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5	Corresponding Author	Family Name	**Duong**
6		Particle	
7		Given Name	**Hung Cong**
8		Suffix	
9		Organization	University of Technology Sydney
10		Division	Centre for Technology in Water and Wastewater
11		Address	Ultimo 2007, NSW, Australia
12		Organization	Le Quy Don Technical University
13		Division	
14		Address	Hanoi, Vietnam
15		e-mail	hungduongcong@gmail.com
16	Author	Family Name	**Ansari**
17		Particle	
18		Given Name	**Ashley Joy**
19		Suffix	
20		Organization	University of Wollongong
21		Division	Strategic Water Infrastructure Laboratory, School of Civil, Mining and Environmental Engineering
22		Address	Wollongong 2522, NSW, Australia
23		e-mail	
24	Author	Family Name	**Hailemariam**
25		Particle	
26		Given Name	**Ruth Habte**
27		Suffix	
28		Organization	University of Science and Technology (UST)
29		Division	Department of Civil and Environment Engineering

	Address	217, Gajeong-Ro, Yuseong-Gu, Daejeon 34113, Republic of Korea
	e-mail	
Author	Family Name	**Woo**
	Particle	
	Given Name	**Yun Chul**
	Suffix	
	Organization	University of Science and Technology (UST)
	Division	Department of Civil and Environment Engineering
	Address	217, Gajeong-Ro, Yuseong-Gu, Daejeon 34113, Republic of Korea
	Organization	Korea Institute of Civil Engineering and Building Technology
	Division	Department of Land, Water and Environment Research
	Address	283 Goyang-Daero, Ilsanseo-Gu, Goyang-Si 10223, Gyeonggi-Do, Republic of Korea
	e-mail	
Author	Family Name	**Pham**
	Particle	
	Given Name	**Thao Manh**
	Suffix	
	Organization	Le Quy Don Technical University
	Division	
	Address	Hanoi, Vietnam
	e-mail	
Author	Family Name	**Thi**
	Particle	
	Given Name	**Lan Ngo**
	Suffix	
	Organization	Le Quy Don Technical University
	Division	
	Address	Hanoi, Vietnam
	e-mail	
Author	Family Name	**Dao**
	Particle	
	Given Name	**Duong Thanh**
	Suffix	
	Organization	University of Science and Technology of Hanoi (USTH)
	Division	Department of Water, Environment and Oceanography

	Address	Hanoi, Vietnam
	e-mail	
Author	Family Name	**Nghiem**
	Particle	
	Given Name	**Long D.**
	Suffix	
	Organization	University of Technology Sydney
	Division	Centre for Technology in Water and Wastewater
	Address	Ultimo 2007, NSW, Australia
	e-mail	

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Abstract

Purpose of Review: Membrane distillation (MD) has been known as a promising water treatment process for many years. However, despite its advantages, MD has never been able to compete with other processes for industrial water treatment and supply. Instead, it has been orientated towards several unique strategic water treatment applications. This review aims to uncover the opportunities and technical challenges pertinent to the MD process and the current status of its strategic water treatment applications most notably including decentralised small-scale desalination for fresh water provision in remote areas, hybridisation with forward osmosis (FO) for treatment of challenging polluted waters, regeneration of liquid desiccant solutions for air conditioning, and treatment of acid effluents for beneficial reuse.

Recent Findings: Pilot and small-scale MD systems have been demonstrated for decentralised desalination using various renewable energy sources to supply fresh water in remote, rural areas and on ships where other desalination processes are inefficient or unfeasible. For this strategic desalination application, MD is technically viable, but more works on configuration modification and process optimisation are required to reduce the process energy consumption and water production costs. For the three other strategic applications, the technical viability of the MD process has been proved by extensive lab-scale researches, but its economic feasibility is still questionable due to the lack of large-scale evaluation and the uncertain costs of MD systems.

Summary: The orientation of MD towards strategic water treatment applications is clear. However, huge efforts are required to facilitate these applications at commercial and full scale.

Keywords separated by ' - '

Membrane distillation (MD) - Decentralised desalination - Draw solution regeneration - Liquid desiccant solution regeneration - Acid effluent treatment - Beneficial reuse

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Membrane Distillation for Strategic Water Treatment Applications: Opportunities, Challenges, and Current Status

Hung Cong Duong[1,2] · **Ashley Joy Ansari**[3] · **Ruth Habte Hailemariam**[4] · **Yun Chul Woo**[4,5] · **Thao Manh Pham**[1] · **Lan Ngo Thi**[1] · **Duong Thanh Dao**[6] · **Long D. Nghiem**[2]

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Keywords Membrane distillation (MD) · Decentralised desalination · Draw solution regeneration · Liquid desiccant solution regeneration · Acid effluent treatment · Beneficial reuse

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Introduction

For many years, membrane distillation (MD) has been known as a promising water treatment process. In 1963, the first patent on MD was licenced to Bodell, and several years later, Findley published the first research article demonstrating the great potential of the MD process [1, 2]. Since its first invention, MD has gone through a long development journey, with three different phases: initiation in the first 30 years since 1960, emergence in the subsequent 20 years, and rapid growth

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✉ Hung Cong Duong
hungduongcong@gmail.com

[1] Le Quy Don Technical University, Hanoi, Vietnam

[2] Centre for Technology in Water and Wastewater, University of Technology Sydney, Ultimo, NSW 2007, Australia

[3] Strategic Water Infrastructure Laboratory, School of Civil, Mining and Environmental Engineering, University of Wollongong, Wollongong, NSW 2522, Australia

[4] Department of Civil and Environment Engineering, University of Science and Technology (UST), 217, Gajeong-Ro, Yuseong-Gu, Daejeon 34113, Republic of Korea

[5] Department of Land, Water and Environment Research, Korea Institute of Civil Engineering and Building Technology, 283 Goyang-Daero, Ilsanseo-Gu, Goyang-Si, Gyeonggi-Do 10223, Republic of Korea

[6] Department of Water, Environment and Oceanography, University of Science and Technology of Hanoi (USTH), Hanoi, Vietnam

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in the last 10 years [1, 3]. These three MD development phases are clearly distinguished by their time span, research interest demonstrated by the numbers of publications, and particularly the trend in MD applications. While in the initiation phase, MD had been primarily applied for desalination [1, 3]; in recent years, the applications of MD have diversified and extended to areas beyond desalination such as brine concentration, recovery of critical resources, and removal of toxic compounds from water [1–7]. Through its long journey, MD has achieved significant development, but it has never been able to compete with other processes for industrial/commercial water treatment applications. Instead, MD has immense potential for some strategic applications where other water treatment processes are not technically or economically viable.

MD is a versatile thermal-based membrane process capable of treating various impaired waters to achieve fresh water of high quality [3, 8, 9]. As a membrane separation means, MD can offer a robust, modularised, and hence compact water treatment platform that can be operated as a stand-alone or combined process for improved treatment capacity or efficiency. Like other thermal distillation processes, MD can theoretically produce pure water from impaired hypersaline waters. Moreover, the MD process can be coupled with low-grade heat sources such as waste heat or solar thermal energy to reduce the operational costs of water treatment [10–14]. However, despite these notable advantages, the MD process is faced with several technical challenges that currently impede its competitiveness with other water treatment processes. As a result, the MD process has been orientated towards several unique strategic water treatment applications.

This paper aims to provide a comprehensive review of the strategic applications of MD for unique water treatment purposes. These strategic applications include decentralised small-scale desalination for fresh water provision in remote areas, hybridisation with forward osmosis (FO) for treatment of challenging polluted waters, regeneration of liquid desiccant solutions used for air conditioning systems, and treatment of acid effluents for beneficial reuse. For each strategic application, the opportunities and challenges pertinent to the MD process are elaborated, and the current status of the application is discussed. Based on these elaboration and discussion, the future research directions on MD for each strategic application are pinpointed.

Working Principles, Opportunities, and Technical Challenges of MD

Amongst many water treatment processes practised to augment fresh water supply worldwide, MD has emerged as a particularly promising means to mitigate the water-energy stress. The compatibility of MD with highly concentrated saline solutions and low-grade heat sources renders it an ideal process for strategic water treatment applications. This section will discuss some fundamentals of the MD process to highlight its opportunities as well as technical challenges relating to its strategic water treatment applications.

Unlike in pressure-driven membrane processes, in MD, the driving force for mass transfer through the membrane is the vapour pressure gradient induced by a temperature difference between two sides of the membrane. The mass flux in the MD process is expressed as below [15, 16]:

$$J = C_m\left(P_f - P_p\right) \quad (1)$$

where C_m is the membrane permeability and P_f and P_p are respectively the water vapour pressure at the feed and permeate side of the membrane. Compared with osmotic pressure, the water vapour pressure of the solution is much less subject to solution salinity. In other words, the mass flux of the MD process is significantly less affected by the feed solution concentration, giving the MD process the ability to treat highly saline waters with which the pressure-driven processes are incompatible [17, 18]. This intrinsic advantage renders MD an ideal process for the treatment of highly saline solutions such as reverse osmosis (RO) brine, FO draw solutions, and liquid desiccant solutions used in air conditioning.

As a thermally driven separation process, MD requires thermal energy (i.e. heating and cooling) to facilitate water evaporation and vapour condensation to achieve the process separation. Electrical energy is also consumed for water circulation in the MD process; however, it is negligible compared with thermal energy [19]. As a result, great numbers of MD studies focus on configuration and process optimisation to enhance the process of thermal efficiency. Largely, the thermal efficiency of the MD process is assessed using two parameters: specific thermal energy consumption (STEC) and gained output ratio (GOR). The calculation of STEC and GOR is as bellow [16, 19, 20]:

$$\mathrm{STEC} = \frac{Q_{\mathrm{input}}}{V_{\mathrm{distillate}}} \quad (2)$$

$$\mathrm{GOR} = \frac{m_{\mathrm{distillate}}\Delta H_v}{Q_{\mathrm{input}}} \quad (3)$$

where Q_{input} is the thermal energy input, $V_{\mathrm{distillate}}$ and $m_{\mathrm{distillate}}$ are respectively the distillate volume and mass, and ΔH_v is the latent heat of evaporation of water. While STEC represents the amount of external thermal energy consumed to obtain a volume unit of distillate, GOR indicates the proportion of useful thermal energy (i.e. the latent heat associated with the transfer of water vapour through the membrane) with the thermal energy input of the process. Given their definitions, STEC is used to evaluate the thermal efficiency of the MD process, whereas the use of GOR is largely relevant to the process with internal heat recovery [16]. It is also noteworthy that using STEC and GOR defined in the Eqs. (2) and (3) for the process thermal efficiency assessment is suitable for seawater desalination in which fresh water is the desired product; for strategic MD desalination applications, alternative indicators to STEC and GOR might be required. This will be discussed further in Section 3.

Considerable merit of the MD process is the ability to use low-grade waste heat or renewable energy to reduce its energy cost. While traditional thermal distillation processes require boiling the feed water, the MD process can be operated at mild feed temperature (i.e. as low as 40 °C) [21]. As demonstrated in Fig. 1, as long as a vapour pressure gradient is maintained across the two sides of the membrane, the mass flux (i.e. water for desalination applications) through the membrane can be achieved. The vapour pressure gradient across the membrane can be induced by heating the feed water while cooling the distillate, applying vacuum, or sweeping gas on the permeate side of the membrane [22, 23]. Thus, the MD process can achieve practical water flux at the operating feed temperature that can be sourced from industrial waste heat or solar thermal energy. With the availability of these heat sources on-site, MD can be a considerably cost-effective process for the treatment of hypersaline solutions and other challenging waters.

The separation (i.e. rejection) of the MD process relies on the hydrophobicity of the microporous membrane inserted between the feed and distillate streams in the membrane module [24–26]. The hydrophobic membrane prevents the permeation of liquid solution while allowing for the transfer of water vapour and volatile compounds through the membrane pores. As a result, in the MD process, dissolved salts and non-volatile compounds are retained in the feed stream, and the process can achieve a complete rejection of these contaminants when the dry condition of the membrane pores is maintained. This attribute of the MD process is particularly essential for strategic applications to obtain super pure distillate or to regenerate valuable solutes/solutions.

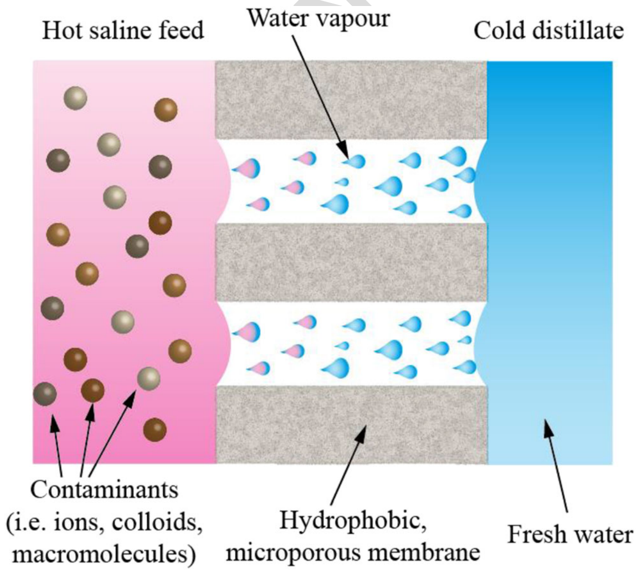

Fig. 1 The schematic illustration of heat and mass transfer during a direct contact MD process

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The non-wetting of membrane pores is critical for the MD process to achieve the complete rejection of dissolved salts and contaminants. When the membrane pores are wetted, liquid water might permeate through the membrane, compromising the membrane rejection and deteriorating the distillate purity or the process separation efficiency. The membrane pore dryness is dependent on the process operating conditions and membrane properties. According to Franken et al. [27], the membrane pores become wetted when the hydraulic pressure difference at the feed liquid-vapour interface exceeds the pore liquid entry pressure (LEP) as expressed bellow:

$$\mathrm{LEP} = \frac{-2B\lambda_L\cos\theta}{r} < \Delta P_{\mathrm{interface}} \tag{4}$$

where LEP is a function of the membrane pore structural geometric factor (B), liquid surface tension (γ_L), membrane hydrophobicity (θ), and pore radius (r), while $\Delta P_{\mathrm{interface}}$ is the pressure difference between the liquid and vapour phase at the membrane pore entrance on the feed side. For the MD process with pure water feed using the membrane with pore radius of 0.1 μm, the LEP value is in the range from 2.8 to 4.6 bar while the $\Delta P_{\mathrm{interface}}$ is 1 bar; thus, the process is intrinsically safe with respect to membrane pore wetting [16, 28]. However, in the MD process with challenging feed waters, contaminants such as organic matters and surfactants might reduce liquid surface tension and deteriorate the membrane hydrophobicity, hence lowering the LEP value [16, 25]. As a result, for the strategic MD treatments of challenging feed waters, membrane wetting can be a serious technical challenge.

The Strategic Water Treatment Applications of MD

Decentralised, Small-Scaled Desalination for Fresh Water Supply in Remote Areas

MD is emerging in the global desalination market which has been led by other desalination processes such as reverse osmosis (RO), multi-stage flash (MSF), and multi-effect distillation (MED). As reported by 2019, these three leading desalination processes account for 97% of the global desalinated water [29], and this trend is hard to change in the foreseeable future [29, 30]. Large-scale seawater and brackish water desalination using these leading processes have been considered a practical approach to augment fresh water supply in many areas around the world. Nevertheless, fresh water scarcity

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remains a critical issue for remote coastal and arid inland areas, where large-scale desalination plants are not viable. While RO is heavily reliant on stable electricity supply and requires intensive operational services and maintenance, MSF and MED are powered by fossil fuels and hence highly susceptible to the fuel provision. Fresh water supply in remote areas requires robust small-scale, stand-alone desalination processes that can be powered by renewable energy. For this niche desalination application, MD is deemed a prospective candidate [2–4].

The most noticeable advantages of MD for decentralised desalination applications are the process compatibility with renewable energy or low-grade waste heat and the ease of process operation. Unlike MSF and MED, the MD process requires mild feed water operating temperature (i.e. 40–80 °C), which can be viably generated by solar radiation and waste heat from co-generation plants, thus eliminating its reliance on fossil fuel. The MD process can also tolerate intermittent and fluctuating operating conditions offered by renewable energy sources, and the MD membrane is resistant to dry-out situations due to its hydrophobic nature [31]. Compared with RO, the MD process is less sensitive to membrane fouling and the feed water pH variation, and it can produce fresh water of super quality regardless of the feed water salinity. Thus, there is no need for intensive chemical pre-treatment, and simple pre-filtration is adequate for the MD desalination process [12, 14]. This significantly reduces the maintenance and operational cost of the MD desalination process.

For remote areas with no or limited electricity access, small-scale solar-powered MD desalination has been explored as a strategic means to meet the demand for drinking water. A great number of pilot solar-powered MD desalination plants with various capacities have been reported in the literature [14, 32–41]. These plants can be assisted or wholly powered by solar energy and treat various sources of saline waters. For example, Chafidz et al. [14] developed a portable, stand-alone solar-powered seawater MD desalination system to provide fresh water in the arid remote areas of Saudi Arabia. The pilot system integrated MD plate-and-frame membrane modules with solar thermal evacuated tube collectors and solar PV arrays and produced a maximum of 35 L of high-quality fresh water per day [14]. Kim et al. [42] reported a solar-assisted MD system with heat recovery to provide 3.4 m^3/day of fresh water from seawater. Larger solar-powered seawater MD systems with fresh water production capacity as high as 50 m^3/day were tested in the MEDESOL project with the aim to provide high-quality potable water in arid and semi-arid regions [36]. Of particular note, Duong et al. [43] trialled a pilot solar-assisted MD system for the treatment of a concentrated brine from an RO process of coal seam gas produced water for simultaneous brine volume reduction and fresh water production. Using the real solar radiation conditions in New South Wales, Australia, the authors demonstrated that MD plants combined with 1 ha of flat-plate solar thermal collectors can produce 94.4 m^3/day of fresh water from coal seam gas RO brine [43].

While the technical feasibility of solar-powered MD desalination systems has been proved, huge efforts are required to improve their economic viability. Abundant solar radiation is free, but systems required to harvest and convey it to power the MD process are costly. Due to their low-energy efficiency, most solar-powered or solar-assisted MD processes require large areas of solar thermal collectors and/or solar PV arrays, resulting in discernibly high fresh water production costs. For example, Banat et al. [44] performed an economic assessment on a small- and large-scale stand-alone solar-powered MD plants for the fresh water supply in remote areas and reported the water production costs varying between \$15/m^3 and \$18/m^3, mostly depending on the plant capital cost. The estimated MD water production costs are exceedingly higher than those of other desalination processes, particularly RO [45, 46]. Thus, extensive researches have focused on membrane module configuration modifications and process optimisation for enhanced process energy efficiency and hence reduced water production cost of solar-powered MD desalination processes. The most notable improved membrane module configurations for solar-powered MD systems can be vacuum multi-effect membrane distillation (V-MEMD) and air gap membrane distillation (AGMD) or permeate gap membrane distillation (PGMD) with internal heat recovery (Fig. 2). In these modified configurations, seawater feed is used as the coolant to condense water vapour and in tandem to be preheated before feeding to the evaporator channels of the membrane module, hence reducing the required thermal energy load on solar collectors. Even with internal heat recovery, the specific energy consumption of most pilot MD processes using modified membrane module configurations is still several orders of magnitude higher than that of RO (Table 1). It is noteworthy that internal heat recovery is not allowed in other MD membrane module configurations including direct contact membrane distillation (DCMD) and sweeping gas membrane distillation (SGMD). Moreover, the pilot MD processes with internal heat recovery are operated at much lower driving force and hence water flux than those reported in the literature for the lab-scale DCMD or SGMD processes [19, 43, 47].

Geothermal energy is another possible renewable energy that can be coupled with MD for practical fresh water supply in remote and rural locations. Compared with solar-powered systems, geothermal energy-driven MD can offer fresh water at lower production costs, and the process operation is less susceptible to intermittence, which is a typical issue for solar energy. However, the geothermal MD process has not been widely developed [3], and so far there have been only few studies on the MD desalination process powered by geothermal energy [48, 49]. For example, Sarbatly et al. [48] evaluated the possibility of the geothermal energy-powered MD

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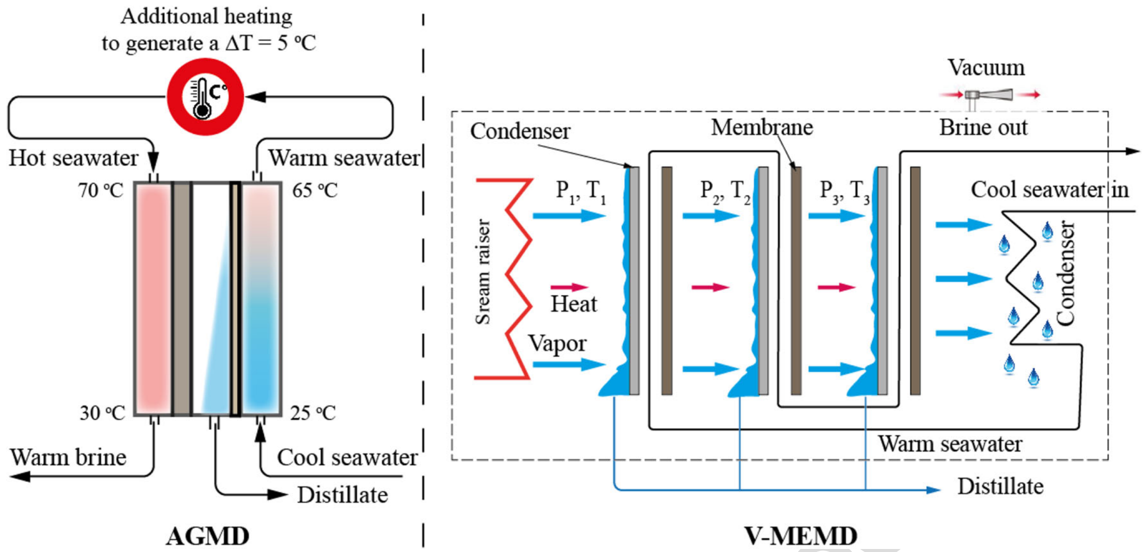

Q4 **Fig. 2** Schematic diagram of the modified MD configurations with internal heat recovery. The latent heat of water vapour condensation is used to preheat the feed water stream prior to the evaporator channels, thus reducing the external thermal energy requirement

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process for desalination purposes. The experimental results and economic analysis manifested that the MD process fed with geothermal saline water could produce drinking water with TDS below 119 ppm at the production cost of \$0.5/m^3, and using geothermal saline water feed helped reduce 90% of the MD process energy consumption. In another study, Bouguecha et al. [49] applied a hybrid MD-fluidised bed crystalliser process for the treatment of geothermal springs in Tunisia. The low-grade heat of the geothermal springs (i.e. temperature in the range 30–70 °C) was exploited to thermally power the MD process, and the fluidised bed crystalliser was applied to reduce the hardness of the geothermal springs [49]. The experimental results showed that the geothermal spring's hardness constrained the MD process

Q5 **Table 1** Capacities and performance of pilot MD systems using modified configurations for improved thermal efficiency

Reference	MD configuration, membrane area, and operating feed inlet temperature	Desalination capacity and process performance
[35]	• AGMD module • Membrane area 7–12 m^2 • Feed inlet temperature 60–85 °C	• Capacity 100 and 500 L/day • STEC 100–200 kWh/m^3 • GOR 3–6
[40]	• AGMD module • Membrane area 8 m^2 • Feed inlet temperature 60–85 °C	• Capacity 0.2–10 m^3/day • STEC 150–200 kWh/m^3 • GOR 4–6
[39]	• AGMD module • Membrane area 10 m^2 • Feed inlet temperature 60–85 °C	• Capacity 120 L/day • STEC 200–300 kWh/m^3 • GOR 0.3–0.9
[38]	• AGMD module • Membrane area 10 m^2 • Feed inlet temperature 60–80 °C	• Capacity 15 L/h • STEC 925–1389 kWh/m^3 • GOR 0.4–0.7
[41]	• PGMD module • Membrane area 5–14 m^2 • Feed inlet temperature 60–80 °C	• Capacity 4.5 L/h • STEC 130–207 kWh/m^3 • GOR: n.a.
[33]	• AGMD and PGMD module • Membrane area 10 m^2 • Feed inlet temperature 60–80 °C	• Capacity 5–120 L/day • STEC 140–350 kWh/m^3 • GOR 2–4
[19]	• AGMD module • Membranes are 7.2 m^2 • Feed inlet temperature 70 °C	• Capacity 7.2 L/h • STEC 90–95 kWh/m^3 • GOR 6–7
[47]	• V-MEMD module • Membrane are 6.4 m^2 • Feed inlet temperature 60–80 °C	• Capacity 400 L/day • STEC: n.a. • GOR 1.5–3.2

n.a., not available

water recovery due to the high risk of membrane scaling [49]. These preliminary studies indicate that geothermal energy-based MD desalination is a promising way for the economical and sustainable fresh water supply in rural and remote areas; however, additional works are required to demonstrate the long-term process reliability. Thus, future researches on geothermal energy-based MD need to focus on strategies to increase the water recovery rate, address the effect of hardness on the system performance, determine the fouling and scaling resulted from the hardness of geothermal water, and investigate the long-term operation.

Small-scale seawater MD desalination systems have also been trialled for fresh water supply on ships to exploit the waste heat generated from the ship engines [50, 51]. So far, fresh water provision on most cruise ships relies on the mature seawater desalination processes including RO and MSF [51]. As a pressure-driven desalination process, RO requires electricity generated from the ship engines to operate high-pressure pumps, thus increasing the carbon footprint and operational cost of cruise ships. Moreover, the RO process water flux and energy consumption are heavily subject to the osmotic pressure and salinity of seawater; thus, the seawater RO desalination performance and efficiency widely fluctuate for ships cruising long trips due to the variation in seawater salinity. On the other hand, the MSF process is less affected by the changing seawater salinity and hence commonly applied to ships, but it needs significantly large space to establish liquid-vapour contact. Largely, the available space on ships is restricted; thus, more compact desalination technologies are required. Therefore, the seawater MD desalination process has been proposed as an attractive alternative to RO and MSF for fresh water supply on ships. For instance, Xu et al. [50] installed a pilot-scale MD system using polypropylene hollow fibre membrane on a cruise ship and studied operational conditions for the desalination of seawater. The seawater feed was heated using waste heat generated from the ship engine. The waste heat could raise the seawater feed temperature to 55 °C, and the MD process achieved a water flux of 5.4 L/m^2 h and excellent salt rejection. The product water from the MD process had salt concentration < 3 mg/L and met the drinking water standard. In another study, Amaya-Vías et al. [51] investigated and compared the performance of different MD configurations (e.g. direct contact, water gap, and air gap) on cruise ships, exploiting the residual heat of the ship engine jacket water. All investigated MD configurations with real seawater feed achieved a nearly complete salt rejection (i.e. 99.99%) and water flux comparable with that of seawater RO [51]. Therefore, the authors suggested that MD desalination could be an additional and sustainable water production for cruise ships [51].

The strategic application of MD for fresh water supply in rural remote areas and on ships has been demonstrated, and MD was found to be a technically viable desalination process to produce high-quality water from different feed solutions. However, these MD applications have not been commercialised mainly because of the high process of thermal energy consumption that results in excessively high operational costs. To promote the commercial realisation of MD for small-scale decentralised desalination applications, more researches are needed on MD membrane module design and configuration improvement to enhance the process of thermal efficiency so that the energy consumption and hence, the cost of the MD desalinated water can be reduced.

Coupling with FO for Treatment of Challenging Polluted Waters

Another strategic application of MD is to combine with FO to allow for the complete treatment of challenging polluted waters. FO has received growing research attention as an energy-saving, low-fouling membrane treatment process of polluted waters in recent years [52–54]. The FO process involves a semi-permeable membrane separating a polluted water feed from a concentrated draw solution. The draw solution provides an osmotic driving force that allows water to permeate from the feed through the membrane to the draw solution [54]. Compared with RO technology, FO holds potential benefits related to low external operating pressure, hence lower membrane fouling propensity and reduced process energy consumption [53, 54]. However, the FO process alone is unable to reclaim fresh water from the polluted water, and it needs to be combined with another process to regenerate the diluted draw solution (i.e. in order to replenish the osmotic driving force) and, in tandem, reclaim fresh water [55, 56]. In other words, the FO process alone is only suitable for the pre-treatment of polluted waters, and an additional process is generally always required to extract fresh water from the draw solution. Therefore, draw solution regeneration plays a critical role in the development and success of the FO process for the treatment of polluted waters. Recently, MD has emerged as a promising solution to achieve highly effective and potential low-energy regeneration of FO draw solutions [57–63].

FO-MD hybrid systems have been strategically developed to overcome the key issues associated with each individual process during the treatment of polluted waters. Essentially, FO-MD hybrid systems consist of FO pre-treatment, followed by MD regeneration of the draw solution as the final step to produce clean water (Fig. 3). In wastewater treatment applications, stand-alone MD is usually unsuccessful due to high foulant concentrations that can lead to membrane wetting and the consequent termination of the MD process. Applying FO pre-treatment has been a successful strategy to contain foulants in the FO feed solution and hence prevent wetting of the MD membrane [57, 61, 63–65]. The foulants in the wastewater feed can accumulate on the FO membrane, but their effects on FO water flux are significantly lower

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compared with those of membrane wetting on MD water flux. Moreover, the inclusion of two high retention membrane processes in FO-MD hybrid systems results in the double-barrier treatment of wastewaters, therefore enhancing the removal of contaminants such as organic micropollutants, oil, and particularly surfactants. For example, the FO-MD hybrid treatment of domestic sewage and urine achieved almost complete removal of total organic carbon (TOC), total nitrogen (TN), and ammonium nitrogen (NH_4^+-N), considerably higher than the removal rates of the same contaminants treated by the standalone FO process [60, 66]. Furthermore, MD allows for the effective regeneration of FO draw solutions of hypersalinity since water flux and the complete salt rejection of the MD process are not susceptible to its feed water salt concentration. Thus, the FO-MD hybrid process can be operated at a higher osmotic driving force and hence be compatible with more concentrated wastewater feeds. More importantly, as heat is the primary power input to MD, FO-MD hybrid systems can offer energy- and cost-saving treatment of wastewaters when waste heat or solar thermal energy is readily available.

Finding proper draw solutions plays a vital role in realising the practical applications of the FO-MD hybrid process. Ideal FO draw solutions are expected to offer high water flux but minimal reverse salt flux and to be effectively regenerated by MD. A great number of draw solutions have been exploited for the FO process alone, but only several of them are suitable for the FO-MD hybrid process as they need to be non-volatile and possess a low risk of membrane wetting to the MD process. The most notable examples of the draw solutions for the FO-MD hybrid process are high charged salts of sodium including ethylenediaminetetraacetic disodium (EDTA-2Na) [62], sodium phosphate (Na_3PO_4) [67], and their mixture [68]. Compared with NaCl, which is the most common FO draw solution, high charged salts draw solutions that exhibit higher water flux but significantly lower reverse salt flux due to their larger ions [69]. Moreover, the diluted draw solutions of the high charged sodium salts can be completely recovered by the MD regeneration process. For example, the MD regeneration process of the diluted draw solutions containing single EDTA-2Na or Na_3PO_4 and their mixture retained 100% dissolved salts in the feedwater, producing distillate of excellent purity. However, it is noteworthy that the pH of the high charged salt draw solutions profoundly affects the salt dissociation in the solution, exerting critical impacts of the FO process water and reverse salt flux. Therefore, the high charged draw solutions might require pH adjustment so that the FO-MD hybrid process can achieve the optimum water separation performance [67, 68].

Although FO-MD systems offer complementary functions, the hybrid process has several challenges. In the merged FO-MD hybrid process (Fig. 3a), the FO draw solution and the MD feed flow in the same module, allowing for compact FO-MD hybrid systems. However, merging the FO draw solution and the MD feed in the same module inevitably leads to a reduction in the driving force for MD [60]. This is due to heat dissipation from the FO draw/MD feed solution to both the FO feed and MD permeate streams, resulting in a lower temperature (and hence reduced partial vapour pressure) gradient across the MD membrane. In the same way, the increased temperature of the FO draw solution might adversely affect the FO membrane integrity and promote the FO reverse salt flux. The issues with heat dissipation and its adverse effects on both FO and MD performance can be prevented when using the side-by-side FO-MD hybrid process whereby the FO draw solution and the MD feed are separated (Fig. 3b). Nevertheless, the side-by-side hybrid process is less compact

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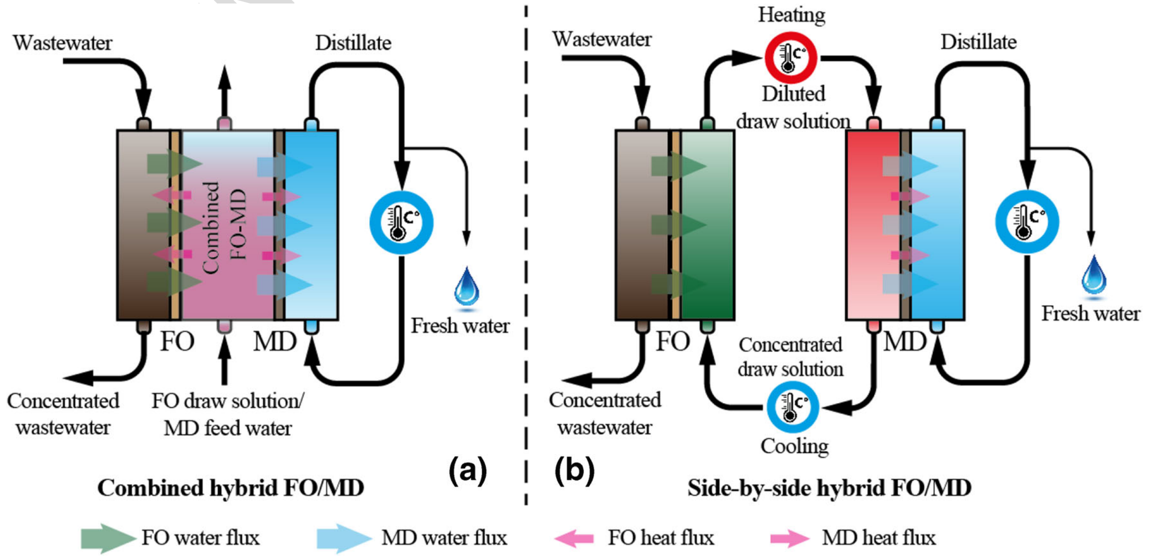

Fig. 3 The schematic diagram of the **a** merged and **b** side-by-side hybrid FO-MD process: the FO process extracts water from the feed solution to the draw solution while the MD process simultaneously recovers fresh water and regenerates the diluted draw solution

and requires additional heating the MD feed and cooling the FO draw solution, thus raising the process of overall energy consumption. Moreover, in both merged and side-by-side FO-MD hybrid processes, the presence of volatile compounds and ammonia in the source wastewaters has a high potential for accumulation at the MD distillate, subsequently contaminating the distillate. Finally, hybrid MD-FO can lead to highly concentrated effluent that needs to be further managed. The demand for post-treatment of the concentrated effluent and the additional treatment to remove volatile organics from MD distillate will invariably require more materials and incur additional operational costs. These factors must be considered if the hybrid MD-FO process is to be economically and practically effective.

Another challenge to the FO-MD hybrid process is to achieve the balance in water transfer rates between the FO and the MD unit to sustain continuous process operation. In general, the feasibility of an FO-MD process for continuous operation requires strict water management; that is, a water balance between the two individual units. So far, most studies on the hybrid FO-MD process have focused only on demonstrating the treatment viability using lab-scale FO and MD units in batch operation, few have trialled continuous operation [2, 70–72]. It is worth emphasising that FO and MD have a different driving force, and the influences of the feed salt concentration on water transfer in these two processes are unique. For example, the FO process is typically applied with asymmetric membranes (including commercial ones) [53]. In such asymmetric membrane systems, the existing internal concentration polarisation (ICP) effect in the draw solution channel—which is regarded as the Achilles heel—exerts great influence on the FO water flux [57]. The draw solution concentration decreases along with water penetration from the feed to the draw side; however, the declining trend in water flux does not have a linear relationship with the salt content owing to the ICP, particularly at a high concentration level. On the other hand, the influence of the draw solution salt content and the concentration polarisation effect on MD water flux is negligible. In this manner, the water equilibration between the FO and MD components becomes rather complicated, resulting in the frequent adjustment of operating temperatures in MD. Thus, future works on the FO-MD hybrid process need to particularly focus on process optimisation to achieve the optimum performance of each single unit as well as the balance in water transfer rates between these two units. Indeed, it appears that imbalances between the FO and MD sides of the hybrid system are not fully understood or addressed in the literature [70].

It is noteworthy that so far hybrid FO-MD systems have been demonstrated only at a lab-scale level, and significant effort is required to facilitate the large-scale and commercial hybrid FO-MD applications. A comprehensive review of hybrid FO systems carried out by Chekli et al. [72] highlighted the FO-MD system as a promising application for producing high-quality water from polluted waters. However, their findings also indicated that several limitations should be overcome before the process can become feasible at a large scale. These include membrane pore wetting, a low feed recovery rate, uncertainty related to the availability of low-cost energy sources for the draw solution regeneration by MD, and economic costs. These limitations appear to be pertinent to the FO and MD process on their own. The comprehensive review of the hybrid FO-MD literature also indicated non-uniformity and non-accuracy of experimental results [72]. This could be due to the use of a large variety of feed and draw solutions, the short duration of experiments, the wide variety of membranes, and non-similar operating conditions.

Regeneration of Liquid Desiccant Solutions for Air Conditioning Systems

One emerging strategic application of MD is for the regeneration of liquid desiccant solutions used in liquid desiccant air conditioning (LDAC) systems. In recent years, LDAC has become an energy-saving alternative to conventional vapour compression-based air conditioners [73–75]. In conventional air conditioners, the air is dehumidified by overcooling to dew point temperature to facilitate the moisture condensation to liquid water; then, the dehumidified air is reheated to the desired temperature. Thus, a significant amount of energy is wasted for overcooling and reheating the air in these systems. On the contrary, the LDAC process dehumidifies the air by directly absorbing the air moisture to a concentrated liquid desiccant solution, thus obviating the need for overcooling and the subsequent reheating of the air. Therefore, the energy consumption of the LDAC process is noticeably lower compared with that of conventional air conditioners [73–75]. Combining MD for the regeneration of liquid desiccant solutions potentially reduces the energy cost of the LDAC process as MD can utilise low-grade waste heat and the abundant solar energy that coincides with the demand for air conditioning.

Regeneration of liquid desiccant solutions is vital to the efficiency of the LDAC process. One typical LDAC system consists of an air dehumidifier and a desiccant solution regenerator. In the air dehumidifier, when the concentrated liquid desiccant solution absorbs moisture (i.e. latent heat) to dehumidify the air, it is diluted by the absorbed moisture, leading to a reduction in the desiccant concentration. As the dehumidification capacity of the liquid desiccant solution profoundly depends on the solution concentration, the diluted liquid desiccant solution needs to be regenerated prior to the next dehumidification cycle. In most current LDAC systems, the diluted liquid desiccant solution is regenerated using the traditional thermal evaporation method, in which the diluted desiccant solution is heated to about 90 °C and flows counter-currently with a hot air stream through packing media

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[76–78]. The evaporation of water requires significant thermal energy; therefore, liquid desiccant solution regeneration contributes over 75% to the total energy consumption of the LDAC process [78]. Moreover, desiccant carryover is an intrinsic technical problem pertinent to the thermal evaporation regeneration method [76, 77]. Due to the direct contact in the packing media, small desiccant droplets are swept away by the hot air stream in the regenerator. The desiccant carryover issue poses a serious risk of corrosion and detrimental health effects and inevitably results in desiccant solution replenishment in the LDAC process. Thus, great effort has been devoted to exploring innovative technologies, including MD, for the regeneration of a liquid desiccant solution to replace the thermal evaporation method (Fig. 4).

Compared with the thermal evaporation regeneration method, MD has several noticeable advantages including the capability of using waste heat or renewable energy and the process resistance to the desiccant carryover issue. As water transfer across the MD membrane occurs when exists a transmembrane vapour pressure difference, the MD regeneration of liquid desiccant solution process can be operated at a mild solution temperature, enabling the utilisation of low-grade waste heat or the solar thermal energy available on site to reduce the energy cost of LDAC. It is worth noting that desiccant solution regeneration primarily contributes to the total energy of the LDAC process. Thus, the exploration of MD for the regeneration of the liquid desiccant solution might considerably drive down the operation cost of the LDAC process. Furthermore, the MD membrane only allows for the permeation of water vapour but not liquid water; therefore, the MD process can achieve a complete salt rejection during the regeneration of liquid desiccant solution if membrane pore wetting does not occur. As a result, the desiccant carryover issue can be eradicated when using MD for the regeneration of the liquid desiccant solution.

The technical feasibility of MD for liquid desiccant solution regeneration has been experimentally demonstrated [79–84]. These experimental works have proven the MD viability for the regeneration of liquid desiccant solutions at discernibly low solution temperature without the desiccant carryover issue. For example, Duong et al. [81, 83] experimentally investigated the direct contact MD regeneration of liquid desiccant solutions of halide salts (e.g. LiCl, $CaCl_2$, and mixed $CaCl_2$/LiCl), and manifested that the direct contact MD process could regenerate the desiccant solutions at feed operating temperature as low as 55 °C, and exhibited a complete rejection of dissolved desiccant salts. However, a great challenge to the MD regeneration of the liquid desiccant solution for LDAC is the hypersaline nature of the solution. The hypersalinity of liquid desiccant solution together with polarisation effects severely restrains water flux and hence the regeneration capacity of the MD process [81, 83]. Indeed, the direct contact MD process at the feed temperature of 65 °C could only concentrate the LiCl solution up to 29 wt.%, above this concentration, the direct contact MD process achieved zero water flux [83]. The limited regeneration capacity of the MD process can be improved by elevating the feed operating temperature and deploying the vacuum configuration: a vacuum MD system at feed temperature of 70 °C could concentrate the LiCl solution up to 40 wt.% [82]. It is, however, noteworthy that the vacuum MD process at elevated feed temperature requires additional equipment (e.g. steam raiser, vacuum pump, and condenser) and heat input, thus entailing increased process complexity and investment and operational costs. Therefore, further studies are needed to develop innovative MD configurations that can regenerate the hypersaline desiccant solutions at reasonable investment and operational costs.

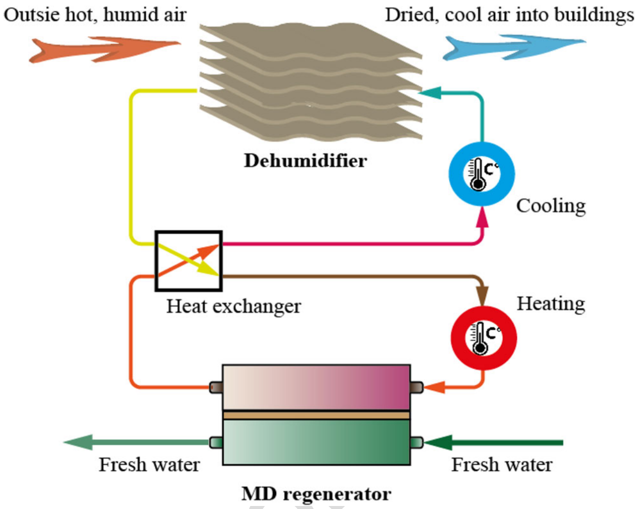

Fig. 4 The schematic diagram of the combined MD/LDAC process: the hot and humid air from outside is dehumidified by the cool liquid desiccant solution in the dehumidifier before circulating to buildings, while the MD process simultaneously regenerates the diluted liquid desiccant and produces fresh water

It is noted that most experimental studies on MD regeneration of liquid desiccant solutions for LDAC report no issue with membrane wetting, and the MD process exhibits a nearly complete salt rejection [79, 81, 83, 85]. This is a marked advantage of MD over the conventional thermal evaporation method for the regeneration of liquid desiccant solutions. Generally, the MD process with liquid desiccant solution feeds is more resistant to membrane wetting than that with wastewaters because of two reasons. Firstly, liquid desiccant solutions are prepared from fresh water and pure desiccant salts; thus, they are largely free of organic matter and surfactant that can cause membrane wetting. Secondly, desiccant salts at high concentration in liquid desiccant solutions increase the solution surface tension [86], hence elevating the liquid entry pressure (LEP) and reducing the risk of membrane wetting as expressed in the Eq. (4). Few studies have reported the issue with precipitation of corrosion products on the membrane surface during the MD regeneration of LiCl and $CaCl_2$

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desiccant solutions [81]. Nevertheless, membrane wetting did not occur and the mostly pure distillate was obtained during these MD tests with the liquid desiccant solutions.

Another direction for future researches on MD regeneration of liquid desiccant solutions is process modelling and simulation. It is noteworthy that while there have been a great number of modelling and simulation works on the MD process for seawater desalination applications, very few studies have been devoted to simulation and optimisation of the MD regeneration of liquid desiccant solutions. As mentioned above, the MD process of liquid desiccant solutions suffers severely from the solution hypersalinity and polarisation effects, resulting in much lower water flux compared with the seawater MD desalination process. Mass and heat transfer along the membrane inside MD membrane modules during the regeneration of liquid desiccant solutions might considerably deviate from that during MD desalination of seawater and other diluted saline solutions. Indeed, Duong et al. [81, 83] have stressed the need for modelling and simulation of the MD regeneration of liquid desiccant solutions with the inclusion of concentration polarisation in heat and mass transfer calculation. Moreover, the simulation of a pilot-scale direct contact MD process of seawater feed has shown marked declines in solution temperature and water flux along the membrane [87]. These reductions are envisaged to be more significant during the MD process for liquid desiccant solution regeneration given its hypersalinity and severe polarisation effects. Therefore, it is critical to simulate and optimise the MD process of liquid desiccant solutions, particularly at the pilot or large-scale operations.

The majority of researches on MD regeneration of liquid desiccant solutions so far have focused on the process technical feasibility demonstration, while the economic viability of the process for this application has not been examined. This can be attributed to the fact that MD has just been recently proposed for the regeneration of liquid desiccant solutions for LDAC and the lack of commercial MD membranes and systems. Even with the primary MD application for seawater desalination, there have been widely dispersed reports on the costs and the economic viability of the process as summarised in [88]. Moreover, the costs of the MD process for regeneration of liquid desiccant solution might by far differ from that for seawater desalination because the main product for the former is the concentrated desiccant solution while that of the latter is fresh water. For seawater MD desalination, the process-specific energy consumption is the amount of energy (i.e. thermal and electric) required to obtain one volumetric unit of fresh water (i.e. kWh/m^3). A different approach is required to assess the specific energy consumption of MD regeneration of liquid desiccant solution as fresh water is only a process by-product. Thus, future researches on MD regeneration of liquid desiccant solutions need to address the economic aspects of the process.

Treatment of Acid Effluents for Beneficial Reuse

As a strategic desalination process, MD has been applied in unique areas where other desalination processes are not technically or economically viable. One of those areas is the treatment of acid effluents from mining and metal-plating industries for the recovery of valuable minerals and fresh water. Given the rapid development of these industries, acid effluents have become a serious source of environmental issues worldwide. Largely, acid effluents have discernibly low pH and high contents of toxic metals and sulphates (Table 2), causing severe water pollution if directly discharged into the environment [93, 94]. The most common treatment chain of acid effluents consists of alkali addition for pH adjustment followed by a conventional process such as coagulation, flocculation, or precipitation. However, these conventional processes are considered inefficient and environmentally unfriendly for the treatment of complex acid effluents [95]. Pressure-driven membrane filtration processes including nanofiltration (NF), FO, and RO have been evaluated for acid effluent treatment, but the strong acidic nature and high concentrations of dissolved metals in the effluents pose detrimental impacts on the separation efficiency and integrity of the membranes [92, 96–98]. Unlike the pressure-driven membrane processes, MD is resistant to the negative impacts of acid and much less affected by dissolved metals and sulphates; therefore, it has been explored as a potential technology platform for the treatment of acid effluents. Moreover, as MD is capable of concentrating the effluents to their saturation limits, it can facilitate the recovery of valuable minerals together with fresh water from the acid effluents.

Acid mine drainage (AMD) is one of the most common acid effluents worldwide, and its treatment by MD has been demonstrated in several recent studies [95, 96, 99–101]. For example, real AMD from Tinto River in Spain has been experimentally treated by the MD process with two different configurations: air gap and water gap [95]. Although the real AMD had noticeably high concentrations of metals (e.g. iron, zinc, copper, manganese, cobalt, and nickel) and sulphate, the MD process achieved high water flux (i.e. 16.8 and 10.16 L/m^2 h respectively for the water gap and air gap configuration) and produced excellent distillate with average electrical conductivity (EC) below 19 µS/cm in all tests [95]. Particularly, the acidic nature of the AMD feed water did not pose any impacts on the MD membrane, and the MD process obtained fresh distillate with neutral pH of 7.6, despite the AMD feed had a low pH of 2.1 [95]. The authors of the study [95] also assessed the STEC and GOR of the MD process and highlighted the potential of MD for the sustainable treatment of AMD. In another study, Hull and Zodrow [100] examined the feasibility of MD treatment of an acid rock drainage (i.e. one type of AMD) feed at a high water recovery ratio (i.e. 80%) with respect to membrane scaling under two scenarios:

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Table 2 Characteristics of several acid effluents reported in the literature

Water characteristics and element compositions	References			
	[89]	[90]	[91]	[92]
General characteristics				
pH	2.4	3.3	2.6	1.3
Conductivity (mS/cm)	3.6	n.a.	n.a.	18.2
Element compositions (mg/L)				
SO_4^{2-}	n.a.	1950.0	n.a.	18.2
Ca	561.2	1070.0	500.0	347.0
Fe	835.0	186.0	443.0	467.0
Mg	384.8	384.8	771.0	n.a.
Na	192.0	14.0	158.0	18.6
Ni	1.8	n.a.	1.3	102.0
Zn	0.9	n.a.	410.0	82.0
Cu	0.2	n.a.	35.3	95.7

n.a., not available

with and without thermal pre-treatment. The experimental results demonstrated that the MD process operated at the 34 °C temperature difference between the hot feed and the cold distillate could achieve an initial water flux of 38.4 L/m^2 h and dissolved salt rejection > 99% [100]. The MD process operated at 80% water recovery suffered from membrane scaling caused by iron hydroxide, leading to a slight decrease in water flux with operating time [100]. However, a chemical-free thermal precipitation pre-treatment was effective at preventing membrane scaling and the resulting water flux decline during the MD treatment of acid rock drainage [100]. At the water recovery ratio of 80%, the MD process concentrated the mineral concentrations in the acid rock drainage by 5-folds, bringing valuable minerals to their saturation limits and thus facilitating their recovery in a subsequent process [100].

Acidic effluents from metal-plating processes have also been treated by MD for beneficial reuse of minerals and fresh water [102–104]. As a growing industry, metal-plating generates a huge volume of acidic wastewaters with high concentrations of heavy metals such as chromium and nickel. These heavy metals are toxic in aqueous environments, but they are valuable minerals and their resources have been depleted. As a result, the treatment of metal-plating effluents for beneficial reuses has been prioritised over direct discharge into the environment. The MD process has been demonstrated for the treatment of metal-plating effluents and showed promising results. Duong et al. [102] trialled the MD process for the treatment of nickel electroplating effluent to simultaneously facilitate the recovery of nickel and obtain fresh water (Fig. 5). The MD process at the mild feed operating temperature of 60 °C increased the nickel concentration in the effluent by > 100-folds, from 0.3 to 33.0 g/L (i.e. near the nickel saturated concentration), and produced fresh water with quality comparable with that of RO permeate [102]. At such a high concentration, nickel sulphate precipitated on the MD membrane but only led to a slight reduction in the process water flux (i.e. by 20%) and did not cause membrane wetting [102]. The authors also conducted a preliminary analysis of thermal energy saving when combining MD with chemical precipitation/electrodeposition for the treatment of nickel electroplating effluent for beneficial reuses and reported that the thermal energy consumption of the nickel recovery process could be significantly reduced by the utilising the sensible heat of the MD concentrated effluent [102]. In another study, Tomaszewska et al. [104] deployed MD to treat a real metal pickling solution for the recovery of metals (e.g. copper, iron, zinc, and magnesium) and hydrochloric acid (i.e. HCl). At the feed and distillate temperatures of 70 and 20 °C, the MD process could extract nearly all HCl from the pickling solution and at the same time increased the concentrations of the metals in the solution more than two-fold, hence promoting the recovery of those metals in the crystalline form [104]. The technical viability of MD for the treatment of real metal-plating effluents was also demonstrated in the study conducted by Zoungrana et al. [103] using the modified direct contact MD configuration.

Beyond the desalination applications, MD has also been tested for the recovery of acids from acidic wastewaters. Kesieme et al. [105] experimentally assessed the feasibility of direct contact MD for acid and water recovery from real sulphuric acid (i.e. H_2SO_4) or HCl leach solutions disposed of a hydrometallurgical plant. Experimental results showed that the direct contact MD process with the H_2SO_4 leach solution retained > 99.9% sulphate in the concentrate and recovered > 80% fresh water from the solution. The acid was then extracted from the concentrate using solvent extraction [101]. On the other hand, the direct contact MD process with the HCl leach solution captured the acid on the permeate side at a concentration of 1.10 M, leaving behind only 0.41 M in the feed. These experimental results confirmed that MD is technically viable for the recovery of H_2SO_4 and HCl from their leach solutions.

From the lab-scale works, MD has proved its applicability for the recovery of acid and critical minerals from various acid effluents. However, it must be noted that there remain several key challenges that need to be overcome prior to the industrial realisation of MD for this strategic desalination. These challenges include the relatively low MD water flux (i.e. compared with pressure-driven membrane processes), flux reductions caused by concentration polarisation, membrane wetting in long-term operation, high MD module and system costs, and the significantly high thermal energy consumption [4, 25, 106]. These challenges are similar to those faced by the strategic applications of MD for the regeneration of FO draw solution and liquid desiccant solutions for LDAC systems. It

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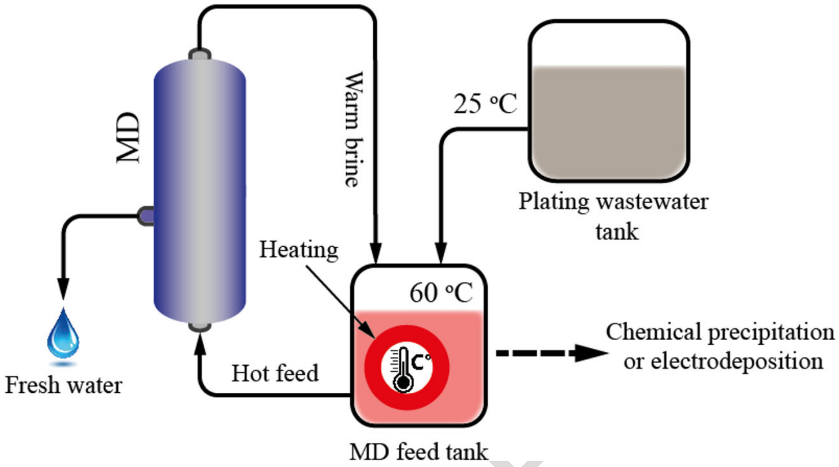

Fig. 5 The combined MD/chemical precipitation process for the treatment of the nickel electroplating effluent for beneficial reuse (from [102]). The sensible heat of the MD concentrate can be exploited in the chemical precipitation, hence reducing the thermal energy consumption of the treatment process

is also worth emphasising that the application of MD for the strategic treatment of acid effluents has not been demonstrated at the pilot or large-scale levels; therefore, future researches on the MD process of acid effluents need to focus on the pilot and large-scale demonstrations particularly regarding the challenges pointed out above.

Conclusions

This paper comprehensively reviewed four strategic water treatment applications of MD, including decentralised small-scale desalination for fresh water provision in remote areas, hybridisation with FO for treatment of challenging polluted waters, regeneration of liquid desiccant solutions for air conditioning, and treatment of acid effluents for beneficial reuses. For each strategic application, the opportunities and technical challenges pertinent to the MD process were analysed, and current status as well as future directions of the MD development were discussed. Amongst the four strategic water treatment applications reviewed here, decentralised small-scale MD desalination for fresh water supply in remote areas has advanced the most, confirmed by a great number of pilot and small-scale demonstrations. Small-scale renewable energy-driven MD desalination systems are technically viable to provide fresh water in remote areas where other mature desalination processes are not practical. However, more works on MD configuration modification and process optimisation are required to enhance energy efficiency and reduce water production costs. For the regeneration of FO draw solution and liquid desiccant solution, the technical viability of MD has been proven: MD can regenerate these hypersaline solutions without any issue of membrane wetting and achieve high-quality fresh water. Future works on MD regeneration of these hypersaline solutions need to focus on the pilot and large-scale demonstrations to evaluate the economic viability. Finally, the MD process has also been successfully demonstrated for the treatment of acid effluents for critical minerals and fresh water recovery by extensive lab-scale studies. The MD process is resistant to the strong acidic nature and high contents of metals and sulphates in the effluents; thus, it can effectively treat the acid effluents for beneficial reuse. Like for the strategic regeneration of FO draw solution and liquid desiccant solution, the economic practicality of MD for the treatment of acid effluents requires more future studies on the pilot and large-scale demonstrations.

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Compliance with Ethical Standards

Conflict of Interest On behalf of all authors, the corresponding author states that there is no conflict of interest.

Human and Animal Rights and Informed Consent This article does not contain any studies with human or animal subjects performed by any of the authors.

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