



Review

# Membrane Distillation for Wastewater Treatment: A Mini Review

Zhongsen Yan <sup>1</sup>, Yuling Jiang <sup>1</sup>, Lingshan Liu <sup>1</sup>, Zhongsheng Li <sup>2</sup>, Xiaolei Chen <sup>1</sup>, Mingqian Xia <sup>1,\*</sup>,  
Gongduan Fan <sup>1</sup>  and An Ding <sup>3,\*</sup> 

- <sup>1</sup> College of Civil Engineering, Fuzhou University, Fuzhou 350116, China; zhongsen.yan@fzu.edu.cn (Z.Y.); jylfzuer@163.com (Y.J.); 051903205@fzu.edu.cn (L.L.); 051803221@fzu.edu.cn (X.C.); fgdfz@fzu.edu.cn (G.F.)  
<sup>2</sup> Zijin International Holdings Co., Ltd., Sanya 572000, China; fjshlzs@163.com  
<sup>3</sup> State Key Laboratory of Urban Water Resource and Environment, School of Environment, Harbin Institute of Technology, Harbin 150090, China  
\* Correspondence: mq.xia@fzu.edu.cn (M.X.); dinganhit@163.com (A.D.)

**Abstract:** Water serves as an indispensable part of human life and production. On account of the overexploitation of traditional water sources, the demand for wastewater recycling is expanding rapidly. As a promising water treatment process, membrane distillation (MD) has been utilized in various wastewater treatments, such as desalination brine, textile wastewater, radioactive wastewater, and oily wastewater. This review summarized the investigation work applying MD in wastewater treatment, and the performance was comprehensively introduced. Moreover, the obstructions of industrialization, such as membrane fouling, membrane wetting, and high energy consumption, were discussed with the practical investigation. To cope with these problems, various strategies have been adopted to enhance MD performance, including coupling membrane processes and developing membranes with specific surface characteristics. In addition, the significance of nutrient recovery and waste heat utilization was indicated.



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**Keywords:** membrane distillation; wastewater treatment; membrane fouling; membrane wetting; nutrient recovery; waste heat

## 1. Introduction

With the development of human activities, a huge volume of municipal, industrial, and agricultural wastewater has been discharged into the aqueous environment. Most wastewater intake from municipal wastewater treatment plants contains high concentrations of nutrients, mainly nitrogen and phosphorus, which can be used for nutrient recovery [1,2]. Industrial wastewater involves more than seven hundred organic and inorganic micropollutants; some of them are highly toxic and carcinogenic, while some others remain in the environment for a long time and can neither be biodegraded nor biotransformed [3]. Without proper treatment, those stubborn and non-biodegradable compounds in wastewater would pose a serious threat to the humans and environment. Agricultural wastewater contains fertilizers rich in macronutrients, i.e., phosphorus, nitrogen, and potassium, as well as a variety of pesticides [4,5]. The former can result in the eutrophication of surrounding lakes, while the latter can cause serious harm to the health of living things including plants, animals, microorganisms, and even humans.

With the increased shortage of freshwater, a growing number of countries are searching for suitable technologies for wastewater recycling [6]. As a newly developed technology, membrane distillation (MD) exhibits increasing potential for wastewater treatment [7,8]. Compared with other pressure-driven membrane technologies, operating at low pressure endows MD with high fouling resistance. Theoretically, since only gaseous substances are allowed to pass through the membrane, 100% of macromolecules and ions in the wastewater could be rejected by MD [9]. Besides, MD is able to utilize low-grade waste heat or alternative energy sources such as solar and geothermal energy, thus making it a

more promising separation technology [9,10]. Therefore, considerable attention has been paid to the advancement of MD processes for wastewater treatment and water reuse [11].

Even though MD has been widely tested in various wastewater treatments, the practical application of MD is still obstructed by multifarious factors. Typically, membrane fouling inevitably occurs during wastewater treatments, which significantly impacts the treatment efficiency. Moreover, membrane wetting is the key factor to determine whether MD can maintain the long-term operation. To address these challenges, developing high-performance membrane materials and coupling with other processes have been investigated [12–14]. So far, however, a comprehensive review of the applications of MD in wastewater treatment and the obstacles and solutions has not been undertaken.

Therein, the basic information of MD, such as MD configurations and key parameters, is introduced first. A systematic review of the specific application of MD in different wastewaters is further presented. Based on the present research, the problems, such as membrane fouling, membrane wetting, and high energy consumption, are summarized. Finally, up-to-date strategies in MD wastewater treatment are presented.

## 2. MD System

### 2.1. MD Configuration

MD is a thermally driven process in which only vapor molecules are transported through a porous hydrophobic membrane. The liquid to be treated is in direct contact with the hydrophobic side of the membrane, which prevents the liquid from passing through the membrane pores and forms a liquid/vapor interface at the inlet of the membrane pores due to its hydrophobicity [15]. According to the different permeation sides, it can be divided into the following four types [15–21]:

- (1) Direct contact membrane distillation (DCMD) is an MD system with both sides in contact with the membrane surface, which does not require the structure of an external condenser (Figure 1a). The most suitable main permeable components are water and non-volatile components.
- (2) Air-gap membrane distillation (AGMD) is an MD system with a permeation side separated with an air gap, and an external condenser is not needed (Figure 1b). AGMD can be used to remove trace volatile components from an aqueous solution because the osmotic solution does not contact directly with the membrane surface.
- (3) Sweeping gas membrane distillation (SGMD) is an MD system in which the cold inert gas sweeps the permeable side of the membrane carrying the vapor molecules and condenses outside the membrane assembly (Figure 1c). This type of configuration is suitable for the removal of volatile components.
- (4) Vacuum membrane distillation (VMD) is an MD system with a vacuum permeation side (Figure 1d). A vacuum is applied to the permeable side of the membrane assembly through a vacuum pump at a pressure lower than the saturation pressure of the volatile molecules to be separated from the feed solution. In this case, condensation occurs outside the membrane module. This MD configuration is suitable for the removal of volatile organic components.

### 2.2. MD Membrane

The MD membrane is one of the most critical parts of an MD system. At least one side of the MD membrane must be hydrophobic to ensure that only the vapor phase is allowed through the membrane pores. Polyvinylidene fluoride (PVDF), polypropylene (PP), and flat polytetrafluoroethylene (PTFE) are the most used polymer materials for membrane preparation in MD applications. In particular, the characteristics needed for membranes are as follows:

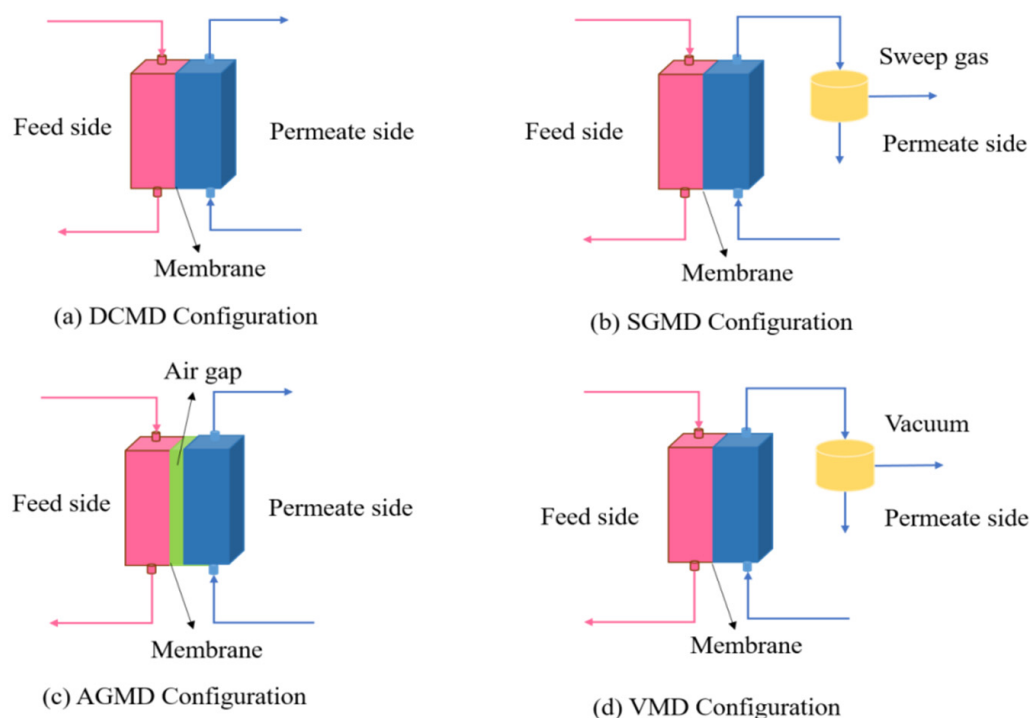
- (1) High wetting resistance. The liquid entry pressure (LEP) is the minimum hydrostatic pressure that must be applied to the feed solution before it overcomes the membrane hydrophobic force and enters the membrane pores [14]. For maintaining high rejection performance, a large LEP value is preferred.

- (2) High permeability. The molar flux through a pore is related to the membrane's other characteristic parameters by:

$$N \propto \frac{(r^\alpha) \cdot \varepsilon}{\tau \cdot \delta} \quad (1)$$

where  $\varepsilon$  is the membrane porosity,  $\tau$  is the membrane tortuosity,  $\delta$  is the membrane thickness,  $(r)$  is the average pore size for Knudsen diffusion (when  $\alpha = 1$ ), and  $(r^\alpha)$  is the average squared pore size for viscous flux (when  $\alpha = 2$ ) [17].

- (3) Low fouling tendency. Membrane fouling is one of the major problems in the application of MD. Therefore, a membrane with high fouling resistance is critical for the MD operation.



**Figure 1.** Different types of MD configurations.

Due to the complexity of wastewater, reactions may happen between the solvent and the membrane material, which will affect the membrane matrix and surface structure. Besides, membrane materials may degrade at high temperatures between 280 and 420 °C. Therefore, good thermal stability and excellent chemical resistance to feed solutions are required in wastewater treatment [22].

### 3. Applications of MD in Wastewater Treatment

#### 3.1. Desalination Brine

Desalination brine is the by-product of the desalination process, and the total dissolved solids (TDS) usually exceed 55,000 mg/L. In addition to its high salinity, it may also contain some dangerous pretreatment chemicals, organics, and heavy metals, which are detrimental to the environment when discharged directly into the environment [23,24]. Therefore, minimizing its impact on the environment or achieving zero liquid discharge (ZLD) is indispensable for disposal.

The most widely used desalination brine treatment technologies include distillation (evaporation and cooling), membrane separation, electrodialysis (ED), ion exchange, eutectic freezing, and chemical processes [25–30]. Distillation is the most common salt recovery method, but the operating temperature is relatively high, inducing a large requirement of

energy consumption. For membrane separation, when the feed solution is highly concentrated or near saturation, these processes would be limited by operating pressures (RO and NF) or applied voltages (ED) and membrane scaling [31]. Since the vapor pressure is not significantly affected by salinity, MD can handle desalination brine with extremely high TDS (up to 350,000 mg/L). Therefore, MD has been widely investigated in desalination brine treatment [32–37].

Bouchrit et al. [38] applied DCMD in hyper-saline solution treatment, and the feed was concentrated to a super-saturation degree until crystals were produced. The membrane flux decreased by 90% after 20 h of operation due to the membrane fouling. In high-salinity desalination brine treatment, inorganic fouling, termed scaling, is normally the dominant foulant in the treatment [39]. A combination of aeration and acidification is regarded as effective pretreatments for the carbonate scaling control, and the maximum concentration ratio can be achieved [40]. In addition, desalinated brine can also lead to membrane wetting, which is the result of complex interactions between various compounds [41,42]. Once the membrane is wetted, it is no longer selective and therefore cannot achieve desalination [9]. To solve the problems of membrane fouling and membrane wetting, it is necessary to improve the hydrophobicity, porosity, thermal stability, chemical resistance, and mechanical rigidity of the membrane [11]. Zhu et al. [43] fabricated a superhydrophobic-omniphobic membrane with outstanding mechanical rigidity to treat high-salinity wastewaters. The membrane had ~100% salt rejection, excellent fouling resistance, and sufficient wetting resistance when treating a high-salinity solution, i.e., simulated RO brine. To advance membrane durability, Zhu et al. [44] chose the polyimide fibrous membrane (PIFM) as the substrate, designing a hierarchically roughened omniphobic membrane. The membrane had excellent fouling and wetting resistances and showed a high initial flux of  $48 \text{ L m}^{-2} \text{ h}^{-1}$  under optimal membrane operating.

Safavi et al. [45] investigated the influential factors on VMD membrane flux, and the flux was found to increase with the increase in the feed temperature, flow rate, and vacuum pressure, but it decreased with the increase in the feed concentration. Alkhudhiri et al. used four different salts (NaCl,  $\text{MgCl}_2$ ,  $\text{Na}_2\text{CO}_3$ , and  $\text{Na}_2\text{SO}_4$ ) and three different pore sizes of flat PTFE microporous hydrophobic films (0.2, 0.45, and  $1.0 \mu\text{m}$ ) to conduct the operation of AGMD. The increase in the salt concentration and the decrease in membrane pore size decreased the flux [46]. The vapor pressure was closely related to the temperatures on both sides of the membrane. Currently, MD researchers typically focus their studies on solutions with temperatures below  $80 \text{ }^\circ\text{C}$ . As the temperature rises, however, the vapor pressure increases almost exponentially. Singh et al. [47] studied the MD experiments of PTFE microporous membranes in the temperature range of  $80\text{--}130 \text{ }^\circ\text{C}$ , achieving a vapor flux of up to  $195 \text{ kg/m}^2 \text{ h}$  at the highest temperature.

Besides the laboratory-scale investigation, pilot-scale experiments have been performed in the desalination brine treatment. Schwantes et al. operated a novel pilot-scale AGMD (the air-gap distillate was actively evacuated by an air jet) for a total of 881 h in about five months, and a feed salinity that ranged from 0 to  $253 \text{ g/kg NaCl}$  was tested. The AGMD was able to concentrate the highly saline brine to near saturation very well [48]. Single-stage MD can handle high-salinity brines, but the huge energy consumption impedes its application. Developing multistage MD can substantially reduce energy consumption [49]. A pilot-scale vacuum multi-effect MD unit utilizing low-grade waste heat and hot concentrated brine from power plants achieved the goal of low energy consumption for the MD system and achieved a rejection rate of  $>99.9\%$  for most salts [50].

### 3.2. Textile Wastewater

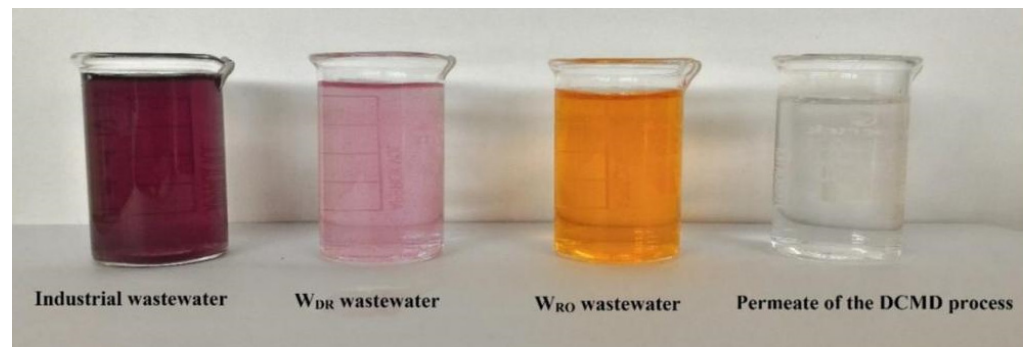
Huge quantities of textile wastewater are regarded as the main environmental obstacle to the development of the textile industry [51]. According to the World Bank estimation, about 17–20% of industrial water pollution comes from textile dyeing and finishing treatment given to fabric [52]. The textile industry uses a variety of synthetic dyes and discharges large amounts of high-color wastewater. In addition to dyes, textile wastew-

ater contains detergents, stabilizers, degradable organic matter, inorganic salts, desizing agents, and heavy metals [53]. This high-colored textile wastewater not only affects plant photosynthesis, but also causes serious harm to aquatic life due to its resulting low light transmittance and oxygen consumption, and may even be fatal to some forms of marine life (due to its metal content) [51]. Therefore, textile wastewater must be treated before discharge.

Current solutions for textile wastewater treatment include oxidation (cavitation, photocatalytic oxidation, ozone, H<sub>2</sub>O<sub>2</sub>, Fenton process), physical processes (adsorption and filtration), and biological processes (fungi, algae, bacteria, microbial fuel cells) [51,54–56]. However, the application of these processes is often limited owing to poor decolorization efficiency and high price [57,58]. In terms of membrane processes, ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO) are the most extensively used [59]. Nevertheless, UF typically presents low rejection performance, while NF and RO are limited by the high osmotic pressure generated at high concentrations [60]. The MD process boasts a great deal of merits for textile wastewater treatment, such as small area coverage, high efficiency, facile implementation, scalability, and recycling dyes from concentrate. More importantly, considering the discharge temperature of the textile wastewater (50–80 °C), the MD process can also directly gain access to the wastewater without further energy used for the heating step [61,62].

Calabrò et al. [63] first investigated the performance of MD in dye solution concentration in 1991, and the potential of MD to produce high-purity water in textile wastewater treatment was demonstrated. Barnett et al. [64] used tubular modules with PP membranes to treat methylene blue water mixtures through VMD, and the results showed that dye molecules were totally rejected on the feed side. A similar result was reported by Criscuoli et al. [65] that pure water was recovered at the distillate side during the VMD concentrating of solutions with different amounts of dye (25–500 PPM). Li et al. [66] compared the performance of two hydrophobic membranes (PTFE and PVDF) in textile wastewater with DCMD. Comparatively, the PTFE membranes with the same pore diameters had a higher permeability flux and rejection rate of contaminants due to their enhanced hydrophobicity and excellent wetting resistance. The PTFE membrane demonstrated 90% COD removal and 94% color removal over 48 h of continuous operation. Textile wastewater can also be purified by AGMD. Leaper et al. [67] demonstrated that as a one-step process, AGMD could concurrently remove all salts, surfactants, and dyes. After 70 h of continuous testing, the dye rejection rate was maintained at 100%, whereas the salt rejection rate decreased to 91%, indicating that partial wetting occurred.

Pilot-scale experiments of DCMD were reported to have been run in textile mills, and plant integration evaluation found that the integration of waste heat into the MD for textile wastewater treatment appears to be feasible [68]. However, textile wastewater also contains surfactants that can damage the basic hydrophobic properties of the membranes, leading to membrane wetting. To tackle this problem, Villalobos et al. [69] tested a custom-made PTFE membrane with a polyurethane coating in a pilot MD and compared it with a conventional hydrophobic PTFE membrane. The conventional PTFE membrane showed an increase in permeation conductivity, while the hydrophilic coating membrane exhibited a continuous decrease in conductivity, indicating that the hydrophilic coating membrane was intact and resistant to surface wetting. A novel double-layer nanofiber membrane was fabricated and used in DCMD for the treatment of industrial textile wastewater by Meshkani et al. [70]. The results showed that the COD was reduced by almost 99%, and the color rejection rate was 100%. A visual view of the feed sample and the penetrant obtained during this work is shown in Figure 2.



**Figure 2.** A visual view of the textile wastewater (industrial wastewater, wastewater by dispersing red dye ( $W_{DR}$ ) and the wastewater by reactive orange dye ( $W_{RO}$ )) and the permeate after DCMD treatment [70].

### 3.3. Oily Wastewater

The rapid development of the shale oil and gas industry, the petroleum industry, the metallurgical industry, and the catering industry has led to a dramatic increase in the discharge of oil-bearing wastewater [71–75]. The main constituents of the oily stream are oil, surfactant, and salt. Francesca et al. [76] investigated different polymer (PVDF and PP) membranes in the oilfield-produced water treatment by DCMD. The overall salt rejection rate was greater than 99%, and the total carbon removal rate was greater than 90% [76]. Zhang et al. [77] developed a new two-stage VMD process for a natural gas extraction wastewater treatment. After 130 h of operation, the water recovery rate reached 88.6%, but heavy contamination occurred on the inner surface of the membrane. Due to strong and long-term hydrophobic interaction, the hydrophobic membrane is easily polluted by hydrophobic pollutants inherently, and conventional MD membranes are not feasible for oily water separation processes [78,79]. For the oily wastewater treatment, the membrane surface modification of the hydrophobic membrane was widely applied in improving the anti-oil and anti-wetting performance [80]. Related works about the membrane modification for oily water treatment are summarized in Table 1. So far, most of the studies on oily wastewater treatment by MD have been based on DCMD configurations, and the modification had negative impacts on the osmotic flux. Therefore, there is a trade-off between the antifouling (antiwetting) and flux of the membrane.

**Table 1.** An overview of the investigation of MD in oily water treatment.

MD Configuration	Composite/Modified Membrane	Wastewater Content			MD Performance	Ref.
		Oil (g/L)	Surfactant (mg/L)	Salt (wt%)		
DCMD	PVDF membrane modified with silica nanoparticles (SiNPs), chitosan hydrogel, and fluoropolymer	1	\	3.5	1. The salt rejection rate was 100%; 2. Stable wetting resistance; 3. No obvious fouling during the 36-h operation.	[81]
DCMD	PVDF membrane modified with SiNPs and fluoroalkylsilane (perfluorodecyltrichlorosilane)	0.08	0.2 (mmol/L)	5.8	1. The salt rejection rate was 99.99%; 2. Stable wetting resistance; 3. No fouling for 8-h operation.	[82]
DCMD	PVDF membrane with low-surface-energy perfluoroalkyl functional groups	1	\	3.5	1. The salt rejection rate was 99.9%; 2. No wetting; 3. Improved the fouling resistance.	[83]

Table 1. Cont.

MD Configuration	Composite/Modified Membrane	Wastewater Content			MD Performance	Ref.
		Oil (g/L)	Surfactant (mg/L)	Salt (wt%)		
DCMD	PVDF membrane modified with oxidation-induced dopamine polymerization	0.5	50	3.5	1. The salt rejection > 99.99%; 2. No wetting and fouling during 80-h operation.	[84]
DCMD	PVDF membrane modified with SiNPs/polydopamine (PDA)	1	\	3.5	1. Salt rejection > 99.9%; 2. Effective and robust fouling resistance.	[85]
DCMD	PTFE membrane modified with polyacrylonitrile (PAN) coating	1	\	3.5	1. The salt rejection was 100%; 2. Robust anti-oil-fouling	[86]
DCMD	PVDF membrane grafted with polyethylene glycol and later coated by TiO <sub>2</sub>	0.01 (wt%)	\	3.5	1. No wetting and fouling during 24-h operation.	[87]
DCMD	PVDF membrane modified by graphene oxide (GO)	\	50	3.5	1. Excellent salt rejection; 2. Excellent wetting and fouling resistance.	[88]
DCMD	PVDF membrane with macro-corrugated and nano-patterned hierarchically structured	1% (v/v)	1 (mmol/L)	5.8	1. No membrane wetting for 24 h.	[89]
DCMD	PVDF membrane modified with hydrophilic polydopamine (PDA)/polyethyleneimine (PEI)	0.45	50	3.5	1. The salt rejection rates > 99.9%; 2. No wetting and fouling after 137-h operation.	[90]
DCMD	PVDF membrane prepared by the coating of the hydrophobic silica nanoparticles	1	\	3.5	1. The salt rejection is 100%; 2. Superior water-repelling ability; 3. No fouling for 30 h.	[91]
DCMD	Omniphobic PVDF membrane of hierarchical structure and having silica nanoparticle coating (MSF-PVDF)	10	10,000	4	1. No observable wetting; 2. No fouling.	[92]
DCMD	PVDF membrane modified by multiwall carbon nanotube (CNT) through hydrophilization and further functionalized by polyvinyl alcohol and glutaraldehyde	1	\	0.5	1. Salt rejection > 99.9%; 2. Excellent oil fouling resistance.	[93]
DCMD	Polysulfone (PSf) membrane with double Re-entrant structure	\	12	3.5	1. Salt rejection was 99.99%.	[94]
DCMD	PTFE/PP membrane coated with Teflon and PDA	0.5	0.4	3.5	1. Salt rejection ~100%; 2. Excellent wetting and fouling resistance.	[95]
AGMD	Novel fluorosilane-grafted ceramic membranes	0.11	\	0.7–0.9	1. The salt rejection was 99%.	[96]
AGMD	New tubular fluorosilane-grafted sand membrane	46 ± 1	\	\	1. The salt rejection was 99.65%.	[97]

Table 1. Cont.

MD Configuration	Composite/Modified Membrane	Wastewater Content			MD Performance	Ref.
		Oil (g/L)	Surfactant (mg/L)	Salt (wt%)		
VMD	PTFE membranes coated with PDA	0.5	\	3.5	1. The salt rejection was 99.9%; 2. Excellent wetting and fouling resistance.	[98]
VMD	PTFE membranes with a thin Nafion layer	\	0.1	3.5	1. Salt rejection was 99.99% for 60 h; 2. Excellent wetting and fouling resistance.	[99]
SGMD	Porous SiOC ceramic membrane made up of polydimethylsiloxane (PDMS) and polysiloxane (PSO) polymers	1	10	0.4–1.6	1. Salt rejection was 99.9%; 2. Wetting resistant; 3. Possibly fouling-free.	[100]

### 3.4. Radioactive Wastewater

After the Fukushima accident in Japan, the treatment of radioactive wastewater has attracted more and more attention throughout the world to support the sustainable development of the nuclear energy industry [101]. To date, radioactive wastewater treatment methods mainly include evaporation, adsorption, ion exchange, and membrane separation [102]. As a thermal-driven system, MD may have an inherent advantage for radioactive wastewater treatment because nuclear power plants have a large amount of waste heat [103]. From the perspective of treatment performance, the permeate of MD in the radioactive solution treatment can be reused or discharged directly [104–106].

Table 2 summarizes the research work on radioactive wastewater treatment by MD. The decontamination factor (DF), which is defined as the ratio of the concentration of nuclides in the feed solution and the distillate, is commonly used to represent the rejection performance. It can be seen from Table 2 that DFs for the feed stream were very high. Specifically, some even reached infinity, indicating that the corresponding element could not be detected on the permeation side. The DF results of the feed solution with Cs were surprisingly satisfactory, since this radioactive isotope is difficult to remove from treated waste by most known methods [107]. It was proved that MD is feasible to process low- and medium-level radioactive wastes, resulting in high DF in a single stage.

Even so, the chemical stability of the membrane material is one major concern in radioactive wastewater treatment. After exposure to radiation for a long time, some polymers in the membrane will degrade and lose their chemical properties, which will endanger the stable operation of MD. It is worth noting that only a small number of membrane manufacturers have their products certified by the International Atomic Energy Agency [108]. Therefore, improving the chemical and radiation stability of the membrane is another key issue that should be considered besides the separation characteristics in the radioactive wastewater treatment.

Table 2. Summary of the investigation works in radioactive wastewater treatment by MD.

MD Configurations	Membrane	Scale	Pollutants	DF	Ref.
DCMD	PVDF hollow-fiber membrane	Lab	Cs <sup>+</sup>	Infinity	[105]
			Sr <sup>2+</sup>	Infinity	
			Co <sup>2+</sup>	Infinity	



Table 2. Cont.

MD Configurations	Membrane	Scale	Pollutants	DF	Ref.
DCMD	Commercial hydrophobic polypropylene hollow fiber membrane	Lab	Cs <sup>+</sup>	10 <sup>5</sup> –10 <sup>6</sup>	[109]
			Sr <sup>2+</sup>	10 <sup>5</sup> –10 <sup>6</sup>	
			Co <sup>2+</sup>	10 <sup>5</sup> –10 <sup>6</sup>	
VMD	Commercial polypropylene hollow fiber membrane	Lab	Cs <sup>+</sup>	333.3	[110]
VMD	Commercial polypropylene hollow fiber membrane	Lab	Co <sup>2+</sup>	550	[111]
VMD	Commercial polypropylene hollow fiber membrane	Lab	Sr <sup>2+</sup>	200	[112]
VMD	Commercial polypropylene hollow fiber membrane	Lab	Cs <sup>+</sup>	7600	[113]
			Co <sup>2+</sup>	8900	
			Sr <sup>2+</sup>	7800	
DCMD	Surface modified membranes by surface modifying macromolecules (SMMs)	Lab	<sup>60</sup> Co	>300	[114]
			<sup>137</sup> Cs	>888	
			<sup>85</sup> Sr	>400	
DCMD	MFFK membranes by compositing microfiltrational fluoroplastics	Lab	<sup>137</sup> Cs	90–11,000	[106]
			<sup>90</sup> Sr	140–15,000	
			<sup>90</sup> Y	180–18,000	
DCMD	Hydrophobized PET track-etched membrane by photografting of styrene	Lab	<sup>60</sup> Co	85	[115]
			<sup>137</sup> Cs	1727	
			<sup>241</sup> Am	5	
DCMD	Spiral-wound PTFE membrane	Pilot	<sup>60</sup> Co	4336.5	[107]
			<sup>137</sup> Cs	43.8	
			<sup>65</sup> Zn	Infinity	
			<sup>110</sup> Ag	Infinity	
			<sup>133</sup> Ba	Infinity	
			<sup>134</sup> Cs	Infinity	
			<sup>170</sup> Tm	Infinity	
<sup>192</sup> Ir	Infinity				
VMD	PTFE hollow-fiber membrane	Pilot	<sup>133</sup> Cs	10 <sup>4.85</sup>	[116]

### 3.5. Other Wastewaters

#### 3.5.1. Urine

Urine (also called yellow water) contributes 90% of the N, 60–65% of the P, and 50–80% of the potassium (K) in sanitary sewage, which can cause eutrophication of the environment without appropriate treatment [117]. At the same time, urine is a promising resource in terms of nutrient recovery. Besides, extracting pure water from urine is essential on the space station. Many researchers are working to develop and optimize different physicochemical processes for extracting pure water from urine, and MD is promising for recovering water on the space station in the future [118,119]. Zhao et al. [120] demonstrated that VMD is a low-cost and efficient process for the efficient removal of organic and inorganic salts from urine, with a COD removal rate of nearly 99%. Tun et al. [121] applied DCMD in urine concentration, and pre-filtration (1.2 µm) of acidified urine was helpful in preventing membrane contamination. In addition, MBR and UF can also be used as the pretreatment of MD to postpone the occurrence of membrane fouling [122,123]. Membrane modification is considered a method to improve the efficiency of water recovery from the

urine feed stream. Khumalo et al. [124] prepared PVDF/PTFE membranes modified with methylated functionalized silica nanoparticles for prolonged longevity in MD application, and approximately 80% water recovery was obtained from urine samples used in this study.

### 3.5.2. Landfill Leachate

Landfill leachate contains high concentrations of harmful pollutants, such as organic matter, ammonia, heavy metals, and toxic substances [125]. High-concentration ammonia recovery is the main challenge of landfill leachate treatment. The recovered ammonia has special economic benefits and can be applied to the fertilizer market [126]. At present, the investigation of applying the MD process to landfill leachate treatment is still rare. Zoungrana et al. [127] first investigated the feasibility and influential factors of DCMD for landfill leachate treatment in 2017. The COD removal rate of the permeate fluids reached 99%, while the removal rate of ammonia nitrogen was not high (72.6%) at pH = 8. This was because ammonia nitrogen is more easily converted into  $\text{NH}_3$  and volatilized to the permeable side during the heating process of the alkaline feed solution [128]. Zico et al. [129] tested a solar-powered DCMD system in ammonia recovery from landfill leachate, and 98% of ammonia removal and 59% of ammonia recovery were achieved under optimal experimental conditions. Due to the high concentration of contaminate in the landfill leachate, pretreatments, such as NaOH/PAM polyacrylamide pretreatment and Fenton oxidation, have been applied to mitigate membrane fouling in the MD process [130,131].

In our recent work, MD treatment performances of the landfill leachate were investigated under three pH values (5, 7, and 9), and the potential of combination with an incineration plan was evaluated [132]. The rejection rates of organic, heavy metal, and phosphate almost exceeded 99%, and wetting was not detected during the short-term operation. After theoretical calculation, the incineration waste heat was sufficient to drive the MD operation for the daily landfill leachate treatment. Although acidification of the feed solution can alleviate membrane fouling, it is still an obstacle for the application.

### 3.5.3. Anaerobic Digestate

Wastewater from the anaerobic digestion process usually contains high concentrations of dissolved ammonium nitrogen, phosphorus, and suspended and colloidal solids [133,134]. Traditional treatment techniques for anaerobic digestate mainly focus on pollutant removal [135]. MD technology can simultaneously recover the nutrients and pure water in one step, which is of vital significance for the anaerobic digestion treatment. By studying the MD treatment of anaerobic digestate, Jacob et al. [136] found that although increasing temperature would greatly improve membrane flux, the removal rate of ammonia nitrogen would also decrease correspondingly. Our previous work indicated that pH significantly impacted the rejection of ammonia in a cow dung anaerobic digestate treatment, and that feed acidification significantly increased the ammonia removal rate (from 66% to 99%) [8]. By contrast, Kim et al. [137] found that the rejection rate of TN was dominated by the extent of the cake layer during the treatment of the anaerobic digestate from livestock wastewater. Membrane fouling is also a critical concern of the application of MD in anaerobic digestate treatment. Due to the complex component in the digestate, synergetic fouling of organic and inorganic fouling would occur. Our previous work indicated that feed acidification was able to decrease organic fouling by inhibiting inorganic scaling [138]. Amine et al. [139] optimized the cleaning method for membrane fouling control in an anaerobic digestate treatment. The optimized conditions could allow 75.5% of the initial flux to be retained after 96 h of operation. Ershad et al. [140] examined the feasibility of using AGMD to separate anaerobic digestion. The results showed that the removal rates of COD, P, S, and K were all > 98%, and the removal rate of TAN was close to 100%. The team further quantified the industrial application of MD for anaerobic digestion by using the biogas plant as a waste heat recovery source and evaluated the technical and economic feasibility of thermal integration between the biogas plant and MD [141]. Although the laboratory-scale experiments have achieved great performance in the anaerobic digestate treatment, the

pilot-scale application of MD is rarely reported, and long-term performance should be evaluated for the industrial application

#### 3.5.4. Pharmaceutical Industry Wastewater

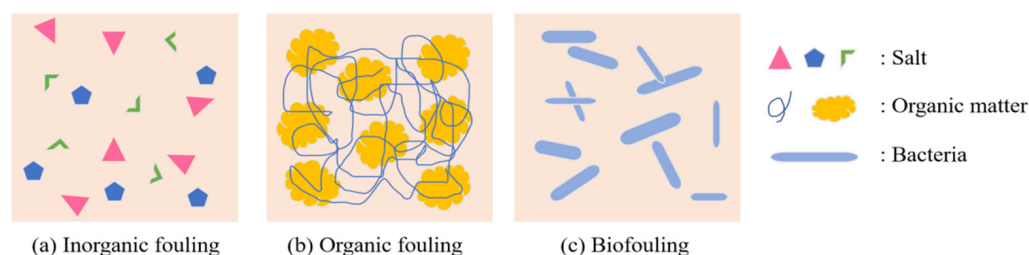
Among many emerging water pollutants, drugs are the most crucial category, including antibiotics, analgesics, steroids, antidepressants, antipyretics, stimulants, antimicrobials, analgesics, hormones, anti-inflammatory drugs,  $\beta$ -blockers, lipid modulators, contrast agents, and impotence drugs [142]. The persistence and potentially toxic effects of these drug residues make them a potential long-term threat to discharged water. So far, various treatment technologies have been studied to eliminate drug residues in wastewater, but the removal efficiency is not always satisfactory [143]. Based on the high rejection rate of non-volatile compounds and low operation temperature, MD might be an option for the pharmaceutical industry wastewater treatment [144,145].

However, the membrane flux of MD is generally smaller than that of other membrane technologies. Ken Gethard et al. [146] used carbon nanotube immobilized membrane (CNIM) to enhance the MD performance to produce both pure water and concentrated pharmaceutical wastewater, and the active ingredients of four drugs (ibuprofen, dibucaine, acetaminophen, and diphenhydramine) were adopted in the investigation. The carbon nanotubes in CNIM were reported to act as sorbent sites and to provide an additional pathway for enhanced water vapor transport. The results proved that CNIM is clearly more efficient than conventional MD membrane, achieving a higher flux level with the same energy consumption level. Besides the membrane hydrophobicity, the membrane surface charge would also affect the treatment performance in the pharmaceutical industry wastewater by MD. Guo et al. [147] used negatively charged PVDF membranes in the treatment of antibiotic wastewater with different charges. When positively charged antibiotics were presented in the feed, the membrane fluxes decreased significantly and wetting occurred during the MD treatment. To control the fouling and wetting by the positively charged antibiotics, surface charge adjustment via feed alkalization and water flushing was proven to be an efficient method [148].

### 4. Obstacles of Applying MD in Wastewater Treatment

#### 4.1. Membrane Fouling

The accumulation of unwanted materials on the membrane surface or within the membrane pores is termed membrane fouling, which will result in the decline of the permeate flux and salt rejection [149]. Similar to other membrane processes, membrane fouling is one of the critical issues in MD. The fouling involves a variety of mechanisms, such as adsorption, accumulation, or precipitation, which can occur simultaneously to cause fouling. As Figure 3 shows, membrane fouling can be classified into inorganic fouling (scaling), organic fouling, and biological fouling. In wastewater treatment, the component of membrane fouling would be significantly different.



**Figure 3.** Schematic diagram of three membrane fouling types: (a) Inorganic fouling, (b) Organic fouling, and (c) Biofouling.

##### 4.1.1. Inorganic Fouling

Inorganic fouling is ubiquitous in wastewater treatment by MD. Specifically, inorganic scaling plays a dominant role in the desalination brine treatment by MD [39]. With the

cumulation of inorganic salt on the feed side, the concentration of slightly dissolved salt would exceed the equilibrium solubility product, inducing a supersaturation condition and crystallization. In the MD process, the supersaturation is mainly caused by water evaporation and temperature change [150]. Under the supersaturated condition, ions attract each other and form crystals in the solution [151]. These crystals can form on the membrane surface or inside the membrane pores by gravity settling or particle transfer. The accumulation of these crystals will eventually block the pores and impede the transport of vapor molecules [152]. In addition, deposit layers formed on the membrane surface add additional thermal resistance and increase the temperature polarization [150]. These two reasons lead to a reduced driving force across the membrane and consequently compromise the permeate flux.

According to the alkalinity of scaling, inorganic fouling is divided into three categories: alkaline, non-alkaline, and uncharged molecular scales (based on silica) [153].

1. Alkaline scales. The most typical alkaline scale is carbonate, which mainly presents in the form of bicarbonate and calcium ions [9]. These salts are called alkaline salts because they produce more alkaline solutions when they dissolve.  $\text{CaCO}_3$  is one of the most common alkaline scales and one of the primary components of desalination brine [154]. The three anhydrous crystalline polycrystalline forms of  $\text{CaCO}_3$  are classified as spherical aragonite, calcite, aragonite, and three other hydrated forms. Viader et al. found that the  $\text{CaCO}_3$  was the easiest inorganic salt when integrating MD as volume reduction technology for in-land desalination brines [40].  $\text{CaCO}_3$  scale causes a rapid decrease in permeate flux. Gryta et al. [155] proposed that increasing the feed flow rate could reduce crystallization growth and make carbonate scales more porous and looser. However, this only works for calcium carbonate pollution alone. In the actual feed solution, the fouling pattern was more complicated with the presence of other impurities.
2. Non-alkaline scales. Non-alkaline salts are ions that dissolve in water but do not cause the pH of the water to rise.  $\text{CaSO}_4$ ,  $\text{Ca}_3(\text{PO}_4)_2$ , and  $\text{NaCl}$  are examples of non-alkaline scaling. Calcium sulfate is the most common non-alkaline scaling in desalination brine, which mainly takes three forms: anhydrite ( $\text{CaSO}_4$ ), semi-hydrate (Bassanite- $\text{CaSO}_4 \cdot 0.5\text{H}_2\text{O}$ ), or dehydrate (Gypsum- $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ ) [156,157]. Since its solubility is insensitive to pH and the scaling of gypsum is more difficult to be prevented and removed, the investigation of gypsum scaling attracts great attention in MD [158–160]. Zhou et al. studied the performance of submerged VMD to treat desalination brine and proved that  $\text{CaSO}_4$  crystallization, which needs to be eliminated through proper feed pretreatment, is the most important cause of flux attenuation through the combination of experiments and model calculation [161]. Dosing antiscalant is the common method for gypsum scaling control [162], but phosphate-containing antiscalants may induce calcium phosphate scaling during MD operation [9]. Although  $\text{NaCl}$  has been extensively studied in various wastewaters, the effect on membrane scaling appears to be negligible due to its high solubility [163].
3. Uncharged molecular scales. Silica presents in the natural water supply as colloidal silica, particulate silica, or dissolved silica. In the supersaturated feed solution, the insoluble silica begins to polymerize to form a gelatinous layer and precipitate on the film surface [164]. Silica deposits clog the membrane holes and thus reduce the osmotic flux by up to 70% [165]. However, acidification is not very successful in cleaning silica scaling due to its uncharged molecules [166].

#### 4.1.2. Organic Fouling

Organic fouling refers to the deposition of natural organic matter (NOM), oils, protein, organic salts, and polyacrylic polymers [74,167–170]. NOM fouling is widely investigated in membrane technology, but the MD membrane shows great fouling resistance to the modeled NOM, such as humic acid, bovine serum albumin, and sodium alginate [170]. Even though the membrane flux was not affected by the organics, the deposition of the

organic fouling layer was visible. In general, hydrophobic membranes are used in MD. Due to the existence of strong hydrophobic interaction, chemical affinity, and electrostatic mechanisms, the adsorption of organic matter on the surface of the hydrophobic membrane occurs easily. In oily wastewater treatment, the high concentration of hydrophobic oil will deposit on the membrane via the hydrophobic interaction and block the membrane pore. When surfactant or salt are present in the oily wastewater, membrane fouling significantly decreased [171]. In textile wastewater treatment, chemical bonds play an important role in the mechanism of dye fouling on the hydrophobic membranes. Due to physical and chemical interactions, dye molecules can be adsorbed to the membrane surface to form fouling, which affects the membrane permeability [172]. Similarly, the charged nature of pollutants in pharmaceutical wastewater can affect the affinity that exists on the membrane surface and thus affect fouling production [147].

#### 4.1.3. Biological Fouling

Since higher temperature and salinity of feed are not favored for the retention of microorganisms, MD had lower biological fouling (biofouling) compared to other membrane processes. The biofouling is mainly caused by bacteria, fungi, sludge, algae, yeast, and other microorganisms, referring to the proliferation and accumulation of bacteria or living microorganisms on membrane surfaces. With the development of MD membrane bioreactors, understanding and control of biofouling in MD has begun to receive more attention [173]. Gryta [174] observed the growth of fungi and anaerobic bacteria on the membrane surface in MD, but aerobic bacteria did not have appropriate conditions for their growth. Bogler et al. [175] explored the situation of biological fouling in MD with a thermophilic *Anoxybacillus* sp. at 47 (below the optimum temperature for bacterial growth), 55 (at the optimum temperature for bacterial growth), and 65 °C (above the optimum temperature for bacterial growth). At 55 °C, the biofilm covered large membrane areas, causing the most serious permeate water flux decline. In addition, bacteria multiplied continuously in the feed water, and endospores or even elongated bacterial cells formed on the contaminated membrane surface can penetrate the membrane pores, resulting in membrane wetting or pore blockage.

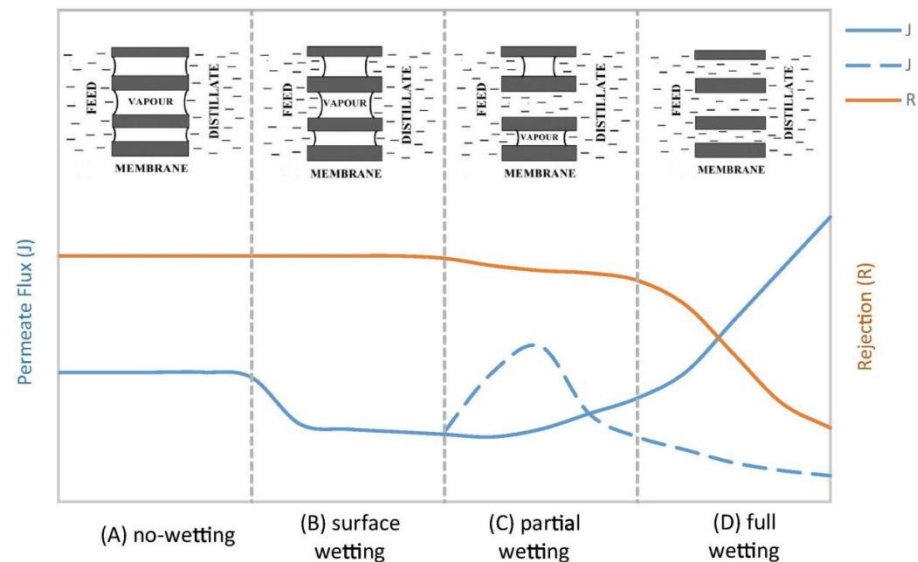
Although membrane fouling is classified into inorganic, organic, and biological fouling, in most cases, a combination of different fouling and mechanisms occur in the practical application of the MD process. So far, membrane fouling is one of the most critical issues for MD, especially in the actual wastewater treatment with high recovery. Identifying their interaction would provide a guide for membrane fouling control, but the present work is still rare and more focus should be emphasized.

#### 4.2. Membrane Wetting

Membrane wetting refers to the permeation of feed water through the membrane pore, which would decrease the membrane flux and rejection performance [176]. As Figure 4 shows, the membrane wetting state can be divided into four grades: (a) non-wetted; (b) surface-wetted; (c) partially wetted; and (d) completely wetted [177]. The non-wetted membrane shows that all the membrane pores are filled with steam. The surface-wetted membrane shows that the membrane pores are partially filled with liquid, but the liquid has not yet crossed the entire pores. Partial wetting means that the local membrane pores are filled with liquid. Completely wetted is when the whole membrane has lost hydrophobicity, and all of the membrane pores are filled with liquid.

For organic-rich wastewater (e.g., oily wastewater, industrial wastewater, yellowwater), membrane wetting is frequently observed during MD treatment [124,178,179]. Due to the presence of amphiphilic organic compounds in the wastewater, membrane pores would be wetted, with a loss of selectivity over time [42]. When low surface energy organic compounds such as alcohols are present in the feed solution, the surface tension of the solution will decrease, which will significantly reduce the LEP of the membrane and induce instant pore wetting [180]. Different from alcohols, surfactants induce membrane wetting

in a more progressive way. Due to the hydrophobicity of the MD membrane surface, the hydrophobic head (usually an alkyl group) will interact with membrane surface via hydrophobic interaction, and the hydrophilic head (carboxyl or hydroxyl groups) attached to the membrane pore would interact with the feed through strong hydrogen bonding [181]. Along with the water vapor gradient, the alcohols or surfactants also transfer continuously into the membrane pores [182]. The water molecules bind to a large amount of alcohols or surfactants remaining in the membrane pores, and some liquid water may become trapped in the membrane pores, which can cause the membrane to be wetted.



**Figure 4.** Membrane wetting degrees: (A) non-wetted; (B) surface wetted; (C) partially wetted; and (D) completely wetted [177].

The accumulation of inorganic sediments on the membrane surface in desalination brine concentration or acid mine drainage treatment can also lead to pore wetting [36,183]. The growth of inorganic scaling (usually mainly composed of calcium carbonate, calcium sulfate, and sodium chloride) on the membrane surface reduces the hydrophobicity of the membrane, eventually leading to complete wetting of the membrane pores [165,184]. When dealing with complex water matrices, wetting will be the result of complex interactions between the various compounds found in the wastewater [41].

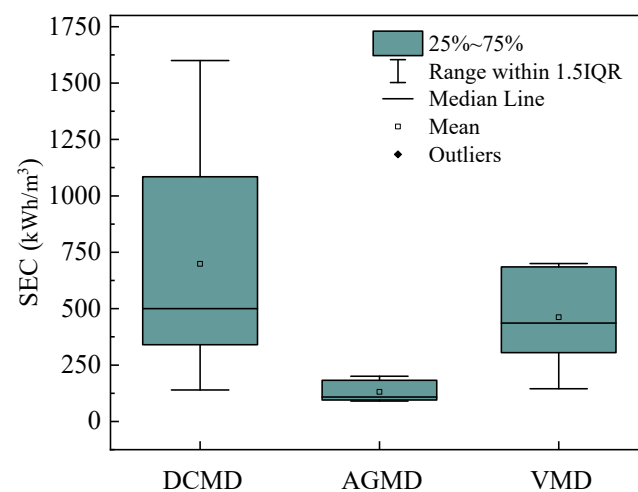
The membrane wetting greatly impacts the separation efficiency and operation stability of the MD process [185]. The detection of membrane wettability is therefore necessary to provide information about the appropriate time for membrane replacement. The traditional method of detecting membrane wettability is to monitor the conductivity of the permeate, because the increased diffusion of the solute through the liquid-filled pores leads to a decrease in the desalting rate and an increase in the conductivity of the permeate [185–187]. Despite its advantages due to simplicity, the traditional method can only detect wettability after the membrane has failed [186]. In addition, the presence of dissolved gases such as ammonia and carbon dioxide can also interfere with the detection, as they can also increase the osmotic conductivity [132,187]. More recently, Ahmed et al. [185] applied conductive layers to DCMD, combined with electrochemical systems to detect wettability. The membrane acts as an electrode and a barrier to salt. When wetting occurs, water and salt ions pass through the membrane to complete the circuit, thereby increasing the electrolytic conductivity and the current through the electrochemical system. Another in-situ measurement method is based on measurements of transmembrane impedance to detect pore wettability [188]. As mentioned earlier, the water and salt ions complete the circuit through the wetting membrane, and the gap filled with air before wetting prevents the completion of the circuit. As the wetting progresses, the air gap decreases, thus reducing the overall system impedance. Table 3 details the many causes of MD wetting.

**Table 3.** Membrane pore wetting mechanisms in MD.

Cause	Mechanism	Reference
Membrane fouling	Inorganic fouling: inorganic salts deposition on the surface and crystallization inside the membrane pores, thereby reducing the hydrophobicity of the membrane	[189]
	Organic fouling: due to the adsorption between the hydrophobic film and hydrophobic organic matter, thus reducing the hydrophobicity of the membrane	[190]
	Biofouling: the formation of biofilm on the membrane surface reduces the hydrophilicity of the membrane	[191]
Membrane damage	Chemical degradation of the membrane: the chemical oxidation degradation of the membrane formed on the surface of the hydrophilic groups	[192]
	Mechanical damage: Sharp crystals can damage the surface of the polymer membrane and accelerate the wetting	[193]
Feed solution	Low-surface-tension fluid decreases the LEP	[194]

#### 4.3. Energy Consumption

As a thermally driven process, MD is still regarded as an energy-intensive technology [195]. To evaluate the energy efficiency of the MD process, we introduced the concept of specific energy consumption (SEC), which is the energy consumed to produce 1 m<sup>3</sup> of distillate water [196]. The SEC of MD reported in the literature varies from 1 to 9000 kWh/m<sup>3</sup>, and this range was mainly attributed to whether heat recovery or other energy is adopted in the system [197]. In a single-stage MD system, part of the heat is discharged via the rejected brine with a temperature higher than the inlet temperature of the feed. Therefore, energy recovery can be achieved through a multi-stage system [198]. The SEC of pilot-scale MD is shown in Figure 5. The SEC of DCMD system was around 140–1600 kWh/m<sup>3</sup> with an average value around 500 kWh/m<sup>3</sup>. AGMD and VMD have significantly low energy consumption than DCMD, especially for AGMD with an average value around 120 kWh/m<sup>3</sup>. Although SEC could be reduced by heat recovery and system optimization, there is a trade-off between SEC and membrane flux [198]. Therefore, how to reduce the energy consumption without compromising other performance needs further investigation.



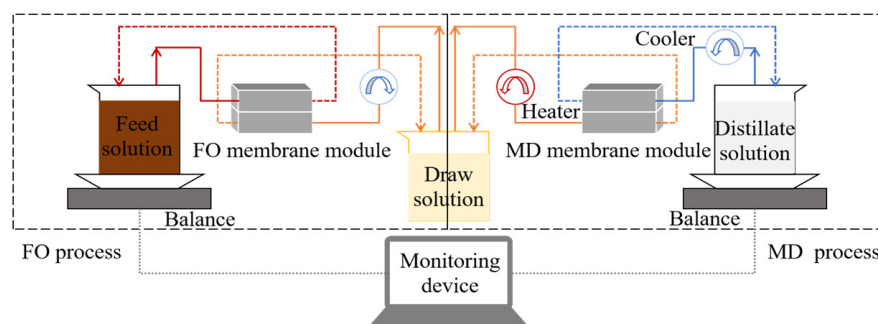
**Figure 5.** A minor review of the pilot SEC of MD with different configurations. Data adapted from Yan et al. [132]. Reprinted with permission from Elsevier (2021).

## 5. New Developments of MD for Wastewater Treatment

### 5.1. Integrated MD System

#### 5.1.1. Forward Osmosis—Membrane Distillation (FO-MD)

As Figure 6 shows, forward osmosis (FO) is an osmotic pressure-driven osmotic technology that uses a semi-permeable membrane to separate and purify water from dissolved solute [199]. FO has higher resistance to membrane fouling than the traditional pressure-driven membrane process because FO hardly needs a pressure drive. However, the FO performance is still affected by dilution of the extracted solution during process operation. Commonly, the extracted solution consists of a high concentration of salt to endow FO with a high membrane flux. As an efficient technology in high salinity water treatment, a combination of MD with FO can not only solve the problem of decreasing flux in the FO but also increase the fouling resistance of the process [14].



**Figure 6.** Schematic of the FO-MD system.

Kwon et al. [200] indicated that the optimal operating conditions and the selection of appropriate driving solutions with a high diffusion coefficient, and low hydrophilicity makes the FO-MD combination process a promising candidate for desalination. Ge et al. [201] applied FO-MD technology to treat dye-containing wastewater, observing a complete rejection of salt. Zhang et al. [202] applied an integrated FO-MD system to oily wastewater, and a recovery rate of >90% could be achieved with almost completely exclusion of oil droplets smaller than 10  $\mu\text{m}$ . The FO-MD system was also applied for human urine treatment, and the FO-MD system could almost completely reject the contaminants in the yellowwater and showed superior rejection performance than MD alone [203]. Zhou et al. [14] first combined FO-MD to treat high-salinity hazardous waste landfill leachate. The salt rejection rate was higher than 96%, and the rejection rate of toxic ions was higher than 98% under optimum conditions. FO-MD showed excellent performance in high-nutrient sludge [204], protein solutions [205], small-scale distributed sewer mining [206], and sidestream from anaerobic digestion [207] treatment.

#### 5.1.2. Membrane Distillation Crystallization (MDC)

As Figure 7 shows, the membrane distillation crystallization (MDC) is a combination of MD and crystallization, with the ability to simultaneously recover valuable compounds and freshwater [208]. Typically, MDC was applied in the crystal recovery in the high-salinity wastewater treatment. Edwie et al. [209] developed hollow fiber membranes to recover water and salt from high-concentration brine by DCMD and crystallization. They also developed synchronous membrane distillation crystallization (SMDC) to treat saturated brine, which solved the problem that the production of water and salt was limited by equilibrium solubility in the early stage [210]. Jia et al. [211] studied the recovery of boric acid from simulated radioactive wastewater by using VMDC. The system was stable in the whole process of concentration, and the rejection rate of boric acid was about 99.5%. In addition, MDC has been shown to have great potential to recover pharmaceutical compounds from wastewater and to recover active pharmaceutical compounds in crystalline form from wastewater solutions [212].



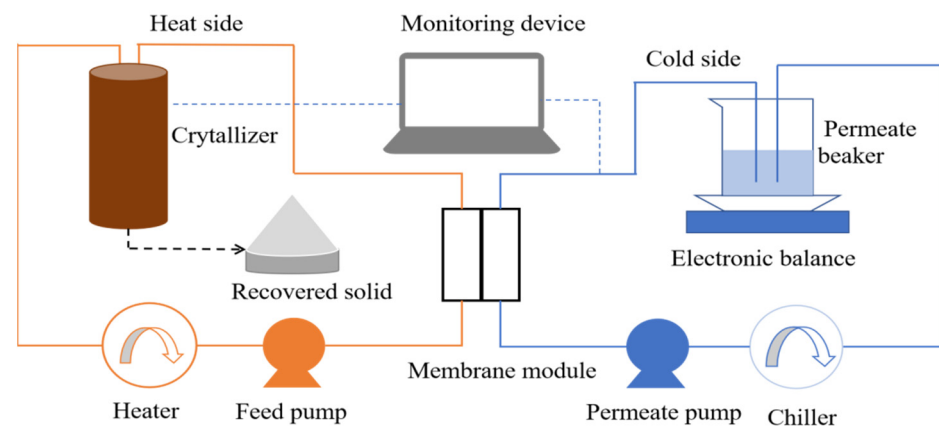


Figure 7. Schematic of the MDC system.

Besides the recovery of crystal and freshwater, the combination of crystallizer was regarded as an effective way for membrane scaling control. Kim et al. [213] applied MDC in shale gas-produced water treatment, and the inorganic scaling was reduced. Julian et al. [214] used vacuum membrane distillation crystallization (VMDC) to treat inland brine and improved mass transfer through transverse vibration and feed aeration, greatly accelerating membrane fouling caused by the crystal precipitation. To control the scaling of gypsum in the MD process, induced crystallization with seeding and heating was able to significantly enhance the MD performance in both membrane gypsum scaling and wetting control [160]. Further, the crystallization mechanism of SMDC has been elucidated through experiments [215]. The results show that a moderate flow rate leads to the lowest scaling tendency. Both surface and bulk crystallization at low flow rates and rapid bulk crystallization due to secondary nucleation at high flow rates lead to decreased flux.

Although MDC has been successfully used to recover freshwater and valuable resources from challenging solutions, membrane surface crystallization and high energy consumption of the crystallization process need to be urgently solved [160].

### 5.2. High Antifouling and Antiwetting Membrane

A hydrophobic membrane is commonly used in the MD process, but the hydrophobic interface is highly susceptible to fouling and/or wetting by the hydrophobic and/or amphiphilic constituents, which are ubiquitous in the various wastewaters. For example, oil in the oily wastewater is prone to induce membrane fouling and wetting in MD. To cope with fouling or wetting of oil, two strategies have been applied. By the combination of surface tension modification and surface re-entrance structure construction, omniphobic membranes with oleophobic surfaces were developed to resist the wetting and fouling of oil [216]. The introduction of a hydrophilic layer in the feed side, usually termed a Janus membrane, showed great improvement in fouling and wetting resistance to the oil [95]. However, when amphiphilic agents, such as surfactants were present in the oily wastewater, the omniphobic membrane was fouled, and the Janus membrane was inefficient in resisting the wetting [217]. Lin et al. [217] constructed a Janus membrane by integrating an omniphobic substrate and an in-air hydrophilic and underwater oleophobic skin layer, endowing MD with the ability to simultaneously resist wetting and fouling.

Besides the Janus or omniphobic membrane, a superhydrophobic membrane has received wide attention in inorganic fouling control. Inspired by the lotus leaf, construction of a membrane surface with similar surface characteristics (high water contact angle ( $>150^\circ$ ) and low sliding angle ( $<10^\circ$ )) endows the membrane with a self-cleaning ability [218]. Karanikola et al. [219] demonstrated that MD membranes with a superhydrophobic, slippery surface was helpful in increasing the water recovery in high-salinity industrial wastewater treatment. He et al. [220–222] comprehensively classified the antiscaling mechanism of a superhydrophobic membrane (slippery membrane), and wetting state and slip length were proved to play a critical role in determining the anti-scaling behavior.

### 5.3. Nutrient Recovery

With the principles of sustainability, the recovery of valuable resources and energy is particularly important in the wastewater treatment industry, as nutrient recovery may generate revenue to offset the cost of wastewater treatment. The optimized application of MDC in the production of high-quality organic and protein crystals has made a vital contribution to the pharmaceutical and biotechnology industries. These include cross-linked enzyme production with large surface-to-volume ratios [223]; high-quality tetragonal protein (lysozyme crystals) [224]; and commercial paracetamol [225] and l-glutamic acid using controlled polymorphism. In contrast, recovery of inorganic crystals/salts using MDC is limited to those present at high concentrations in water sources with mixed ingredients. Table 4 lists the recovery of various salts from different mixtures.

**Table 4.** Resource recovered by MDC from a variety of water sources.

Resource Recovered	Source	Recovery	Yield	Purity	Ref.
NaCl	Produced water	37%	16.4 kg/m <sup>3</sup>	>99.9%	[226]
NaCl	Saturated brine		34 kg/m <sup>3</sup>		[210]
NaCl	Desalination brine	88–89%	21 kg/m <sup>3</sup>		[26]
Na <sub>2</sub> CO <sub>3</sub>	Na <sub>2</sub> CO <sub>3</sub> salt solution			99.5%	[227]
Struvite	Wastewater treatment plant	70%		High	[228]
Struvite	Phosphate-rich feed solution	82%		>99%	[229]
CaCO <sub>3</sub>	Shale gas-produced water	75–84%	2.72 kg/(m <sup>2</sup> d)	>94.4%	[230]
CaSO <sub>4</sub>	Saturated calcium sulfate feed solution		12–16 kg/(m <sup>3</sup> d)		[160]
Boric acid	Boron containing radioactive wastewater	50%		>99%	[211]
LiCl	Single LiCl salt solution	8.3%	15.1 kg/h		[231]

Not only economic factors and appropriate source water identification but also the feasibility of resource recovery is highly dependent on appropriate element extraction methods, as most source water is a complex solution containing a mixed composition of various elements [232]. As can be seen from Table 4, in general, almost all the resources recovered from MDC have extremely high purity, which makes the MDC process very competitive. However, there seems to be no unified expression method for yield, which determines its economic feasibility, and the comparability between different studies is limited. In addition to the recovery of metal ions from seawater, the potential for the recovery of valuable resources from other water sources has also been identified. However, it is worth noting that most of these studies (Table 4) were conducted through chemical modeling simulations and synthetic single-solute aqueous solutions. Selective recovery of valuable elements is challenging, mainly because it is difficult to separate target elements from other major ions. Therefore, there is still a lot of room for development in the research of nutrient recovery based on actual wastewater.

### 5.4. Combination with Waste Heat

As a heat intensify membrane process, exploiting an economical heat source is critical for MD application. Due to the low operation temperature, various heat energies could be used in MD. In addition to the most convenient electric power supply, it can also come from renewable resources such as solar energy [233] and geothermal energy [234], as well as low-grade waste heat such as industrial waste heat [178] and ship waste heat [235].

A lot of waste heat is available in industries, power stations, etc. Compared to other utilization methods, in-place utilization of low-grade waste heat is most economical. Khraisheh [236] studied the feasibility of DCMD for desalination of industrial low-grade waste heat. The investigation used heat integration and heat recovery to screen the suitable low-grade waste heat, demonstrating a hypothetical MD plant with the requirements.

Dow et al. [178] used waste heat from gas-fired power plants for DCMD wastewater treatment, testifying that the 500 MW electric rated power station matched with 8000 kL/d for desalination water produced by MD. This case was done in three months without replacing membranes or components, achieving a water recovery rate of 92.8%. Ali et al. [237] demonstrated the potential application of DCMD in treating flue gas desulfurization (FGD) plant wastewater. In all cases, an excellent quality permeate was produced with a stable flux over the duration of the experiment (for each membrane, the experiment was conducted for four days, approximately 8 h per day). By experimental calculation, the SEC of MD used in wastewater treatment is between 946 and 2830 kWh/m<sup>3</sup>. MD allows more than 80% freshwater to be extracted from FGD wastewater streams while maintaining high (>99.60%) electrical conductivity rejection. The exhaust flow from a natural gas compressor station (NGCS) has been identified as a potential waste heat source that could be used to operate the DCMD, thus providing an economically viable option for the treatment of high-salinity produced water [238]. Energy analysis showed that the waste heat from the gas compressor station was sufficient to concentrate all generated water to 30 wt%, despite the higher initial salinity. Morciano et al. [239] developed small desalination plants (at temperatures below 80 °C). Field experiments have shown that for the engine tested, up to 1.12 kWh/m<sup>2</sup> can be recovered under standard operating conditions, producing nearly 2.61 kg/(m<sup>2</sup> h) for freshwater production from seawater. Silva et al. [240] believed they could reduce the water footprint by using a heat-recovery MD system. The system produced freshwater without any high energy costs. High solute rejection rates and stable permeation fluxes were observed throughout the test. MD membranes showed good chemical stability even after 210 days of exposure.

In the past, desalination on ships was mainly carried out by a multistage flashing system/multiple effect distillation (MSF/MED). However, MSF/MED is powered by advanced thermal energy (temperatures generally above 70 °C), which is much higher than most global waste heat temperatures [241]. Therefore, the focus has moved to recover the heat from the engine cooling system to drive MD desalination. Xu et al. [242] operated a pilot VMD system by recovery using waste heat from ships, and a desalination degree of 99.99% and membrane flux of 5.4 kg/(m<sup>2</sup> h) were achieved under a feed temperature of 55 °C and pressure of 0.093 MPa. Comparative studies of various MD configurations and hydrophobic films to allow for the possible use of waste heat generated in the engine cooling system have been carried out on cruise ships [236]. High permeate quality (99.99% salt rejection rate) and average flux (approximately 13 kg/(m<sup>2</sup> h) were achieved. Koo et al. [243] attempted to improve the efficiency of VMD equipment by combining waste heat generated by ships with experiments under a variety of operating conditions. By recovering heat from ship engine cooling water for multistage AGMD operation, SECs of 1.58 and 2.63 kWh/m<sup>3</sup> were achieved [244].

Although recovery waste for MD utilization seems promising for solving the energy problem, recovery of the low-grade temperature will require a huge investment in heat exchangers, and further research and development in terms of experimentation and modeling are urgent.

## 6. Conclusions

As a promising wastewater treatment technology, MD is able to sustainably alleviate global water stress by recovering freshwater from wastewater. So far, various types of wastewaters, such as desalination brine, textile wastewater, oily wastewater, radioactive wastewater, and landfill leachate have been successfully purified by MD. However, membrane fouling, membrane wetting, and energy consumption remain important factors that impede the practical application. To enhance the treatment performance, the development and modification of novel membranes have been widely investigated. The development of intergraded MD processes, such as FO-MD and MDC, shows great potential in wastewater purification and resource recovery. Combined with low-grade waste heat, the MD system can save energy to a large extent and can achieve low-energy operation, demonstrating its

advantages over other traditional membrane technologies. The capacity of MD wastewater treatment is expected to expand in the coming period.

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