using an MDC [131].

Meanwhile, Ye et al. [132] reported that sodium carbonate crystal's product achieved 99 % purity by using MDC with a mixed solution of sodium sulphate and sodium carbonate. According to Salmon et al. [127], the highest carbon dioxide removal of 97 % could be achieved at the highest feed concentration of 1 mol L⁻¹ and gradually decreased to 80 and 50 % when the feed concentrations were reduced to 0.5 and 0.2 mol L⁻¹, respectively. This condition indicates that the higher the feed concentration used, the higher the efficiency of carbon dioxide removal.

5 Current Trend and Future Perspectives of Membrane Distillation Crystallization

Tab.1 compiles recent reports on MDC, which provides an overview of the state-of-the-art research. MDC has been emerging as an important topic since 2013, in which most reports demonstrated its effectiveness for simultaneously recovering the water and the minerals. The range of reported water recovery is from 80 to 95 %, while the salts recovery range is from 37 % [87] to salt recovery 100 % [133]. Moreover, MDC can achieve near ZLD, hence significantly reduce the impact on human health and the environment [134]. ZLD is a design approach focusing on recovering all water content leaving no wastewater [135]. It is found out that hybrid desalination systems have the advantages of high plant capacity, high quality of water production, flexibility in operation and low specific energy consumption [136].

A combination of NF-MDC was reported to achieve salt rejection of 99.6 %, allowing a high possibility to achieve near ZLD [113]. Several studies claimed that vacuum membrane distillation (VMD) and air gap membrane distillation (AGMD) are often chosen to commercialize MD processes [100] as both have high thermal energy efficiency [137]. However, it is mentioned by Ahmed et al. [136] that the integration of energy systems in hybrid and multi-hybrid desalination technologies is still in its early stages and unexplored. Several studies have been reported about renewable energy sources for desalination [1, 138], however, only limited studies focus on the energy requirement for renewable energy resources for desalination and their comparisons with conventional energy resources

Table 1. Summary of reports for membrane-based and MDC processes for various applications.

Type of configuration	Feed solution	Operational data	System performance	Energy consumption	Ref.
MF-NF-MD-crystallization	Industrial effluent	Feed temperature: 60 °C Permeate temperature: 20 °C	Flux: 15–20 L m ⁻² h ⁻¹ Water recovery: 80 %	72.5 kWh m ⁻³	[5]
MD + crystallization	Industrial effluent	Feed temperature: 37.4 °C Distillate temperature: 24.1 °C Feed flow rate: 200 L h ⁻¹	Flux for feed with Ca and Mg: 0.0005 L m ⁻² h ⁻¹ Pa ⁻¹ Flux for feed with only Na ₂ SO ₄ : 0.0005–0.0015 L m ⁻² h ⁻¹ Pa ⁻¹	–	[18]
RO-MCr	NaCl and MgSO ₄ ·7H ₂ O solution	Feed temperature: 20 °C, Feed pressure: 5.5 MPa, Feed flow rate: 432 000 m ³ d ⁻¹	MCr recovery rate: 98 % RO recovery rate: 50 % RO salt rejection: 99.6 %	27.3 kWh m ⁻³	[25]
NF-MDC	Sulfate waste	Feed temperature: 30 to 40 °C	Rejection rate: 99.1–99.6 % Flux increment: 156 % Water recovery: 87 %	–	[113]
Fractional MDC	NaCl, KCl, NH ₄ Cl, Na ₂ SO ₄ , and MgSO ₄	CG ratio for inorganic compounds: 1.51, 1.83 CG ratio for lower molecular weight compound: 0.94, 1.46	Flux reduction: 12.56 % (Na ₂ SO ₄), 55.93 % (MgSO ₄)	–	[24]
DCMD + Crystallization	Raw shale gas produced water	Critical temperature: 30 to 50 °C	Salt production rate: 2.72 kg m ⁻² d ⁻¹ Recovery rate: 84 %	28.2 kWh m ⁻³	[30]
MDC + MD-OD	Concentrated CaCO ₃ solution	Transmembrane temperature: 12 °C Feed temperature: 64 °C	Flux: 6 L m ⁻² h ⁻¹ bar ⁻¹	–	[33]
MDC	Produced water	Feed temperature: 35 to 55 °C Permeate temperature: 10 °C	Salt production: 354 258 mg L ⁻¹ Salt recovery: 37 %	–	[87]
DCMD + crystallization	Synthetic shale gas produced water	Feed temperature: 60 °C Permeate temperature: 20 °C	Salt recovery: 62.5 %	–	[116]
SMDC	Raw seawater	Feed temperature: 70 °C	Flux: 5–20 kg m ⁻² h ⁻¹	–	[122]
MCr	Vitrofur solution	Feed temperature: 20 °C	Flux: 0.0012–0.0025 L m ⁻² min ⁻¹	–	[131]
DCMD + crystallization	LiCl solution	Feed temperature: 52 °C Permeate temperature: 20 °C	Salt recovery: 100 %	–	[133]
DCMD + crystallization	Na ₂ SO ₄ solution	Feed temperature: 70 °C Permeate temperature: 25 °C Feed and permeate flow rate: 2 L h ⁻¹	Salt recovery: 80 % Salt production: 100 kg m ⁻³	–	[148]

Table 1. Continued.

Type of configuration	Feed solution	Operational data	System performance	Energy consumption	Ref.
RO-MDC	(NH ₄) ₂ SO ₄ solution	Feed temperature: 15 °C Permeate temperature: 50 °C	Retention factor: 0.974 %	–	[149]
DCMD + Crystallization	Seawater	–	Recovery factor: 92.8 %	1.6–27.5 kWh m ⁻³	[150]
MDC	NaCl solution	–	Flux: 6–16 L m ⁻² h ⁻¹	–	[151]
DCMD	Synthetic seawater solution	Trans-membrane pressure: 30 to 55 bar Temperature: 30 to 50 °C	Flux: 1.14–2.05 L m ⁻² h ⁻¹	–	[152]
MDC	NaCl solution	–	Flux: 4–35 kg m ⁻² h ⁻¹ Water recovery factor: 92.8 %	–	[153]
RO + MCr	Standard seawater	Feed temperature: 20 °C	Flux: 0.0035–0.006 kg m ⁻² min ⁻¹	–	[80]
MDC	NaCl solution	Feed temperature: 25 °C	Flux: 0.1–0.3 kg m ⁻² h ⁻¹	–	[154]
MCr	Fumaric acid	Feed temperature: 15 °C Retentate temperature: 14 °C Permeate temperature: 32 °C	Flux: 0.60–0.65 kg m ⁻² h ⁻¹	–	[155]
MCr	NaCl solution	Trans-membrane temperature: 25 °C	Flux: 0.024–0.09 L m ⁻² h ⁻¹	–	[156]

Type of Configuration – MDC: Membrane Distillation Crystallizer; DCMD: Direct Contact Membrane Distillation; RO: Reverse Osmosis; MF: Micro-filtration; NF: Nanofiltration; OD: osmotic distillation; SMDC: Simultaneous Membrane Distillation Crystallizer; MCr: Membrane Crystallizer; MaC: Membrane-assisted Crystallizer; MD: Membrane Distillation, FC-VMD: Freeze Crystallization with Vacuum Membrane Distillation; Materials – NaCl: Sodium Chloride; KCl: Potassium Chloride; NH₄Cl: Ammonium Chloride; Lithium Chloride: LiCl; Na₂SO₄: Sodium Sulfate; MgSO₄: Magnesium Sulfate; CaCO₃: Calcium Carbonate; MgSO₄·7H₂O: Epsomite, Ca: Calcium; Mg: Magnesium; CaSO₄·2H₂O: Calcium Sulphate Dihydrate; (NH₄)₂SO₄: Ammonium Sulfate.

[139,140]. For example, integrating the MD hybrid system with solar thermal energy could be beneficial for its performance. However, the primary constraint is the cost of heat supply would affect the entire system's operational costs [141].

MDC is still immature and thus still requires extensive research on the performance indicators, optimization of operating parameters, analysis of scaling mechanism, and estimation of energy consumption [30]. Despite the promising results reported so far, further studies on performance efficiency and economic viability for full-scale MDC are required to gauge its true potential. Furthermore, the MDC process's most significant drawback is the high energy consumption [134], mainly to supply heat in the MD. According to You et al. [142], the energy consumption in MDC is about 97.8 % of the total operation energy and mainly caused by the heat requirement due to the temperature difference in MDC [143]. Energy also required for a cooling system to allow crystallization [89, 121]. However, Pantoja et al. [144] reported that MDC's energy consumption could be reduced by increasing the membrane thickness. It is said that at the membrane thickness of 50 and 100 μm, the energy consumption for MDC are 1.3 and 1.2 kWh kg⁻¹,

respectively [144]. Further increment in membrane thickness (>100 μm) has no effect towards the energy consumption but would cause decrement in flux [144].

On the other hand, the energy consumption is found out to be reduced as the transmembrane flux increases under a turbulent flow [114]. In addition, increasing the feed flow rate while decreasing feed temperature would increase energy consumption. According to Elmarghany et al. [145], an increase in feed flow rate would reduce energy consumption from 2064 to 1037 kWh m⁻³. An increment in feed temperature would increase energy consumption from 689 to 1037 kWh m⁻³. Besides, freeze desalination and vacuum membrane distillation (FD-VMD) is also reported as an option to overcome the limitation of conventional desalination as it is suitable for high feed concentration [146]. The FD-VMD hybrid system requires cold energy and electricity. It is proven that FD-VMD is an energy-efficient system for seawater desalination, which has a high recovery of water (74 %) [147].

6 Conclusions

MDC is an emerging process that has been proven effective in recovering high-quality clean water and high-purity salt crystal with excellent yields. The process intensification that combines an otherwise standalone MD and crystallization to form MDC helps overcome the limitations of MDC operations of both processes standalone wise. A substantial amount of MDC research showed that this combination process has a high potential to treat challenging water scarcity issues and address climate change. The state-of-the-art MDC is still premature, thus requires extensive fundamental as well as applied research. Particular focus must be given to energy, sustainability, and life-cycle impact, as well as techno-economic analysis. The advancement of MD can take advantage of the more advanced development of microfiltration (MF) process and by focusing on system optimization to minimize the energy input.

Conflicts of Interest

The authors declare no conflict of interest.



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Abbreviations used

AGMD	air gap membrane distillation
CaSO ₄ ·2H ₂ O	calcium sulphate dihydrate
DCMD	direct contact membrane distillation
ED	electrodialysis
F-SMDC	fractional submerged membrane distillation crystallizer

FC	freeze crystallization
FC-VMD	freeze crystallization with vacuum membrane distillation
FD-VMD	freeze distillation and vacuum membrane distillation
MCr	membrane crystallizer
MD	membrane distillation
MDC	membrane distillation crystallizer
MED	multi-effects distillation
MF	microfiltration
Mg(OH) ₂	brucite
MgSO ₄ ·7H ₂ O	epsomite
MSF	multistage flashing
Na ₂ Ca(SO ₄) ₂	glauberite
Na ₂ CO ₃ ·10H ₂ O	Sodium carbonate decahydrate
NF	nanofiltration
(NH ₄) ₂ SO ₄	ammonium sulfate
OD	osmotic distillation
PIS	process intensification
RO	reverse osmosis
SD	solar still distillation
SMDC	simultaneous membrane distillation crystallizer
VCF	volume concentration factor
VMD	vacuum membrane distillation
ZLD	zero liquid discharge

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This review focused on how the MDC process intensification improves the limitations of those standalone units' (membrane distillation and crystallization). The potential applications of MDC in addressing some pressing issues of water scarcity and climate change are also evaluated. The current trends in the MDC research are also discussed to project the required future developments.

A Review on Recent Progress in Membrane Distillation Crystallization

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