Recent Progress in Fabrication of Eco-membranes for Membrane Distillation: A Mini-Review

Mostafa M. Sayed
Chemical and Petrochemicals Engineering Department, Egypt-Japan University of Science and Technology

Hamouda M. Mousa
Mechanical Engineering Department, Faculty of Engineering, South Valley University

Ahmed H. El-Shazly
Chemical and Petrochemicals Engineering Department, Egypt-Japan University of Science and Technology

Abdelrahman Zkria
Department of Applied Science for Electronics and Materials, Kyushu University

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Recent Progress in Fabrication of Eco-membranes for Membrane Distillation: A Mini-Review

Mostafa M. Sayed[1, 2, 3*], Hamouda M. Mousa[4,5], Ahmed H. El-Shazly[1,6], Abdelrahman Zkria[3,7], Tsuyoshi Yoshitak[3] and Marwa ElKady[1,8]

[1]Chemical and Petrochemicals Engineering Department, Egypt-Japan University of Science and Technology, Alexandria, 21934, Egypt

[2]Materials Engineering and Design, Faculty of Energy Engineering, Aswan University, Aswan, 81528, Egypt,

[3]Department of Applied Science for Electronics and Materials, Kyushu University, Kasuga, Fukuoka, 816-8580, Japan

[4]Mechanical Engineering Department, Faculty of Engineering, South Valley University, Qena 83523, Egypt

[5]Faculty of Technological Industry and Energy, Thebes Technological University, Thebes, 85863, Luxor, Egypt

[6]Chemical Engineering Department, Faculty of Engineering, Alexandria University, Alexandria, 21544, Egypt

[7]Department of Physics, Faculty of Science, Aswan University, Aswan, 81528, Egypt

[8]Fabrication Technology Department, Advanced Technology and New Materials Research Institute (ATNMRI), City of Scientific Research and Technology Applications, Alexandria, 21934, Egypt

Corresponding author e-mail: mostafa_sayed91@energy.aswu.edu.eg

Abstract: *Membrane distillation (MD) is an emerging water purification and desalination technology that offers several advantages over conventional thermal and membrane-based processes. As environmental concerns grow, there is a significant interest in developing eco-friendly membranes for MD applications. This mini-review investigates recent progress in the fabrication of eco-membranes for membrane distillation. Key advancements include using natural polymers like chitosan and cellulose, biodegradable synthetic polymers, and inorganic materials to create more sustainable membrane options. Fabrication techniques such as electrospinning and phase inversion have produced high-performance eco-membranes with enhanced permeability, selectivity, and stability. While challenges remain in scaling up production and optimizing long-term performance, the potential of eco-membranes to significantly reduce MD processes' environmental impact is a promising development that cannot be overlooked. This review provides an overview of eco-membrane materials, fabrication methods, performance characteristics, and future research directions to guide further development in this rapidly evolving field.*

Keywords: Membrane distillation; Eco-friendly membrane; Sustainable; Biodegradable.

1. INTRODUCTION

The water-energy-environment (WEE) nexus framework addresses the complex challenges around essential resources in the 21st century. Rapid population growth, urbanization, and climate change have raised concerns about water scarcity, energy depletion, and environmental damage [1]. By 2050, half of the global urban population is predicted to experience water scarcity, while energy demand is expected to increase by 50%, putting additional pressure on water resources and contributing to greenhouse gas emissions [2]. The WEE nexus highlights the importance of integrating approaches to manage interconnected resources, recognizing that actions in one sector can significantly impact others. As freshwater resources face growing strain, alternative water sources like desalination and reclamation have become essential for urban water supply systems. Conventional desalination technologies, such as reverse osmosis (RO), multi-stage flash (MSF), and multi-effect distillation (MED), dominate the current market but face challenges related to high energy consumption, operational complexity and environmental impacts [3–5]. In response, sustainable desalination technologies are gaining attention for their potential to maximize freshwater output while minimizing energy use, costs, and environmental footprint.

Membrane distillation (MD) is a thermally-driven separation process that has gained significant attention in recent years as a promising technology for water purification and desalination [6]. In MD, a hydrophobic microporous membrane acts as a barrier between a hot feed solution and a cold permeate stream, allowing only water vapor to pass through while rejecting non-volatile contaminants [7]. The driving force for mass transfer is the vapor pressure difference created by the temperature gradient across the membrane. MD processes can be classified based on the structural arrangement of the membrane unit and the specific mechanisms used for heat and mass transfer. MD processes have four main configurations, all differing in design on the permeate side: direct contact membrane distillation (DCMD), air gap membrane distillation (AGMD), sweeping gas membrane distillation (SGMD), and vacuum membrane distillation (VMD) [8], [9], [10], [11]. **Fig. 1** displays the four main configurations of the MD process. MD presents several advantages over conventional thermal distillation and pressure-driven membrane processes such as reverse osmosis. These benefits include the capability to treat highly concentrated brines, operate at lower pressures and temperatures, and harness low-grade waste heat or renewable energy sources. Additionally, MD achieves high rejection rates of non-volatile contaminants, making it a highly effective separation technology.

Despite MD's benefits, challenges such as wetting, fouling, low permeate flux, efficiency concerns, the environmental impact of membrane materials, and the limited availability of specialized membranes remain. Overcoming these requires advancements in membrane materials and module designs for broader adoption of MD technology [12]. Petroleum-based polymer membranes are extensively used in MD process due to their high porosity, low thermal conductivity, stability, cost-effectiveness, and hydrophobic properties [13]. Among the most widely studied polymeric membranes for MD applications are polyvinylidene fluoride (PVDF), polydimethylsiloxane (PDMS), polytetrafluoroethylene (PTFE), and polypropylene (PP), with PVDF being the preferred choice due to its advantageous characteristics [14–17]. Despite these advantages, the low biodegradability of these polymer materials presents a significant environmental concern. Their resistance to microbial degradation, attributed to their high thermal and chemical stability, impedes the breakdown of their carbon linkages once disposed of in natural environments [18–20].

The research community in membrane technology has become increasingly aware of the environmental impact of petroleum-based polymers [18,20]. In response, there is a growing push to develop sustainable products using bio-based materials, with biopolymers expected to replace petroleum-based polymers gradually [21]. Biodegradable polymers such as cellulose acetate (CA), polyvinyl alcohol (PVA), polylactic acid (PLA), chitosan, and polycaprolactone (PCL) have emerged as promising alternatives due to their biocompatibility and eco-friendliness [21–24]. Biopolymers have found applications in various fields, including wastewater treatment, where they have been used in processes like microfiltration and pervaporation [25–27]. While biopolymers can be used in pressure-driven membrane processes, they have limitations due to their low mechanical properties and wettability. However, these properties, particularly low wettability, can be advantageous in non-pressure-driven applications such as MD. Despite their potential, biopolymeric-based membranes for wettability control still need to be explored in the literature, especially for MD applications [24]. This presents an opportunity for further research and development in this area.

This mini-review aims to address these challenges by focusing on the development of eco-friendly membranes for MD applications. The main objectives of this paper are to explore the use of natural and biodegradable synthetic polymers, as well as advanced fabrication techniques, to create sustainable membrane options. Key advancements include using natural polymers like chitosan and cellulose, biodegradable synthetic polymers such as polylactic acid (PLA) and polycaprolactone (PCL), and innovative fabrication methods like electrospinning and phase inversion. These eco-membranes are designed to minimize environmental impact while maintaining or enhancing performance characteristics, such as permeability, selectivity, and stability. By addressing these aspects, this review seeks to guide the development and adoption of eco-membranes for more sustainable MD processes.

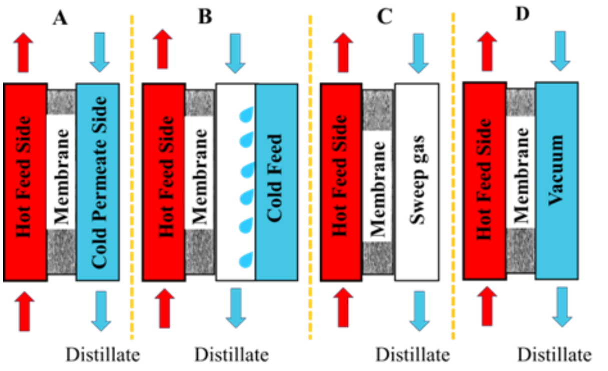

Fig. 1. Schematic illustration of the four main MD configurations: (A) DCMD, (B) AGMD, (C) SGMD, and (D) VMD.

2. ECO-MEMBRANE MATERIALS

In recent years, there has been an increasing focus on developing sustainable fabrication processes in MD. This involves exploring environmentally friendly alternatives to traditional toxic solvents [18,28] and incorporating biopolymers to reduce the environmental impact associated with petroleum-based polymers [19,20]. Eco-membranes for MD applications are designed to minimize environmental impact while maintaining or even enhancing performance characteristics. Critical features of eco-membranes include biodegradability, use of renewable resources, and reduced energy consumption during production. Recent research has identified several promising eco-membrane materials.

2.1 Natural Polymers

Research on chitosan (CS) and its derivatives has revealed promising results in MD applications. A study by Li et al. developed a triple-layer composite membrane (TL-M) with a PVDF-PTFE hydrophobic layer, a PET support layer, and a chitosan-polyethylene oxide (CS-PEO) hydrophilic layer [29]. The TL-M outperformed a double-layer membrane (DL-M) in DCMD with a 26.7% higher average flux (19 $kg/m^2.h$ vs 15 $kg/m^2.h$) and demonstrated better stability and slightly higher salt rejection (99.92% vs 99.88%). Additionally, in another study by Liu et al., a hydrogel-like coating of chitosan and sodium alginate (CS/SAH) was found to enhance membrane performance by preventing fouling, enhancing vapor flux from 23.6 to 32.3 $L/m^2.h$, and controlling pore size from 0.22 µm to 0.08 µm while maintaining high salt rejection [30]. These findings underscore the potential of chitosan-based membranes for water treatment applications. Furthermore, Cellulose materials have been investigated for eco-membrane production [31]. Ritika Joshi et al. developed a superhydrophobic cellulosic membrane for MD utilizing microfibrillated cellulose (MFC), achieving a contact angle of $>$ 152° [32]. It displayed exceptional wetting resistance and robust mechanical properties, with a permeate flux of 14.6 $kg/m^2.h$ and over 99.9% salt rejection during testing (see **Fig. 2 A and B**). This underscores its potential as an eco-friendly substitute for conventional petroleum-based membranes.

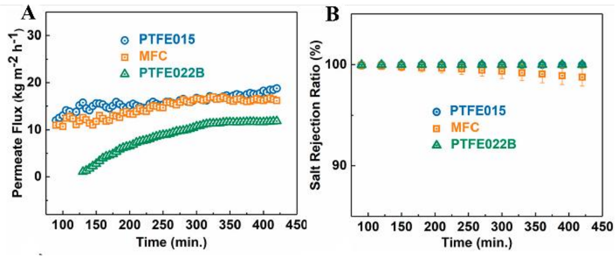

Fig. 2. Composite cellulosic (MCF) and commercial PTFE membranes DCMD performance. A) Permeate mass flux and B) Salt rejection ratio. (DCMD conditions: a feed temperature of 50°C, a permeate temperature of 20°C, and 8 g/L NaCl solution as the feed) [32].

2.2 Biodegradable polymers

Polylactic acid (PLA) is a biodegradable polyester derived from renewable resources. A research study has developed sustainable and efficient membranes for MD using PLA nanofibrous membranes [19]. The PLA membrane was created with nanoscale fibers measuring 700 nm, heat treatment, a hierarchical structure, and PVDF support layers. These advancements resulted in a stable flux of approximately 2 kg/m^2.h and high salt rejection of over 99%, offering a sustainable alternative for desalination. Moreover, Polycaprolactone (PCL) is another biodegradable polymer that has been investigated for MD membranes. In a previous study, we developed a new dual-layer membrane for the DCMD system [20]. This membrane comprises a hydrophobic PCL top layer and a hydrophilic cellulose acetate (CA) bottom layer. Coating silica nanoparticles (SiO_2) into the PCL layer increases surface roughness and hydrophobicity, resulting in a superhydrophobic surface (152.4°) (see **Fig. 3**). The CA layer aids in water vapor absorption, minimizing pore wetting and enhancing membrane efficiency. This eco-membrane exhibits potential for sustainable water desalination, demonstrating a durable permeate flux of 15.6 kg/m^2.h and a salt rejection rate of 99.97%.

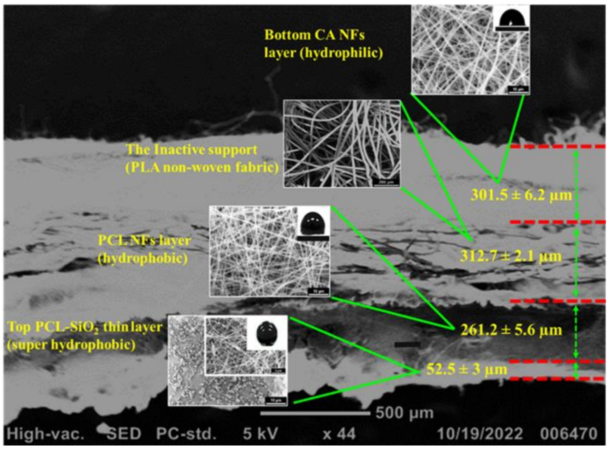

Fig. 3. A cross-sectional SEM image of the PCL-SiO2/CA eco-membrane layers [20].

2.3 Inorganic materials

Ceramic membranes offer excellent thermal and chemical stability for MD applications. In a recent study, Javad et al. developed an environmentally friendly superhydrophobic ceramic membrane for water desalination [33]. The membrane, which utilized affordable materials such as kaolin and calcium carbonate, was treated with hexadecyltrimethoxysilane (HDTMS) to improve its hydrophobic properties, resulting in a contact angle of 160°. This modified membrane exhibited impressive salt rejection rates of 99.62% for synthetic NaCl solutions and 99.81% for seawater. Additionally, it maintained an average permeate flux of 3.15 kg/m²·h for NaCl and 2.37 kg/m²·h for seawater over a 20 h test period.

2.4 Composite membranes

Combining multiple materials can create eco-membranes with enhanced properties. For example, as discussed previously, a research study developed sustainable PLA nanofibrous membranes for MD with a stable flux and salt rejection, making it a viable alternative for desalination [19]. The PLA membrane features nanoscale fibers, heat treatment, a hierarchical structure, and PVDF support layers. Additionally, a dual-layer membrane combining a hydrophobic PCL top layer and a hydrophilic CA bottom layer, enhanced with SiO_2 nanoparticles, achieved a superhydrophobic surface and high efficiency in water desalination [20]. These eco-membrane materials offer promising alternatives to conventional petroleum-based polymers, potentially reducing the environmental impact of MD processes while maintaining high-performance standards.

3. FABRICATION TECHNIQUES

Advancements in fabrication techniques have played a crucial role in developing high-performance eco-membranes for MD applications. A notable method is phase inversion, which remains a versatile and scalable technique for membrane fabrication. Recent innovations include the use of green solvents like dibasic esters (DBEs) such as dimethyl succinate (DMS), dimethyl glutarate (DMG), and dimethyl adipate (DMA) for manufacturing polyvinylidene fluoride (PVDF) membranes [34]. These environmentally friendly solvents replace traditional toxic ones, facilitating the creation of PVDF membranes with robust mechanical strength and high performance in wastewater treatment. DBEs modify the phase diagram's monotectic point, resulting in membranes with a desirable bicontinuous structure via liquid-liquid phase inversion. These membranes exhibit a superior flux of 42.40 kg/m^2.h and rejection rates (>99%) in purifying synthetic nuclear wastewater, suggesting a promising path for advancing high-performance, eco-friendly industrial applications (see **Fig. 4**). A recent study found that adding cellulose nanofibers (CNF) to PVDF membranes improved their performance [35]. With higher CNF concentrations, the hybrid membranes became more porous and hydrophilic, leading to better stability, permeate flux, and solute retention. This enhancement indicates that CNF holds promise for sustainable and efficient improvement of PVDF membranes in water treatment.

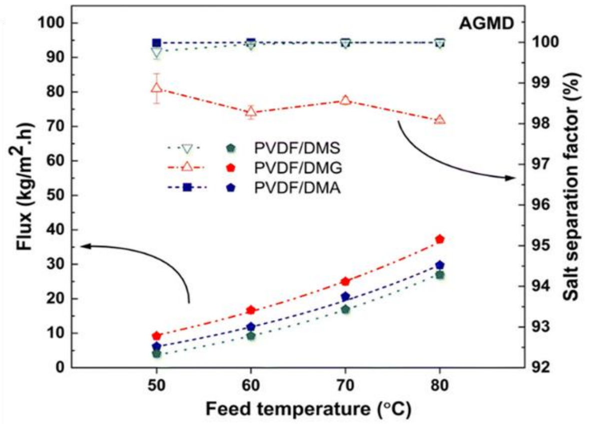

Fig. 4. AGMD performance for the prepared membranes at a feed solution concentration of 35 g/L NaCl [34].

Electrospinning is considered one of the most effective methods for producing hydrophobic polymeric membranes. This technique allows for precise control over the morphology and size of the fibers, making it possible to tailor the membranes to meet specific application requirements by adjusting the processing parameters. Electrospinning has become increasingly popular for creating nanofiber membranes with high porosity and surface area. **Fig. 5** shows a schematic representation of the electrospinning technique. Research conducted by Sayed et al. focused on developing and evaluating environmentally friendly, biodegradable polycaprolactone (PCL) membranes for water distillation [36]. The study found that the 12 wt% PCL membrane exhibited exceptional performance in DCM, with a flux of 41.9 kg/m²·h and a salt rejection rate exceeding 94%. These results indicate that the 12 wt% PCL membrane is highly suitable for sustainable water desalination. Guo et al. developed a novel biaxial electrospinning method to create a green superhydrophobic membrane for membrane distillation using PVDF-co-HFP polymer [37]. The membrane achieved superhydrophobicity (153.8°), improved tensile strength, reduced thermal conductivity, and demonstrated long-term test performance (rejection rate of 99.8% and flux of 29.6 LHM). The biaxial electrospinning approach allows for one-step fabrication and enhances surface properties, effectively addressing MD challenges and promoting environmentally friendly processes.

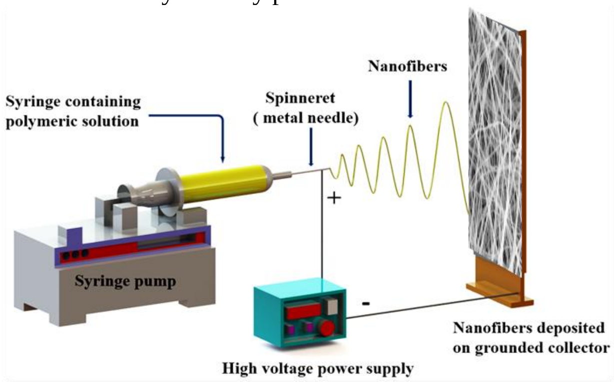

Fig. 5. Schematic representation of the electrospinning technique.

4. PERFORMANCE EVALUATION

It is crucial to assess eco-membrane performance to determine its suitability for MD applications. In order to compete with conventional membranes, eco-membranes must achieve adequate water vapor flux and high salt rejection. Recent studies have produced promising results, as summarized in **Table 1**. Most eco-membranes reported in recent literature have exhibited salt rejection rates exceeding 99%, which is on par with the performance of conventional membranes. These values are comparable to or even superior to those of PVDF membranes.

Table 1. Provides a comparison of key performance metrics for selected eco-membranes and PVDF membranes.

Membrane	Flux (kg/m².h)	Salt Rejection (%)	Reference
PCL-SiO_2/CA	15.6	99.97	[20]
Three layers of PLA/PVDF	2.5	99	[19]
PVDF-PTFE/PET/(CS-PEO)	19	99.92	[29]
PTFE@CS/SAH	32	99.99	[30]
Alumina layer on the composite mullite $CaCO_3$ synthesis membrane	2.2-3.6	99.96	[33]
PVDF Membranes	5-4.7-2.3	>99	[38,39]

This comparison demonstrates that eco-membranes can achieve comparable or superior flux and selectivity to commercial membranes, though mechanical properties may require further improvement.

5. ENVIRONMENTAL CONSIDERATIONS

The development of eco-membranes for MD aims to reduce the environmental impact of membrane production and disposal while maintaining membrane performance. A study on PLA membrane degradation under alkaline hydrolysis revealed significant weight loss over time[19]. Fiber morphology changed drastically under harsh conditions, becoming fragmented and wrinkled, unlike the cylindrical shape of pure PLA. Increased NaOH concentration reduced fiber diameter until destruction. Despite petroleum-based polymers in the support layer, about 90% of the membrane degraded due to biopolymeric materials. PLA biopolymers, derived from starch, show promise for replacing petroleum-based polymers, but further research is needed to improve biodegradability in MD membranes.

6. CHALLENGES AND FUTURE DIRECTIONS

Despite significant advancements, eco-membranes for MD processes face several challenges, including insufficient long-term durability, difficulties in scaling laboratory techniques to industrial production, and the need for further performance optimization in flux, selectivity, and fouling resistance. The lack of standardized testing methods also complicates performance comparisons across studies. Future research should focus on advanced materials, scalable green manufacturing processes, bio-inspired surface modifications, and integrating eco-membranes with sustainable technologies like solar thermal systems. Comprehensive life cycle and techno-economic analyses are essential to understand their environmental and economic impacts fully. As research progresses, eco-membranes are expected to play a crucial role in sustainable MD processes, enhancing environmentally friendly water treatment and desalination solutions.

7. CONCLUSION AND RECOMMENDATIONS

This review has examined recent progress in fabricating eco-membranes for membrane distillation (MD) applications. Significant advancements have been made in developing membrane materials from natural polymers, biodegradable synthetics, and inorganic compounds, as well as improving fabrication techniques to enhance membrane performance. Key findings from recent studies on eco-membranes for MD processes indicate that these innovative membranes can achieve flux and selectivity on par with or exceeding conventional membranes. However, further enhancements in eco-membrane properties are necessary. Novel fabrication techniques, such as green electrospinning and phase inversion using eco-friendly solvents, have shown promise in creating high-performance, sustainable membranes. Life cycle assessments highlight the potential environmental benefits of eco-membranes, though more extensive research is required for a comprehensive understanding. Despite challenges in scaling production, optimizing long-term performance, and standardizing evaluation methods, eco-membranes present substantial potential for mitigating the environmental impact of MD processes.

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