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Grand Challenges in Membrane Distillation for Desalination and Water Recovery

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ABSTRACT

Seawater desalination and water recovery from wastewater are potential solutions to meet the ever-growing water demand. Membrane distillation (MD) is a next-generation membrane technology that can be harnessed for sustainable water production. The advantages of the MD process and the various operating configurations are discussed. The challenges in membrane development are highlighted, and the various state-of-the-art approaches for improving membrane performance, fouling resistance, anti-wetting character, and minimizing concentration/temperature polarisations are included. The process design is another crucial aspect of the successful implementation of the MD. Response surface methodology and Analysis of Variance (ANOVA) have been explored to identify the optimal operating conditions. Machine learning and computational fluid dynamics analysis (CFD) that have been used to predict the performance and influence of the process parameters are discussed. The energetics and economics of the MD process have also been discussed. The MD process could become sustainable if it utilizes renewable energy sources (solar, geothermal) for bulk heating of the feed. This article highlights the various challenges associated with MD technology and provides an overview of the strategies researched to overcome them.

Keywords: Membrane distillation, hydrophobicity, configuration, wetting, energy efficiency

1.0 INTRODUCTION

The availability and access to fresh water are a significant concern globally. Membrane-based separation processes have garnered attention for the desalination of brine/seawater and wastewater recovery. Membrane distillation (MD) is a membrane separation process that produces high-purity water from saline and brackish feed solutions. Even though the MD concept was proposed 55 years ago, the process is still under continuous research and reshaping for its commercial application [1].

In MD, the volatile components of a feed solution evaporate and cross a porous hydrophobic membrane. The membrane allows the transport of vapour molecules through the membrane, thus achieving separation—the process results in a highly pure distillate and concentrated feed solution where the non-volatile solutes are retained. MD is a non-isothermal process involving simultaneous mass and heat transfer. The driving force of MD is the vapour pressure difference between the feed and distillate solutions, which causes evaporation at the feed side of the membrane and condensation at the distillate side. The membrane is designed to keep the pores dry throughout the operation to ensure maximum vapour transport. Hence, inherently hydrophobic membranes or

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those modified with hydrophobic, low surface-energy materials are employed for the MD process [2].

MD is far superior as opposed to competing technologies due to the following reasons: (1) MD can achieve theoretically 100 % rejection of all non-volatile components, including inorganic ions, macromolecules etc. (2) can handle feeds at higher temperatures (while operating at lower temperatures as compared to thermal desalination processes like evaporation) (3) lower operating pressures as compared to Reverse Osmosis (RO) (Where applied pressure must be several times greater than osmotic pressure) (4) can handle high salinity feeds, (5) mechanical stability requirement is lower as compared to RO/Nanofiltration (NF) membranes, (6) able to utilize waste-heat and low-grade energy sources and (7) can be integrated with low-grade energy sources or renewable energy [3]. MD has been widely explored for desalination and brine treatment and is now being investigated for extraction of medicinal extracts and pharmaceutical products, treatment of textile dye effluents, the concentration of fruit juices and dairy streams, etc., [4].

Direct contact membrane distillation (DCMD), air gap membrane distillation (AGMD), vacuum membrane distillation (VMD) and sweeping gas membrane distillation (SGMD) are some of the standard configurations in which MD is operated. Though the most straightforward configuration, DCMD suffers from the highest heat loss due to thermal conduction as the membrane is in direct contact with the feed and permeate solutions. This heat loss can be overcome by opting for an AGMD configuration, in which a stagnant air gap is maintained and serves as an insulating layer for heat [5]. The permeate vapours can also be collected using a vacuum (VMD) or a non-condensable sweep gas (SGMD), and these require an additional step for condensing the transported vapours.

Despite its advantages, challenges such as temperature polarization, fouling, wetting, cost and energy efficiency, etc., restrict the application of MD at large-scale levels leading to reduced flux and high energy consumption [6].

This article provides an overview of the various facets of MD – (i) membrane design, (ii) process design and (iii) energetics and economics. The first part discusses different membrane fabrication techniques, strategies to improve membrane hydrophobicity and state-of-the-art advances in membranes with enhanced anti-wetting, self-cleaning and photothermal properties. The process design section discusses techniques for optimizing the operating parameters, novel membrane modules, and the pretreatment strategies employed. The last section discusses the thermal efficiency and costs of producing high-quality permeate. The advantage of using renewable energy sources for pre-treating the feed is also included in this section.

2.0 MEMBRANE DESIGN

The hydrophobic/omniphobic membrane is the heart of the MD process. An ideal membrane must possess (i) high vapour permeability, (ii) optimum-sized pores (0.1 to 1μm), (iii) high porosity (> 60-70 %) with narrow pore size distribution, (iv) excellent thermal stability (at least up to 90 °C), (v) high liquid entry pressure (>2.5 bar), (vi) high hydrophobic character (water contact angle (WCA) > 90°/ better > 150°) and (vii) low thermal conductivity (0.04-

0.06 W/m K) [7], [8], [9], [10].

Though ceramic membranes are thermochemically and mechanically stable, they are expensive, intrinsically hydrophilic, brittle and exhibit large conductive heat loss due to high thermal conductivities [11, 12]. Hence, polymeric membranes dominate usage in MD compared to ceramic membranes. Polymers such as polyvinylidene fluoride (PVDF), polytetrafluoroethylene (PTFE) and polypropylene (PP) are widely employed for the fabrication of membranes due to their inherent hydrophobicity, mechanical stability and excellent thermal/chemical stability. However, the most commonly available membranes are microfiltration membranes and are not exclusively designed for MD. Depending on their configuration, MD membranes can be flatsheet or hollow fibre membranes. Flat sheet membranes are easier to fabricate, clean, maintain, and inexpensive. These membranes have comparatively higher porosities and larger pore sizes and are more advantageous than other configurations in specific energy consumption [13]. Currently, commercial flat sheet membranes are widely used in MD applications. Hollow fibre and capillary configurations have the advantages of high packing density and compactness and are less prone to wetting than flat sheet membranes [14].

MD membranes are generally prepared by dry/wet phase inversion, leading to larger pores and lower porosity, resulting in lower flux and membrane wetting during MD [15]. The electrospinning technique is being explored to fabricate membranes with tuneable pores, higher porosities and lower tortuosity. The polymer solution is subjected to an electric field that draws out charged polymer strands collected on a collector. The diameter of the fibres can range up to hundreds of nanometers [16]. Various post-treatment techniques can improve these electrospun nanofibrous membranes' mechanical strength and pore-size distribution.

Irrespective of the fabrication technique, the membranes suffer from low hydrophobicity, which leads to a lower permeate flux and a faster wetting of the membrane's pores. Three strategies are generally employed to overcome these limitations:

(1) Blending hydrophobic polymers

Incorporating poly(vinylidene fluoride-co-hexafluoropropylene) (PVDF-co-HFP) or fluorinated surface modifying macromolecules improved liquid entry pressure (LEP) and membrane performance [17, 18].

(2) Surface grafting using alkylsilanes

The fluoroalkyl silanes such as 1H,1H,2H,2H-perfluorooctyltriethoxysilane, 1H,1H,2H,2H-perfluorodecyltriethoxysilane, octadecyl trichlorosilane or fluorine-free alkylsilanes such as polysiloxanes, dimethylsiloxane and hexadecyltrimethoxysilane are widely investigated to impart superhydrophobicity or oleophobic character to the membrane surface making them suitable for MD applications [19].

(3) Incorporation of nanoparticles

The addition of functionalized nanomaterials such as metal oxides, carbon nanotubes, metal-organic frameworks, inorganic materials, etc., within/on the polymer membrane improves the surface roughness of the membranes forming hierarchical microstructures that can act as air pockets and reduce membrane fouling and provide constant vapour permeate flux [20].

The membranes exposed to high concentration feed solutions containing complex contaminants or low surface tension compounds can aid the fouling or wetting of the membrane, which is detrimental to the long-term operation (permeate quality and quantity). Membrane fouling can be classified as organic fouling (due to humic acids, oils, extracellular polymeric substances, proteins and polysaccharides), inorganic fouling (due to deposition of mineral salts such as $CaSO_4$, $CaCO_3$, silica) or biofouling (due to microorganisms). Some specific membrane modifications to overcome these limitations are discussed as follows. An asymmetric super-wettable Janus skin and hydrophobic nanofibrous membranes exhibited an underwater superoleophobicity (underwater oil contact angle (CA) 164°), and in-air superhydrophobicity (WCA 166°) was robust and recovered water from oil-in-saline water emulsion (1000ppm oil) in DCMD mode [21]. Novel self-cleaning, anti-wetting membranes fabricated by electrospraying BiOBr/Ag photocatalyst onto electrospun membrane were found to reject 99.9 % of dyes, and these membranes could be regenerated by exposure to visible light [22]. Another approach is the fabrication of multi-layered membranes where each layer could be specifically tuned for the required application. Dual-layer membranes consisting of fluoroalkyl silane (FAS) grafted zinc oxide nanoparticles on FAS-modified electrospun PVDF-co-HFP exhibited lower sliding angles with excellent rejection towards salt and 0.1mM SDS in hypersaline solutions [23]. Triple-layered membranes comprising hydrophobic SiO_2-PVDF (top layer), polyacrylonitrile / MOF-808 (middle layer) and hydrophilic SiO_2-PVDF (bottom layer) exhibited a high contact angle of 140.8°. When applied for desalination by DCMD mode, these triple-layered membranes showed a flux of 4.40 LMH with permeate conductivity of 3.8 µS/cm when operated for 5 hours at ΔT of 30°C [24].

The addition of photothermal materials minimized the temperature polarisation effect (improves the thermal efficiency of the MD) due to the plasmon-induced hot electron transfer mechanism when irradiated with light. Silver nanoparticle incorporated PVDF electrospun membranes used in MD exhibited a photothermal effect and reached a temperature of 92.3° upon 60 seconds of Ultraviolet illumination. Thiol modification of these membranes exhibited very high hydrophobicity with a water contact angle of 148±2.1° and showed superior durability up to 60 hours [25]. Another contactless heating technique involves the induction-based heating of iron oxide-carbon nanotubes modified (dual-layer prepared by spray coating on PTFE membranes) membranes in the presence of an electromagnetic field [26].

3.0 PROCESS DESIGN

The MD process design parameters must be optimized for different configurations and applications to obtain maximum flux, high rejection, high thermal efficiency and long operational life. In one study that used a three-level RSM design to optimize feed temperature (50 to 70°C), feed flow rate (2 to 4.2 L/min), feed NaCl concentration (up to 20%), coolant flow rate (2 to 6.5 L/min) and air gap

(0.9 mm) in an AGMD used for the recovery of dye wastewater. The ANOVA analysis revealed that the feed temperature had the strongest influence on the permeate flux, and when the temperature was increased from 50 to 75°C, mass flux increased from 12 to 55.1 LMH and thermal efficiency improved from 11.5 to 52.7% was observed [27]. A multi-layer feed-forward artificial neural network was developed with input parameters such as feed temperature, feed velocity, and membrane type with over 18 data points to predict the permeate flux in the DCMD for treating palm oil mill effluent. Among 18 data points, the network was randomly trained, validated and tested based on a 70:15:15 (12 samples: 3 samples: 3 samples) ratio, and the optimization predicted that feed temperature and feed velocity significantly influence the permeate flux [28]. A CFD analysis of the MD process for further concentrating RO reject suggested an increase in membrane roughness, flow rate and temperature gradient improved the flux by reducing concentration and temperature polarization. However, excessive flow rate and temperature could also accelerate the chances of membrane scaling as it promotes the nucleation of salt crystals [29].

Recently, novel process configurations have been developed to improve vapour flux and energy efficiency. *Flashed feed-VMD* is one such advanced configuration in which feed is flashed through a small orifice, and as the hot feed is not in direct contact with the membrane, temperature polarisation effects are minimized. Compared to conventional VMD, flashed feed-VMD exhibits a 3.5-fold higher flux under similar operating conditions [30]. *Dead-end filtration* is another advanced configuration in which heating of feed solution is done using a localized heating element. Such heating provides uniform heat transfer and a stable temperature regime to the membrane surface. Intermittent flushing is usually provided to minimize the temperature polarisation effects and fouling. This configuration showed a 133% increase in vapour production and a GOR increase up to 132% compared to bulk heating [31].

Using actual wastewater directly in the MD process can be challenging due to the complex interactions between the foulants and the membrane, resulting in faster membrane fouling and, ultimately wetting. Therefore, choosing appropriate pretreatment technologies such as coagulation, ion exchange and pressure-driven membrane processes (microfiltration, nanofiltration, ultrafiltration) prior to MD operation is vital to control the extent of fouling. Periodic flushing with deionized water [32], flow and temperature reversal [33] and usage of antiscalants such as weak acids to inhibit the nucleation inhibitors of scaling are a few mitigation strategies for inorganic scaling [34]. Due to the absence of transmembrane pressure, MD experiences much less biofouling and can be overcome by adding disinfectants.

4.0 ENERGETICS AND ECONOMICS

Although the MD process requires lower temperature and pressure for operation than thermal-driven or pressure-driven (RO) desalination technologies, the energy efficiency is still low. The Gain Output Ratio (GOR) defines the heat utilization efficiency of a thermally driven process. It is defined as the ratio of latent heat of evaporation for water production to the input heat energy provided to that system, generally expressed in kg water produced/ kg steam supplied. The GOR of MD processes reported is generally below 1, making it attractive only when a large quantity of low-grade waste is available. The GOR can be improved by (i) improving the thermal efficiency of the membrane (ratio of the amount of heat transferred via vapour transport to the total amount of heat transferred via both vapour transport and thermal conduction), (ii) reducing trans membrane temperature difference, (iii) increasing the vapour pressure gradient (increase bulk temperature difference) and (iv) increasing number of stages with heat recovery [35].

The specific energy consumption (SEC) is another energy efficiency metric defined as the overall energy consumed to produce 1m^3 of pure water [36], and this value reported in the literature vary anywhere between 1 to 9000 kWh/m^3. Therefore strategies such as waste heat recovery and recycling of streams are necessary to make the MD process sustainable [37]. Exergy analysis is also carried out to understand the thermodynamic efficiencies of the MD process. The major share of the energy is used for bulk heating the feed solution, and utilizing renewable energy (solar, geothermal, etc.) for this can reduce. Waste heat from engines and generator exhaust, coolant heat, natural gas compressor stations, etc., can also be utilized as the driving energy for the MD process to produce water at a significantly lower cost [38]. In one instance, the integration with solar energy lowered the cost of producing water from 6.80 \$/m^3 to 1.6 \$/m^3.

Solar-powered membrane distillation (SPMD) uses solar collectors to meet the thermal as well as electrical requirements of the MD process [39]. Paraffin wax can be used to store solar thermal energy, which could be used at night time, and this increased the cumulative yield by 43.2% and improved the GOR by 34.4% [40]. However, solar radiation varies seasonally and during each day, leading to a permeate flux variation which cannot be predicted. Nano-photonics-enabled solar MD is more energy efficient due to the nanoparticle's light absorption and spatial distribution of the absorbed energy [41]. Integrating MD with other hybrid membrane processes, such as forward osmosis, electrodialysis, membrane crystallization, etc., increases water production and helps achieve zero liquid discharge [38].

5.0 CONCLUSION

(1) Membrane Distillation can be further explored as a sustainable technology for wastewater recovery and seawater desalination.

(2) Membrane development is carried out by blending with hydrophobic polymers and surface grafting using fluoro/alkylsilanes or hydrophobic nanoparticles.

(3) Superhydrophobic and omniphobic membranes are designed as a strategy to prevent membrane fouling (organic, inorganic or biofouling). Multi-layer electrospun membranes and hierarchical nanostructures are efficient in preventing membrane wetting, even in the presence of surfactants or low-surface tension liquids.

(4) Photothermal nanomaterials reduce temperature polarisation by heating the feed solution in contact with the membrane surface when exposed to the light of a suitable wavelength. This also makes the process more energy-efficient.

(5) Modified process design,

intermittent water flushing and pretreatment of feed solutions are promising strategies to obtain constant high-quality permeate flux.

(6) The GOR and SEC of the MD process can improve if low-grade waste heat or renewable energy sources or heat recovery (for successive stages) is carried out.

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