



# Effects of hydrophilic silica nanoparticles and backing material in improving the structure and performance of VMD PVDF membranes



Mohammadali Baghbanzadeh, Dipak Rana, Christopher Q. Lan\*, Takeshi Matsuura

Department of Chemical and Biological Engineering, University of Ottawa, 161 Louis Pasteur Private, Ottawa, Ontario K1N 6N5, Canada

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## ABSTRACT

Effects of hydrophilic silica nanoparticles and Non-Woven Fabric (NWF) backing material on the properties and performance of the Polyvinylidene Fluoride (PVDF) membrane have been studied in this work. The nanocomposite membranes were prepared by the phase inversion method and characterized in terms of surface pore size, cross-sectional morphology, thickness, porosity, surface roughness and hydrophobicity. The fabricated membranes were subjected to Vacuum Membrane Distillation (VMD) experiments and their performance was evaluated through measuring the pure water flux and salt rejection. According to the results, effects of the hydrophilic nano-additives and NWF polyester enabled much higher VMD flux than the neat PVDF membrane when an appropriate amount of the nano-filler was added, possibly due to increase in the surface pore size and the reduction of the sponge-like layer thickness. The supported nanocomposite membranes possessed appropriate Liquid Entry Pressure (LEP<sub>w</sub>) and mechanical strength, which make the membrane applicable in VMD process. When 7.0 wt.% of the silica nanoparticles was incorporated in a NWF supported membrane, the pure water flux became 12749.6 g/m<sup>2</sup> h at feed temperature of 27.5 °C and permeate side pressure of 1.2 kPa, representing a 2456% increase from the neat PVDF membrane. Almost complete NaCl rejection was also achieved when tested with 35 g/L NaCl aqueous solution.

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## 1. Introduction

Saline water desalination has attracted extensive attentions during the last few decades. Water shortage due to the population and agricultural growth, and global warming in different areas of the world has forced people to find an alternative approach to alleviate the lack of fresh water. To this end, the desalination technology has demonstrated to be one of the most effective options [1–6]. Many processes could be used to remove the salty materials from the saline water, e.g., thermal processes such as Multiple Effect Distillation (MED) and Multi-stage Flash Distillation (MSF), Mechanical Vapor Compression (VC), and membrane-based

processes. Currently desalination market is mainly dominated by the membrane-based processes [7–9], which include Reverse Osmosis (RO), Forward Osmosis (FO), Ultrafiltration (UF), Microfiltration (MF), Nanofiltration (NF), Membrane Distillation (MD) and the combination of two or more of them, due to the relatively low cost and energy consumption. While RO is presently placed on the top of these membrane processes [10–13], it has its own limitations such as strong tendency to fouling and scaling, limited product water recovery, huge electrical energy consumption required to work at extremely high operating pressures [14–17], which opens the way for the other membrane processes to enter into the competition in the desalination market. Particularly, MD has been demonstrated to be potentially applicable [18–22] provided a high performance durable membrane is available with high flux at relatively low operating temperatures to increase membrane life time and enable the utilization of low-quality thermal energy.

It is known that the membrane morphology and surface chemistry play a major role in determining the performance of the membranes, and they can be manipulated via different strategies in the manufacturing process, e.g., addition of nano-additives. The incorporation of nano-additives into neat membrane has been

*Abbreviations:* CNIM, Carbon Nanotube Immobilized Membrane; DCMD, Direct Contact Membrane Distillation; DMAC, N,N-Dimethylacetamide; EDS, Electron Dispersion Spectroscopy; FO, Forward Osmosis; LEP, Liquid Entry Pressure; MD, Membrane Distillation; MED, Multiple Effect Distillation; MF, Microfiltration; MSF, Multi-stage Flash Distillation; NF, Nanofiltration; NP, Nanoparticle; NWF, Non-Woven Fabric; PP, Polypropylene; PVDF, Polyvinylidene Fluoride; RO, Reverse Osmosis; SEM, Scanning Electron Microscopy; UF, Ultrafiltration; VC, Mechanical Vapor Compression; VMD, Vacuum Membrane Distillation.

\* Corresponding author.

E-mail address: [Christopher.Lan@uottawa.ca](mailto:Christopher.Lan@uottawa.ca) (C.Q. Lan).

shown to be an appropriate approach in this direction as shown by the enhancement of the performance of the UF, MF, NF, RO, and FO membranes by numerous researchers [23]. However, the use of nanocomposite membranes in MD has been limited to only a few studies.

Gethard et al. [20] used Carbon Nanotube Immobilized Membrane (CNIM) in Polypropylene (PP) in a Sweep Gas Membrane Distillation (SGMD) configuration and reported a higher flux than the neat PP membrane. In a similar work, Bhadra et al. [24] also observed an improvement in the PP membrane permeability through the addition of carboxylated carbon nanotubes. The functionalized carbon nanotubes, especially the single walled ones, are very expensive to be used in commercial membrane production. In another attempt, Hou et al. [25,26] showed hydrophobic modified  $\text{CaCO}_3$  nanoparticles to be effective in increasing the permeability of Polyvinylidene Fluoride (PVDF) membrane in Direct Contact Membrane Distillation (DCMD). In our recent works, it was shown that the addition of both superhydrophobic silica nanoparticles [27] and hydrophilic CuO nanoparticles [15] could significantly increase the flux of Vacuum Membrane Distillation (VMD), but due to different mechanisms. Nevertheless, the fluxes obtained in these works, despite of the significant improvements in comparison to neat membranes, were still quite moderate and efforts dedicated to exploring more effective nano-additives are warranted.

The objective of this study is to investigate the effects of hydrophilic amino modified  $\text{SiO}_2$  nanoparticles on the VMD performance of PVDF membranes. To the best of our knowledge, this work is the first attempt demonstrating that enclosure of hydrophilic silica nanoparticles could drastically increase the VMD flux of MD membranes without compromising hydrophobicity or salt rejection.

This work was motivated by our earlier works, investigating the effects of hydrophilic CuO nanoparticles to lower the mass transfer resistance by increasing the fraction of the finger-like layer structure within the membrane layer and the compatibility of the silica nanoparticles with the dope solution, besides their mechanical strength and non-toxic nature [28]. The effects of backing materials on both the flux and stability of the membranes were also studied.

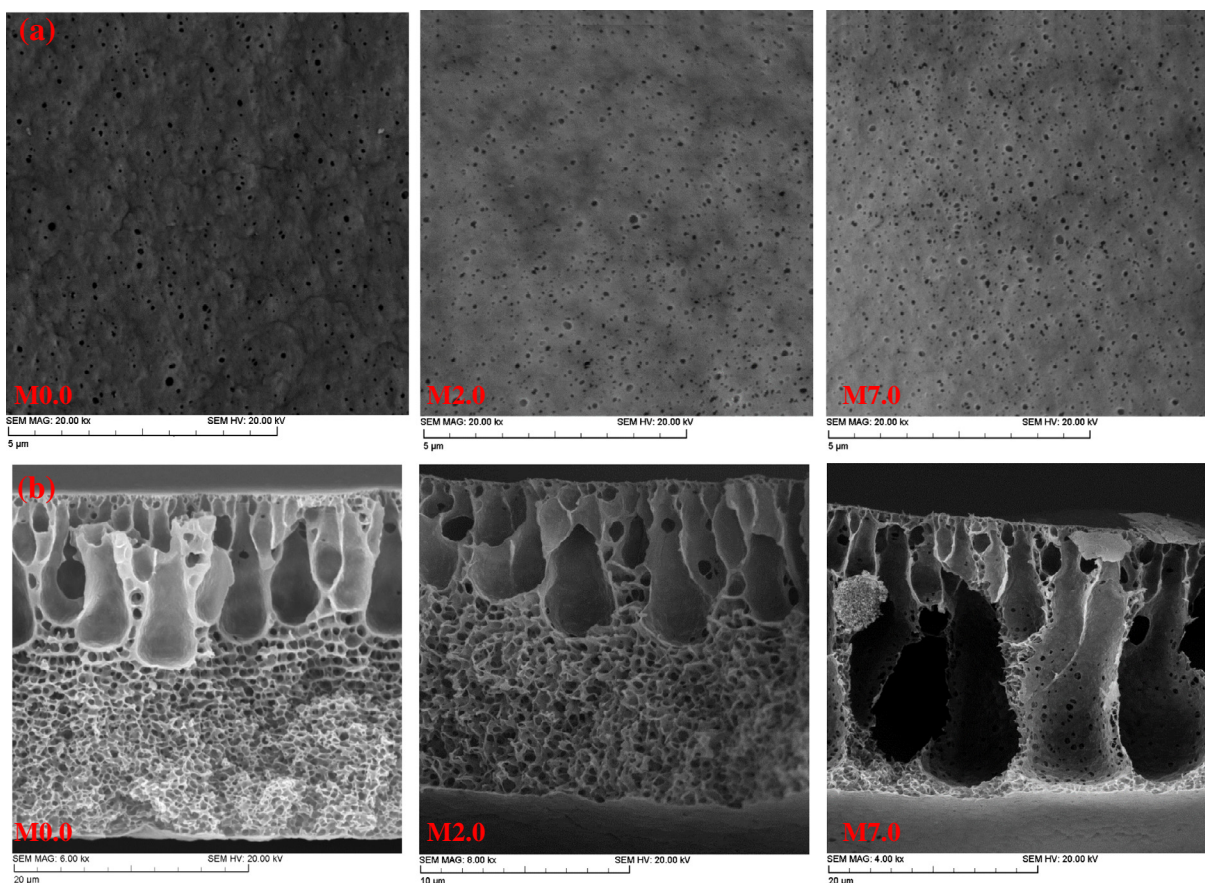
## 2. Experimental methods

### 2.1. Materials

Two kinds of PVDF, Kynar<sup>®</sup> 740, (Pellet, melt viscosity:  $1850 \pm 250$  Pa s; melting temperature:  $160.1^\circ\text{C}$ ) and Kynar<sup>®</sup> HSV900 (Powder, melt viscosity:  $4930$  Pa s; melting temperature:  $165.1^\circ\text{C}$ ) generously supplied by Arkema Inc. (Philadelphia, PA) were blended. DMAc and silver nitrate with the purity of 99.9% and 99+%, respectively, were supplied by Sigma–Aldrich. Hydrophilic amino modified  $\text{SiO}_2$  nanoparticles with the size of 10–20 nm and 99.8% purity were purchased from Skyspring Nanomaterials Inc. NWF polyester, Hollytex<sup>®</sup> 3396 was supplied by Kavon Filter Products Co. (Farmingdale, NJ) and used as the backing material. Sodium chloride and n-butanol were provided by Fisher Scientific and used as received.

### 2.2. Membrane fabrication

To make the membranes, phase inversion method was used. First, 1.25 wt.% water and 15 wt.% PVDF (HSV900:740 = 2:3



**Fig. 1.** (a) Top surface and (b) cross-sectional SEM images of the  $\text{SiO}_2$ /PVDF membranes (M0.0, M2.0, and M7.0), the surface pores are specified by the black spots in top surface.

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