



Nanodiamond immobilized membranes for enhanced desalination via membrane distillation



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HIGHLIGHTS

- Detonation nanodiamond as novel nanomaterial for immobilizing hydrophobic membrane
- Detonation nanodiamond membrane (DNDIM) showed vast potential for desalination.
- DNDIM showed more significant flux enhancement than conventional hydrophobic membrane.
- DNDIM membrane was highly stable over a long operational period.

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ABSTRACT

Recent years have witnessed the emergence of detonation nanodiamonds (DNDs) as a new member of the nanocarbon family. This paper reports the immobilization of DNDs within a hydrophobic membrane for desalination applications. A dispersion of DNDs in polyvinylidene fluoride polymer was injected through a hydrophobic membrane, which incorporated the DNDs within the membrane. Embedding only a minimal quantity of (approximately 2%) the nanocarbon favorably altered the water vapor–membrane interactions to enhance water vapor permeability while preventing liquid penetration into the membrane pores thereby enhancing water vapor flux. Overall, desalination was consistently better with DND immobilized membrane than with conventional polypropylene, with flux enhancement as high as 118% in the sweep gas membrane distillation mode.

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1. Introduction

In the last two decades, detonation nanodiamonds (DNDs) which are typically produced by detonation method have attracted much attention. DNDs are carbon nanoparticles [1] with a truncated octahedral architecture that is typically about 2 to 8 nm in diameter. The DNDs have tetrahedral network structures and comprise a diamond core (sp^3), a middle core ($sp^2 + x$) and a graphitized outer core (sp^2) that is often partially oxidized [1,2]. They also have large grain boundary density, and low to negative electron affinity which has made them suitable for electronic applications [3]. They exhibit some interesting characteristics such as chemical stability, small size, inertness, large surface area, and high adsorption capacity [1]. Additionally, one of the distinct features of DNDs compared to carbon nanotubes and other carbon nanoparticles is the presence of a large number of different functional groups on the surface [4]. Consequently, they are being used in a variety

of applications including coatings, polymer composites, lubricants, sensors, imaging, drug delivery, solar cells and electronics [4–14].

It has been previously demonstrated by our group that by immobilizing carbon nanotube in different types of membranes alters the solute–membrane interactions, which is one of the major physicochemical factors affecting the permeability of a membrane [15]. Carbon Nanotube Immobilized Membrane (CNIM) has been used in extraction, pervaporation, and preconcentration as well as desalination [15–19]. In line with this research, an important consideration is whether the DNDs could be introduced as a new class of nanomaterial for enhanced separation. The outer core of DNDs is somewhat similar to that of nanotubes and can serve as sorption sites that could potentially be useful in enhancing permeation flux. For example, the immobilization of DND within conventional hydrophobic membrane for membrane distillation (MD) could lead to specific vapor–membrane interactions thereby enhancing water vapor permeation. The objective of this research is twofold. Firstly to immobilize the DNDs within the hydrophobic membranes and then study the modified membrane for sweep gas membrane distillation (SGMD) applications.

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2. Experimental

The DND immobilized membrane (referred to as DNDIM) was prepared using Celgard type X30-240 (Celgard, LLC, and Charlotte, NC, USA) hollow fiber as the starting material. For the preparation of DNDIM, each of 10 mg of DND was dispersed in a solution containing 0.1 mg of polyvinylidene fluoride (PVDF) in 15 ml of acetone by sonicating for 3 h. The PVDF-nanotube dispersion was forced under controlled vacuum into the bore of the polypropylene hollow fiber membrane. The PVDF polymer served as glue that held the DNDs in place within the membrane. The membrane was flushed with acetone to remove excess PVDF. The original polypropylene membrane was also immobilized with sonicated PVDF solution in acetone without the DNDs, and this served as the control. The morphologies of unmodified polypropylene and DNDIM membrane were studied using scanning electron microscopy (SEM, Model LEO 1530), and thermal gravimetric analysis (TGA) was performed using a Perkin Elmer Pyris 7 TGA instrument to study the thermal stability of the membranes.

The membrane modules were constructed in a shell and tube format using 1/4 in. polypropylene tubing. Ten, 16.6 cm long hollow fiber strands were used in the module. Each module contained approximately 12.50 cm² of effective membrane contact area (based on internal surface). The ends were then sealed with epoxy to prevent leakage into the shell side. Vacuum was applied to one drain port to draw dry air through the other port, which created a higher pressure differential and provided a sweep air.

The schematic of the experimental system is shown in Fig. 1. This was the equivalent of sweep gas membrane distillation (SGMD). The feed used in these experiments ranged from 3000–34,000 ppm NaCl solutions (Sigma Aldrich). This was pumped through the module using a Master flex 7519-10 peristaltic pump. The preheated hot feed solution traveled through a heat exchanger which was used to maintain the desired temperature throughout the experiment. Dry air was passed into the shell side and the permeate was collected in a trap. Air flow was maintained at 1 L min⁻¹. The ionic strength of the original solution, the permeate and the concentrate were measured using a Jenway Electrode Conductivity Meter 4310. Each experiment was repeated in triplicate for reproducibility.

3. Results and discussions

3.1. Characterization of the DNDIM

The dispersion of DNDs in PVDF-acetone solution after sonication, which was utilized for fabrication of DNDIM, is depicted in Fig. 2A. As shown, PVDF was chosen because the DNDs dispersed well in it. Scanning electron micrographs of the unmodified membrane, DNDIM and pristine DND crystals are shown in Fig. 2B (a, b and d) respectively. As compared to the unmodified membrane (Fig. 2B (a)), the incorporation

of the DNDs in DNDIM is clearly evident in Fig. 2B (b) and (c) which depicts that the DNDs were uniformly distributed within the membrane. Thermal gravimetric analysis (TGA) was performed to determine the thermal stability of the DNDIM membrane. The TGA curve is shown in Fig. 3 and the amount of DND was found to be approximately 2% by weight. As observed, the thermal degradation of unmodified polypropylene membrane started at around 260 °C. The presence of DNDs increased the degradation temperature by 40 °C. This implies that the DNDs were highly stable and enhanced the thermal stability of the membrane. Incorporation of carbon nanotubes has shown similar behavior in terms of enhancing thermal stability [15]. This is an important factor for MD, where the elevated temperatures can be used for desalination [15]. Additionally, the surface chemistries of the pristine DNDs were characterized by FTIR spectroscopy and from the spectra it was confirmed that surface functional groups such as hydroxyl-, carboxylic, and amines were present on the DNDs surface [20]. The results are not presented here for brevity.

3.2. Desalination using DNDIM

The DNDIM membranes were tested for MD. The water vapor flux, J_w , across the membrane can be expressed as:

$$J_w = \frac{w_p}{t \cdot A} \quad (1)$$

where, w_p is the total mass of permeate, t is the permeate collection time and A is the membrane surface area. Also, J_w can be denoted as:

$$J_w = k(C_f - C_p) \quad (2)$$

where, k is the mass transfer coefficient, C_f and C_p is the water vapor concentration in feed and permeate side. Usually C_p is close to zero, since dry air was utilized as sweep gas. So overall mass transfer coefficient was calculated as:

$$k = \frac{J_w}{C_f} \quad (3)$$

SGMD experiments were carried out at the range of 70–90 °C at feed flow rate 10 mL min⁻¹.

As can be observed in Fig. 4a, increasing temperature increased permeate flux for both membrane types. This was due to the fact that the increased temperature difference created higher vapor pressure difference, thus enhancing overall water vapor flux. The DNDIM membrane showed enhanced flux at all temperatures. For example at 70 °C feed temperature, the flux using DNDIM was 10 lit/m²h and was nearly the same (9.67 lit/m²h) as that accomplished at 90 °C

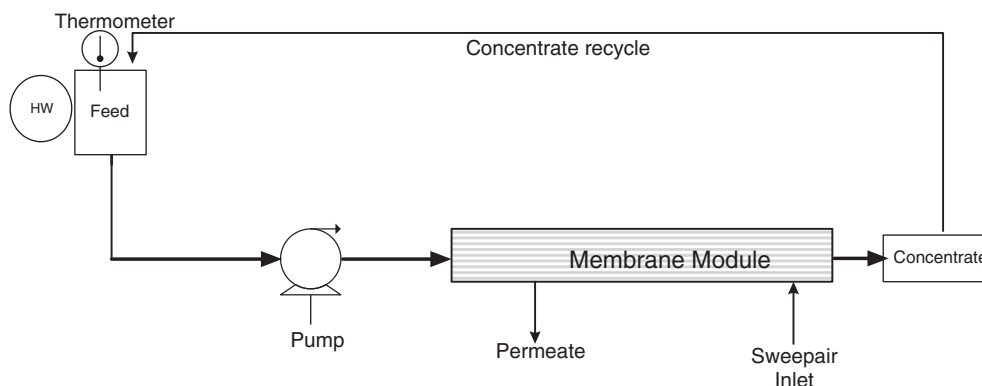


Fig. 1. Schematic of experimental set up of sweep gas membrane distillation.

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