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Recent Progress in Membrane Distillation

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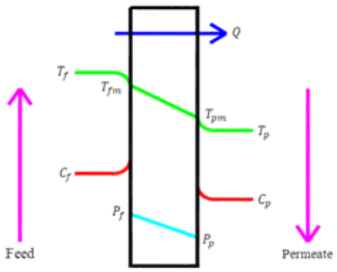
Graphical abstract



Abstract

Membrane distillation (MD) is an emerging membrane separation technique which provides a competition for the conventional separation process such as reverse osmosis (RO) and thermal distillation. The MD process was first developed in the 1960s, but only recently garnered the interest from academics and industry due to the advancement of membrane fabrication technique. The MD is a thermal-driven process which has an ability to be integrated with renewable energy and/or waste heat. The driving force of the MD process is vapor pressure difference where the feed vapor is transported through the non-wetted hydrophobic porous membrane to the permeate regime where permeate will be collected via condensation. As such, the MD possesses a theoretical rejection rate of nearly 100%. This review addressed the recent progress of the MD process in terms of membrane fabrication, integration with renewable energy and/or other membrane separation process as well as applications of MD in various industries. This paper may serve as an update of the recent progress of MD which in some way, is able to help the researchers explore the new investigation field in MD for it to be commercially more viable.

Keywords: Membrane distillation; desalination; renewable energy; hybrid

1.0 INTRODUCTION

Membrane technology has been widely discussed in the industry and academics particularly on its applications on seawater desalination and wastewater treatment. Membrane distillation (MD) is one of the promising membrane separation techniques which adopts thermal separation process compared to conventional pressure separation process, such as reverse osmosis (RO), nanofiltration (NF) and ultrafiltration (UF) [1]. Thermal vapor pressure difference between feed and permeate solutions acts as a driving force of the MD process and requires lower energy consumption. As such, the integration of MD with renewable energy accelerates the growth of research interest from the industry and academics to further explore its viability on the commercial applications [2].

The first MD process was patented by Bodell in 1963 using silicon rubber as membrane in desalination. In 1967, Findley enhanced the MD process and published his research finding in International Journal Industrial and Engineering Chemistry Process Design Development which was regarded as a great breakthrough of this process [3]. The MD process concept proposed by both Findley and Bodell was direct contact membrane distillation (DCMD) in which various materials were utilized as membrane such as paper hot cup and nylon, but the most of the materials used in their MD failed to perform. Since then, the MD interest was diminishing owing to the low flux performance relative to other membrane separation techniques such as RO. With the advancement of membrane technology in the novel membrane production in the early 1980s, the interest in MD research was recovered when a membrane with better MD characteristics was fabricated [1]. To date, the MD research is still being actively carried out with great focus on the membrane fabrication and enhancement of permeate flux performance.

The separation mechanism in the MD process involves the thermal pressure difference in which the feed liquid solution is heated to the liquid-vapor coexist phase, enabling the vapor to gain adequate energy to transport across the pores of hydrophobic porous membrane [1, 3]. The feed liquid molecules will be resisted by the membrane due to high surface tension force of hydrophobic membrane. The vapor will be condensed in the permeate regime by permeate liquid, vacuum or sweeping gas and lead to the clean distilled water. Generally, there are four common MD configurations according to the permeate condensation process, namely direct contact membrane distillation (DCMD), air gap membrane distillation (AGMD), sweeping gas membrane distillation (SGMD) and vacuum membrane distillation (VMD) [1, 3]. Notably, DCMD is the most popular configuration in the literature owning to its simplicity in setup and excellent heat transfer capability [1, 2].

Membrane material plays a pivotal role in the MD process which requires a good hydrophobic characteristic to only allow the vapor to transport across the membrane. In view of this requirement, the membrane is fabricated by hydrophobic polymeric materials such as polyethylene (PE), polypropylene (PP), polytetrafluoroethylene (PTFE) and polyvinylidene fluoride (PVDF) [4]. Further to the hydrophobicity characteristic of the membrane, it has been extensively documented in the literature that a good MD membrane shall exhibit several characteristics for better permeate flux performance. Among the characteristics are firstly, the membrane shall have a high liquid entry pressure (LEP) which can be acquired by high contact angle or low surface energy materials [4, 5]. A high-LEP membrane can prevent the liquid molecules from transporting through the membrane pores and lead to a better wetting resistance characteristic. Secondly, the pore size of the membrane shall be in the range of nanometer to micrometer, whereas the pore size distribution should be as narrow as possible [4, 5]. A fine pore size and narrow pore size distribution membrane will always exhibit an excellent rejection rate with the good control on the passage of large particles through the membrane [4, 5]. Thirdly, the membrane shall have a high porosity where more void areas for the vapor to transfer across the membrane. Both Bourawi *et al.*[1] and Alkhudhiri *et al.* [2] reported that the porosity had a positive effect on the permeate flux performance in which the higher the membrane porosity, the better the permeate flux performance, possibly attributed to the increasing spacing for the evaporation process of feed vapor. Fourthly, the heat and mass transfer rate in the MD process is inversely proportional to membrane thickness. Hence, the membrane thickness in MD process shall be as thin as possible to facilitate a high heat and mass transfer rate across the membrane. Lastly, the membrane shall exhibit a good thermal stability, fine chemical and fouling resistance for it to have an excellent stability in both permeate flux performance and reject rate under long term operation [1, 4, 5].

■2.0 MD CONFIGURATIONS

The heat and mass transfer mechanism in membrane distillation is illustrated in Figure 1. Feed solutiuon that contains volatile compound with certain concentration C_f, will be heated to the feed temperature, T_f with the typical temperature range between 30 to 90ºC which is usually below the boiling point of the feed liquid. The liquid phase in the feed solution will be changed to a mixed region of liquid vapor co-exist phase with the increase of thermal energy in the solution and there will be heat transfer across the membrane, Q. When the feed vapor gains adequate energy from the heating process, the thermal driving force will drive the vapor to transport acorss the membrane through the membrane pores. The vapor will be condensed in the permeate solution due to the temperature difference between feed temeprature and permeate temeprature, T_p. Clean distilled water as the product will be collected from the permeate solution as a result of the condensation process [6].

As previously mentioned, four configurations of the MD process including DCMD, VMD, SGMD and AGMD are illustrated in Figure 2. The permeate flux performance of respective configuration is summarized in Table 1. In the DCMD configuration, the feed solution is in direct contact with the permeate solution where both of the solutions are separated by hydrophobic membrane. Both feed and permeate solutions are circulating either in parallel or counter flow by the assistance of pump or stirrer. The heated vapor molecules induced by the vapor pressure difference will transport across the membrane to permeate regime through the membrane pores [6]. Later, the vapor will be condensed in the permeate regime and the clean distilled water can be collected. The AGMD has a similar working principle as the DCMD; however, one additional air gap between feed and permeate solution is added[6]. The feed vapor transporting through the membrane pores to the permeate regime will be condensed at this air gap where the distilled water as the end product will be collected outside of the membrane module. There are several notable AGMD studies utilizing, such as solar-driven desalination pilot plant in Gran Canaria, Spain [7].

The VMD introduces a vacuum gap between the feed and permeate solutions with a vacuum pump [8, 9]. The vapor molecules transport across the membrane pores to the vacuum gap and they will flow out from the system due to the vacuum pressure difference [8, 9]. Permeate will be collected outside the membrane module by a storage tank for further application. Several researches have been applying the VMD to remove 2,4-dichlorophenol [8] and ,1,1-tricholoethane [9] in the wastewater treatment. In the SGMD configuration, a cold inert gas is introduced to sweep through the vapor permeate in the gap between feed and permeate solutions. Similar to the VMD, the vapor will be condensed outside the membrane module by cold inert gas. This is the least studied method in the MD process owing to the system complication and the cost of using inert gas as the condensate medium [5].

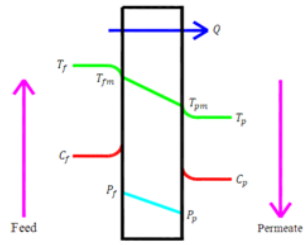


Figure 1 Heat and mass transfer mechanism in MD [6]

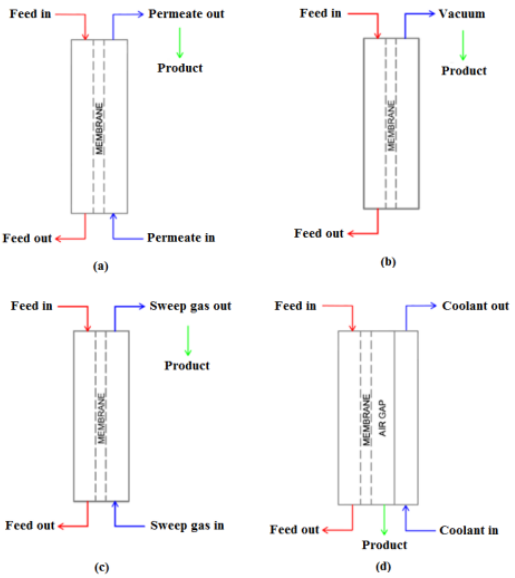


Figure 2 Configuration of membrane distillation: (a) Direct contact membrane distillation, (b) Vacuum membrane distillation, (c) Sweeping gas membrane distillation, (d) Air gap membrane distillation [6]

Table 1 Permeate flux of different MD configurations

Configuration	Membrane material	Solution	Feed temperature (°C)	Permeate flux (kg/m²h)	Reference
DCMD	PVDF	3.5 wt% NaCl	40 – 55	≈2 – 14	[10]
DCMD	PVDF	3.5 wt% NaCl	55 – 80	≈10 – 25	[11]
DCMD	PVDF	10.0 wt% NaCl	60	≈8 – 19	[12]
DCMD	PVDF	3.5 wt% NaCl	50 – 70	≈2 – 8	[13]
DCMD	PVDF	3.5 wt% NaCl	40 – 80	≈2 – 20	[14]
VMD	PES	0.7 wt% NaCl	20 – 70	≈6 – 15	[15]
VMD	PVDF	3.0 wt% NaCl	25 – 75	≈2 – 12	[16]
VMD	PVDF/PTFE	3.5 wt% NaCl	40 – 70	≈2 – 30	[17]
VMD	PVDF	3.5 wt% NaCl	50	≈15 – 22	[13]
AGMD	PTFE	46 - 84 wt% NaCl	5 – 25	≈5 – 13	[18]
AGMD	PTFE	Produced water	40 – 80	≈5 – 25	[19]
AGMD	PVDF	3.5 wt% NaCl	30 – 80	≈2 – 15	[20]
SGMD	PTFE	15.0 wt% NaCl	10 – 70	≈15 – 56	[21]
SGMD	PTFE	Fruit juice	10 – 45	≈0.27 – 4	[22]
SGMD	PTFE	1 - 5 wt% Glycerol	45 – 65	≈4 – 20	[23]

■3.0 RECENT TREND OF MEMRBANE ADDTIVES

The membrane used in the MD process must be a porous hydrophobic membrane and can fulfill certain MD characteristics. Throughout the development of membrane fabrication specifically for particular requirements of the MD process, there are many novel proposals by adding non-solvent additives into the membrane dope solution in order to fabricate a membrane that exhibits excellent MD characteristics. Several notable additives reported in many literature studies are surface modifying macromolecule (SMM), lithium chloride (LiCl) and ethylene glycol (EG) as these additives are able to enhance certain membrane characteristics [24].

The addition of the SMM materials can increase either the membrane hydrophilicity or hydrophobicity. The SMM materials consist of amphiapathic structure of hydrophobic materials with both hydrophilic (polyurea or polyurethane) and hydrophobic (fluorine based polymer chain) parts [25, 26]. This macromolecule structure is able to modify the membrane surface property by forming the nano-scale agglomerates that change the membrane surface into a more hydrophilic or hydrophobic and vice versa due to the elimination of SMM materials during the phase inversion process [25, 27]. The migration concept of SMM materials during the membrane forming process is depicted in Figure 3.

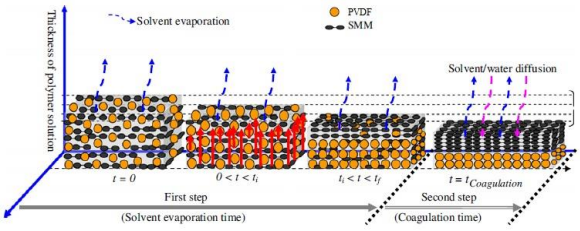


Figure 3 SMM migration during membrane formation [26]

LiCl is produced from the treatment of lithium carbonate with hydrochloric acid. It is frequently described as a pore forming materials in membrane fabrication [10]. The function of LiCl in membrane dope solution is to dissolve rapidly in the water during the phase inversion process which is able to raise the diffusion rate of the solvent from the dope solution, leading to the increase of porosity [27, 28]. With the increment of porosity of a membrane, the membrane possesses a higher void area ratio which allows more vapor to transport across the membrane and hence enhances the permeate flux. However, it is noteworthy to mention that the use of the LiCl additive should be optimized as the further increase of the LiCl additive leads to an adverse effect on reducing the mechanical strength of the membrane as a result of the increasing cavities and porous structure formation [28]. A PVDF hollow fiber membrane with LiCl as additive is presented in Figure 4 which possess a diameter of 400 µm, 85% porosity and 76° contact angle [10].

EG is an organic compound which is one of the non-solvent additives used to produce thin skin layer. The involvement of EG in the rapid diffusion of the solvent into the coagulation during phase inverse process enhances the arrangement of the molecule in the skin layer which eventually results in a more organized and thinner skin layer. Several literature reviews reported that with the reduction of skin layer of a MD membrane, the permeate flux will increase, possibly attributed to the reduction of the barrier of membrane skin layer during the evaporation of feed solution [29, 30].

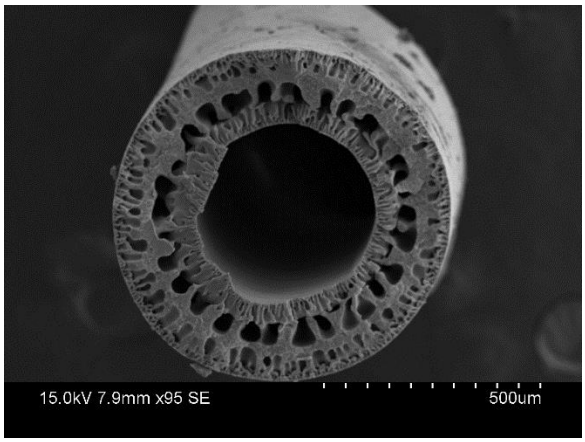


Figure 4 SEM morphology of the cross section of PVDF hollow fiber membrane with LiCl as additive [10]

4.0 MD WITH RENEWABLE ENERGY

The MD has a potential to be integrated with renewable energy source as low-grade energy is required for heating the feed solution. For instance, solar energy can be integrated with the MD system either as a standalone system by generating electricity required for the process equipment such as circulation pump, or producing thermal energy as the heating source of feed solution [31, 32]. In addition, thermal energy source such as the waste heat generated by the diesel engine is also one of the viable sources of thermal energy to be applied in the MD process to generate driving force for the heat and mass transfer process.

Few notable MD processes with the integration of renewable energy are the MD system integrated with diesel engine waste heat in Pantelleria, Italy and the MD system coupled with solar thermal flat plate collector in Gran Canaria, Spain [33, 34]. The MD system in Pantelleria was built beside a diesel power station where the cooling circuits were able to provide waste heat with a temperature up to 90°C for 24 hours per day. The capacity of the MD system was designed to be 5 m^3 per day under 24-hour operation using 12 MD membrane modules with a total membrane effective area of 120 m^2. The schematic diagram of the MD system in Pantelleria is displayed in Figure 5. The heat exchanger of the diesel engine in the cooling circuit transferred the waste heat to the intermediate circuit of the MD system which served as the thermal energy for the evaporator to heat the seawater. The heated vapor will be condensed on the cold water loop.

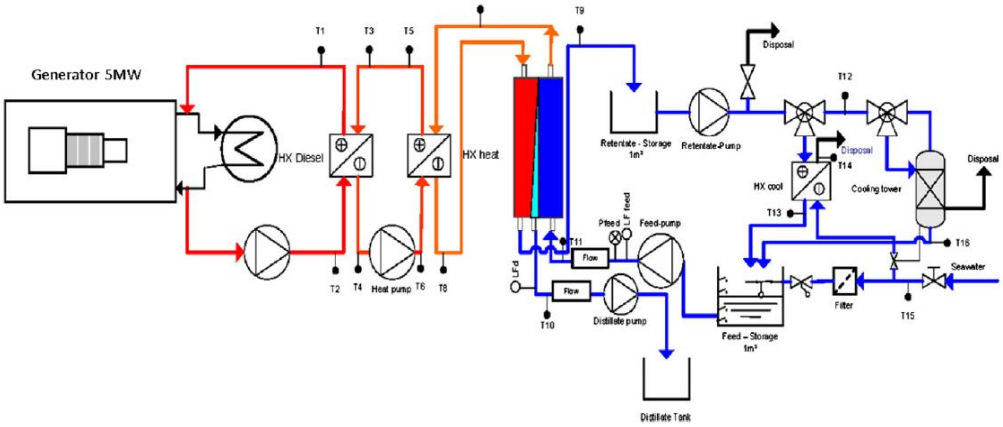


Figure 5 Schematic diagram of membrane distillation system in Pantelleria [33]

The MD system with a membrane effective area of 120 m^2 in Gran Canaria used thermal energy as the heating source for the feed solution with a water production up to 3.5 m^3 per day (Figure 6). The thermal energy for heating feed solution in this MD process was supplied by the solar thermal collector with an effective collecting area of 182 m^2.The MD system adopted two heating circuit with an heat exchanger. Water was heated by solar collector in the heat circuit which its thermal energy was transferred to feed circuit through heat exchanger. The feed solutions received heat from the heat circuit to increase the feed temperature up to 80°C. The condensed permeate solution consisting of clean distilled water was collected in the distillate circuit and stored in the distillate tank.

Figure 6 DCMD plant in Gran Canaria [33]

5.0 HYBRID MD SYTEM

A hybrid system is defined as a combination of different unit operations which are interlinked with each other to provide an optimum performance for a specific task. In a hybrid MD system, it can be integrated with other membrane separation process such as RO, UF and NF for a better separation performance under long-term operation [5]. However, the hybrid MD systems are still under the laboratory scale and pilot plant study, and there is no commercial application of the hybrid MD system to date [5].

The oily wastewater remains a serious problem in the worldwide and poses a major treat to environment and ecology if the wastewater is discharged to the river or sea without proper treatment. The conventional method to treat the oil emulsion contained in the wastewater is by either chemical or mechanical which are not effectively and high in operation cost. Gryta *et al*. [35] had proposed a hybrid MD system which combined UF and MD processes in treating the oily wastewater in laboratory as shown in Figure 7. The oily wastewater with oil content of 120 to 360 ppm was pre-treated by UF process. The permeate produced by the UF process typically contained 5 ppm which was subsequently purified by the MD process. The MD process demonstrated astonishing rejection rate of 99.5% which could almost remove all the oil content from the UF permeate.

Similar to the hybrid UF-MD process, Mericq *et al*. [36] applied the MD method as the secondary purification further from the RO process in the brine water desalination. Laboratory synthesized brine water with a concentration of up to 291.1 g L^{-1} was used in the study. Prior to the first stage purification by RO process, the brine water concentration was reduced significantly

to 50 gL^{-1} and the RO permeate was directed to the MD circuit for second phase separation. The MD process in this study demonstrated a rejection rate of 99%. The overall hybrid RO-MD system was able to archive a high rejection rate relative to the RO process. In the meantime, for the permeate pressure of 6 kPa, feed temperature of 50 °C, the hybrid RO-MD exhibited a 2 fold increase of permeate flux compared to the RO process.

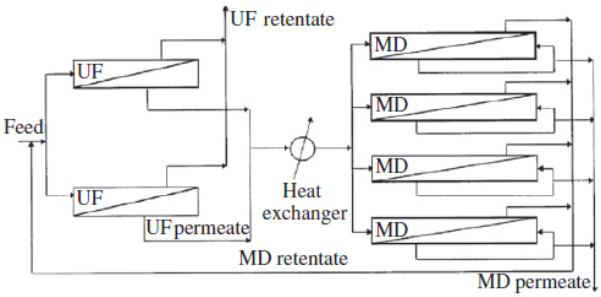


Figure 7 Schematic diagram of hybrid UF-MD process for oily wastewater treatment [35]

It is expected the hybrid MD system can help enhance the permeation flux and rejection rate. The additional membrane process usually acts a pre-treatment prior to the MD process to achieve the above two purposes and mitigate the fouling. If a high pressure-driven membrane process, such as RO, is added prior to the MD process, the energy consumption can also be reduced.

■6.0 CURRENT APPLICATIONS OF MD

Table 2 summarizes the some current MD applications in separating different non-volatile components in laboratory scale. In the food processing industry, the MD was successfully applied in the separation of skim milk and fruit juice concentration [1, 2, 5]. Hausmann *et al*. [37] separated the skim milk of 20% dry matter concentration from the dairy solution using DCMD under the feed temperature range of 35 to 55°C. The result from this study showed that the DCMD presented a rejection rate of 99.5% with a permeate flux of 12 kg/m^2hr which was comparable to the RO process in the dairy industry and it was suitable for the future application in the dairy industry owing to the low flux sensitivity towards the concentration of the processed fluid. On the other hand, Jørgensen *et al*. [22] and Jensen *et al*. [38] adopted the MD process in the recovery of black currant and berry fruit juice aroma. The experimental study conducted by Jensen *et al*. revealed that the recovery of black currant juice was at a rate of 99% and the flux was measured between 1.5 to 10 kg/m^2hr under the temperature range of 20 – 70°C [38]. In the meantime, the study conducted by Jørgensen *et al*. reported a recovery rate of 73–84 vol% with the permeate flux of 5 kg/m^2hr under the feed temperature of 45°C for berry fruit juice aroma using the MD process [22].

In addition, the MD also demonstrates the potential in textile industry for the treatment of dye solution. Criscuoli *et al*. [39] reported that different commercial dye materials such as remazol brillant blue R, reactive black 5, indigo (vat blue 1), acid red 4 and methylene blue were removed from the dye solution using the VMD process. The permeate flux were recorded in the range of 14–40 kg/m^2hr under the feed temperature of 40–60°C with a dye rejection rate of 62%. The study of the removal of RB5 dye with a concentration of 0.05 gL^{-1} was conducted by Mokhtar *et al*. [40] using the DCMD process. Their study revealed a dye rejection rate of 99.78% and a permeate flux of 5.64 kg/m^2hr. Interestingly, their rejection rate was found to be more superior to those using RO, UF and NF processes.

In addition to food processing and textile industries, the MD is also widely applied in the wastewater treatment, nuclear and chemical industries as summarized in Table 2. It is believed that more potential MD applications in various types of industries, such as pharmaceutical industry, environmental protection, energy and etc. will be continuously explored by academicians and industrial experts.

Table 2 Some current applications of MD process

Area	MD Configuration	Solution	Reference
Food process	DCMD	Black currant juice	[38]
Food process	SGMD	Berry juice	[22]
Food process	VMD	Berry juice	[22]
Textile	DCMD	Textile dye	[39]
Textile	DCMD	Textile dye	[40]
Nuclear	DCMD	Radioactive	[41]
Chemical	DCMD	Humic acid	[42]
Chemical	DCMD	Arsenic	[43]
Chemical	SGMD	Ammonia	[44]

■7.0 CHALLENGES IN MD

Membrane distillation is considered as a promising technique in the membrane separation process which possesses a high possibility of commercialization. However, there are only a few industrial/practical applications to date. Furthers for MD to become commercially viable, there is much challenges to be tackled which particularly as follow,

- The design and fabrication of the membrane specifically for the MD application are still scarce. Most of the membranes used in the MD process reported in the literature are based on the commercially available membranes which are originally intended for the usage of other membrane processes. An ideal membrane for the MD process has optimum performance under long-term application with minimum fouling. More investigations are required for the development of required MD membrane, such as the fabrication of hydrophilic/hydrophobic membrane, nanostructure membrane and/or plasma and surface modified membrane with low production cost.
- Most of the current investigations of MD process in various applications and on the study of the effect of operating conditions are still remained in laboratory scale. A pilot plant study based on the industrial application is required for performance monitoring before it can be practically utilized.
- The energy consumption in the MD process should be further reviewed as the current cost of the MD in the production of fresh water from seawater desalination and wastewater is still relatively high compared to the existing available technique. A more innovative MD process should be developed for the purpose of cost reduction, such as heat recovery system in order for it to be commercially economical and feasible.

8.0 CONCLUSIONS

MD has a significant advantage over other membrane separation techniques such as RO and NF owning to its low energy consumption and ability to be integrated with renewable energy. Nonetheless, from the MD development since the 1960s to present, this method is still not widely implemented in practice and/or industry where most of the investigations are still conducted in the laboratory scale. Along with the implementation issue of MD for it to be practically and industrially feasible, a higher permeate performance should be enhanced in the MD process, as comparable with conventional pressure driven processes such as RO. In addition, a more extreme system should be developed, such as the integration with other membrane separation techniques coupled with renewable energy source. This integrated system may lead to a better overall performance on separation and energy saving, as well as in turn reduction on the capital investment and operating cost. In the near future, a commercial viable MD system is expected to possess a low capital and operation cost as well as a high permeates flux performance that is on par with the pressure driven process.

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