



Review Paper

Membrane Distillation for Water Recovery and Its Fouling Phenomena

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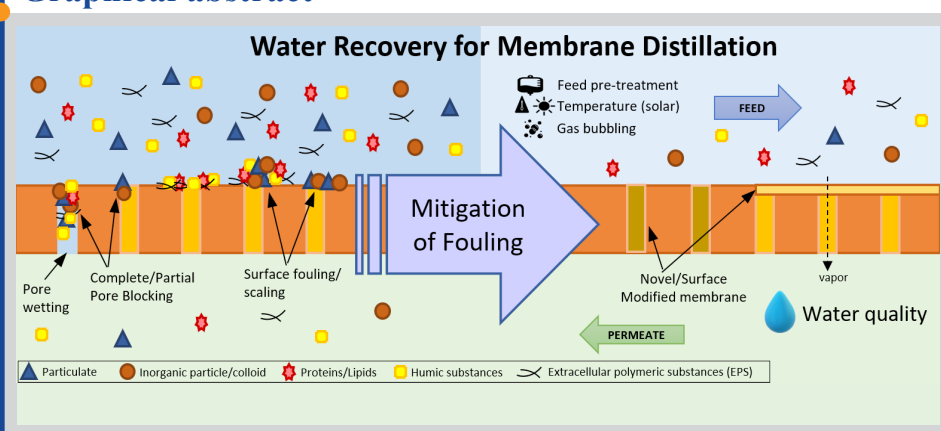
Keywords

Membrane distillation
 Fouling
 Hydrophobicity
 Wetting

Highlights

- Seawater and wastewater water recovery via MD and its fouling phenomena
- Fouling mitigation of MD via material and process design
- Balance of wetting and fouling and its architectural design of hydrophobic/hydrophilic membrane

Graphical abstract



Abstract

The total volume of water on Earth is circa 300 million cubic miles, with close to 98.0% being salt water and the remaining 2.0% fresh water. It has been increasingly more challenging to harvest fresh water from surface water, seawater and even from wastewater due to the combination of factors, viz. burgeoning population growth, rapid industrialization and climate change. Recently, membrane distillation (MD) emerges as a promising cost-effective thermal driven sustainable water recovery technology when integrated with renewable energy sources. However, one of the major challenges for MD is the membrane fouling, which has been gaining popularity in the recent literature, as well. The membrane fouling propensity for MD is very much depends on the type of feed water, suitability of membrane and the operating conditions. The objective of this review is to investigate the fouling phenomena of membrane distillation in wastewater treatment and desalination. The design of membrane and its system from the perspective of material and process design were discussed to provide an insight on the current and future advancement in MD technology for water recovery. Finally, the future trend of MD is projected based on the state of the art development of MD process.

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1. Introduction

Arising from the rapid increase of world's population and development of industry, the global fresh water demand has been putting on a spurt in the recent years. As time goes by, the existing water resources are being polluted by climate change and human activities. Desalination technology is popularly served as an alternative solution to harvest large capacity of fresh water supply. Among the desalination technologies, membrane distillation (MD) has been touted as a promising thermal driven separation technology. It requires lower energy or lower operating temperature which can be tapped from the waste heat compared to the relatively high energy demand pressure-driven process such as reverse osmosis (RO), microfiltration (MF), ultrafiltration (UF), etc. Besides, MD was claimed to have higher fouling resistance than RO and has approximately 100% salt rejection. Nonetheless, MD is regarded as an energy intensive process which is not feasible without tapping at the waste heat. Besides energy requirement, it is evident that membrane fouling is significantly affect the performance of MD for different source of feed water. In spite of its higher fouling resistance, the literatures about the fouling phenomenon in MD compared to RO process remains scarce [1-3].

In the non-isothermal MD process, the hot saline water is circulated on one side of a microporous membrane and cold fresh water is harvested in the permeate side. The temperature gradient across the membrane results in a vapour pressure difference that drives the desalination process. The hydrophobic membrane allows water vapor to evaporate through the pores but hinder the flow of liquid water due to surface tension. MD is not constricted by feed salinity level and requires low thermal energy [4-6]. De-salting of seawater to produce clean water are the most common MD applications due to its advantage of retaining non-volatile particles.

The current review provides a comprehensive overview of MD application for water recovery and its fouling phenomena. As fouling is inevitable in all membrane separation methods, comprehension of the fouling mechanism is crucial towards developing effective strategies for fouling mitigation. This review also summarizes types of foulants, their fouling phenomena on MD and its mitigation method from the perspective of material and process design. Finally, the future outlook of MD in water recovery is also presented.

2. Membrane distillation for water recovery

MD is gaining higher popularity in the application of water recovery process. The advantages and disadvantages of MD when compare with others separation processes such as pressure driven reverse osmosis (RO), concentration driven forward osmosis (FO) and electric potential gradient driven electro dialysis are summarized in Table 1. In general, it is perceived that MD is a low to high energy intensive process with low fouling propensity when it is applied in water recovery treatment due to its lower operating temperatures compared to thermal distillation but higher thermal energy compared to RO process [7]. Higher thermal energy consumption makes it less attractive to be applied in the smaller system capacity (<100 m³/day) compared to other thermal separation technologies such as multi-stage flash (MSF) and multi-effect distillation (MED) [8-13]. In addition, permeate flux decay deters the feasibility of MD application due to the unavoidable thermal polarization and pore wetting phenomena which is aggravated if the membrane is prone to fouling by the complex nature of the water source.

MD is commonly used in separating or concentrating non-volatile components, such as acids, colloids, ions and macromolecules from aqueous solutions. It can also be used to remove traces of volatile organic compounds from water. It is also used to extract organic components for example extraction of alcohols from aqueous solutions. However, for water recovery,

the permeate will be the product stream with other non-volatile components should be retained on the feed side. To date, the applications of MD for water recovery are mainly for desalination and wastewater treatment.

Table 1
Advantages and disadvantages of MD in water recovery

Advantages	Disadvantages
Can be operated at low temperature and pressure with the utilization of renewable energy (e.g. solar, geothermal) or waste heat	Permeate flux is low when compared with pressure-based membrane separation processes
The feed normally does not require pre-treatment	Thermal polarization leads to decrease in flux
High suspended particle or heat-sensitive components are able to be treated using MD	Sensitive to volatile waste components
High membrane surface per volume ratios	Conduction occurs through polymeric membrane
Better for the environment because of the use of simple and non-harmful materials.	Pore wetting and fouling problem
Easy to scale up	MD modules are expensive, where costs are set according to different types of MD configuration and its application

2.1. Wastewater treatment using MD process

2.1.1. Direct contact membrane distillation (DCMD) process

Direct contact MD is the most widely used mode in MD process for both desalination and wastewater treatment due to its simplicity of operation mode. It is the most common reported configuration in MD literature and laboratories fouling studies, concentration of solutions and water or wastewater recovery. However, when compare with other MD configurations, DCMD encounter greatest heat loss via conduction.

In the study by Li et al. [14] who assessed the performance of MD to remove dyes from textile wastewater, small scale of DCMD was used to treat the highly polluted textile effluent. The DCMD test unit was run with microporous hydrophobic membranes made of polytetrafluoroethylene (PTFE) and polyvinylidene fluoride (PVDF). Different industrial dyeing wastewaters were used as feed for the DCMD studies. Results showed that PTFE membrane gave higher flux and rejection performance compared to PVDF membrane. In addition, the rejection efficiency for the organic acid were more than 95% at the feed operational temperature of 50. Nonetheless, PVDF membrane showed higher hydrophobicity (water contact angle = 133.7°) and reduced wettability compared to PTFE membrane (water contact angle = 126.4°). PTFE membrane was further tested on its wetting and fouling properties and it was found that particle such as SiO₂ and dyes might be the main pollutant that promote wetting and fouling. Generally, their findings suggested that it is possible to adopt DCMD in treating dyeing wastewater provided the membrane surface properties need to be improved to

mitigate wetting and fouling phenomena.

In a textile wastewater treatment plant studied by Dow et al. [15], in order to achieve zero liquid discharge, foam fractionation on the untreated waste was applied to avoid MD wetting phenomena. During 3 months of operation, no wetting was observed after pre-treatment using foam fractionation was applied. However, permeate flux start to decline from 5LMH to 2LMH. Chemical cleaning was applied to the membrane and the flux restored to 4LMH. The overall water recovery was 91.6% with >99.9% of non-volatile sulphate rejection. Their works suggested the possibility of using DCMD to treat textile waste and it was suggested to combine with other separation technology such as RO and NF.

2.1.2. Air gap membrane distillation (AGMD) process

AGMD was introduced due to the limitation of DCMD in high heat loss rate through membrane conduction and extra cost for an extra condenser in SGMD. In AGMD, the hot feed stays in direct contact with membrane while the evaporated vapour will pass through a stagnant air gap before it condenses on a cooled plate. AGMD configuration provides less heat loss due to membrane conduction. However, the air gap design has increase mass transport resistance, hence, permeate flux of AGMD is normally lower than DCMD or VMD configurations [16]. As the air gap was introduced and distillate was condensed without contacting with membrane surface, AGMD can be applied in the areas where applications using other MD configurations were restricted. One of the applications is on treatment of produced water.

Produced water is brought to surface during oil and gas production [17]. It is considered the largest waste stream produced in oil and gas industries. In addition, the usage of water in oil and gas industry was high, hence, treatment and reuse of produced water is an alternative way for water resources in oil and gas industry. Alkhudhiri et al [18] has implemented AGMD in treating produced water. In their work, flat sheet PTFE membranes with different pore size were used and 80% of water recovery can be achieved using AGMD. Besides, they also found that energy consumption for AGMD was almost same regardless of the membrane pore size.

In a pilot scale application study of AGMD using solar energy to produce water from saline oily wastewater by Asadi et al. [19], an average water production rate of 1.3 L/m².day was obtained. From the produced drinking water quality analysis, high percentage of contaminant removal was achieved. In another pilot plant study by Woldemariam et al. [20] using AGMD to treat 19 pharmaceutical residues, it was found that the water recovery of 85% could be achieved but the purity of the vapor permeate is very much depends on type of pharmaceutical product with low boiling point compounds could still significantly found in the permeate (Table 2).

2.1.3. Vacuum membrane distillation (VMD) process

In VMD process, the distillate is collected outside the MD module in a condenser which transported by vacuum suction. Normally, a vacuum pump is used on the distillate site which helps to increase membrane permeability by reduced mass transfer resistance. In addition, heat loss through conduction was negligible in VMD configuration. VMD are often used to remove VOCs from aqueous solutions [21].

Application of VMD has been applied in the studies of Sikamumar et al. [22] to treat effluent discharge from coal mine. During coal mining, huge amount of brackish need to be pumped to the surface from mine workings and at the same time fresh water also required for others coal cleaning purpose. Hollow fiber membrane was used in their study and in their work, they managed to achieve 99.9% removal of TDS from mine water. They had demonstrated different operating parameters such as feed temperature, flow rate, salinity and vacuum pressure in affecting distillate flux rate. From the four variables that they studied, they have concluded that the most affecting parameter in VMD process was vacuum pressure followed by feed temperature, flow rate and salinity. Although the distillate quality managed to meet the standard for portable use however, it is still not suitable for human consumption.

Another VMD work has been studied by Mohammadi and Kazemi [23] to optimize phenolic wastewater treatment. In their work, a flat sheet PTFE membrane was used. They had also studied the effect of parameters such as feed temperature, feed pH and vacuum pressure. In contrast with the VMD study on phenolic wastewater treatment by Sikamumar et al. [22], instead of vacuum pressure, they have found that feed temperature was the most influence parameter in their VMD process followed by waste water composition. From the result obtained, they found that water separation factor will increase with decreasing feed temperature and it was almost independent from vacuum pressure. VMD was also adopted by Jia et al. [24] to remove strontium ions from simulated radioactive wastewater. From their study, there was more than 99.6% of strontium ion can be removed from the wastewater

by using hollow fiber membrane.

2.1.4. Sweep gas membrane distillation (SGMD) process

At the distillate side of SGMD, an external condenser is needed to collect vapour because condensation takes place outside the membrane module. Cold inert gas was normally used to create the sweeping flow and collect vapour at permeate side. Due to the sweeping flow, SGMD has lower heat loss with higher mass transport compared to AGMD [25, 26]. Feed temperature and sweeping gas flow rate in SGMD was the significant operating parameter in governing the distillate flux [27].

In glycerol refining, dewatering is one of the most difficult stages due to high boiling point of glycerol at 290°C. Shirazi et al. [28] has study dewatering of glycerol from dilute glycerol wastewater using SGMD process. Operating parameters such as feed temperature, concentration, flow rate and flow rate of sweeping gas has been studied. From the obtained result, concentration of glycerol was successful whereby more than 99% of glycerol was rejected.

In recent year, MD has gained great attention to removed volatile compound such as ammonia due to its low energy condition. Xie et al. [29] studied the application of the SGMD to remove ammonia from wastewater. In the work, temperature and flow rate on the feed side and sweeping gas flow rate on the distillate side were varied to study the effect of SGMD operation parameters on removal of ammonia, distillate flux and ammonia selectivity. They have found that feed temperature was the most important operating parameter for removal of ammonia. 97% of ammonia was successfully removed by using SGMD process in their study.

In another comparison study of using SGMD and VMD to recover fruit juice aroma compound by Bagger-Jørgensen et al. [30], it was found that the concentration factor can be increased by increasing feed temperature in SGMD or increasing feed flow rate in VMD process. High temperature evaporation was the most used techniques in industrial scale for juice concentration and aroma recovery. However, MD offers lower energy consumption for aroma stripping. In the mentioned work, both SGMD and VMD are reassuring methods for aroma recovery and concentration in fruit juice and can be considered as alternative methods in future.

2.2. MD for desalination

Compare to MD for wastewater treatment, MD for desalination has higher literature study. As shown in Table 3, various works have been done to develop membrane distillation and its system for desalination in both lab and pilot scales. Although MD has higher thermal energy consumption than the other membrane technologies as aforementioned in Section 2.0, which limits the implementation of commercial membrane for long-term desalination process, this thermal energy required for heating the feed water can be minimized when the integration of renewable energy sources such as solar energy or waste heat is employed in MD process [31-33]. In view of this, Schwantes et al. [34] introduced a solar driven pilot scale permeate gap membrane distillation (PGMD) unit in Namibia. The unit which consists of 12 parallel multi-modules with each area of 14 m² was directly connected into 100 flat plate collectors with total area of 232 m² for heat absorption. The highest gained output ratio (GOR) value of 4.4 indicated that the system energy consumption is low. This is because the extended module channel improved the internal heat recovery and increased the module efficiency.

Meindersma et al. [35] estimated that the energy cost of the waste heat integrated AGMD using Memstill® technology was 0.14 USD/m³ lower than RO process. Dow et al. [36] powered DCMD with the waste heat generated from a gas fired power station to treat the waste from saline demineralisation plant. This pilot plant achieved stable flux at 3 L/m²h, 92.8% of water recovery and 99.9% of total dissolved solids (TDS) rejection without replacing the membrane and module over three months of operation. Since this work is focused on water recovery of MD, flux decline which caused by membrane fouling is more critical than energy consumption in MD process when considering about the water recovery rate which is defined as distillate production rate over hot feed mass flow rate. Therefore, mitigation of membrane fouling is emphasized in this work in order to achieve more stable flux and higher water recovery rate, at the same prolonged MD operation.

Currently, the performance of membrane with 99% rejection of the salt has reached the highest flux of more than 100 L/hr.m² using PE membrane with pore size of 0.2 µm [37]. However, the results presented in Table 3 do not show the flux stability data especially for the MD with permeability higher than 100 L/hr.m². The possibility of these membranes being wetted and performed high permeation flux cannot be excluded as compared to the current established membrane with average flux of 30 L/hr.m².

3. Fouling phenomena of membrane distillation

Fouling is a phenomenon that allows the build-up of undesired material on solid/membrane surfaces [38]. Generally, fouling materials consist of either living organisms or a non-living substance. Membrane surface or its pores will be blocked via accumulation, attachment or adsorption of foulants during mass transport of membrane distillation process. Fouling will cause

permeate flux dropped especially under prolonged operation. The concept of fouling in MD process is illustrated in Figure 1. As seen in Figure 1(a), initial membrane fouling only happens on membrane surface by deposition or adsorption of foulants. At this initial stage, the water vapor flux can be maintained as there is available open pore area that allows water evaporation to occur in the feed side.

Table 2
Published literature reports on MD and its wastewater treatment applications

MD setup	wastewater	Type of membrane	Scale	Pore size (µm)	Porosity (%)	Thickness (µm)	Membrane area (m ²)	Feed/Permeate Temperature, T _f /T _p (°C)	Vacuum pressure, p _v (kPa)	Feed flow (L/min)		Permeate flux (kg/m ² ·h)	Rejection / Water Recovery	Ref.							
										F	P										
DCMD	Textile wastewater	PVDF flat sheet	bench	0.22	75.0	125	2.75 x 10 ⁻³	T _f : 70 T _p : 20		500 rpm	500 rpm	Initially: 21.9 Decline to: 9.8 (day 6)	Dyes rejection (>99.73%)	[38]							
		PTFE flat sheet	bench	0.22	85.1	180	6 x 10 ⁻⁴	T _f : 60 T _p : 20		0.35	0.25	Initially: 20.7	COD removal (96%) Colour removal (100%) Dyes rejection (99.95%)	[14]							
		PTFE flat-sheet	pilot	0.50	-	-	6.38	T _f : 60 T _p : 20		45-47	45-47	Initially: 5 Decline to : 2 (day 65) Finally : 4 (after cleaning)	Sulphate (>99.9%) Water recovery (91.6%)	[15]							
AGMD	Pharmaceutical wastewater	PTFE with PP support	pilot	0.20	80.0	200	2.3	T _f : 55-90 T _p : 15-50		20	20	2-7	Diclofenac, Atenolol (99%) Carbamazepine, Hydrochlorothiazide (99-100%) Ciprofloxacin (37-99%) Estradiol (70-98%) Estril (76-87%) Estrone (66-86%) Ethinylestradiol (72-90%) Ibuprofen (95-98%) Ketoprofen (92-98%) Metoprolol (100%) Naproxen (62-95%) Norfloxacin (60-98%) Progesterone (67-83%) Propranolol (96-100%) Ranitidine (89-100%) Sulfamethoxazole (92-99%) Trimetoprim (80-99%) Water recovery (85%)	[20]							
								Produced water	PE flat sheet	pilot	0.3	85	76	7.2	T _f : 60 T _p : 3.9		7.5	7.5	Initially: 2.1 Decline to: 1.4	Conductivity rejection (>99.0%) Salinity rejection (>99.0%) Water recovery (80%)	[17]
															Saline oily effluent	Hydrophilic non-porous polymeric material	pilot	-	-	-	40
VMD	Radioactive wastewater	PP hollow fibre	bench	0.18	60	860	T _f : 30-70 p _v : 10.13-99.30		0.175	NA	6.71	Sr ²⁺ ion removal (99.6%)	[24]								
							Phenolic wastewater	PTFE flat sheet	bench	0.22	85	230	1.661x10 ⁻³	T _f : 45 p _v : 6		1.17	NA	31.85	Separation factor (63.64)	[23]	
														Mine Water	PTFE hollow-fiber	bench	0.2	-	0.8	T _f : 65 p _v : 5	

SGMD	glycerol wastewater	PTFE flat sheet	bench	0.22	70	175	-	T _f : 65	0.4	0.4	20.93	Glycerol rejection (99%)	[27]
	Ammonia wastewater	PTFE flat sheet	bench	0.45	70	100	5x10 ⁻³	T _f : 65	0.25	3	12	Ammonia removal (97%)	[29]
	Fruit juice aroma compound	PTFE flat sheet	bench	0.1	-	260	1.59x10 ⁻²	T _f : 45	6.67	20-33	5.04	Aroma recovery (73-84vol.%)	[30]

Table 3
Published literature reports about the application of MD in desalination

MD set-up	Material and Type of membrane	Pore size, d_p (μm)	Porosity, ϵ (%)	Thickness, δ (μm)	Membrane area, A (m^2)	Salt rejection, R (%)	Feed/Permeate temperature, T_f/T_p ($^{\circ}\text{C}$)	Vacuum pressure, p_v (abs kPa)	Feed flow, v (m/s); or Φ (L/min) F: Feed; C: Cold permeate	Permeate flux, J_p ($\text{kg}/\text{m}^2\cdot\text{h}$)	Significant outcomes	Ref.
DCMD		0.45	80	619	0.062	>99.95	40-90/5-25		F: 4.65 L/min C: 3.65 L/min	10-100	The increase of temperature and flow rate in feed side showed more significant in flux improvement compared to the increase of cold permeate temperature and cold permeate flow rate.	[39]
	PTFE Flat-sheet	0.2-0.45	75	100-310	0.014	99.99	70/30		ϕ : 1.8 L/min	9-28 after 1400 min of operation	The distillate with low conductivity (<10 $\mu\text{S}/\text{cm}$) was produced from thermal brines (70,000 ppm TDS content).	[40]
		0.22-0.45	80-82	140-160	0.0050	N/A	30-70/20		F: 0.6 L/min C: 0.5 L/min	10-75	The non-woven support membrane showed better flux compared to the scrim-backing as it is more porous and less exposure to the active membrane surface.	[41]
		0.22-0.45	N/A	160	0.014	99.93	55/25		F: 0.45 L/min	27.7-40.7	The hydrophobic PTFE membrane with 0.45 μm pore diameter showed the best performance.	[42]
		0.22-0.45	N/A	100	0.014				C: 6 L/min	11.3-19.4		
	PVDF Flat-sheet	0.27-0.33	60-80	170-290	0.0058	99	25-60/25-40		0.24	10.2-11.5	The incorporation of 2 wt.% nanocrystalline cellulose (NCC) into the electrospun polyvinylidene fluoride-co-hexafluoropropylene (PVDF-HFP) membrane increased its LEP by 8 psi.	[43]
	PVDF Hollow fiber	0.11-0.75	73-85	130-150	N/A	99.99	80.5/20		F: 0.5 C: 0.15	46.3	PVDF composite hollow fiber with the addition of CaCO_3 nanoparticles showed good permeate flux enhancement, performance stability and thermal efficiency.	[44]
	PE Flat-sheet	0.05-0.20	50-66	45-65	0.001	>99.9	50-80/17		F: 0.5-1.5 L/min C: 0.4 L/min	83.3-123	The PE membrane with larger pore size and porosity showed better flux and energy efficiency.	[37]
	PP Flat-sheet	0.22	75	200	0.0169	99	80/20		F: 0.8 L/min C: 0.4 L/min	37	PP membrane showed higher water contact angle and thickness than PVDF membrane, thus leading to higher salt rejection.	[45]
	PP Hollow fiber	0.65	60-80	150	0.28	N/A	40-92/25-35		F: 0.038 C: 0.475	2-56	The water vapor flux significantly depends on feed temperature, but independent of salt concentration.	[46]
	PP Capillary	0.2-0.4	73	200-440	N/A	>99	60-85/20		F: 0.45-0.84 L/min C: 0.48-0.84 L/min	17-39	Increase in both feed flow rate and temperature improved permeate flux and thermal efficiency.	[47]
PGMD	Unknown polymer hollow fiber	0.22	70	150	0.1-0.5	N/A	40-90		F: 0.5-2.0 L/min	2-35	Higher flow rate and smaller membrane area yielded better flux but reduced heat recovery.	[48]
	PTFE spiral wound	0.2	80	70	5-14	N/A	80/25		F: 3.3-8.3 L/min	3-25 (kg/h)	Larger active membrane area can reduce the specific energy demand using lower saline feed water.	[49]

	PTFE spiral wound	0.2	80	35	10	>99.2	60-80	F: 16.7-20.8 L/min	1.45-2.05	The energy efficiency of the solar driven MD pilot plant unit increased due to the vapor condensation allows internal heat recovery which can be used to preheat feed water.	[50]
AGMD	PVDF Flat-sheet	0.18	85.3	92.7	0.0021	>99	60/20	0.2 L/min	6.2-15.5	The PVDF co-hexapropylene membrane has smaller pore size and its flux was 1.5 higher than commercial PVDF.	[51]
		0.22	70	175	0.00036	>98	60/15	F: 0.92 L/min	Ground water: 21.87 Seawater: 12.11	The permeate flux declined when the coolant temperature and air gap thickness increase.	[52]
VMD	PTFE Flat-sheet	0.22	70	175	0.00036	>99.9	40-60 p_v : 1.5-6 kPa	F: 0.5-0.92 L/min	12-28	Increase in permeate pressure reduced the permeate flux.	[52]
		0.35-0.49	69-72	92-98	0.00785	99.9	40-70 p_v : 3-15 kPa	F: 0.3-1.5 L/min	2-35	The superhydrophobic PVDF-PTFE nanofibrous membrane showed stable performance due to its bigger fiber diameter, pore size, LEP and contact angle.	[53]
	PVDF Flat-sheet	0.49	78	82	0.00264	99.9	73 p_v : 31.5 kPa	F: 0.9 L/min	22.4	VMD has better permeate flux and thermal efficiency compared to DCMD.	[54]
	PVDF Hollow fiber	0.18	86	250	0.8	99.8	68-88 p_v : 17-47 kPa	F: 0.033-0.05	3-14	The efficiency of VMD can be improved by optimising the feed flow rate and temperature solution as well as the vacuum degree.	[55]
	Alumina Hollow fiber	0.7	42.8	200	0.004	99.5	80 p_v : 4 kPa	N/A	42.9	The membrane was prepared via the method of phase inversion extrusion, sintering and surface grating with fluoroalkylsilane (FAS).	[56]
	PP hollow fiber	N/A	N/A	53	12.3	99.99	55 p_v : 8 kPa	F: 8.3 L/min	5.4	Low grade heat from the vessel engine was used to preheat seawater feed.	[57]
VMEMD	PTFE flat sheet	0.2	N/A	N/A	5	N/A	75 p_v : 9 kPa	F: 8-27.7 L/min	7	Increasing the number of effects is limited by the hydraulic resistances of the feed flow and the boiling point elevation with salinity	[58]
SGMD	PTFE Flat-sheet	0.2	75	600	0.035	N/A	50	F: 0.8 C: 6.3-11.3	1.5	Compared to PP and PVDF, highest flux was obtained when the feed temperature increased by 30°C.	
	PVDF Flat-sheet	0.2	75	600	0.035	N/A	50	F: 0.8 C: 6.3-11.3	1.25	Distillate flux was achieved up to 4.2 when the feed temperature increased by 30°C.	[59]
	PP Flat-sheet	0.2	75	600	0.035	N/A	50	F: 0.8 C: 6.3-11.3	1.0	As feed temperature increased from 25°C to 60°C, the temperature polarization was induced.	

The deposits will then continuously cover the membrane pores, which partially or fully blocks the membrane pores as shown in Figure 1(b, c and d). Some pores are partially wetted or covered due to the attachment of hydrophilic foulants onto the inner wall of the pores. The fouling exacerbation affects the flow pattern of bulk feed and thus significantly increases the resistance of heat and mass transfer between the bulk feed and evaporation interface. As a result, the temperature and concentration polarizations are induced at the evaporation interface. Consequently, water vapour flux is declined due to smaller water vapour pressure gradient across the membrane. Besides, the polarizations will cause the hot brine feed solution to become supersaturated and gradually exhibit crystallization growth in the pores, which significantly leads to complete pore wetting toward the permeate side of the membrane, as illustrated in Figure 1(e and f).

Even though literature has reported that MD is less prone of fouling, however the direct contact of hydrophobic membrane to high concentrated solution under elevated temperature, in fact, makes MD process more susceptible to membrane fouling [60]. In general, membrane fouling can be affected by:

(i) Surface charge

Deposition of solute or particle on the membrane surface can be

contributed by surface charge of membrane and foulants. When the membrane and foulants have counter-charges, fouling is more susceptible due to electrostatic interactions. For instance, the polyamide based NF and RO membranes which are commonly negatively charged is prone to fouling by the multivalent cations [61]. However, it was expected that the effect of surface charge on the membrane distillation should be lower under the conditions of minimum contact of the solution with the omniphobic membrane surface.

(ii) Membrane Hydrophobicity

Hydrophobicity has mixed effect on membrane fouling. Increasing membrane hydrophobicity may renders higher fouling phenomena because foulants like proteins are hydrophobic which favours its adherent via hydrophobic interaction. The hydrophobic interactions enable the wicking of protein or oil into the pores and caused pore blocking. Pore blocking hinders the vapor permeation, therefore reduced its productivity. On the other hand, superhydrophobic membrane or omniphobic membrane could minimize the contact of the water or the foulants on the membrane surface due to the higher surface tension. The roll off effect of the foulants can reduce the chances of foulants deposition and indirectly membrane fouling.

(iii) Surface Roughness

A rough membrane surface will experience more fouling because foulant particles are more likely to be entrapped on the rough surface. When the membranes have a rough surface, foulants can deposit within the rig and valley structure and hinder it from the hydrodynamic cleaning action [61-63]. However, for membrane distillation, the hierarchical surface roughness provides beneficial features in avoiding the fouling. It can be achieved by inducing the surface omniphobic characteristic which could prevent the direct contact of the solution with the membrane surface.

There are four types of foulants that lead to membrane fouling namely i) biological foulants such as microalgae, colloidal and flocs, ii) organic foulants such as polyelectrolytes and iii) scalants such as precipitates of mineral. Microalgae is abundant in the nutrient rich organic wastewater effluent whereas minerals can be commonly found in the brackish or seawater. Compared to fouling in pressure-driven membrane system, fouling study in MD is still scarce [3, 64]. In MD, the foulants generally interact with each other under the induction of temperature effect and might deposit on membrane surface and caused initial pore wetting followed by the irreversible pore blocking.

3.1. Organic fouling

Organic fouling is normally related to natural organic matter (NOM). NOM is prevalent in natural waters and poses the risk for MD. Humic substances are the main components in ground/surface water followed by carbohydrates and protein [65-67]. High organic contents in the feed would lower the hydrophobicity of membrane [68] due to adsorption of organic matters which then contribute to membrane wetting. Organic fouling analysis in MD studies is still deficient and fouling detection are mainly based on permeate flux decrease trend [3, 69-72]. Compared to the polymeric organic materials, fine organic foulants are expected to cause more serious wetting problems due to its micro-sized [73].

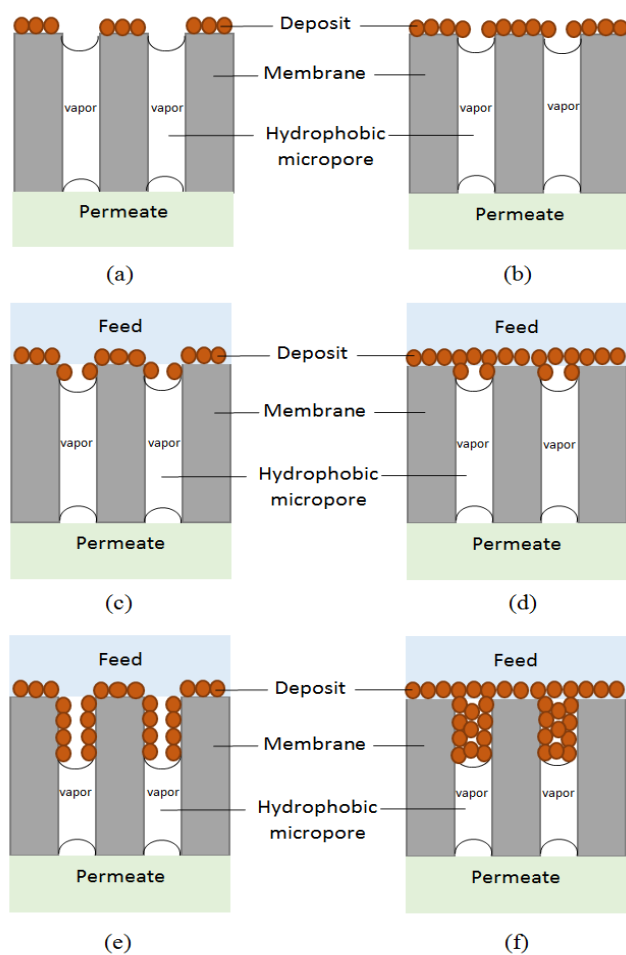


Fig. 1. Membrane fouling in MD process (Adapted from [74])

There are contradicting observation pertaining the organic fouling on MD. In one of the study, the presence of organic foulants did not affect the permeate quality [3, 69] even though the phenomenon of surface wetting was obvious. Another study reviewed that organic foulants could decrease the membrane contact angle and increased the organic concentration in the permeate [75, 76]. In fact, both studies showed that different type of organic foulants will have different degree of wetting on the membrane surface. For instance, Goh et al. [76] found that MD bioreactors that contains dense accumulated extra polymerase has very low fouling rate. Likewise, study carried out by Naidu et al.'s [76] showed that humic substances induced wetting faster compared to protein.

There are a lot of factors that organic compounds can contribute to membrane fouling such as hydrodynamic condition, ionic strength, membrane surface structure and solution pH [77]. From the perspective of chemical properties, organic fouling should be more prevalence for hydrophobic membrane compared to hydrophilic membranes (non MD membrane) due to the presence of hydration layer that could prevent the adherent of organic foulant directly onto the membrane surface [78, 79]. There are three main organic compounds that cause membrane fouling in MD which are discussed below:

i. Polysaccharide (AA)

Phattaranawik et al. [80] monitored the fouling phenomenon in the MD bioreactor by polysaccharide. It was found that the repulsion force between hydrophilic AA and hydrophobic membranes result in less attachment of AA on membrane surfaces [66]. Similar results were reported by Naidu et al. [75] who found that AA has lower fouling rate when compared to humic substances and proteins. AA appears to be the important components in microalgae in most of the water source. The low AA fouling rate is in fact encouraging for employing MD for water recovery.

ii. Humic substance (HA)

Fouling of humic substance for MD has been widely studies [69-71, 81]. HA is a type organic acids with its reactivity are comes from phenolic and carboxylic groups [82]. It is obtained from the biodegradation of organic matter from the rivers water with the appearance of yellowish and brown colour [83]. It was highlighted that HA will nutrify bacteria, and lead to significant bacterial fouling [84]. According to Jucker and Clark [79], humic substances are more favourable absorb onto hydrophobic membrane. This finding has been proven by Khayet et al. [71] who carried out the DCMD study using synthetic HA as feed. In the experiment, two commercial MD membranes were used and the results showed that the more hydrophobic commercial MD membranes has higher fouling rate compared to the hydrophilic membrane. In addition, they have proved that fouling of humic substance is an irreversible process when they cannot recover the initial permeates flux by water flushing during membrane cleaning.

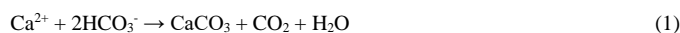
In an HA fouling study by Naidu et al. [75], they reported that the organic concentration on the permeate will increase gradually. These low molecular weight humic substances are able to penetrate through membrane pores and increases permeate organic concentration. In the DCMD study with seawater by Naidu et al., they found that humic substance was the major organic foulants. It was found that at high temperature, molecular size of humic substance decrease and disaggregate occurred when humic substance was observed using dynamic light scattering (DLS) and ultrasonic velocimetry [85, 86]. However, there is no change on structural characteristics of HA compound up to 200–400°C. Hence, when temperature increased, rate of humic aggregation increased and thus increase the fouling on membrane distillation [67].

iii. Protein (BSA)

MD operated at high feed temperature is one of the causes for protein fouling [87]. Feedwater containing proteins has a high propensity to accumulate on the hydrophobic membrane [3]. BSA fouling can caused 60%-70% decrease in permeate flux [72]. In a DCMD study comparing between BSA and HA foulants by Naidu et al. [75], they reported that BSA compound has higher organic mass accumulation on the membrane when compared with HA compound. However, there is no pore wetting observed when BSA fouled on the membrane while HA foulants on the membrane will cause pore wetting. In addition, BSA compound showed higher fouling reversibility compared to HA compound when flushing with DI water. For fouling of BSA compound, high feed temperature of 85°C will decline the permeate flux as much as 72% by forming the gel-like structure on the membrane surface [68, 88].

3.2. Inorganic fouling

Inorganic fouling is generally known as scaling. It involves salt precipitation from feed solution deposited onto the membrane surface. Consequently, it affects the water vapor transport across the membrane and significantly cause a flux reduction [6]. MD scaling occurs when supersaturation is reached due to water evaporation and temperature changes via crystallization growth on the membrane surface [89]. The scales tend to tackle the larger pores and consequently induce pore wetting as illustrated in Figure 1 (e and f) [5]. Deposition of the crystal on the membrane surface will increase temperature polarization and consequently causing the permeate flux decline [4]. Common calcium-based scales such as calcium sulfate, calcium carbonate and calcium phosphate are widely found in water source like seawater. Besides, the presence of silica, iron oxide and barium sulfate are others contributing foulants for MD operations. Table 4 shows some literature reports on inorganic fouling in DCMD [81, 90-98], AGMD [99-103] and VMD [104-108]. According to Guillen-Burrieza et al. [103], PTFE flat sheet membrane module in the pilot-scale solar powered AGMD unit which was partially soaked in saline water feed could induce salt crystallization to happen during dry-out periods. The membrane should be rinsed with pure water after operation and then kept under freshwater or completely dry. Based on Table 4, Calcium carbonate is found to be the major scale in MD process and most prevalent to be supersaturated in feed solutions [10]. Calcium carbonate precipitate is dominantly formed after breaking down the bicarbonate, HCO_3^- [12], as shown in equation below:



Increase of pH and carbonate concentration promote calcium carbonate precipitation [109]. Qin et al. [110] reported that flux declination of DCMD is lower than that of VMD and SGMD based on calcium carbonate fouling impact. This is because the decomposition of dissolved $\text{Ca}(\text{HCO}_3)_2$ in VMD and SGMD is faster than that of DCMD which will shift the reaction to the right to produce CaCO_3 upon CO_2 removal. Besides, solubility of CaCO_3 is inversely proportional to temperature. High temperature of feed solution will induce CaCO_3 crystal formation (see Figure 1). Many studies reported that pure calcium carbonate scales causes rapid flux decline up to 66% due to its non-porous nature [111, 112]. However, previous works studied that higher feed flow rates will reduce crystallization growth and make the carbonate scales more porous and loosen [111]. Besides, many researchers found that the penetration of calcium carbonate scales into the membrane pores could induce wetting and contamination of permeate [113].

CaSO_4 is the second most common encountered non-alkaline scales in MD especially for desalination. CaSO_4 scale is reported as a recalcitrant scale in thermal distillation, and also in MD processes too [113]. The different crystal structures of CaSO_4 and CaCO_3 will definitely give different compactness of the cake layer. The morphologies of the CaCO_3 [81, 97], CaSO_4 [97, 114] and silica [97, 115] scaling on different membrane types can be visualized by a collection of SEM images as illustrated in Table 5. According to Gryta [113], the needle-like gypsum crystals is prevalent to penetrate into membrane pores, thus consequently, inducing more pore wetting and membrane damage that could reduce the flux as much as 29%.

Cleaning of CaSO_4 is complex compared to other alkaline scales, therefore some studies have focused on scale-mitigation conditions [109]. Naidu et al. [116] observed that a slight permeate flux decline (18–20%) by the loosely-deposited CaSO_4 crystals was found in the V-MEMD system after 920 mins of operation under the conditions of using low feed temperature and high feed flow. Periodic washing using deionised water was efficient for scaling removal and permeate flux recovery. For desalination, seawater pretreatment to remove the sulfate can be crucial to prevent the CaSO_4 deposition.

Sodium chloride is the main component presents in the seawater. NaCl, as non-alkaline scale, is very soluble in water at high temperature. Therefore, it is lack of concern in MD fouling studies. However, some scaling experiments reported that increase of NaCl concentrations could induce the concentration and temperature polarization phenomena that tend to form small crystal on the membrane surface [117, 118]. Besides, some studies reported that during membranes drying out, NaCl salts deposited on the membrane surface and causing pore wetting [95].

Silica is another common inorganic scalants found during membrane desalination. In natural waters, silica can be in the colloidal, particulate or dissolved form. Once the supersaturation is reached (100 ppm above at pH 7.0), insoluble silica polymerizes in the form of precipitates and deposits can be found on the membrane surface [119, 120]. pH also plays an important role in ionising and polymerising silica. Silica is more likely unionized at pH less than 5 or higher than 10, reducing the risk of scaling [121, 122]. Since silica solubility increases at elevated temperature, scaling risk by silica for MD is relatively low. As reported by Qin et al. [110], silica feed has negligible impact on both DCMD and SGMD fluxes because both the silica scales on the DCMD and SGMD were porous in nature. On the other hand, Karakulski et al. [112] found that silica precipitation was very significant for capillary membrane. Flux decline is usually caused by the clogging of capillary membrane inlets [112].

3.3. Biofouling in membrane system

The adherence of microscopic organism and the formation of biofilms layer are called biofouling. In membrane processes, biofilm growth and biofouling will promote reduction in permeate flux due to reduction of transfer mechanisms and thus affect membrane performance, especially during long-term operation [123-126]. Microbial growth and adherence rate are governed by variety of factors such as flow rate [127], hydraulic pressure [128-130], membrane properties [131-135], microbial cells properties [136] and feed solution source [5, 6, 137] which presented in Table 6.

Most microorganisms form specialized surface attached communities called biofilms. Biofilm formation includes several phases that take place steps by steps or simultaneously.

a) Formation of conditioning film

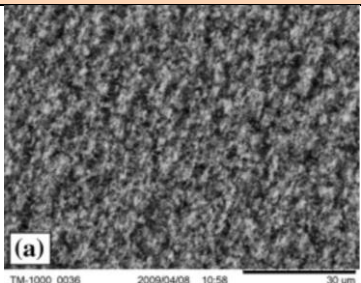
Existing organic colloids and dissolved organic carbon in aqua are adsorbed on the surface of membrane and coat the surface with nanometre-thin (<300nm) organic conditioning layer or bacterial primer within minutes that comprised of polysaccharides, proteins, lipids, nucleic acids and humic acids [136, 138-140]. The existence of conditioning film will promote the adhesion of microorganisms [5, 141].

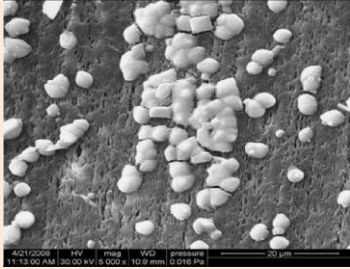
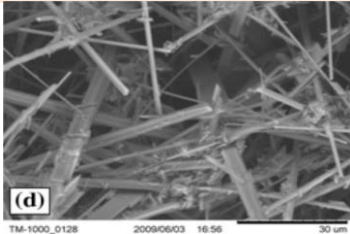
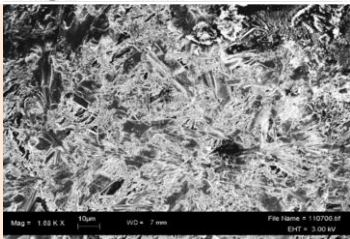
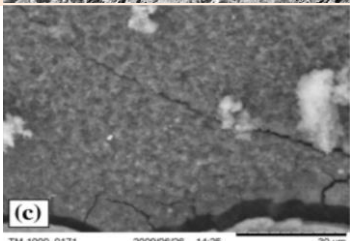
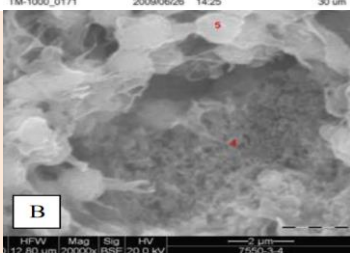
Table 4
Published literature reports about inorganic fouling in the MD application

MD set-up	Material and Type of membrane	Pore size, d_p (μm)	Porosity, ϵ (%)	Thickness, δ (μm)	Foulant	Feed/Permeate temperature, T_f/T_p ($^\circ\text{C}$) Vacuum pressure, p_v (abs kPa)	Feed flow, v (m/s); or Φ (L/min) F : Feed; C : Cold permeate	Permeate flux, J_p ($\text{kg}/\text{m}^2 \cdot \text{h}$)	Observations	Ref.
DCMD	PTFE Flat-sheet	0.2	75	60	CaSO_4 and MgSO_4	40-50-60/25	0.03-0.06	12-18	Increasing feed temperature doubled up the initial water flux of the process. However, it also magnified polarization effects and promoted membrane scaling due to supersaturated CaSO_4 .	[92]
		0.2	70	175	CaCO_3 , CaSO_4 , Na_2SiO_3	40/20	ϕ : 1.0 L/min	11-12 1-11 7-11	CaSO_4 scaling more severe than that of CaCO_3 and Na_2SiO_3	[97]
		0.2	N/A	175	CaCO_3	70/20-40	ϕ : 1.5 L/min	20-35	Flux did not decline after 17 h of operation with NaCl solution, rejected brine and seawater. Only small fouling layer of CaCO_3 was found on membrane operated on seawater.	[94]
		0.2	65	41	NaCl	30-50/24	0.32	28 at $\Delta T = 15$	The PTFE membrane surfaces had scales with the presence	[95]

PVDF Flat-sheet	0.2	80	197	MgSiO ₃ , MgCO ₃ , CaCO ₃			°C	of crack and smaller crystals.	
	0.45	N/A	125	Gypsum, CaCl ₂ , Na ₂ SO ₄	60/20	0.085-0.17	2.3-9.3	PVDF membranes surface had larger crystals and its hydrophobicity reduced. The modified slippery surface PVDF membrane showed the most stable MD performance in the presence of gypsum scales. [91]	
PVDF Hollow fiber	0.34	83.31	221	N/A	60/20	F: 0.016 C: 0.01	8-11	Deposits formed at inner surface of doped membrane with PVP additive, and pore wetting was induced. [93]	
PP Flat-sheet	0.1	65-70	100	CaCO ₃ , CaSO ₄ , SiO ₂	60/20	φ: 0.6 L/min	30	Membrane scaling caused both flux and salt rejection decline. [96]	
PP Hollow fiber	0.2	70	N/A	CaCO ₃	40/20	φ: 7 L/min	1.4-2.05	Vaterite found on the membrane surface at high concentrated factor of 4-6. [81]	
	0.46	80	250	MgSO ₄ , CaSO ₄	55-60/30	φ: 2 L/min	4.3-8.7	The deposits consist of organic components combined with inorganics and were found to be hardly flush off by water rinsing. Ca and Mg content could make deposits more compact. [90]	
PP capillary	0.22	73	N/A	Iron oxides	60-80/20	F: 1.01 C: 0.38	32-19	A porous structured rust found on the membrane surface without flux decline. Concentrated HCl solution used during rinsing will induce membrane wettability. [98]	
AGMD	LDPE Flat-sheet	0.3	85	76	CaSO ₄ and MgSO ₄	35-60/25-50	0.03	2.5 to zero at T _f /T _p = 60/50 1.5 to 0.9 at T _f /T _p = 60/50	Water flux rapidly decreased as the feed salinity increased due to salt deposition. [99]
	PVDF Flat-sheet	0.2	N/A	200	CaSO ₄	70/20	20-23	20-22	The filters with 20 μm significantly reduce scaling and anti-wetting. The heterogeneous nucleation of crystals more likely formed in the bulk and on the surface. [101]
	PTFE Flat-sheet	0.2-0.45	80	135-139	NaCl	45-82/25	F: 1.7 L/min	3.8-36	Salts were deposited over the membrane surface with significant flux decline rate of 2.37%/h and 4.10%/h at 82°C for the membrane of 0.2 μm and 0.4 μm respectively. [100]
		0.18	64	54	NaCl, Mg, Fe, Al oxides	60-80/15-37	F: 20 L/min	4.5	Porous NaCl deposited on top surface membrane with agglomeration of metal oxides. The fibrils were slightly damaged and defected. [103]
	0.45	90	100	CaCO ₃	40/10	F: 5 L/min C: 6.7 L/min	3.2-4.54	Crystals growth fouled the membrane with pore clogging. [102]	
VMD	PTFE Flat-sheet	0.22	40	175	CaCO ₃ , CaSO ₄	25-75 p _v : 0.1-10	0.4-2.0	9.3-8.3	Calcium carbonate and calcium sulphate precipitated on the membrane surface with minor impact on permeate flux. [108]
	PTFE Hollow fiber	0.46	63.4	700	NaCl, CaCO ₃	70 p _v : 5	φ: 1.0 L/min	11.7-13.8	Crystals deposited on the membrane surfaces after 20 days of operation. [106]
	PVDF Hollow fiber	0.25	79	150	CaCO ₃	52-68 p _v : 5	0.14	8.96-25	Microwave irradiation inhibited calcite scales. [107]
	PP capillary	0.22	N/A	N/A	NaCl, CaCl ₂	70/25 p _v : 3	0.075	7.62-8.06	Serious wetting occurred after 22 h of operation. [105]
0.2		73	450	MgSiO ₃ , CaCO ₃	70 p _v : 6	N/A	5.5	Calcites and magnesium silicates found on the membrane surface and caused rapid flux decline. [104]	

Table 5
SEM images of showing scaling morphologies by use of various membranes under different operating conditions.

Scaling	MD set up	Membrane type	Feed/ Permeate temperature (°C)	Feed/ Permeate flow rate (L/min)	SEM images	Adapted from
CaCO ₃	DCMD	Flat sheet PTFE	40 / 20	1 / 1		[97]

		Hollow fiber PP	40 / 20	7 / 7		[81]
CaSO ₄	DCMD	Flat sheet PTFE	40 / 20	1 / 1		[97]
		Hollow fiber PP	86 / 22	0.465/ 0.138		[114]
Silica	DCMD	Flat sheet PTFE	40 / 20	1 / 1		[97]
		Flat sheet PVDF	75 / 50	0.5-0.9/ 0.5		[115]

b) Attachment of microorganisms

Environmental signal will attract microorganisms to come closer to the conditioning film and attached via weak van der Waal forces whereby the attachment was reversible and can be removed by mild shear forces. Besides van der Waal forces, other factors such as surface hydrophobicity, surface charges, electrostatic repulsive force and hydrogen bonding will affect the attachment bonding between the conditioning film and microorganisms. It usually takes place within 1 hour with 10% of the microorganisms' population irreversibly attaching to conditioning layer.

c) Excretion of Extracellular Polymeric Substances (EPS)

However, as the attachment time of microorganisms become longer on the surface, they will excrete EPS which is crucial for the establishment and continuance of micro-colonies and oxygen-free conditions in the development of biofilm. Excretion of EPS act as a bridge between microorganisms and conditioning layer which will result in irreversible attachment of microorganisms by covalent, electrostatic and hydrogen bonding, further with van der Waals, dipole-dipole and hydrophobic interaction between microorganisms and surfaces [141]. EPS are basically made up of high molecular weight of polysaccharides and proteins [130] where they were responsible for the structural and functional integrity [142].

d) Biofilm formation

Biofilm develop with the growth of microorganisms and continuous excretions of EPS which will then allow larger organisms that flowed over trapped within biofilm and contribute to additional nutrient source and increase in biofilm size. Normally after 8 hours of biofilm development, there will be more than 91% of microorganisms are irreversibly attached. The continuous nutrients sources will enhance the biofilm formation. Under this condition, a mature biofilm structure will normally form within 1 day. The colonization of larger organisms is referring as biofouling which will then affect the performance and mechanical strength of membrane [142-145] by inducing membrane wetting, which assisted the movement of microorganisms and endospores through the membrane structure into the distillate

e) Biofilm aggregation

Fragment of the biofilm may shed from time to time. The released microorganisms may be moved to a new spot to develop new biofilm or remain inside the liquid as a contaminant. Hitherto, limited effort has been put in the study of biofouling in the membrane distillation mainly due to its higher operating temperature that retard the growth of microorganism [84]. MD desalination in biofouling studies normally includes algae, bacteria, biofilm and fungi [6]. Microbial are widespread in waters [146] and becomes

an issue to the performance of membranes [147]. Nevertheless, highly feed temperature and salinity during MD operation can restrict the microbial growth. Hence, biofouling in MD will be less severe than other membrane separations process for example NF, RO and UF etc.

Table 6
Factors that affect microbial growth and adherence rate on membrane surface

Factor	Further explanation	Ref.
Flow rate	The lower the flow velocities, the thicker the hydrodynamic boundary layer. The laminar flow will affect membrane surface shear stress diminished which then increases the attachment of bacteria and thicker biofilm formation.	[127]
Hydraulic pressure	Biofilms that build up under high hydraulic pressure were normally homogeneous, densely packed and covered with EPS which were hard to remove physically while biofilms that build up in unpressurized condition were thicker and scattered.	[128-130]
Membrane properties	Interaction between foulants and membrane depends on membrane properties like surface roughness, surface charge and membrane hydrophobicity. Rough surface membranes tend to attract more bacteria to attach on membrane surface due to larger surface area compared to smooth surface membranes. Besides, positively charged membranes were more susceptible to the adhesion of bacterial due to electrostatic attraction of negatively charged bacterial. Furthermore, microorganisms tend to attach on hydrophobic and nonpolar surface rather than hydrophilic surface because hydrophobic surfaces will reinforce attachment of bacterial cells which is a reversible process.	[131-135]
Microbial cells properties	Structure of microbial cell, microbial cell hydrophobicity and EPS production were examples of properties that will affect the rate of attachment on membrane surface. A bacterium consists of fimbriae which contribute to cell surface hydrophobicity and attachment between the cell and membrane surface by overcoming the electrostatic repulsion barrier.	[136]
Feed solution source	Different source of feed solution have different properties in term of amount of bacterial, nutrients levels, present of organic and inorganic material, ionic strength, dissolved oxygen, pH etc. High concentration level of microorganisms and nutrients in feed enable rapid formation of biofilm.	[5, 6, 137]

From the study by Krivorot et al. [84] with hollow fiber MD, during the process at 40°C, a conditioning biofilm was formed within 4 hours and a noticeable biofilm was clearly visible after 28 hours. However, biofouling was reduced when the process was run at 70°C. Hence, operate at higher temperature can reduce biofilm formation. In another study done by Gryta [125] showed that Streptococcus bacteria will pass through the hollow fiber and enter to permeate side during DCMD. In the experiment, bacteria and fungi were detected on the surface of the membrane when it was operated at 80°C with 300,000 ppm of sea salts. However, there is no bacterium found on the membrane surface when the temperature was elevated to 90°C. It can be noticed that by increasing the operating temperature, increasing the concentration of salt, and operating at low pH values can prevent the growth of bacteria. Both studies have suggested that increasing in operating temperature can prevent biofilm formation.

In biofilm formation, microbial cells will die at high temperature. However, high temperature cannot get rid of EPS that bacteria have already produced. Bogler and Bar-Zeev [148] have a comprehensive study of biofouling at different bacterial growth temperature at 47°C, 55°C and 65°C using PVDF membrane in DCMD system whereby 55°C is the optimum temperature for the growth of bacteria. From the result obtained, feed temperature operate at 47°C has a 30% flux declined while optimum bacterial growth temperature of 55°C has 78% of flux declined after 3 days operation. Although the bacteria proliferation was weakened at 65°C but the

production of EPS and extensive endospores formation had caused serious pores wetting in MD system. In another experiment done by Zdrorow et al. [129] to compare the biofouling between MD and RO had suggested that bacteria can live at high temperature by forming endospores which will later adhere on the membrane surface when the temperature was lowered. They have observed that although bacteria concentration decreased but there is still biofilm formation.

The conclusion given by Krivorot et al. [84] and Gryta [125] stating that biofouling is not significant due to higher operating temperature may impede biofouling formation is different from the experiment results from Bogler and Bar-Zeev [148] and Zdrorow et al. [129] where they suggest that biofouling still occur at high temperature due to EPS produced by bacteria. Different conclusions were made by researchers regarding the significance of biofouling is because biofouling studies conditions were dynamic. Biofouling can be varied in terms of feed sources, water quality, membrane properties, operating parameters, bacteria species and duration of experiment conducted. In future, experiment on factors affecting biofouling need to be conducted such as effect of water conditions, operational conditions and bacterial conditions to have a strong comprehension on biofouling studies in MD process.

4. Mitigation of membrane distillation fouling via material and process enhancement

Based on the discussion in the previous section, membrane fouling for desalination or water treatment is inevitable due to the presence of various foulants. Intensive efforts have been made to mitigate the fouling issue. The promising fouling mitigation approaches can be categorised into material design strategies which includes formulating new materials and membrane modification, and also process design such as operating condition optimisation and feed pre-treatment.

PVDF membrane, in spite of widely used for MD application, its synthesis route of pristine PVDF membrane still does not meet the MD requirement in terms of liquid entry pressure as well as wettability. The suitable MD membrane is still limited to the composite membrane with surface roughness enhancement via nanoparticle dosing. In view of this, most of the works have been focused on membrane modification and material development to mitigate the membrane fouling and wetting during MD process. For example, Khan et al. [149] successfully functionalized polyethersulfone membrane with hybrid organic-inorganic material by dip-coating the perfluorodecyl triethoxysilane and polydimethylsiloxane modified silica nanoparticles under vacuum filtration. The modified PES membrane possessed amphiphilic nature and performed an excellent flux, anti-wetting and anti-fouling behaviour, better salt rejection and longer durability compared to commercial PP and PVDF membranes. Besides, the particular membrane also achieved consistent flux at 17 kg/m²h using high saline salt as feed with concentration of 1 M containing 10 ppm of humic acid (HA).

Shao et al. [150] also fabricated a composite PP membrane with silica nanoparticles and fluorine coating in order to improve the membrane surface roughness and super-hydrophobicity. The fabricated PP membrane presented excellent fouling and wetting resistances and able to achieve consistent permeate flux at lower feed rate of super-saline solution during the long-term operation of VMD. Politano et al. [151] proved that temperature resistance can be minimised by manipulating thermal collective effects exhibited by an UV-irradiated composite PVDF membrane with silver nanoparticles. Based on the experimental results, with the presence of excited plasmonic modes, PVDF membranes with 25% of silver nanoparticles loading exhibited stable permeate fluxes of 25.7 L/m²h in the VMD process using a 0.5 M of salt feed solution, which was about 10 times higher than the unloaded membrane.

4.1. Material design

(i) Hydrophobic membrane

In designing the material for membrane distillation, hydrophobicity and its structural stability are the utmost important design parameters. Membrane with low hydrophobicity is prone for surface wetting which will lead to the unwanted fouling problem. Based on the literature, hydrophobic membrane materials commonly used for MD are Polyethylene or polythene (PE), Polypropylene (PP), Polyvinylidene difluoride (PVDF) and Polytetrafluoroethylene (PTFE). Unfortunately, most of the hydrophobic materials cannot be made into membrane via the facile phase inversion method due to its solvent resistance characteristic. Nonetheless, surface modification is introduced to lower down its surface energy that suitable for its application.

Membrane surface modification was adopted to improve the surface

properties. Eykens et al. [152] suggested that surface modification methods can be achieved via chemical modification, plasma treatment and addition of surface modifying macromolecules. It is crucial to synthesize a membrane with low surface energy to ease the cleaning of membrane surface. In view of this, Shahkaramipour et al. [153] has outlined the desired materials with antifouling properties which can be realized by coating or grafting the membrane surface. Materials chosen for coating or grafting must have the characteristics that could avoid interactions between the foulants and membranes. Hydrophilic materials, such as Polydopamine (PDA), Poly(ethylene glycol) (PEG), and zwitterions will form dense hydration layers on the surface and this hydration layer will act as energy a physical fencing that help to prevent foulants from binding on the membrane surface. However, the hydrophilic nature of the coating layer is not suitable for MD system as it may induce pore wetting. Hydrophobic materials such as fluoropolymers that has CF_3 part on the surface is introduced to ensure its low-adhesion and low energy properties [154]. It was found that the longer the fluorinated side chains, the lower the surface energy. Low energy surfaces ranging between 10-20mN/m [154, 155] will impede the adhesion of foulants such as precipitated salts, micro- and macro-molecules and bacteria [155-157]. However, there are evidence that oil adsorption on superhydrophobic surface is still taken place.

In addition, accumulated foulants can be easily washed off from the low energy surface due to the weak binding forces between foulants and membrane surface. This was proven in the studies by [158, 159] who coated the poly(perfluoroacrylate) onto a glass substrate. The coating demonstrated a uniform and low surface energy surface which was <13 mN/m. In addition, the coating also showed resistance towards the adhesion of bacteria. Another similar example can be seen from the cross-linked Perfluoropolyethers (PFPEs) that prepared from dimethacrylate. It possessed low surface energy which was almost 14 mN/m with little settlement of zoospore [160]. Therefore, it was presumed that fluorinated membrane that has low surface energy can lessen fouling propensities [161]. Nonetheless, coating or grafting of the fluorinated moieties resulted in high material cost.

Amphiphilic are materials that has distinct polar which is hydrophilic and non-polar which is hydrophobic part in the molecules. For example, hydrophilic PEG and hydrophobic fluoropolymers have illustrated the result in obtaining better antifouling properties. However, hydrophilic part of amphiphilic will induced pore wetting in MD system.

(ii) Superhydrophobic membrane

Polythene (PE), Polypropylene (PP), Polyvinylidene difluoride (PVDF) and Polytetrafluoroethylene (PTFE) are hydrophobic polymers with low surface energy and usually used as membrane materials for MD process. Mosadegh-Sedghi et al. [162] has reported that these synthesized hydrophobic membranes still undergo pore wetting by some aqueous solution and affect MD performance especially during long hour operation. Hence, it is crucial to produce superhydrophobic membrane whereby its surface is very resistant to wetting. Superhydrophobic membrane is always indicated by the Water Contact Angle (WCA) which is allegedly <150° [163, 164].

Direct processing can be achieved by blending, improvement on phase separation and electrospinning. However, there is very limited studies on blending method to produce superhydrophobic membrane [165]. The highest water contact angle for hydrophobic membrane was 148° as synthesized by Kuo et al. [166] by preparing the PVDF through diffusional induced phase separation process. It was found that WCA higher than 150° only can be achieved through electrospinning. Kang et al. [167] electrospun the membrane using polystyrene (PS) solution together with N,N-dimethylformamide (DMF) as solvent, the water contact angle as high as $154.2 \pm 0.7^\circ$ was produced.

Surface modification of hydrophobic to superhydrophobic membrane can be achieved using plasma treatment, deposition of rough polymer surface, chemical vapor deposition, sol-gel method, immersion grafting and two step surface modification. Fluorine-containing plasma gases are commonly used for hydrophobic enhancement. For instance, with 20 minutes time of 150 W plasma treatment, superhydrophobic PVDF flat sheet membranes with high WCA of 162.5° can be produced via CF_4 plasma surface treatment [168]. Besides, superhydrophobic membrane can be produced by depositing rough polymeric layer on the surface of membrane. For instance, in Ju et al.'s membrane fabrication work, hexamethyldisilazane-modified SiO_2 was used to deposit on the PVDF membrane which increased the water contact angle up to 158° [169].

In CVD, chemical reactions of gaseous will result in formation of a thin film on the membrane surface [170]. In the work of Zheng et al. [171], CVD method was applied to modified PVDF membrane surface from methyltrichlorosilane (MTS) solution. The modified PVDF membrane has lotus-leaf-like and a high water contact angle of 155° which then exhibited

superhydrophobic and self-cleaning features.

Other than that, sol-gel method has been adopted by Sun [172] who successfully fabricated superhydrophobic PES membrane by using TEOS and dodecafluoroheptyl-propyl-trimethoxysilane (DPT-12). The fabricated membrane had water contact angle of 154°. Also, direct grafting method that used hydrophobic molecules is also adopted to provide hydrophobicity of an inorganic membrane. In one of the work, Khemakhem et al. [173] proposed to graft the C8 molecules onto the hydrophilic inorganic zirconia membrane. Amazingly, the membrane has an increased of water contact angle from 20° to 160°. Superhydrophobic membrane can be produced via the two-step method. It was started with membrane surface roughening further by hydrophobization using low surface energy material or vice versa. For example, superhydrophobic polymeric membrane was prepared through immersion of nascent membrane in SiO_2 nanoparticles polymer solution accompanied by modification with fluoroalkylsilane [174-176].

(iii) Omniphobic membrane

Omniphobic membrane was introduced via creating the hierarchical roughness on the membrane surface which showed both anti-wetting properties against water and low surface tension organic solvents. In the work of Lin et al. [177], omniphobic membrane was achieved by adsorption of silica nanoparticles to create hierarchical re-entrant structures on hydrophilic glass fiber membrane. Silica nanoparticles interact with the polymer via electrostatic attraction, it was then followed by surface fluorination and polymer coating. The synthesized omniphobic membrane was used to compare with pristine PTFE membrane in DCMD experiment. The membranes were challenged with sodium dodecyl sulfate (SDS) to reduce its surface tension. Result showed that PTFE membrane prone to the wetting phenomenon but not the modified omniphobic membrane.

Boo et al. [178] treated the shale gas produced water using omniphobic membranes with hydrophobic polyvinylidene fluoride (PVDF) based polymer modified with silica nanoparticles. The modified membrane underwent fluorination and polymer coating to decrease the membrane surface energy [177]. The modified membrane has a contact angle of >150° for water and >130° for mineral oil. During DCMC experiment, the modified omniphobic membrane showed stable water flux which suggests the absence of membrane fouling, complete salt rejection in both feed solution containing sodium dodecyl sulfate (SDS) and mineral oil.

In another recent work by Chen et al. [179], omniphobic membrane was fabricated using chemical bath deposition technique. ZnO nanoparticles which have richest growth morphologies were deposited onto the hydrophilic glass fiber (GF) membrane accompanied by surface fluorination and polymer coating. The modified membrane was tested for water and ethanol which gave contact angle of 152.8° and 110.3° respectively. When compared with pristine hydrophobic GF membrane, omniphobic membrane has higher wetting resistance towards low surface tension substances in the feed during DCMD experiments where SDS was added every 2 hr during the experiment. In addition, the salt rejection was maintained at 99.99% throughout the experiment.

It can be concluded that, the MD performance using omniphobic membrane is relatively stable in terms of flux and rejection. The results suggest that omniphobic membrane has the potential to be applied in desalination for wastewaters that contains low surface tension contaminants. However, with the presence of oil without surfactant, omniphobic membrane posed the risk of fouling due to the hydrophobic interaction between the omniphobic membranes and the oil. The omniphobic is stable for the in-air oil droplet but still suffer from low stability in the oil rich wastewater stream.

(iv) Janus membrane

Janus membrane is uniquely defined as a membrane with different surface properties on each side [180]. Examples of different surface characteristics are hydrophilicity/hydrophobicity, superhydrophobicity/superoleophobicity and positive/negative charges, whereby it can be produced through chemical or physical approaches. In general, Janus membrane can be potentially applied in MD system to minimize the effect of fouling. There are two mechanisms on how the Janus membrane can mitigate fouling. In a way, the outer most hydrophobic outer layer will prevent wetting and the hydrophilic inner layer will prevent fouling. In another way, the hydrophilic outer layer will prevent fouling but the wetting cannot penetrate the hydrophobic inner layer.

Conventionally, there are two methods to produce Janus membrane which are asymmetric fabrication where Janus structure formed during formation of membrane and asymmetric decoration where modification was applied on the membrane. There are three different techniques in asymmetric fabrication such as sequentially electrospinning, sequentially vacuum

filtration and molecules migration to fabricate Janus membrane. Thickness of each layer (electrospinning and sequentially vacuum filtration) can be controlled precisely. In the work by Wu et al. [181], polyurethane (PU) which is hydrophobic and cross-linked poly (vinyl alcohol) (c-PVA) fibrous film which is hydrophilic were fabricated by a two-step electrospinning method. The first step was the electrospinning of PVA/glutaraldehyde aqueous on the steel wire gauze followed by electrospinning of PU fibrous film on the PVA film. When tested with WCA, PU fibrous film has a WCA of $142.2^{\circ} \pm 1.5^{\circ}$ while c-PVA film has a WCA of $22.1^{\circ} \pm 1.3^{\circ}$. In another work Hu et al. [182], Janus structure membrane was fabricated by depositing an ultrathin layer of polydopamine coated single walled carbon nanotubes (SWCNT) onto a mixed cellulose ester (MCE) filter substrate through vacuum filtration technique. The resulted membrane has hydrophobic-superoleophilic surface.

In contrary, Janus structure was formed due to migration of components between the membrane matrixes during membrane formation using phase inversion technique where the casting solutions can be prepared by blending two membrane components. For example, Zhang and Barboiu [183] reported that hydrophilic/hydrophobic polymers were synthesised through template partial phase segregation, to form asymmetric membranes. PEG-diamines, alkane-diamine and Trialdehyde were blended in the casting solution. During solvent evaporation, hydrophilic PEG-diamines segments will migrate to the upper layer and hydrophobic alkane-diamine segments will migrated to the bottom layer spontaneously where two elements connected via core-centres Trialdehyde. In another work of Essalhi and Khayet [184], hydrophilic/hydrophobic Janus membrane was synthesised by using a fluorinated surface modifying macromolecule (SMM) and polyetherimide(PEI) polymer which is hydrophilic base via phase inversion technique. During membrane formation, SMM will migrate to the membrane surface and two distinct layers were formed. They observed that the modified membrane is more appropriate for DCMD for desalination when compared with AGMD.

There are three different techniques in asymmetric decoration of Janus Membrane such as photodegradation, photo-crosslinking, vapor treatment, coating, floated deposition and sequential surface modification. Photodegradation technique was conducted by Lin et al [182] for recovery of hydrophilic side of ZnO-coated fabric membrane. In contrary, photo-crosslinking was also a technique to produce a Janus membrane. For instance, Wang et al. [180] soaked a cotton fabric into a solution that consists of hydrophobic block while the other side of the fabric was susceptible to UV irradiation for crosslinking purpose after that hydrophilic PDMAEMA (poly(N,Ndimethylaminoethyl methacrylate)) was grafted onto it. Besides, Janus membrane can be obtained using vapour deposition method. For example, Tian et al. [185] used perfluorooctyl-trichlorosilane vapour to treat the exposed side of cotton fabric to obtain Janus structure. Vapour deposition time will determined the thickness of the hydrophobic layer. In coating technique, Liu et al. [186] obtained a superhydrophobic-hydrophilic fabric using the fluoropolymer foam. The highly viscous fluoropolymer will impede diffusion through membrane pores and create a hydrophobic side. In the work of Yang et al. [187], Janus membrane was obtained by hydrophilization of a hydrophobic surface by floating a hydrophobic membrane surface on top of dopamine solution where Janus structure was obtained due to hydrophobic interaction. When a dried membrane was floated on solution for deposition, a hydrophilic-on-hydrophobic membrane will be achieved and vice versa. Chen and co-workers [188] grafted poly(N,N-dimethylaminoethyl methacrylate) which is hydrophilic and polystyrene which is hydrophobic onto opposite faces of a carbon nanotube/polymer and graphene/polymer composite membrane using sequential surface modification technique.

(v) Stimuli-responsive membranes

Stimuli-responsive membranes constitute an enhancement of existing membranes in responding to its post-fouling cleaning mechanism. They can alter their properties when there is an external stimulus like electrolyte, pH, light, magnetic field and temperature [189]. Significant efforts have been placed in the last decade to develop smart membranes with self-actuation capabilities [190] for surface cleaning. A thermoresponsive membrane can mitigate fouling problem by removing foulants that accumulate on the membrane surface with temperature stimulation. Amongst the polymer, the thermoresponsive Poly(N-isopropylacrylamide) (PNIPAM) is a potential candidate to be introduced onto the surface for self-cleaning purpose. At the temperature lower than LCST, thermo-responsiveness of PNIPAM will undergo entropy change by absorbing water molecules from the hydrophobic alkyl-chain [191] and swell. However, as temperature increases, dehydration of the CH₃ groups, diffusion and aggregation of the chains, and the transition of hydrogen bonds takes place and lead to shrinkage of the hydrogel that caused by the entropy elasticity of the PNIPAM network.

In the work of Ou et al. [192], thermo-responsive membrane was

produced by combining a polyurethane (TPU) and poly (N-isopropylacrylamide) (PNIPAM) which tested to have good mechanical, and flexible properties. This TPU-PNIPAM membrane was able to alter its superhydrophilicity and superhydrophobicity by swelling and shrinking in 3 minutes from 25°C to 45°C. The TPU-PNIPAM membrane was successfully applied separate both the oil-in-water emulsion and water-in-oil emulsion at 25 and 45 °C, respectively. The successfully of PNIPAM hydrogel coated on the membrane and showing antifouling properties can apply to modification of membrane to have antifouling properties for membrane distillation process.

4.2. Process enhancement

Besides material design, the process enhancement also plays an important role in mitigating MD fouling. The frequency and intensity of fouling can be soothed by proper feed pretreatment, operational parameters of MD process with the help of processing aids [3]. However, proper pretreatment is the key factor in ensuring the durability of the membrane in long run operation.

4.2.1. Feed pretreatment

Feed pretreatment is one of the common applications used in most water separation systems depending on incoming water quality. Conventional pretreatments used are filter or membrane typed filtration, dosing of anti-scalants, coagulation/flocculation, and chlorination [193]. MF and NF are common filtration upstream before the MD process; it can remove particulates, colloids, suspended solids as well as bulky scalants in the feed [35, 194]. NF with negative surface charge tends to hinder the permeation of sulfate which is a key contributor to the development of inorganic scales such as CaSO₄. Qu et al. [195] introduced accelerated precipitation softening (APS) method to treat RO concentrate in the DCMD operation by adjusting pH of the feed with sodium hydroxide, followed by calcite seeding and MF, respectively. This pretreatment method was highly effective in removing calcium and significantly alleviating both CaCO₃ and CaSO₄ scaling. Besides, feed acidification with HCl to pH 4 or 5 is efficient to mitigate CaCO₃ deposition [64, 196]. Gryta [197] reported that a 70 mesh net filter which assembled directly at the membrane module inlet can overcome membrane scaling by removing heterogeneous crystallization. Minier-Matar et al. [198] reported that a two-step filtration system (1 µm cartridge filter + Granular Activated Carbon) was used to pretreat the thermal brine to remove particulates and contaminants such as process chemicals and antifoam agent.

Some researchers suggest to use chemical coagulant following by sand filtration or MF in order to minimise membrane fouling [196, 199]. Coagulation process alters the stability of colloidal particles, which may improve permeate flux and reduce membrane fouling rate. Li et al. [200] found that the use of poly-aluminum chloride (PACl) as a pre-coagulant aid effectively reduced the organic pollutants in the operation of DCMD with the feed solution using biologically treated coking wastewater (BTCW) and significantly mitigated membrane fouling and wetting. Anti-scalants has potential to suppress inorganic scaling. It can remove carbonate and sulfate scales, as well as disperse colloids and metal oxides [85, 201]. Anti-scalants slow down the precipitation rate, distort crystal structure and nucleation growth [202]. They are typically common for scaling control due to low cost and low dosage. He et al. [85] claimed that the addition of a nitrogen containing organo-phosphorus anti-scalant compound could inhibit the calcite precipitation at a low dosage of 0.6 mg/L with less than 5% of crystallization. However, other studies showed different findings, for example, Gryta [201] reported that the use of sodium polyphosphates as an anti-scalant can reduce the growth of CaCO₃ crystallization, however it also changes the deposit layer to become low porous and amorphous. The flux decline was worsened when the concentration of anti-scalant increased. In view of this, dosing of anti-scalants is very much depends on type of foulants appeared in the solution.

4.2.2. Operational parameters

4.2.2.1. Feed temperature

An increase in temperature at the membrane surface can lead to permeate flux increase as temperature polarization is reduced. However, as discussed before, temperature is the main factor that can affect scale formation as the salt solubility varies with temperature. Common scales such as calcium carbonate are insoluble at higher temperature will induce temperature polarization and consequently contributing to scaling. Gryta [111] suggested to use feed water temperature below 80°C in order to decelerate the decomposition of bicarbonate ions and thus reduce the CaCO₃ scaling. On the other hand, Gilron et al. [117] found that silica scaling showed more rapid flux decline than gypsum and calcite scaling as silica is insoluble at lower temperature. The changes of feed temperature greatly impact the fouling rate due to concentration polarization induced as a result of the formation of

insoluble salts in feed solution [6]. Phosphate inhibitor showed lower efficiency in removing CaCO_3 precipitation from hardwater at 50°C due to its acceleration of hydrolysis reaction at elevated temperature [203]. The effect of temperature is not only limited to the solubility of the salt and anti-scalants, but its impact on the polymer cannot be neglected. For example, the stability of the PVDF membrane under elevated temperature should be a concern due to its lower modulus. As a consequence, it might induce wetting problem due to the strain on the membrane that create defect structure.

4.2.2.2. Feed flow rate

Increasing feed flow rate is able to improve the heat and mass transfer coefficients and therefore provides higher driving force for vapor permeation through the membrane. In addition, higher flow rates can induce shear force, consequently, higher deposit removal, and reduce the scaling level and fouling rate. According to Gryta [111], the formation of salt crystals was much smaller and more porous when the feed velocity was above than 0.6 m/s. These fine deposits will not cause severe pore blocking and wetting. Besides, vapor permeability was not significantly affected.

4.2.2.3. Applied pressure

Compared to high pressure-driven RO process, the superhydrophobic nature as well as small pore size of the membrane and the low feed pressure applied in the MD can prevent pore wetting and scaling [6]. Some works were studied on the effect of vacuum pressure applied in VMD towards its membrane fouling resistance [204, 205]. In VMD process, vacuum is applied to withdraw the vapor permeate. The vacuum pressure is typically lower than the saturated vapor pressure of volatile particles, so that condensation can occur externally. To suppress pore wetting, the pressure difference must be lower than the maximum LEP of membrane. In addition, membrane wetting can be reduced when membrane pore size is smaller and both liquid surface tension and water contact angle are higher [76].

According to Zhao et al. [205], by lowering the vapor pressure difference between permeate and feed, the permeate flux was kept stable in VMD using aqueous solution of concentrated ginseng extracts whereby membrane fouling was not observed. Zhao et al. [205] claimed that high initial flux can be obtained when lower vacuum pressure is used. However, the flux decline rate was faster, inhibited by fouling on membrane surface. It is consistent with another study by Zhao et al. [204] who operated VMD under temperature of 60°C using human urine at feed flow of 30 L/h. The flux increased and kept stable with the vacuum degree hold below its critical value, 82.6 kPa with no apparent membrane fouling. However, the flux decline rate was faster as the vacuum degree was further increased beyond its critical value; significant membrane fouling was observed even though the initial flux was obtained. The membrane surface was fouled by organics together with NaCl crystals as revealed by the SEM analysis results [204]. Fouling happened because the strong suction force allowed the solute particles dwell on the membrane when the vacuum was too strong. Concentration and temperature polarization were induced by the suction force at higher vacuum degree. In addition, the increase of salt concentration will induce crystallization growth on membrane surface and consequently causing rapid flux decline [204]. To mitigate membrane fouling, the vacuum degree should be taken into account as it has significant impact towards permeate flux and membrane fouling.

4.2.3. Processing Aids

Gas bubbling has been explored to mitigate fouling of membrane by a number of studies [206, 207]. A gas-liquid flow improves the surface shear rate and fluid dynamics thus effectively reduces temperature and concentration polarization near the membrane surface. Recently, microbubble aeration (MBA) was introduced to control membrane scaling and improve permeate flux in VMD for desalination of high salinity seawater [208]. Ye et al. [208] reported that larger number of the smaller size of microbubbles were produced at higher pump pressure (0.4 MPa), which led to increase permeate flux from $1.3 \text{ kg/m}^2\text{h}$ to $2 \text{ kg/m}^2\text{h}$ and reduce salt precipitation. In spite of its advantages, two-phase flow makes the studies more complex.

Some studies have focused on pulsating and intermittent flows [209, 210]. According to Ali et al. [211], at smaller feed flow rate, the flux of using helical fibers employed in the VMD system was 47% higher than straight fibers, while wavy fibers showed 52% increase in flux. This is because the temperature polarization at two sides was reduced. The lower feed flow also led to energy saving for pumping and at the same time maintaining the membrane performance. Besides, compared to steady flow, intermittent flow showed 30% increase in flux and 180% improvement in energy efficiency. This is because the low frequency of back and forth flow at lower flow rates allows proper fluid mixing near the membrane surface with the bulk and increases retention time of the fluid across the membrane, which enhances the

cooling rate.

5. Future outlooks

Membrane fouling is still a key issue in preventing the widespread of MD for water recovery due to its propensity of fouling for different water resources. At present, efforts to prepare membrane with superior antifouling/antiscaling properties are still limited. Current research trend is still focusing on the development of combined fouling and wetting resistive polymeric membranes through the incorporation of nanoparticles in the membrane matrix. Besides, membrane modules design using hollow fibre system is still very limited which hinder the scale up applications of MD. The membrane stability of the hollow fibre membrane with superhydrophobicity is remains a challenge. Membrane modification to make it with omniphobic and Janus characteristic is crucial to avoid fouling problem by both hydrophilic and hydrophobic foulants. Nonetheless, the proper operational design with suitable physicochemical pre-treatment processes are still limited in pertaining to different solution chemistry. Filter aid and anti-scalants for scaling control are crucial for desalination but its effect on long term operation is scares mainly due to limited industrial scale study. The reported flux of MD higher than 100 L/hr.m^2 and the water recovery rate of more than 90% are still facing the long-term stability problem even though its high flux is attractive for industrial scale operation. The balance between superhydrophobicity and the liquid entry pressure should be studied in detail to ensure its practicability.

6. Conclusions

Although membrane distillation is a relatively energy intensive process, it is gaining higher popularity in wastewater treatment and desalination due to its operational flexibility. However, its high thermal energy requirement makes it impractical for industrial scale application unless it is combined with the waste heat or renewable energy resources like solar heat. MD can be applied to recover water from wastewater or via desalination, however, due to its different solution chemistry, the extent of membrane fouling can be very different. Amongst the membrane fouling mechanism, biofouling of MD is less significant compared to the organic and inorganic fouling, however, it is limited to the operation near boiling point ($>90^\circ\text{C}$). At elevated temperature, the fouling of MD by inorganic material will become serious due to its deteriorating ions solubility. This explains why inorganic fouling a more dominating phenomenon as compared to biofouling during desalination. Superhydrophobicity of membrane surface is no longer the sole factor to be considered in the real application of MD. The architectural structure of Janus membrane with both hydrophilic and hydrophobic characteristics should be designed to balance between wetting and fouling. Besides, the fouling and wetting resistance for MD requires further understanding of the solution and membrane properties coupled with optimum operating conditions. The processing aid seems inevitable for the long run MD process to mitigate the fouling phenomena. However, coupled with proper system design, this problem can be solved in a more systematic way.

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