Development of a novel submerged tubular direct-contact membrane distillation system for saltwater treatment

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ABSTRACT

Membrane distillation (MD) is emerging as a promising alternative to current desalination systems because of its low energy consumption and investment cost. This study is aimed to develop a direct-contact membrane distillation (DCMD) system with a simple novel submerged tubular membrane module. The effect of feed temperatures (40°C, 50°C, 60°C and 70°C) on DCMD performance was investigated in the study. Additionally, pore wetting and surface scaling during the long-term operations were observed. As a result, the permeate flux is dependent on the feed temperature, with the flux increasing from 1.00 to 5.24 L/m²h as increased temperature from 40°C to 70°C. Such findings indicated the optimal feed temperature is at $60°C \pm 0.5°C$, being the permeate flux of 4.09 L/m²h under the cooling temperature of $28°C \pm 1°C$. Total dissolved solids, chloride, and sulfate rejection efficiency were 99.36%, 99.91%, and greater than 98.74%, respectively. After 2 months of operation, pore wetting and surface scaling were detected through contact angle measurements and scanning electron microscopy and energy-dispersive X-ray spectroscopy. For flux enhancement, proper membrane materials and module arrangement should be further explored to develop a submerged tubular MD module for saline water treatment in developing countries.

Keywords: Submerged direct-contact membrane distillation; Tubular module; Brackish water; Saltwater; Temperature

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1. Introduction

Currently, freshwater is an essential demand, but the lack of freshwater and the low quality of resources have been much more attention. In detail, climate change, excessive groundwater use, and sea-level increment are the main factors influencing both the quality and quantity of freshwater resources [1]. Notably, the salt intrusion was reported in Asian Nations such as Bangladesh, Bhutan, India, Nepal, and Vietnam, and this issue caused the increase in salinity in both groundwater and surface water; thereby influenced human life [2]. For example, the salinity of above 4 g/L went deeply into the river, which caused water shortage in Mekong Delta (Vietnam) [3]. Moreover, Thi Nhung et al. [4] indicated that 6 g/L of salinity threshold intruded into Sóc Trăng, Bac Lieu and Kien Giang provinces, and water shortage was happening in the dry season in Ca Mau, leading to the limit of accessing clean water of local people for daily demand. Evidently, groundwater with high salinity has become a serious issue in these regions. Accordingly, water purification systems have developed during the past few years [5], being approximately 19,000 desalination plants which were built with a total capacity of 60 million m³/d in 2014 [6]. However, developing a desalination technology being proper for the inherent conditions in each region, that is, salt intrusion areas and low income is extremely crucial. Whilst major technologies such as multistage flash distillation (MSF), multi-effect distillation, reverse osmosis (RO), forward osmosis (FO), electrodialysis (ED), nanofiltration (NF) are being developed [7], such technologies have drawbacks in the cost (energy consumption), treatment cost, membrane fouling (Table 1).

Among desalination technologies, membrane distillation (MD) has been paid much more attention because of its low cost, easy installation, and high rejection rate [9,17,18]. Compared to conventional distillation and RO, MD offered not only a lower feed temperature but also a lower pressure, respectively [19]. In case feed heat energy is optimized using solar energy or waste heat, it may become a good candidate for desalination in poor regions [20]. MD combines distillation and membrane separation into a single process, which is more powerful while possessing less disadvantages than its standalone components [21].

MD has emerged as a potential technology for desalination [22-24]. It is a thermally driven separation process that entails the use of a hydrophobic membrane, which only permits the transport of water vapor while rejecting the permeation of liquid water. For the MD process, the dissolved solutes (i.e., inorganic salts that cannot be evaporated) or suspended solids can be rejected completely [25]. In detail, MD is driven by the vapor pressure difference across the membrane; thus, it is completely influenced by temperature differences. Moreover, the salinity of the feed water has a negligible effect on water flux in MD [26]. The past studies have introduced several membrane types such as tubular, hollow-fiber, and flat-sheet membranes. Each type has its own characteristics and performance that are suitable for certain processes. For example, the hollow-fiber membrane has a high packing density [27,28]. However, the membrane fibers had a high potential of membrane fouling, resulting in the reduction of permeate flux. According to Warsinger et al. [29], the decline of permeate flux in MD using hollow-fiber membranes was caused by clogging membrane capillaries, leading to an increase in concentration and temperature polarization. Moreover, the findings indicated partial wetting always occurs in fibers; in particular, heat loss frequently occurs in hollow-fiber membranes [30]. Therefore, the flat-sheet membrane is commonly used in MD systems [28] because of its ready replacement and cleaning. However, flat-sheet membranes require the use of a support layer because of its thin active layer [31]. Additionally, the flat-sheet membrane frame must be tight to avoid water transportation through the membrane, thereby leading to high investment costs. As reported, the tubular membrane has been little attention since it exhibits a lower flux under the same operating conditions compared to the other membrane types [32]. Nevertheless, a few recent studies showed several advantages of tubular membranes. For instance, cleaning the membrane surface is easier [33] because tubular

Table 1	
Current status of desalination technologies	

Technology	Issues	Reference
MSF	High energy consumption, 20–27 kWh/m ³	[8]
	Large area of desalination	[9]
NF	Treatment cost of 1.34 USD/m ³ , the energy consumption of 0.61 kWh/m ³	[10]
	Fouling control must be regularly cleaned	
RO	Low removal of non-degradable substances with low molecular weights (e.g. boron removal of 35% to 40%)	[11]
	Trans-membrane pressure increased because of fouling	[12]
	It is necessary to regularly conduct chemical cleaning which reduces the membrane lifetime	
	High energy consumption (over 4–5 kWh/m ³ in seawater desalination plant)	[13]
	Saltwater above 65 g/L cannot be treated because of high pressure (over 80×10^5 Pa)	[14]
	Requiring a high-pressure pump (~100 bar) for the RO system	[8]
ED	Cannot treat non-charged contaminants	[15]
FO	Reverse flow of draw solutes reduced osmotic driving force	[16]

MSF: multistage flash; NF: nanofiltration; RO: reverse osmosis; ED: electrodialysis

membranes can reduce membrane fouling and polarization phenomena with a high flow rate of feed stream [34], especially simple design, installation, and replacement [35,36]. Moreover, as the tubular membrane is capable of supplying water with high turbulence, the rapid increase in the temperature of the cooling stream can be reduced. This limits the decline of the thermal gradient between two sides of membranes, thus restricting the drop of permeate flux.

Clearly, tubular membranes may be potential candidates in developing countries (e.g., Vietnam and Thailand) for providing fresh water to poor areas with highly saline water (e.g., Can Gio District). Previous studies on DCMD have revealed a general trend of increasing flux as a function of increasing feed temperature [14,21,22,26,37–40].

Therefore, with those premises, our work focused on developing a DCMD system with a low-cost submerged tubular polytetrafluoroethylene (PTFE) membrane for developing countries. Different thermal conditions and various feed waters were investigated to evaluate treatment performance whilst the effect of scaling and fouling on the membrane performance was observed accordingly.

2. Materials and methods

2.1. Feed water

Feedwater in this study was taken at two different sources, including groundwater (high salinity) and seawater.

The former was taken at Ba Tri District, Ben Tre province, Mekong Delta, Vietnam. The latter was also collected at Can Gio beach. Detailed characteristics of feed water were shown in Table 2.

2.2. Lab-scale DCMD system and experimental conditions

Fig. 1a presents a schematic diagram of the DCMD system. The system includes a 5 L feed tank, an 18 L membrane tank, a blower, a distillation tank, and a cooling tank.

Table 2 Feedwater characteristics

Parameters	Value		
	Groundwater	Seawater	
рН	7.8 ± 0.2	7.8 ± 0.2	
TDS, mg/L	$27,000 \pm 1,000$	$30,000 \pm 1,000$	
Salinity, g/L	23 ± 1.9	25 ± 1.2	
Chloride, mg/L	$15,400 \pm 360$	$18,350 \pm 1,767$	
Sulfate, mg/L	530 ± 26.1	$2,130 \pm 583$	
Calcium, mg/L	921 ± 2.3	727 ± 2.3	
Total organic carbon (TOC),	3.26 ± 0.08	11.26 ± 0.19	
mg/L			



Fig. 1. (a) Schematic diagram of direct contact membrane distillation (DCMD) system and (b) principle of membrane module operation.

An automatic feed pump and electric float were installed to control the constant water level in the membrane tank. This one contains heating equipment (800 W) and a thermal sensor to retain the desired temperatures. The blower (18 W) was installed bottom tank to provide homogenization through the tank. The water was heated in the membrane tank, after which it was evaporated and passed through the submerged tubular membrane module (Fig. 1b). To investigate the effect of temperature on the performance, the following temperatures were set: 40°C, 50°C, 60°C, and 70°C. Subsequently, the vapor was condensed in the cool side of the membrane, in which the temperature was kept at 28°C ± 1°C as the water temperature. To maintain the water temperature, a cooling stream was continuously circulated to the distillation tank using a pump with a flow rate of 0.45 L/min. A valve was installed at the end and a cylinder was used to define the water mass. Flux is defined as the excess distillate flow/membrane area used. The temperature of the distillation tank was controlled by submerging into the cooling tank. The temperature of the cooling tank was retained approximately 20°C using a chiller.

In the DCMD system, the membrane was in direct contact with the cooling water on the permeate side, and its exterior was surrounded by feed water. The construction of submerged MD modules was also carried out in previous work [41,42]. The system used a hydrophobic membrane which only filtered vapor features whilst inorganic salts could not penetrate the membrane. The mass flux is driven by the vapor pressure difference across the membrane and mainly influenced by the difference in temperature between the heating tank and cooling tank [25,26,37,43]. Once the water vapor passed through the membrane, it was condensed in the cooling tank and subsequently returned to the membrane tank to attract water vapor into the distillation tank, thus creating a filtration cycle.

For experimental conditions, the groundwater was diluted to reach the total dissolved solids (TDS) concentration of 5,000 mg/L, which was considered as a low-TDS feed for investigating the effects of various feed temperatures, that is, 40°C, 50°C, 60°C, and 70°C on the performance of submerged tubular DCMD system; thereby defined optimal temperature condition.

Another one, different feed concentrations, that is, diluted groundwater (5,000 mg/L), groundwater (27,000 mg/L) and seawater (30,000 mg/L) was also carried out in this work. For the seawater case, the membrane pore wetting and scaling were also observed after 2 months of operation.

2.3. Membrane module design

This study used a hydrophobic membrane provided by Ray-E Creative Co., Ltd., (Taiwan) (Table 3). The flat PTFE membrane was wrapped around a polyvinyl chloride (PVC) pipe with 1 mm pores in order to make a tubular membrane module. A stainless-steel spacer was placed between the membrane and the PVC pipe to create an air gap in which vapor could directly contact the cooling water to condense distilled water. The membrane was attached to the PVC pipe using glue (ScotchTM 3 M, Japan) produced from Korea. Detailed characteristics of pristine membrane use denoted in Table 3.

Table 3 Characteristics of the pristine membrane (Ray-E Creative Co., Ltd., Taiwan)

Parameter	Membrane
Pore size, µm	0.45
Contact angle, °	123.6 ± 0.8
Membrane thickness, mm	0.2
Material of active membrane	PTFE
Material of support membrane	Polyethylene terephthalate
Membrane area, m ²	0.02

2.4. Membrane wetting and scaling analyses

Contact angle, scanning electron microscopy (SEM) and energy-dispersive X-ray spectroscopy (EDX) measurements were conducted to define the wetting and scaling of the membrane. The former was determined using the sessile drop approach (Kruss G10 goniometer, Kruss, Germany). For latter, the membrane was coated with a platinum layer, after which it was observed under a Hitachi S-4800 microscope, Japan.

3. Results and discussion

3.1. Effect of temperature on the permeate flux and salt rejection

In DCMD processes, the relationship between mass flux and temperature can be presented by the following equation [25]:

$$J = B_m \left(p_{\rm mf} - p_{\rm mp} \right) \tag{1}$$

$$p = e^{\left(23.328 - \frac{3,841}{T - 45}\right)} \tag{2}$$

where *J* is the mass flux, B_m represents the overall mass transfer coefficient, p_{mf} and p_{mp} are the partial vapor pressures of feed and permeate sides, respectively, and *T* is the temperature (in Kelvin scale). Theoretically, an increment of feed temperature exhibits an increase in the partial pressure of the feedwater; thus increase the permeate flux.

Regarding the performance of the DCMD with the submerged tubular membrane module, the water flux was recorded at various temperatures, as illustrated in Fig. 2. The average flux under temperatures of 40°C, 50°C, 60°C and 70°C were 1.00, 2.26, 4.09, and 5.24 L/m²h, respectively. This is attributed to a higher temperature in feed induced higher vapor pressure difference (e.g., higher driving force); thus engendered increased in permeate flux. Such findings were reported from the previous studies [14,38,40,44]. As reported the effect of temperature polarization could occur in DCMD accordingly in which the temperatures in the bulk solution and with thin membrane surface are different [40,45]. This fact caused a decrease in temperature polarization coefficient when temperature increased in bulk feed solution; thereby reducing the effective driving force (i.e., a decrease of permeate flux). Interestingly, in this study, the permeate flux at 50°C, 60°C



Fig. 2. Water flux at different temperatures under the condition: feed TDS of 5,000 mg/L, cooling stream temperature of $28^{\circ}C \pm 1^{\circ}C$.

and 70°C increased by 125%, 307% and 422% compared to at 40°C, respectively whilst these results indicated the flux is proportional to the feed temperature (Fig. 2). This was shown in the linear equation between the permeate flux and the feed temperature with a high regression coefficient of 0.992. Such findings offered a negligible effect of temperature polarization effect on submerged tubular DCMD system, which could be attributed to a proper hydrodynamic condition in feed side (e.g., turbulent flow generated by air diffuser in membrane tank). As expected, the highest permeate flux (5.24 L/m²h) was achieved at 70°C, being five times higher compared with the flux at 40°C. However, an excessively high temperature facilitates membrane dilation; this thus increases the likelihood of pore wetting because the liquid entry pressure decreases. If pore wetting occurs, the liquid is in contact with the membrane; therefore, the membrane becomes impermeable to water vapor [46]. In addition, the membrane hydrophobicity decreases, thus facilitating membrane fouling. According to Tan et al. [31], pore wetting occurs at temperatures higher than 70°C. Therefore, this study considered 60°C to be the optimal feed temperature for operating DCMD systems with submerged tubular membrane modules in saltwater treatment processes.

In comparison to the plate frame membrane studied from previous works [40], the value of 5.25 L/m²h is relatively lower. For example, a past study using a plate frame membrane for Red Sea desalination reported a permeate flux of 10.2 L/m²h under the feed temperature of 80°C and the cooling temperature of 20°C [42]. Another one Qu et al. [47], under the feed concentration of 5,000 mg/L, the feed temperature of 50°C and cooling temperature of 20°C, the permeate flux varied from 5.81 to 7.53 L/m²h when the feed flow rate ranged from 0.23 to 0.58 m/s. Such discrepancy is mainly attributed to a lower feed flow rate (0.027 L/h) used in this study. When the feed flow rate reduced, the Reynolds number in the feed decreased, leading to the drop of the turbulence level, thus reducing permeate flux [40,46]. According to Chan et al. [48], the Reynolds number of submerged configurations using the particular bubble was 720. Besides, another factor causing the low permeate flux in this work is the high cooling temperature $(28^{\circ}C \pm 1^{\circ}C)$. This leads to the drop in the temperature difference between 2 membrane sides, resulting in the reduction of transmembrane pressure difference [21]. Importantly, given studies above the flat-sheet membrane was not submerged in the feed water, which could help to produce more pressure on the membrane, thus resulting in a higher flux compared to the submerged membrane. However, the flat-sheet MD modules required high capital and operating cost because of the low membrane area and requirement of the tight frame [33,49], which is not a proper application for developing areas.

For salt rejection, although the flux increased, the water quality was highly acceptable, with the salinity being low concentration (less than 0.1 g/L). Fig. 3 presents the changes in feed concentration as a function of time and temperature. As a result, the feed concentrations of TDS, chloride, and sulfate gradually increased as the temperature increased from 40°C to 70°C. This indicates that increasing the feed temperature could lead to an increase in the concentration of ions in the feed tank. Such findings are in line with the previous studies [26] and their results reported that feed temperature was higher, more water vapor was generated.

Consequently, the provision of feed water to the membrane tank continued, this fact thus caused the accumulation of the ions, increasing their concentrations over time. Despite the high concentration in the feed water, the permeate water quality remained excellent. The TDS, chloride concentration, and sulfate concentration of permeate were lower 30, 3.5, and 2.0 mg/L, respectively, complied with the standard of drinking water of the World Health Organization (limits for TDS of 1,000 mg/L, chlorine of 250 mg/L, sulfate of 250 mg/L). Clearly submerged tubular DCMD system exhibited sufficient treatment of saltwater despite the change of feed temperature. Moreover, due to its low capital and operating cost, this system could be a potential candidate for developing areas but to increase in flux optimal configuration should be explored.

3.2. Effect of feed concentration on permeate flux

As the results (Fig. 4), when the feed TDS increased from 5,000 to 30,000 mg/L, the permeate flux decreased from 4.09 to 1.26 L/m²h. This is due to the fact that less free water caused by the high salinity in feed water was the cause of the drop in vapor pressure. Thereafter, the difference of vapor pressure between two membrane sides declined, resulting in a decrease of permeate flux [50]. On the other hand, since the membrane system not only operated at dead-end mode but also submerged, there was a remarkable change of the initial concentration of salt over time, probably inducing to the increase of concentration polarization and thus reducing the vapor transmembrane pressure [51,52]. Moreover, the deposition of the solutes on the membrane surface observed was capable of the decline of permeate flux [53]. Although the increase of feed TDS affected permeate flux, the quality of distilled water was not changed significantly. Such results can be found in Table 4.

3.3. Membrane pore wetting and fouling under the long-term operation with seawater

The contact angle was accordingly measured at an initial time and later 2 months of operation (Fig. 5). The former



Fig. 3. TDS, chloride concentration, and sulfate concentration at different feed temperatures (40, 50, 60, and 70) another condition: the cooling stream temperature of $28^{\circ}C \pm 1^{\circ}C$.



Fig. 4. Effect of feed TDS on permeate flux at a feed temperature of 60°C and cooling stream temperature of 28°C ± 1°C.

Table 4
Removal efficiency of TDS, chloride, sulfate, and TOC with the different feed TDS

Parameters	Permeate (mg/L)		Removal (%)			
	Diluted groundwater	Groundwater	Seawater	Diluted groundwater	Groundwater	Seawater
TDS	≤20	≤20	<10	≥99.69	≥99.94	100
TOC	-	<0.58	< 0.41	-	~90	>96
Chloride	<1.8	<4	<3.3	≥99.99	≥99.98	≥99.98
Sulfate	<1.9	<4	<2.1	≥99.03	≥99.17	≥99.91



Fig. 5. Contact angles of (a) pristine membrane and (b) fouled membrane after 2 months of operation (the feed temperature of 60° C, and cooling stream temperature of 28° C ± 1° C).

measurement was for a pristine membrane whilst the latter one was for the fouled membrane. The average contact angle of the pristine membrane was $123.6^{\circ} \pm 0.8^{\circ}$. After operating for 2 months, under conditions, that is, feed temperature of 60°C and a feed TDS of 30,000 mg/L, the contact angle decreased significantly, being of $78.5^{\circ} \pm 0.4^{\circ}$. Such results indicated the presence of membrane pore wetting which caused a reduction in permeate flux (from 1.33 to 1.01 L/m²h). For a typical MD, the contact angle value should be 90°–160° for efficient salt treatments [26] without sustaining pore wetting. A membrane with a higher contact angle is more efficient because it has a more favorable liquid entry pressure, thus resulting in high performance. Another one, the presence of salt was also observed during 2 operation months.

As denoted Fig. 6a, it can be seen that the scaling forming a needle-shaped salt crystal was observed on the membrane surface. This fact is properly confirmed by the SEM, EDX images (Figs. 6b and c). As a result, calcium, oxygen, and carbon were the major salt components on the membrane



Fig. 6. Fouled membrane after long-term operation under conditions (feed TDS of 30,000 mg/L, feed temperature of 60° C, and cooling stream temperature of 28° C ± 1° C). (a) Actual membrane surface scaling, (b) scanning electron microscopy (SEM), and (c) energy-dispersive X-ray spectroscopy (EDX).

surface. The salt crystals are formed on the membrane surface because a high feed temperature engendered ion accumulation, possibly resulting in scaling. Yu et al. [54] reported that under a temperature of 60°C, CaCO₃ precipitates could be formed. As the previous studies, the precipitation of compounds such as CaCO₃ and CaSO₄ on the surface of a membrane not only caused membrane deterioration and but also reduced thermal efficiency [55].

However, compared to the flat-sheet membrane, this submerged tubular membrane system showed a lower influence on membrane wetting. Specifically, Duong et al. [26] indicated that after operating the DCMD system with seawater, the contact angle reduced by 96° (from 116° to 20°) compared to the drop of 45° in this study.

4. Conclusions

A DCMD system with a newly developed submerged tubular membrane module for saltwater treatment was evaluated in this study. The effects of the feed temperature and feed concentration on the permeate flux and salt rejection were confirmed accordingly. For optimal condition (feed temperature of 60°C) the permeate flux could obtain 4.09 L/m²h for treating saline water with TDS of 5,000 mg/L. The novel submerged tubular module-based DCMD performed sufficient rejection on salinity, chloride, and sulfate, which could be a potential candidate for saline treatment in developing countries. However, for the case of seawater (feed water of 30,000 mg/L), a low flux (1.33 L/m²h) was found whilst membrane pore wetting and scaling were observed. Such findings are sound disadvantage but to increase the permeate flux not only an optimal configuration of the submerged tubular membrane module but also new material for hydrophobic membrane should be explored in the future study.

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