A Review on Direct Contact Membrane Distillation

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Abstract: Membrane technology is a rapidly growing separation technique which is spreading its roots in various industries such as desalination, waste water reclamation, fruit juice concentration, ion removal, distillation etc. Being more cost and energy intensive, 'Membrane Distillation', is being researched all over the world as an alternative to conventional separation processes. Direct contact membrane distillation (DCMD) is the simplest of all membrane distillation operations in which either sides of the membrane are in direct contact with the feed and permeate respectively. It is a temperature driven process and is free from osmotic pressure limitation of RO. The scope of using waste heat and low energy sources makes the process more interesting. This review provides a critical analysis of work that has been done so far in DCMD, kinds of membrane used in the process, the heat and mass transfer mechanism and application on DCMD in various industries.

Keywords: direct contact membrane distillation, membrane technology, distillation

1. INTRODUCTION

Worldwide crisis of portable water and decreasing sources of fresh water has led to research in various technologies in desalination and waste water reclamation. Thermal desalination technologies such as multi-stage flash, evaporation and multi-effect distillation, are challenged by membrane based technologies such as reverse osmosis (RO), nanofiltration (NF), electrodialysis (ED) and membrane distillation (MD) [1].A membrane may be defined as a selective barrier, which allows some components to pass through and barricades the other [2]. With the advances in polymeric materials and membrane synthesis techniques, membrane technology has gained a renewed interest among researchers.Membrane based processes are also pave the way for process intensification as they have potential to replace conventional energy-intensive processes, with significant reduction in cost, energy and environmental impact [3].Membrane distillation (MD) is a membrane based separation process in which a heated aqueous feed is contacted with a porous and hydrophobic membrane[4]. The membrane allows the vapors of volatile component and restricts the liquid-mixture from crossing from feed side to permeate side. Thus a liquid-vapor interface is localized at the membrane surface.

The vapors being transported to the permeate side may be condensed inside or outside the membrane module. Depending on the requirement, the desired product may be permeate (e.g., desalination), concentrated feed (e.g., dehydration of fruit juice) or both (e.g., azeotropic distillation) [5]. Similar to conventional distillation process, MD depends on vapor-liquid equilibrium and requires latent heat of vaporization for achieving the phase change [6]. The rate of vapor flow is dependent on the difference in vapor pressure of volatile component at feed side and permeate side. Thus, the partial pressure gradient sustained by the temperature difference is the driving force for the membrane distillation operations [7, 8].

Depending on the permeate condensation mechanism, various MD configurations are possible such as direct contact membrane distillation (DCMD), vacuum membrane distillation (VMD), air gap membrane distillation (AGMD) and sweeping gas membrane distillation (SGMD).

DCMD, is the simplest MD operation in which a porous and hydrophobic membrane is in direct contact with hot aqueous feed and cold-condensed permeate. The membrane acts as a barrier separating hot feed and cold permeate streams.Inside the membrane pores, only the gas phase is present through which the vapors diffuse with partial pressure difference as the driving force. DCMD is ideally suited for operations like desalination and waste water reclamation where water is the volatile component[4, 6, 8-10].

In VMD configuration, the vapors are sucked out of the membrane module using vacuum on permeate side and condensedoutside. This configuration is more suitable for separation of volatile solute from non-volatile solvent. Because of the absence of boundary layer on permeate side, there is less thermal loss, but generation of vacuum required more energy and complicated equipment [11, 12].

In AGMD, an air gap exists between the membrane and the cooling wall on which vapors are condensed. The air gap provides an insulation to the condenser channel, thus minimizing the conduction losses. However, an additional mass transfer resistance is caused because of the gap which leads to reduction in permeate flux. AGMD is well suited for separation of volatile components with low surface tension such as alcohols [13, 14].

In SGMD (also known as air-stripping) the permeate vapors are carried outside the membrane module using a sweeping gas or carrier gas. The condensation takes place in an external condenser. The advantage of SGMD over AGMD is the significant reduction of the mass transfer resistance through forced flow. SGMD is well suited for volatile substances with low surface tension[15, 16].
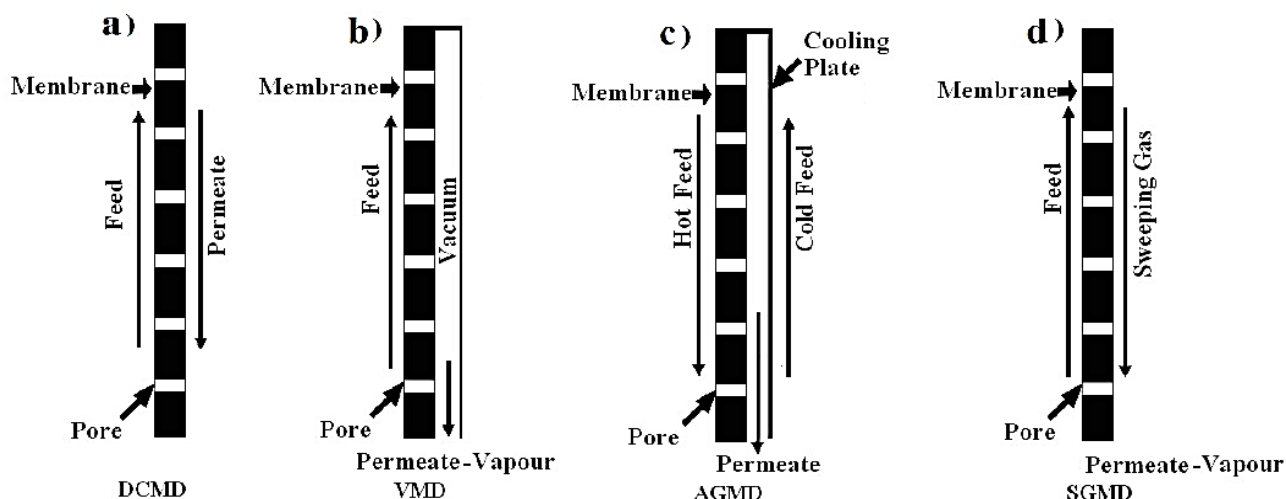

Fig. 1. MD configurations: (a) DCMD, (b) VMD, (c) AGMD and (d) SGMD [4]

Above four configurations are represented in fig 1.

New configurations such as permeate gap membrane distillation (PGMD) [17], liquid-gap membrane distillation (LMD)[18], and material gap membrane distillation (MGMD) [19], are also being investigated.

Any of permeate (demineralization, desalination, waste water recovery) or retentate (concentrated proteins, juices or minerals) can be the desired product after the MD operation depending on the requirement.

2. LITERATURE REVIEW

DCMD operation was first used by Weyl in 1964, for which he was granted a U.S. Patent in 1967. In his experiment, he used an air-filled porous hydrophobic PTFE membrane, 3.2 mm thick, 9 μm in pore size and 42% porosity to recover demineralized water from saline water. The experimental fluxes up to 0.87 Kg/m²-hr, were reported which were in good agreement with theoretic prediction of 1 Kg/m²-hr [20].In 1966,Findley studied evaporation through porous membranes. He used paper, gum wood, glass fibers, cellophane, nylon and diatomaceous earth as membrane materials. These materials were coated with silicone, Teflon or water repellant for hydrophobicity. He compared MD with an infinite-stage flash evaporation system. He suggested improvement in efficiency, by elimination of non-condensable gases through the membrane. He also pointed out disadvantages over conventional evaporation, which were diffusion-resistance through the medium, and heat loss by conduction through the medium. Findley concluded that "If low cost, high temperature, long-life membranes with desirable characteristics can be obtained, this method could become an economical method of evaporation, as well as an important possibility in the conversion of sea water. Considerable improvement over the above calculations should be possible, using more appropriate membranes and eliminating non condensable gases" [21].In 1985, Hanbury et al., experimentally analyzed the performance of PTFE membrane and did cost estimates based on his experimental work. An empirical relation for the distillation heat transfer coefficient, based on average membrane temperature was suggested as under:

$$h(T) = 0.0049 T_m^{1.17} kW/K/m^2$$

Based on the experimental data, an empirical relationship to calculate specific membrane area requirement was also given:

$$\frac{A}{M_d} = \frac{M_r}{M_d} \frac{C_p . \eta}{\frac{0.000833}{\theta}} (T_4^{-0.17} - T_2^{-0.17})$$

Where A is the membrane surface area, M_r and M_d are flow rates of feed and distillate produced respectively, C_p is specific heat of brine, η is distillation efficiency, θ is temperature rise over brine heater and T_2 and T_4 are brine heater entry temperature and feed temperature.

Although very high quality of desalinated water was obtained, process was still uneconomical because of high membrane costs and high area requirements. For making the process economical, two suggestions were made; one was reducing the cost of membrane, by finding substitutes for PTFE. And other was reaching high distillation heat transfer

coefficient by reducing the gas partial pressures within the membrane pores[22].

In 1986, Drioli et al. [23] conducted experiments on capillary microporous polypropylene and PVDF membranes, with NaCl and glucose aqueous solutions as feed. 100% rejection of glucose with fluxes of order 5 l/m^{2}h were obtained with PVDF capillary membranes at T_f =30º C and ΔT=10-15. Drioli observed a decrease in transmembrane flux with an increase in feed concentration and accounted it to decrease in vapor pressure difference due to high solute concentrations. Negative fluxes were observed at 5.3 M NaCl concentration when ΔT~5ºC.

In 1987, Gostoli et al. [24] compared the transmembrane fluxes in DCMD and AGMD systems. The terms 'simple capillary distillation' and 'cold wall distillation' were used for DCMD and AGMD respectively. The fluxes observed for DCMD with 0.5 M NaCl feed solution were much higher than AGMD (by a factor of nearly 4).Chmielewski et al. [25]also compared DCMD and AGMD in a multistage operation for deuterium and heavy oxygen enrichment. The fluxes obtained in DCMD were up to 10 times those of AGMD. Similar comparative study conducted by Alklaibi et al. [26] in 2007 proved that the thermal efficiency of AGMD is higher than that of DCMD by 6% because of the air gap. But permeate flux of DCMD is higher than AGMD upto 4.8 fold.

Ding et al. [27] in 2006 conducted a comparative study of DCMD, VMD and SGMD. It was found that VMD shows highest mass transfer coefficient but the lowest selectivity, DCMD gives highest selectivity and moderate mass transfer coefficient, and SGMD has moderate selectivity and lowest mass transfer coefficient.

Major use of DCMD has been in field of desalination and demineralization. Table 1, provides chronology of major researches in DCMD.

TABLE 1: Major researches in direct contact membrane distillation
PTFE: Polytetrafluoroethylene, PVDF: Polyvinylidene fluoride, PP: Polypropylene

S. No.	Year	Material	Pore Size, d_p (µm)	Thickness (µm)	Porosity (%)	Feed Solution	Ref.
1.	1967	PTFE	9	3200	42	Saline water	[20]
2.	1967	Paper, gum wood, glass fibers, cellophane, nylon &diatomaceous earth as membrane materials	---	---	---	Pure water	[21]
3.	1985	PTFE	---	---	---	Brine of 20,000 ppm NaCl	[22]
4.	1985	PTFE/PP	0.45	178	80	Saline water	[28]
5.	1987	PP	0.10	100	75	Distilled water	[29]
			0.20	140			
		PVDF	0.45	110			
6.	1987	PTFE	0.20	60	60	Salt water	[24]
7.	1987	PTFE	0.20	80	75	Solutions of H_2SO_4, NaOH, HCl, HNO_3, NaCl, acetic acid, formic acid, sucrose, gelatin orange juice, milk	[30]
8.	1987	PP	0.10	160	80	Various organic solutes	[31]
			0.10	300	80		
		PVDF	---	200	75		
			---	250	82		
9.	1988	PP	0.60	400	75	Gelatine containing waste water	[10]
10.	1991	PVDF	0.03	100	81	Waste water from taurine production	[32]
11.	1991	PP	0.45		70	Textile waste water	[33]
12.	1994	PVDF	0.22	140	75	Orange juice	[34]
13.	1995	PTFE	0.84	60	20	Mixture of isotopic compounds 180 ppm D & 1800 ppm O-18	[35]
			0.20	175	70		
14.	1999	PTFE	020	100	80	Low level radioactive waste	[36]
15.	1999	PP	0.20	800	73	Oil-water emulsion	[37]
16.	2000	PP	0.45	120	70	Apple juice	[38]

17.	2006	PVDF	0.45	---	80-85	Apple juice	[39]
18.	2006	PTFE	0.10	80	60	Ammoniated water	[27]
			0.20	60			
19.	2006	PP	0.20	450	73	Tap water	[40]
20.	2008	PP	0.22	800	73	Tap water with alkaline impurities	[41]
21.	2013	PVDF	0.80	80	80	Silica solution (saturation indices 1.5 to 2.2)	[42]
22.	2013	PVDF	0.22	125	70	2 M Na$_2$SO$_4$ and 4.5 M NaCl	[43]
23.	2014	PVDF	0.32	200	90.8	RO brine	[44]

3. HEAT AND MASS TRANSFER

Both heat and mass transfer takes place simultaneously through the membrane, the driving force for mass transfer being the difference in water vapor pressure difference across the membrane. The resistance imposed by the membrane can be described Knudsen diffusion model or Poiseuille flow model, the latter being dominant when membrane pore size is larger than the mean free path length.

As per Knudsen diffusion model:

$$N_K = 1.064 \frac{r\varepsilon}{\chi\delta}\left(\frac{M}{RT}\right)^{0.5}(P_1 - P_0) \tag{1}$$

where r is membrane pore radius, ε is porosity, χ is tortuosity, δ is membrane thickness, M is molecular weight, R is gas constant, T is temperature and $(P_1 - P_0)$ is the partial pressure difference.

Poiseuille flow model, which is based on flow of viscous fluids through a capillary, can be expressed by equation:

$$N_P = 0.125 \frac{r^2\varepsilon}{\chi\delta}\frac{MP_m}{\mu RT}(P_1 - P_0) \tag{2}$$

where μ is the gas viscosity.

Inside the membrane pores, diffusion takes place through a stationary film of air. The mass flux can be expressed by molecular diffusion model [45] :

$$N_D = \frac{1}{Y_{ln}}\frac{D\varepsilon}{\chi\delta}\frac{M}{RT}(P_1 - P_0) \tag{3}$$

where Y_{ln} is the log-mean mole fraction of air.

Based on the above three models, a flux relation of the following form is observed:

$$N = C(P_1 - P_0) \tag{4}$$

or

$$N = C\frac{dP}{dT}\Big|_{T_m}(T_1 - T_0) \tag{5}$$

dP / dT can be calculated from Clausius- Clapeyron equation:

$$\frac{dP}{dT}\Big|_{T_m} = \frac{P\lambda M}{RT^2}\Big|_{T_m} \tag{6}$$

P can be calculated from Antonie equation:

$$P = exp.\left(23.238 - \frac{3841}{T_m - 45}\right) \tag{7}$$

For more concentrated solutions:

$$N = C\frac{dP}{dT}[(T_1 - T_0) - \Delta T_{th}](1 - x_m) \tag{8}$$

where ΔT_{th} is the threshold temperature.

$$\Delta T_{th} = \frac{RT^2}{M\lambda}\frac{x_1 - x_0}{1 - x_m} \tag{9}$$

If $(T_1 - T_0) < \Delta T_{th}$ then, a negative flux is observes because of the reduction of vapor pressure caused by dissolved species in the feed.

Heat transfer in membrane distillation process takes place via two modes: (1) latent heat transfer accompanying vapor flux and (2) heat transfer by conduction through the membrane [29].

$$Q = Q_V + Q_C \tag{10}$$

where,

$$Q_V = N\lambda = C\frac{dP}{dT}\lambda(T_1 - T_0) \text{ and } Q_C = \frac{k_M}{\delta}(T_1 - T_0)$$

where λ is the latent heat of vaporization, T_1 and T_0 are the feed and permeate side temperature of the membrane surfaces respectively.

$$Q = \left(C\frac{dP}{dT}\lambda + \frac{k_M}{\delta}\right)(T_1 - T_0) \tag{11}$$
$$= H(T_1 - T_0)$$

where,

$$k_M = \varepsilon.k_g + (1 - \varepsilon).k_s \tag{12}$$

where k_M, k_g and k_s are conductivities of membrane, gas and polymer material respectively.

but,

$$Q = h_1(T_h - T_1) = h_0(T_0 - T_c) \tag{13}$$

where T_h and T_c are bulk temperatures of feed and permeate side respectively.

$$(T_1 - T_0) = \frac{(T_h - T_c)}{1 + \frac{H}{h_1} + \frac{H}{h_0}} \quad (14)$$

where

$$\tau = \frac{1}{1 + \frac{H}{h_1} + \frac{H}{h_0}} \quad (15)$$

With volatile component vaporizing at the membrane surface, there arises a temperature difference between the bulk and membrane surface. This phenomena causes a significant loss in the driving force. Equation (14), shows that the actual temperature difference across the membrane is less than the bulk temperature difference. This phenomena is called 'Temperature polarization'. τ in equation (15) is called the coefficient of temperature polarization[46].

4. DESIRED MEMBRANE PROPERTIES

Following properties are desired in membrane materials for least mass transfer resistance [4]:

1. Narrow pore size distribution

2. High porosity

3. Strong and stable hydrophobic character

4. Good thermal and chemical resistance

5. Low thermal conductivity

6. High mechanical properties

7. Less fouling

PTFE being an expensive membrane, was replaced by PVDF membranes[47], which were still researched for flux increment by CNT enhancement [48], copolymerization [49], . With the advancements in membrane technology, new membranes for better fluxes are produced. Bonyadi et al. [50] used dual layer hydrophilic–hydrophobic hollow fiber membranes for DCMD process and reported flux as high as 55 kg/(m^{2}h) at 90°C.

5. ADVANTAGES OF DCMD

1. Easy scaling up. Being a modular system, a unit can be extended by adding similar modules.

2. Simplicity of operation.

3. Possibility of high membrane surface/ volume ratio.

4. Possibility of treating solutions with thermosensitive compounds and high level of suspended solids at a temperature much lower than the boiling temperature.

5. 100% theoretical rejection of non-volatile solutes such as macromolecules, colloidal species, ions etc [51, 52].

6. The process can run under very high concentration conditions [32].

7. Low temperatures may be employed without requiring vacuum [10].

8. No entrainment, distillation product is free of particles or bacteria [10].

9. Pure water after concentration process can be reused.

10. Renewable sources of energy and waste heat can be used for heating purposes [4].

11. MD is a potential competent to reverse osmosis (RO), which is a common method for demineralization as it is not limited by osmotic pressure, can be used for high feed concentrations and is free from brine disposal problem.

6. CONCLUSIONS

With the growing need for cheaper methods for desalination and industrial approach towards process intensification, direct contact membrane distillation is an emerging technology. The renewed interest in this process is mainly because of the innovations in membrane synthesis andpolymer technology. Fluxes as high as 55 kg/(m^{2}h) have been reported in the literature. Production of cheaper membranes, elimination of concentration and temperature polarization and coupling with renewable heat sources are few research areas which would decide the future of DCMD technology.

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