



Review Paper

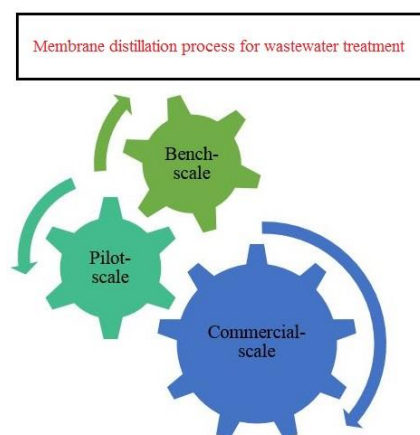
A Review on Applications of Membrane Distillation (MD) Process for Wastewater Treatment

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HIGHLIGHTS

- The water scarcity is driving the implementation of wastewater treatment.
- This review offers a MD state of the art applications for wastewater treatment.

GRAPHICAL ABSTRACT



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ABSTRACT

The growing scarcity of fresh water is driving the implementation of wastewater treatment and water reuse on an increasingly large scale. Various methods have been developed and used for water reuse from wastewater; however, the membrane distillation (MD) process, as a promising separation technology, has recently gained more attention. The MD process is a non-isothermal membrane-based separation used in various applications, especially for desalination and water/wastewater treatment. Compared with other separation processes, the MD process possesses several unique characteristics such as total (100%) rejection, intensive to feed concentration, mild operating conditions as well as stable performance at high contaminant concentrations. Due to the high fresh water demand in recent years, extensive researches have been devoted to the MD process in areas of water/wastewater treatment. The present paper offers a comprehensive MD state of the art review covering the MD applications for wastewater treatment and water reuse.

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1. Introduction

As time goes by, and with the growth of the world population (Table 1), the need for fresh water for various applications is increasing. Figure 1 shows the trend in global water consumption by sector. As can be seen, the

agricultural-related field is the most important water consumer sector. World water resources are mainly salty and some are fresh resources. Saline water is found in seas and oceans (~97.5%) while fresh water (~2.5%) is either stored underground (~30% of 2.5% fresh water) or in the form of ice/snow covered

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mountainous regions, like the Antarctic and Arctic (~70% of 2.5% fresh water), but only 0.3% is accessible by humans [3].

With this limited amount of usable fresh water, introducing an alternative source for fresh water production is a critical subject. Besides the desalination of saline resources [4-6], water recovery by wastewater treatment can be investigated as an emerging and promising resource for the perspective of global fresh water demand. Wastewater has been generated in various industries such as petrochemical, refinery fuel production plants [7-10], agriculture and food processing [11,12], textile and leather industries [13,14], and etc. Among the different wastewater treating methods, membrane-based water recovery unit-operations are highlighted due to their various advantages, which are comprehensively discussed in the literature [14-16].

Membrane separation processes typically used for wastewater treatment include microfiltration (MF) [8], ultrafiltration (UF) [14], nanofiltration (NF) and reverse osmosis (RO) [17], electrodialysis (ED) [18], capacitive deionization (CDI) [5], and etc. Most of these are pressure-driven and use pressure difference as the driving force. Using hydraulic pressure difference as the mass transfer driving force has its own disadvantages. One of the most important weak-points of such pressure-driven membrane processes is the osmotic-pressure limitation [3], especially in the case of brine desalination and hyper saline wastewaters through either RO or NF processes. Therefore, searching for a new water/wastewater alternative is of interest.

Table 1
World population increase trend and its distribution since 1950 to 2050 (millions) [1].

Year	Asia	Africa	Europe	USA	Total
1950	1377	221	296	158	2522
1960	1668	277	316	186	3022
1970	2101	357	341	210	3696
1980	2586	467	356	230	4440
1990	3114	615	365	254	5266
2000	3683	784	376	278	6055
2010	4136	973	376	298	6795
2020	4545	1187	371	317	7502
2030	4877	1406	362	333	8112
2040	5118	1595	349	343	8577
2050	5268	1766	332	349	8909

A new hybrid non-isothermal membrane process familiarized is a combination of distillation and membrane separation called the “membrane distillation” process. Membrane distillation (MD) [19] is a versatile non-isothermal membrane process for separations that is mainly suited for applications in which water is the major component present in the feed to be separated [20]. MD refers to a thermal-driven transport of vapor molecules through a microporous hydrophobic membrane. Among other applications of the MD process, most of the researches have been focused on desalination and water/wastewater treatment [21-23].

The first patent on the MD process was issued in 1963. After this, Lawson and Lloyd conducted an in-depth review on MD and its historical development in 1997. Various MD applications and its theoretical aspects were also reviewed comprehensively by various research teams. In 2011, Khayet reviewed the theoretical modeling and membranes of the MD process [9]. However, application of the MD process for wastewater treatment has not yet been addressed. In this work, a comprehensive MD state of the art review covering the MD applications for wastewater treatment and water reuse is presented.

2. Membrane distillation process

2.1. Basic principles of the MD process

Membrane distillation is an emerging non-isothermal membrane process which uses thermal energy in order to provide a vapor phase of volatile molecules present in the feed stream (i.e. mostly water) and condensing of the permeated vapor in the cold side (Figure 2). The driving force in MD is the partial pressure difference between each side of the membrane pores. The temperature difference leads to a vapor pressure difference across the membrane. Due to the hydrophobic nature of the membrane, only vapor can pass across the membrane and not liquid solution being distilled [19].

There are four major configurations for the MD process, the difference being in the method to impose a vapor pressure difference across the membrane's pores to drive the permeation flux. These major configurations are described in Table 2. In DCMD, an aqueous solution colder than the feed stream is maintained in direct contact with the distillate side of the

hydrophobic membrane. Both the feed and distillate aqueous solutions are circulated tangentially to the membrane surfaces by means of pumps. The DCMD is the simplest and the most studied MD configuration. A stagnant air-gap is interposed between the membrane and a condensing surface in AGMD mode. In this case, the distilled volatile molecules (mostly water molecules) cross both the porous membrane and the air-gap to finally condense over a cold surface inside the membrane module. In the third MD mode, i.e. SGMD, a cold inert gas (mostly dried air) sweeps the distillate side of the membrane carrying the vapor molecules and condensation takes place outside of the membrane module. In this mode, due to the heat transferred from the hot (feed) side via the membrane, the sweeping gas temperature in the distillate side increases continuously along the membrane module length. In order to impose the driving force across the MD membrane, vacuum is also applied in the distillate side by means of a vacuum pump. The applied vacuum pressure should be lower than the saturation pressure of the volatile molecules to be separated from the feed (hot) solution. In this configuration, condensation also takes place outside of the membrane module [9, 19-23].

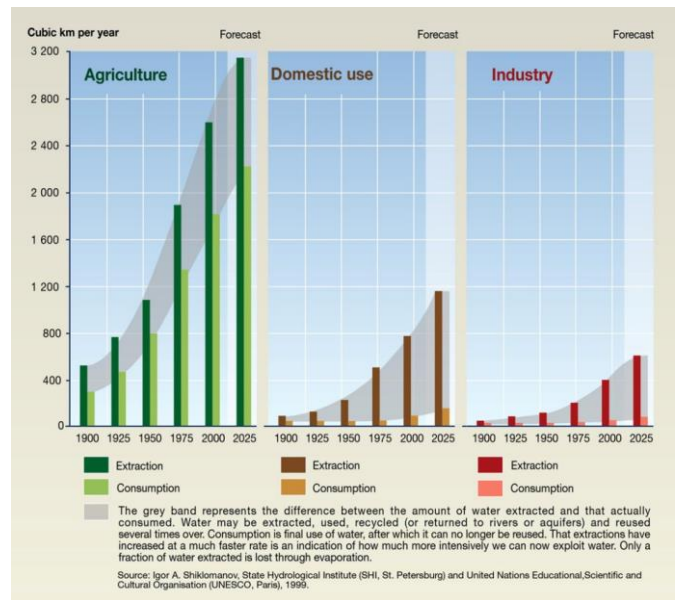


Fig. 1. Global trends in water consumption by sector [2].

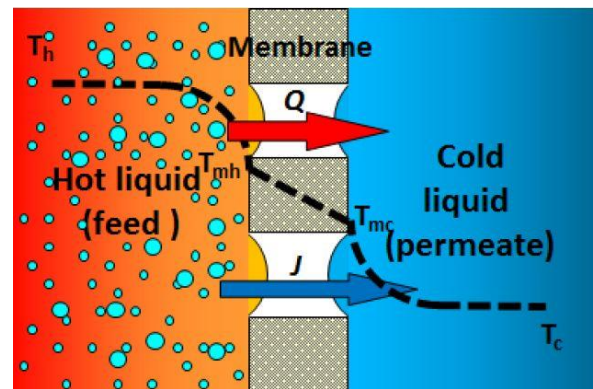


Fig. 2. Basic principles of MD process [24].

2.2. Advantages of the MD process

The MD process was first conceived as a separation process that could operate with a minimum external energy requirement and the least capital and land for the plant [10]. Required equipment for the MD process are much smaller, which translates to a savings in terms of real state; and operating temperatures are much lower, because it is not necessary to heat the process liquid above its boiling temperature. These benefits result in less heat lost to the environment through the equipment surface [23]. On the other hand, the feed temperatures in MD typically ranged from 35 to 85°C. Therefore, low grade, waste and/or alternative energy sources such as solar, wind or geothermal energies can be coupled with MD systems for an economical and energy efficient desalination and water/wastewater treatments [25]. Indeed, MD plants powered by solar energy have been shown to be cost competitive with RO in remote areas [19]. Also, easier operating conditions, lower

operating pressure (usually ambient pressure) which increase safety; and less fouling problems are some other benefits of the MD process [23].

2.3. Membranes for the MD process

Membranes used in the MD process should satisfy some requirements including either the applied membrane being single-layer or multi-layers, at least one of the layers which is within direct contact to the hot stream should be hydrophobic; be thin (since the permeation flux is inversely proportional to the membrane thickness); have reasonably small pore size (in the range of 0.1 to 0.5µm) since the entry pressure difference is inversely proportional to the

pore size; have low surface energy of the membrane material which leads to higher hydrophobicity (a critical property for MD process); be as highly porous as possible, have a low tortuosity factor and high permeability; have adequate chemical, thermal and physical resistance; have high liquid entry pressure (LEP) (a critical property for the MD process); ability to be used in long term performance of desalination and water/wastewater treatment (one of the major weak-points of current MD membranes); high and low heat and mass transfer resistances, respectively; and must be cheaply available. Detailed studies on MD membranes as well as their performance for various applications could be found in the literature [22-28].

Table 2
A comparison of different configurations of MD process.

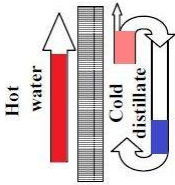
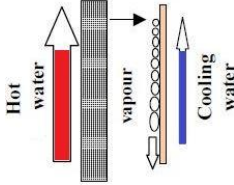
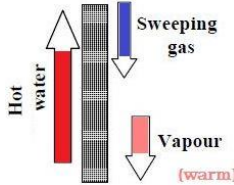
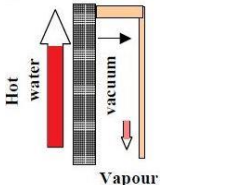
Configuration	General scheme	Specification	Description
Direct contact MD (DCMD)		Membrane is in direct contact with process liquids, i.e. hot and cooling streams	High permeation flux Low energy efficiency Simplest MD mode Most popular MD mode Highest conduction lost
Air-gap MD (AGMD)		A stagnant air-gap in the permeate side is interposed between the membrane and a condensing plate	Highest energy efficiency Low permeation flux Air-gap is around 2-10 mm
Sweeping gas MD (SGMD)		Stripping cold inert gas or air is used as carrier for the produced vapor molecules in the permeate side	Useful for concentrating of non-volatile compounds Condensation happens outside the module
Vacuum MD (VMD)		Permeate side is vapor or air under vacuum	Useful for removal of volatile compounds Permeate is condensed outside the module

Table 3
Timeline of solar-assisted MD systems and their general overview.

Year & Location	MD mode	Membrane	Energy system type	
			Thermal	Electrical
1 2003; Fereiburg, Germany	AGMD	PTFE, Spiral wound	Solar collector	Grid
2 2004; Texas, USA	AGMD	n/a ¹	Solar pond	Grid
3 2007; Irbid, Jordan	AGMD	PTFE, Spiral wound	Solar collector	PV ²
4 2008; Alex, Egypt	AGMD	PTFE, Spiral wound	Solar collector	PV
5 2008; Mexico	DCMD	Hollow fiber	Solar collector	Grid
6 2013; Mahshahr, Iran ³	AGMD, SGMD	PTFE, Flat sheet	Solar collector	Grid

¹ n/a: not available.

² PV: Photovoltaic.

³ Design and constructed in Kargari Research Laboratory (MPRL), Amirkabir University of Technology, Iran.

2.4. Energy for MD process

In a typical MD system, both thermal and electrical energies are required. To provide a hot stream (i.e. feed), the saline solution or water/wastewater should be heated (40 to 85 °C) and in order for re-circulation of hot and cold streams, or in order to provide a vacuum or sweeping gas stream in the permeate side, electrical energy is required. Therefore, the energy source should be used to provide the electricity to operate a MD process. One of the advantages of the MD process is that it could be coupled with a renewable energy source to improve overall efficiency. Various renewable energies such as solar, geothermal, and wind as well as waste thermal energy in the industrial unit have been exercised for coupling with the MD process [18, 29].

Several demonstration projects using renewable energies to drive the MD systems have been constructed; however, most of the researches have been focused on coupling solar energy by the MD process in various regions [30, 31]. These projects appear in temporal order in Table 3. All systems appearing in this table (i.e. Table 3) were solar-assisted either via a solar collector, solar pond or a solar still and were constructed in areas of good solar radiation. Further details could be found in the literature.

3. Water recovery and wastewater treatment using the MD process

3.1. DCMD process

As mentioned earlier, the direct contact MD is the most used mode of the MD process, especially for desalination and water/wastewater treatment. One of the reasons is due to the condensation step that can be carried out inside the MD module enabling a simple MD operation mode [32]. However, it should be noted that the heat transferred by conduction through the membrane, which is considered as the heat loss in MD, is higher than in the other MD configurations [33]. During the DCMD process, evaporation and condensation take place at the liquid-vapor interfaces formed at the pore entrances on the feed and distillate side, respectively. A typical DCMD system used for flat sheet, capillary or hollow-fiber membranes is shown in Figure 3. It is worth quoting that DCMD is mainly suited for applications in which the major component of the feed stream contains nonvolatile solutes such as salt [34].

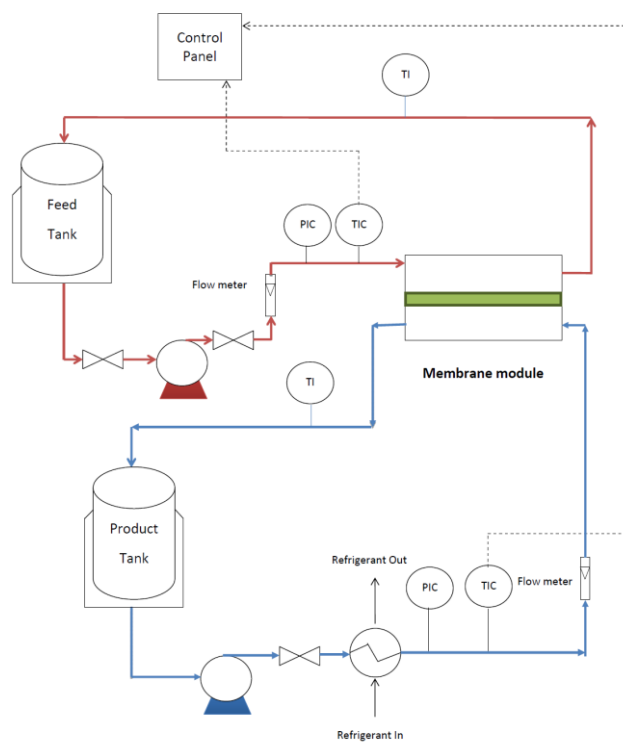


Fig. 3. A general scheme of the DCMD process.

Most of the applied membranes for DCMD experiments are those commercially available, made of hydrophobic polymers and fabricated specifically for microfiltration (MF) purposes [25]. Such membranes consist of composite structures including a thin, microporous, hydrophobic and selective layer (as the top layer) and a porous, less hydrophobic and thicker

support layer. Therefore, the active (selective layer) may have acceptable performance and prevent entering process liquid; however, on the cold side the situation is different. Having more porous structure, being less hydrophobic and thicker leads to amplification of the polarization effect in the distillate side. Consequently, it can reduce the distillate flux which is a serious weak point. Shirazi et al. [3] studied the desalination performance of three typical commercial hydrophobic membranes, e.g. PP, PVDF and PTFE membranes, for real seawater desalination under different operating conditions. Results indicated that the feed temperature may be investigated as the most important parameter and the best performance observed for the PTFE membrane. Moreover, fouling behavior and long term performance of the PTFE membrane is observed. Results of this work were in good agreement with other results in the literature, which used various commercial hydrophobic membranes for DCMD desalination. However, the effect of the membranes' structure was not addressed comprehensively [35]. In another work, Shirazi et al. [23] studied the desalination performance of various PTFE membranes when they were used in the DCMD process. Table 4 shows the characteristics of the applied membranes in this work [23].

In this work, the authors studied the effect of membranes' characteristics, i.e. pore size, type of support layer and thickness. The authors indicated that knowing the effect of support layers in commercial membranes on the distillate flux of the DCMD process is a very important issue for developing specific membranes in order to scale up the DCMD process. Although most of the published papers have indicated that the PTFE membrane may be the best choice for MD purposes, authors comprehensively discussed this fact that only being PTFE membrane material is not enough for a successful DCMD desalination application [23]. It must be noted that as different suppliers have different PTFE membranes (even with the same pore size), very different characters in practice makes some of them completely improper for DCMD desalination. Figure 4 presents the distillate-based performance of the nine different PTFE membranes (Table 4) when they are used in DCMD for simulated seawater desalination.

Table 5 presents the effect of physical characteristics of the typical PTFE membranes (Table 3) on the distillate flux and the salt rejection. During the experiments, all membranes were found to reject salt by more than 99% except for M3 which basically failed due to its large pore size. It is worth quoting that the best pore size range for various MD applications depends on the type of impurity (solute) in the feed stream; however, the pore size value would be investigated in the range of 0.1 to 0.45 μm . Smaller and larger pore sizes may reduce and increase the distillate flux and risk of pore wetting, respectively.

The treatment of olive mill wastewater (OMW) is a major environmental concern in many regions such as Mediterranean countries, where 95% of the total world olive production are produced per year [36]. Low pH and high BOD and COD level as well as low biodegradability and extremely high solid and organic compounds content of such wastewater makes it a dangerous wastewater for the environment. Besides the OMW's phenolic content, it contains many valuable nutrients and has also been referred to possessing soluble dietary fibers and especially pectin materials with excellent gelling ability [37]. El-Abbassi et al. [36] studied the feasibility of the DCMD process for treatment of OMW. The advantage of the DCMD process in comparison to the conventional pressure-driven membrane processes, e.g. MF, UF and NF, rely on its lower operating hydrostatic pressures. Moreover, the DCMD is a non-destructive process regarding phenolic compounds. In their work, three commercial PTFE membranes with different pore sizes (i.e. 0.2, 0.45 and 1.0 μm) were used for treatment of OMW, when the effect of various operating temperatures was investigated. The aim of this study [36] was to investigate the possibility of pure water production and concentration of natural polyphenols from OMW. Results indicated that no significant effect was detected between the pore size and the polyphenols separation coefficient (remains close to 100% after the 8 h DCMD test). The authors concluded that the DCMD processing of OMW using PTFE membranes allows reaching a concentration factor higher than 1.78 after 8 h operating time. They also found that PTFE membranes with larger pore size (i.e. 1 μm) could be used for OMW treatment. Moreover, the obtained OMW concentrate can be used as a source of natural antioxidants such as hydroxytyrosol, which represents ~70% of the total monocyclic phenolic compounds of OMW.

Cooling tower water is typically withdrawn from a freshwater source. Due to evaporation, leakage and wind action, the concentrations of ions, e.g. Ca^{2+} , Mg^{2+} , CO_3^{2-} and HCO_3^- ; microorganisms and chemicals increase, which can lead to scale formation and/or corrosion (Table 6). Hence, concentrated water should be discharged as blowdown stream and freshwater may be supplied as make-up to the tower. For instance, a 300 MW power-plant requires ~20,000 m^3/h circulating cooling water which can potentially lead to 98 m^3/h of the blowdown stream [38,39]. Therefore, treating such wastewater is an interesting subject. Yu et al. [40] studied the application of DCMD for desalting and treatment of cooling tower blowdown (CTB) water. In this

work, the authors used a bench-scale apparatus. A flat sheet hydrophobic PP membrane with 0.1 μm pore size, 65-70% porosity, and $\sim 100 \mu\text{m}$ thickness, was used in this study. When a DCMD process is used for desalting wastewater, distillate flux and its conductivity and also salt rejection are important performance parameters. In this work [40], different compositions of CTB water, especially contents of hardness and silica, were evaluated using the DCMD apparatus. Figure 5 shows the distillate flux and its conductivity for the concentration and desalination of simulated CTB feeds. Moreover, the performance of the DCMD on the concentration of a single silica solution is also shown to highlight the influence of silica. During the

experiments, a distillate flux of $30 \text{ L/m}^2\text{h}$, and a solute (i.e. salt) rejection of 99.95% under feed temperature of $\sim 60 \text{ }^\circ\text{C}$ were achieved. For such wastewater (i.e. CTB), membrane fouling is a certain crisis. Hence, the authors investigated the scaling potential of the applied membranes during the experiments. It was exhibited that the insoluble calcium carbonate scale was formed on the membrane surface for silica-free CTB; however, silica, calcium carbonate and sulfate scaling precipitated together for silica-containing simulated CTB water (Figure 6). The scales resulted in the drop of both distillate flux and salt rejection, while the performance recovered after membrane cleaning [40].

Table 4
Properties of the commercial PTFE membranes used in DCMD desalination [23].

Membrane	Pore size (μm)	Support material	Thickness (μm)	Porosity (%)	Contact angle (o)	LEP (kPa)
M1	0.22	PP ^a	230	80	115.6	117.72
M2	0.45	PP ^a	115	80	120.1	75.67
M3	2.0	PP ^a	300	85	114.7	37.42
M4	0.45	PET ^b	140	75	124.4	82.66
M5	0.45	PP ^b	180	75	124.8	83.33
M6	1.0	PET ^b	175	75	125.2	48.8
M7	0.22	HDPE ^c	175	70	132.2	152.5
M8	0.45	HDPE ^c	175	70	133.5	96.44
M9	1.0	HDPE ^c	175	70	133.6	54.45

Support materials: a) non-woven fabric; b) scrim; c) fiber

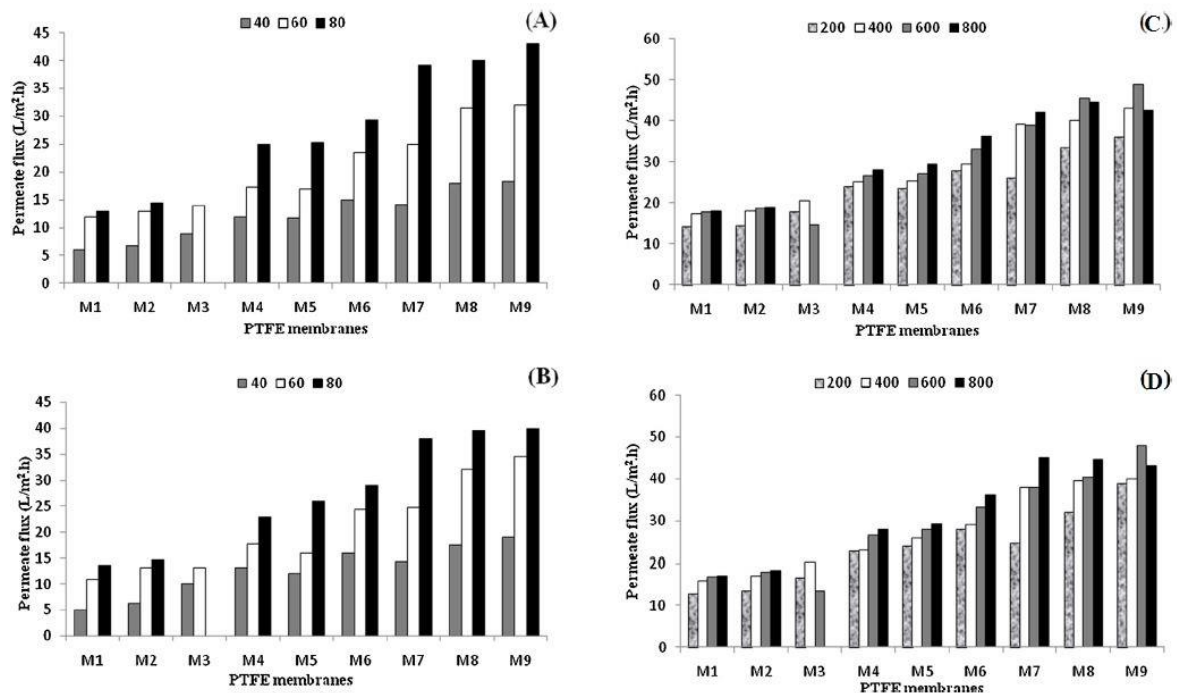


Fig. 4. (A and B) Effect of feed temperature on the distillate flux ($Q_h = 400 \text{ mL/min}$, $Q_c = 200 \text{ mL/min}$, $T_c = 20 \pm 5 \text{ }^\circ\text{C}$, $C_f = 35 \text{ kg/m}^3$ (A) and 45 kg/m^3 (B)); and (C and D) the effect of feed flow rate on the distillate flux ($T_h = 80 \text{ }^\circ\text{C}$, $T_c = 20 \pm 5 \text{ }^\circ\text{C}$, $Q_c = 200 \text{ mL/min}$, $C_f = 35 \text{ kg/m}^3$ (C) and 45 kg/m^3 (D)) [23].

Table 5

The effects of the support layer and membrane thickness on the permeation flux and mass transfer coefficient; ($T_h = 80^\circ\text{C}$, $Q_h = 600 \text{ mL/min}$, $C_f = 45 \text{ kg/m}^3$, $T_c = 20 \pm 2^\circ\text{C}$, $Q_c = 200 \text{ mL/min}$) [23].

Membrane	M1	M2	M4	M5	M6	M7	M8	M9
Flux ($\text{L/m}^2\text{h}$)	16.8	19.4	26.5	28	33.2	38	40.5	48
Mass transfer coefficient (m/s)	0.373	0.431	0.589	0.622	0.738	0.844	0.900	1.067

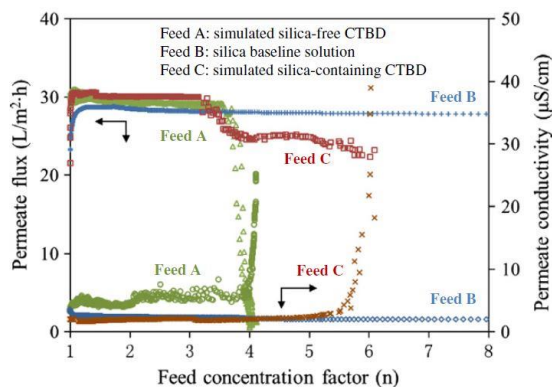


Fig. 5. DCMD flux and distillate conductivity for different types of simulated CTBD feed as a function of feed concentration factor [40].

Ammonia is a common pollutant in industrial and municipal wastewaters and its accumulation in water leads to eutrophication and depletion of oxygen due to nitrification and hence harms the water-borne organisms. Qu et al. [41] worked on the application of a modified DCMD process for ammonia removal from wastewater. In their work, a capillary PVDF membrane with 80% porosity, average pore size of 0.22 μm , LEP of 250 kPa and surface contact angle of 87° was used for the experiments. Feed samples were

prepared by dissolving ammonia chloride into distilled water, and the pH values were adjusted by adding HCl and NaOH to the feed solution. In this work [41], the authors used three different configurations, i.e. a conventional DCMD (a), a hollow fiber membrane contactor (b) and a modified DCMD apparatus (c). In configuration (b), the ammonia stripping was investigated at room temperature without heating and cooling, but the receiving solution containing 0.01 mol/L sulfuric acid was in the permeate. In configuration (a), the feed and the distillate were heated and cooled via a thermostat and cooler, respectively, and no receiving solution was in the distillate. While, in configuration (c), i.e. the modified DCMD process, receiving solution containing 0.01 mol/L sulfuric acid was used in the distillate side. The ammonia removal efficiency by means of the mentioned modules was comparatively studied by investigating the effect of feed pH, temperature, flow-rate, and ammonia concentration. Results showed that ammonia removal efficiency for (a), (b) and (c) was 52%, 88% and 99.5% within 105 min, respectively. The authors indicated that the modified DCMD process was obviously advantageous and could be used as an alternative technique for ammonia removal from wastewater. Results indicated that for configuration (c), feed pH value was proven to be the dominant parameter. In other words, increasing feed pH value was capable of promoting ammonia removal efficiency as well as the distillate flux, but only up to 12.2, after which it gave no noticeable effect. The increase of feed temperature and velocity led to an increase in ammonia removal efficiency, ammonia mass transfer as well as the distillate flux [41].

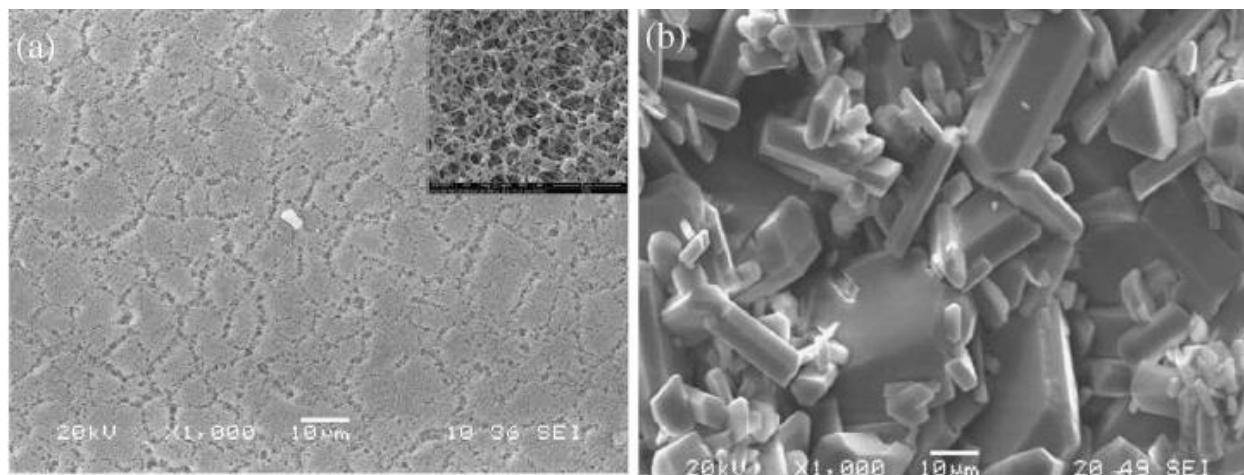


Fig. 6. SEM images for comparison of (a) verging PP membrane and (b) scaled membrane by the simulated silica-free CTBD [40].

3.2. SGMD process

Sweeping gas MD consists of a gas that sweeps the distillate side of the membrane carrying the vaporous distillate away from the permeate side [39]. In this configuration, i.e. SGMD, the condensation of the vapor takes place outside the membrane module. Therefore, an external condenser is required to collect the vapor in the distillate stream. It is worth noting that in SGMD, the gas temperature, the mass transfer and the rate of heat transfer through the membrane change considerably during the gas circulation along the MD module, which can potentially decrease the distillate flux [43,44]. Although, the SGMD process has a great perspective for the future, especially for desalination and water/wastewater treatments, it combines a relatively low conductive heat loss through the membrane with a reduced mass transfer resistance. Similar to the DCMD process, the SGMD can also be used for high-purity water production [9,33] and concentration of ionic, colloid and/or other non-volatile aqueous solutions [28,45]. In SGMD, the feed temperature together with the sweeping gas flow rate was found to be the important operating parameter controlling the distillate flux [45]. The change in partial vapor pressure corresponding to the same temperature change increases as the temperature rises.

As mentioned earlier, the ammonia is a major pollutant in many industrial and agricultural wastewaters, and its elimination is essential in reusing wastewaters for various applications. Various conventional methods have been used for ammonia removal, such as biological treatment, aeration and adsorption. The applicability of ammonia removal technologies generally depends upon several factors. However, investigating an alternative for conventional methods is of current interest. Xie et al. [46] studied the

application of the SGMD process for ammonia removal from wastewater. In their work, wastewater, with 100 mg/L ammonia contaminant at a pH of 11.5 was used for SGMD experiments. The experiments were conducted using commercially available PTFE membranes with 0.45 μm pore size, 70% porosity and, 100 and 200 μm thicknesses. Figure 7 shows a general scheme of the applied SGMD apparatus which was equipped by a MD module made of stainless steel with an effective area of 50 cm² for the membrane surface.

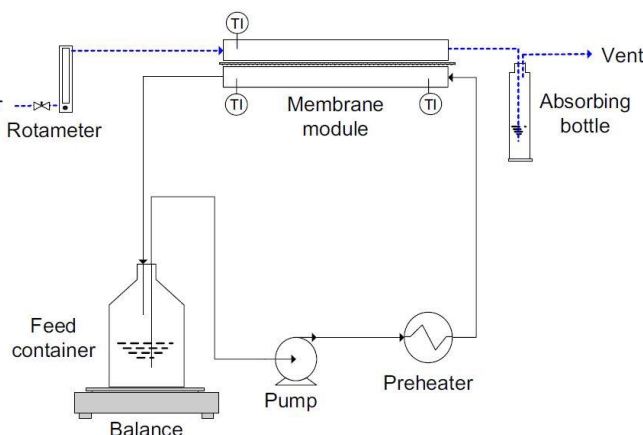


Fig. 7. A general scheme of the applied SGMD set-up in Xie et al. [46] work.

In the mentioned work, i.e. [46], the effects of feed temperature, gas flow rate and feed flow rate on ammonia removal, distillate flux and selectivity were investigated. As expected, feed temperature was found as the most crucial operating parameter, in which with an increase in feed temperature, the distillate flux increased significantly; while the selectivity decreased. These results could be found in Figure 8. The authors concluded that the best performing conditions of highest temperature and fastest sweeping gas flow rate resulted in 97% ammonia removal, resulting in treated water containing only 3.3 mg/L of ammonia. Besides the feed temperature, feed flow rate and gas flow rate were found to be effective on the ammonia removal efficiency; however, sweeping gas temperature had a negligible effect on the distillate flux. On the other hand, the feed flow rate and gas flow rate have less effect on ammonia selectivity [46].

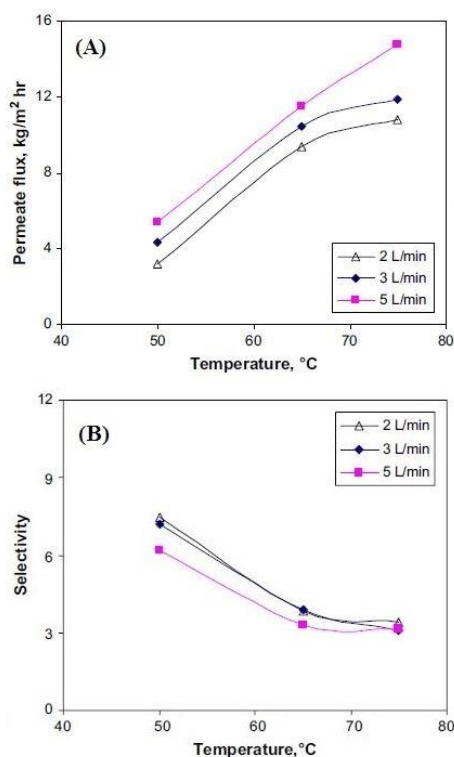


Fig. 8. Effect of feed temperature on the distillate flux (A) and selectivity (B) in ammonia removal using SGMD process [46].

Glycerol is a simple polyol compound, completely soluble in water and insoluble in hydrocarbons such as biodiesel, which has been widely used in food, pharmaceutical and chemical industries [27,44-49]. Dewatering is one of the most critical stages of glycerol refining. Conventionally, the evaporation process has been used; however, due to the high boiling point of glycerol (~290 °C), its downstream processing is difficult and costly. Hence, such a kind of separation process which can achieve water removal at lower operating temperature is attractive. Shirazi et al. [50] studied the feasibility of the SGMD process for dewatering dilute glycerol wastewater. In this work, a PTFE membrane with 0.22 μm pore size and 70% porosity (supplied by Millipore) were used for the experiments. Wastewater samples were prepared by dissolving the analytical grade glycerol in distilled water. The Taguchi optimization method was used in their work in order to carry out a sensitivity analysis study. Figure 9 shows the result of this work, i.e. the main effect of major operating variables, including feed temperature, feed concentration, feed flow rate and sweeping gas flow rate on the distillate flux.

As could be observed (Figure 9), an increase in the feed temperature led to an increase in the distillate flux, but not linearly. This is in good agreement with the results obtained in the literature. With an increase in feed flow rate (up to 400 mL/min), the distillate flux increased and flowed by a decrease in the distillate flux. This is due to the fact that by an increase in the feed flow rate in constant feed channel depth, higher inlet pressure exists for the process liquid which can lead to higher pore wetting risk, as well. Regarding the feed concentration (Fig. 9-c), almost all membrane processes are sensitive to the feed concentration. In this work, increasing the glycerol concentration in the feed up to 3 g/L had a negligible effect on the distillate flux; however, further increase (5 g/L) decreased the distillate flux from about 11.6 L/m²·h to 8.3 L/m²·h. The Taguchi analysis of the data shows that there is some interaction between the operating variables. These interactions are shown in Figure 10.

Further discussion on the parameters' interaction can be found in their paper [50]. The results of this work, which was a new application of the SGMD process, showed that the MD process can be effectively used for dewatering of glycerol. The authors indicated that in all tests conducted, glycerol rejection of more than 99% was achieved.

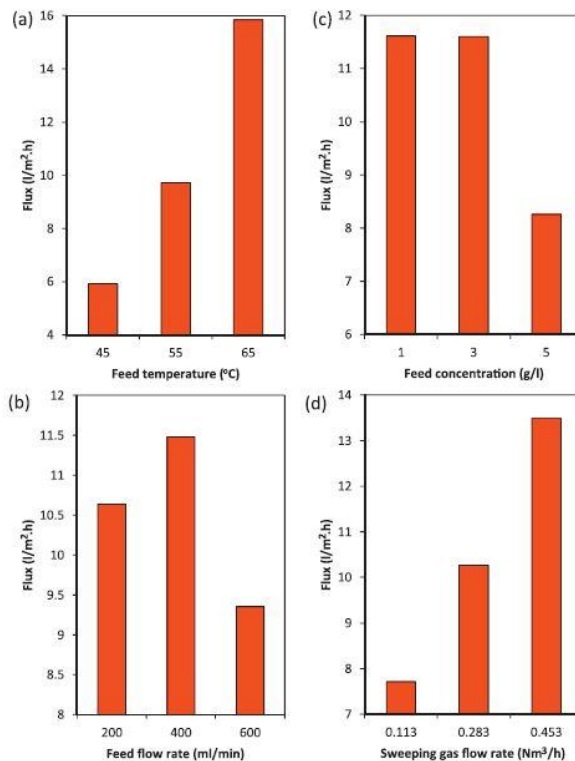


Fig. 9. Main effect of operating variables on the distillate flux; (a) feed temperature, (b) feed flow rate, (c) feed concentration, and (d) sweeping gas flow rate [50].

3.3. AGMD process

As mentioned earlier, the most important drawback of the DCMD configuration is the high rate of heat loss through membrane heat conduction. Furthermore, the need for an outside condenser is the limitation of the SGMD configuration. To solve these drawbacks, a new configuration of MD was introduced, called air-gap membrane distillation (AGMD). In this mode, the temperature difference between the process liquid and the condensing surface is the driving force. As could be observed in Figure 11, mass transfer occurs according to the following four steps, including movement of the volatile molecules from the bulk liquid (i.e. hot feed) towards the active surface of the membrane, evaporation at the liquid-vapor interface (i.e. at the membrane pores), transport of evaporated molecules through the membrane pores and diffusion through the stagnant gas gap, and condensing over the cold surface.

As the distillate is condensed on a cold surface without direct contact with the membrane surface or condensing fluid, AGMD can be used in the fields where DCMD applications are rather limited. Asadi et al. [51] studied the application of AGMD for treatment of an oily-saline wastewater generated in a gas refinery. This work attempted to produce drinking water from high saline oily wastewater. The applied AGMD apparatus in this work was equipped by solar energy (Figure 12). The experimental pilot with a membrane surface area of 40 m² was placed in Sarkhon zone (Bandar Abbas, south of Iran). Results indicated that the average production rate of distillate for the period of spring (2005) was 1.3 L/m²·day, and the total dissolved solid reduced from 1991 to 91 mg/L. Table 7 presents the laboratory analysis of the feed (gas refinery wastewater) and end product of the AGMD process by Asadi et al. [51]. As could be observed, good reduction in the contaminant level is achieved.

As an oil or gas field matures, the rate of production decreases while water production increases. This means that the produced water is the largest waste stream generated in oil and gas industries [52]. Therefore, treatment of this highly polluted wastewater is one of the recent worldwide concerns which should be investigated. Alkudhiri et al. [53] studied the feasibility of produced water (PW) treatment via the AGMD process. In their work, three commercial PTFE membranes were utilized, with a thickness of 175 μm and normal pore sizes of 0.2, 0.45 and 1.0 μm , respectively. The effective area of

the applied membranes in the experimental set-up was 0.003688 m^2 . Figure 13 shows the effect of major operating parameters, e.g. feed temperature, feed flow rate and cooling stream temperature on the distillate flux. As could be observed, with an increase in the pore size, the distillate flux increased. This is in agreement with the results obtained by Shirazi et al. [23], in which authors studied different PTFE membranes with various pore sizes for desalting brine. However, it should be noted that with an increase in the membrane pore size, the pore wetting which is one of the most important MD problems can increase simultaneously. Moreover, the solute rejection for membranes with smaller pore size can be more stable. For example, in the work of Alkhdhiri et al. [53], the salt rejection for membranes with $0.22 \mu\text{m}$

pore size was found to lie between 99.99 to 99.98%; although, the same value for the $1.0 \mu\text{m}$ pore size membrane varied between 97.8 to 97.1% (Figure 13-A). The influence of feed flow rate on the distillate flux was positive. In other words, under constant feed and coolant temperature, increasing the feed flow rate leads to an increase in the distillate flux (Figure 13-B). This can be explained by the fact that using higher feed flow rate under constant conditions and feed channel depth can reduce the effect of polarization effect on the feed-membrane interface. The authors studied the effect of coolant temperature (5 to 25 °C) at constant hot feed temperature and flow rate. Obviously, the distillate flux decreased at a higher coolant temperature (Figure 13-C).

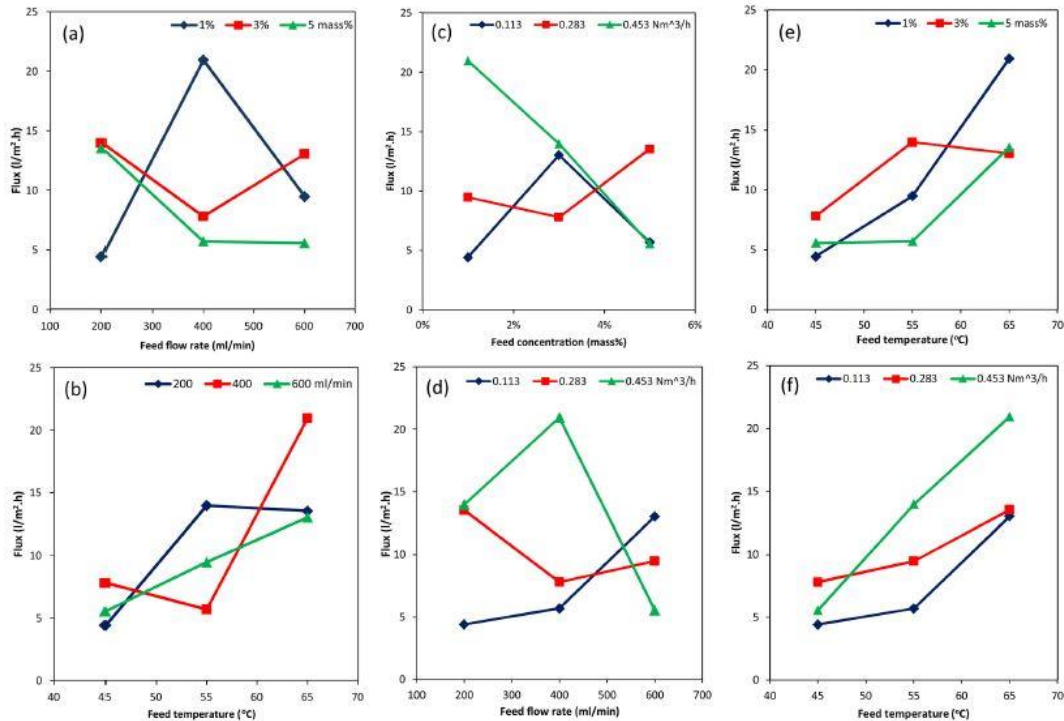


Fig. 10. Plotted interaction lines of operating variables in glycerol dewatering through SGMD process [50].

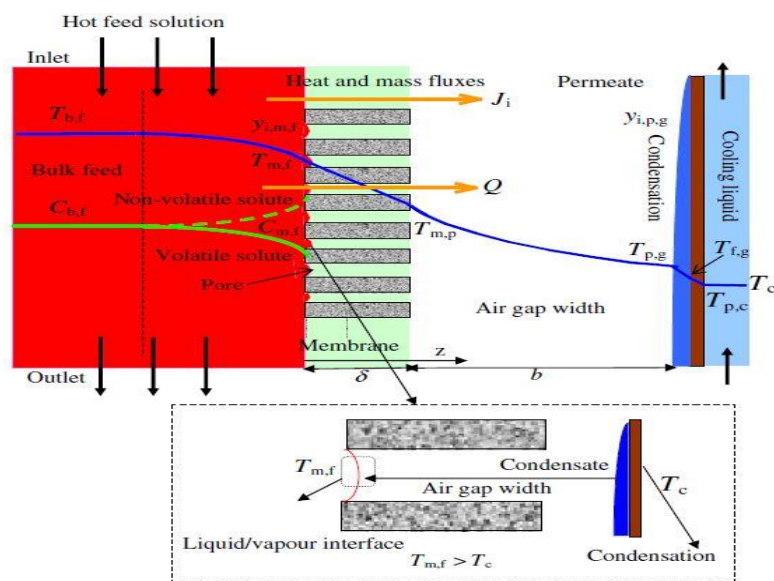


Fig. 11. A detailed scheme of the AGMD process [57].

3.4. VMD process

Another possible way to increase membrane permeability in the MD

process is removing air from its pores, either by deaeration or by using vacuum in the distillate side. It should be noted that this vacuum must be below the equilibrium vapor pressure, i.e. VMD process. In this

configuration, low pressure or vacuum is applied on the distillate side of the module, usually by means of a vacuum pump. As mentioned earlier, condensation takes place outside of the MD module at temperatures much lower than the ambient temperature, and a nitrogen liquid filled condenser is used in the lab scale. There is a very low conductive heat loss in the VMD process. This is due to the insulation against conductive heat loss through the membrane provided by the applied vacuum, in which the boundary layers in the vacuum side are negligible. Moreover, in the VMD process it is a reduced mass transfer resistance.

One of the possible applications of VMD is bioethanol downstream processing. During the bioconversion of lignocellulosic biomass to bioethanol, pretreatment is required to hydrolyze lignocellulose into the corresponding sugars. However, during the hydrolysis process many derivatives, such as aliphatic acids, furans and phenolic compounds are formed. Such byproducts can significantly inhibit fermentation and decrease the ethanol productivity. Chen et al. [54] studied the inhibitors removal from lignocellulosic hydrolyzates by the VMD process. Table 8 presents the composition of dilute acid pretreated hydrolyzates. The experimental apparatus includes a hollow-fiber module and a solar heating system. The authors used distillate flux and removal efficiency to describe the

performance of the VMD process. The effect of operating variables, such as feed temperature and its velocity on the removal efficiency of the inhibitors was investigated. Figure 14 presents the obtained results of Chen et al.'s [54] work. Regarding the feed temperature, high distillate flux and removal efficiency was observed at high feed temperatures. As could be observed, as feed temperature increased (i.e. from 50 to 70 °C), the distillate flux increased by about 158%, i.e. from 2.49 to 6.42 L/m².h. Further, the removal efficiency of acetic acid and furfural increased from 7.26% and 75.47% to 24.79% and 96.25%, respectively. These results can be explained due to an exponential increase in vapor pressure (i.e. MD driving force for mass transfer) of the process liquid (i.e. feed solution) with increasing temperature. On the other hand, the feed velocity was found by the authors as one of the most significant operating variables that influenced the distillate flux of VMD. The authors investigated the feed velocity in the range of 0.45 to 1.05 m/sec at 65 °C feed temperature for 1.5 h operating time. Higher distillate fluxes were achieved at higher velocities likely due to increased heat transfer and temperature/polarization effects (Figure 14-c). Moreover, both acetic acid and furfural removal efficiencies increased slightly with increasing feed velocity, indicating that higher feed velocity promotes mixing at the feed side boundary layers [54].

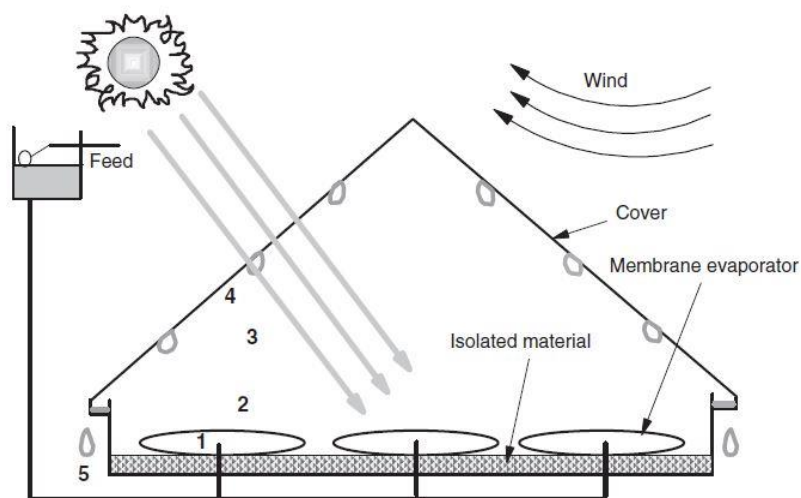


Fig. 12. A general scheme of the experimental pilot in Asadi et al. [51] work.

Mohammadi and Kazemi [55] studied the Taguchi optimization for phenolic wastewater treatment by VMD. In this work, a PTFE membrane with 0.22 μm pore size, 85% porosity and 230 μm thickness, was used for the experiments. The authors investigated the effect of pertinent operating variables, including temperature, vacuum pressure and feed pH. Similar to previous works [9,55], in this application of the MD process, the feed temperature was found as the most effective parameter. To study the separation factor, results show that with decreasing the feed temperature, increasing the phenol concentration and feed pH, the separation factor increased; however, the results show that the water separation factor is approximately independent of vacuum pressure. Based on the Taguchi prediction, a temperature of 45 °C, vacuum pressure of 60 mbar, phenol concentration of 1000 mg/L as well as feed pH of 13 were found as the best operating conditions. In these conditions, the corresponding value for the separation factor was 63.63. It should be noted that in such a feed pH (i.e. 13), the choice for the membrane may be concentrated to the PTFE one. The authors concluded that wastewater containing phenol contaminant can effectively be treated via the VMD process.

4. Conclusions and future perspectives

The MD process has been mainly used for desalination; however, the water recovery from wastewater streams is one of the most promising applications of MD for the future. It has also proven to be a suitable technology for removal of other impurities. While it is capable of treating

many kinds of wastewaters and brines, its ability to compete with current technologies, such as RO and thermal-based water treating technologies, is still limited due to its lack of experimental data in pilot scale and specific membranes and modules. On the other hand, finding new and suitable applications for the MD process currently seems to be one of the major impediments to its commercial use. Moreover, there is another major challenge against MD to be applied for wastewater treatment. Wastewater streams normally include many chemicals that could potentially lead to membrane surface fouling and membrane pore wetting. This is due to the fact that the deposition of these contaminants on the membrane surface could make the membrane less hydrophobic and lead to pore wetting and hence the flux decline. This is the reason that limited works on wastewater treatment using MD are compared with desalination. Therefore, fabricating specific membranes for MD application in wastewater processing is one of the promising future perspectives.

The theory and models of MD are well-known; however, further studies should be investigated for a successful scale-up. Low distillate flux and membrane pore wetting have also been limitations for implementation of the MD process. Therefore, another study on the new and novel membranes, with high porosity and permeability, higher chemical and thermal stability, lower heat conduction capacity, as well as new modules are critical subjects for future MD researches. For the design of new membranes, pore geometry (i.e. lower tortuosity), high porosity, thickness and hydrophobicity are critical variables. Moreover, having lower temperature and concentration polarizations, and lower heat loss are critical parameters for MD module design.

Table 6
Characteristics of a typical CTBD sample [40].

Analytes	Corresponding units	Values
pH	-	8.5
TDS	mg/L	4749
TSS	mg/L	32
Potassium	mg/L	52
Sodium	mg/L	1158
Calcium	mg CaCO ₃ /L	578
Magnesium	mg CaCO ₃ /L	116
P-alkalinity	mg CaCO ₃ /L	5
M-alkalinity	mg CaCO ₃ /L	254
Sulfate	mg/L	2341
Chloride	mg/L	399
Phosphate	mg/L	8.2
Nitride	mg/L	17
Silica	mg/L	96
Conductivity	μS/cm	7132

Table 7
Characteristics of the feed sample and end product in Asadi et al. [51] work.

Parameter	Unit	Feed	Product
TDS	mg/L	1991	91
Sulfate	mg/L	21.9	2
Chloride	mg/L	1565	6.6
COD	mg/L	2173	261
Oil and grease	mg/L	31	1.12
TPH	mg/L	28	0.73
Calcium	mg/L	286.9	30 μg/L

TDS: total dissolved solids

COD: chemical oxygen demand

TPH: total petroleum hydrocarbon

Table 8
Characteristics of the feed stream in Chen et al. [54] work.

Item	pH	Total sugar (g/L)	Glucose (g/L)	Xylose (g/L)	Acetic acid (g/L)	Levulinic acid (g/L)	Furfural (g/L)
Value	4.56	41.92	35.96	5.96	2.62	0.25	0.72

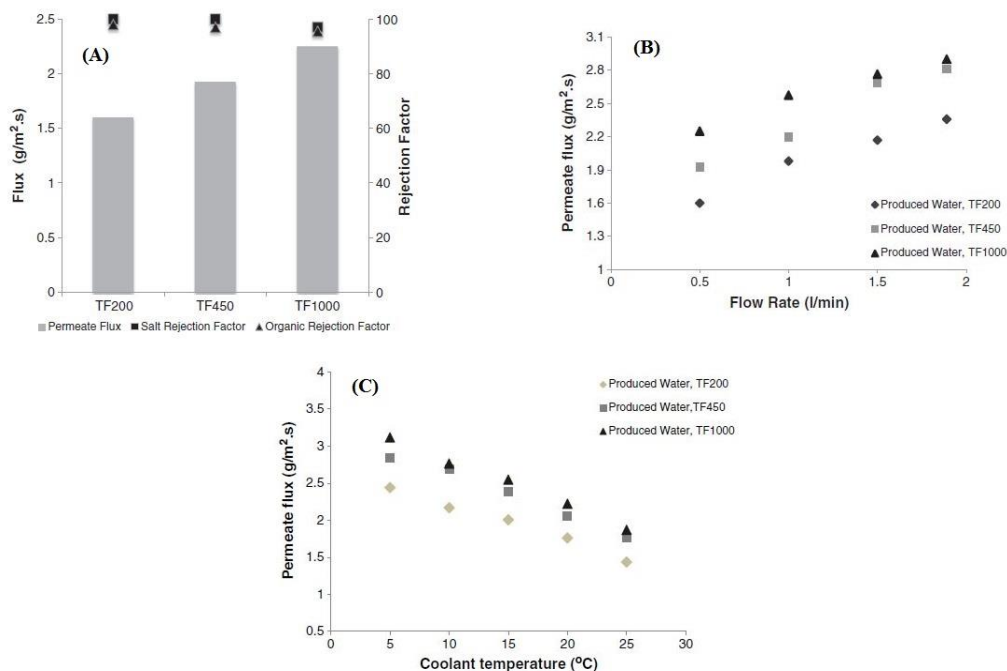


Fig. 13. Effect of pore size ($T_r = 50\text{ }^\circ\text{C}$, $T_c = 10\text{ }^\circ\text{C}$, $Q_f = 0.5\text{ L/min}$) (A); effect of feed flow rate ($T_r = 50\text{ }^\circ\text{C}$ and $T_c = 10\text{ }^\circ\text{C}$) (B); and effect of coolant temperature ($T_r = 50\text{ }^\circ\text{C}$ and $Q_f = 1.5\text{ L/min}$) (C) on the distillate flux in the Alkhudhiri et al. [53] work.

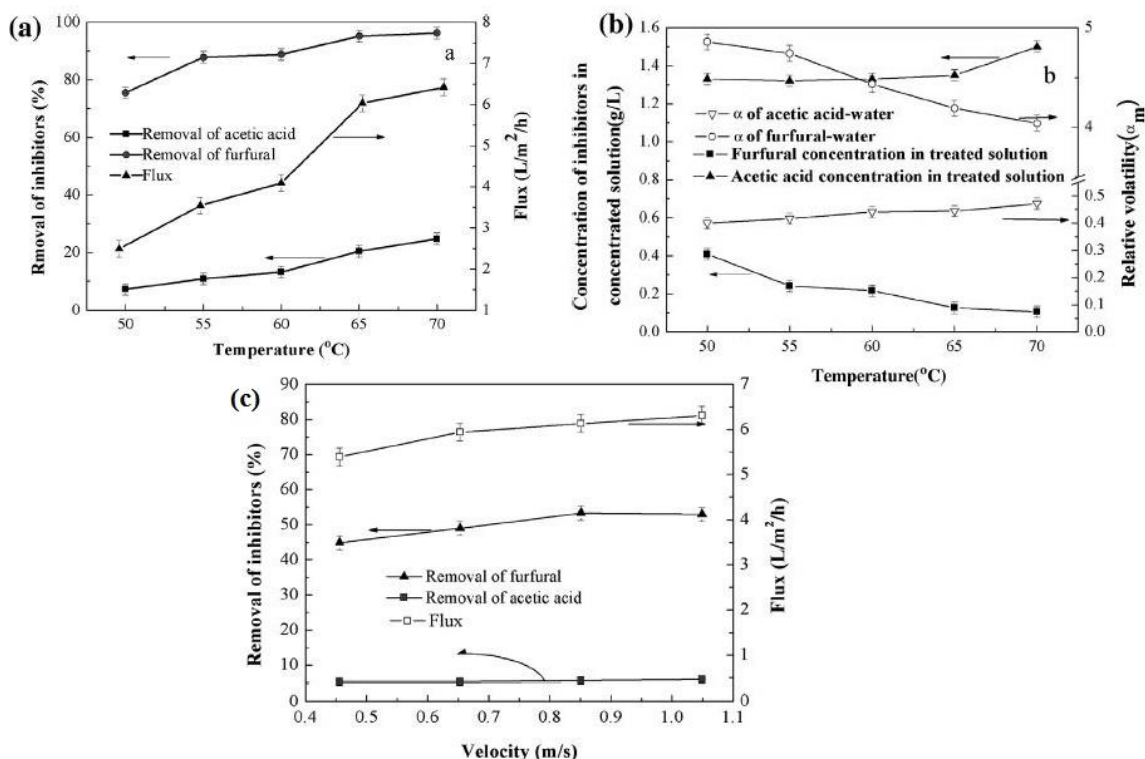


Fig. 14. Effect of temperature on total distillate flux, inhibitors removal (a), treated concentrations of inhibitors and the relative volatility in VMD (b), and effect of velocity on total distillate flux and inhibitors removal (c), in Chen et al. [54] work.

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