



Flux enhancement in direct contact membrane distillation by implementing carbon nanotube immobilized PTFE membrane



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ARTICLE INFO

Article history:

Received 23 October 2015

Received in revised form 28 January 2016

Accepted 29 January 2016

Available online 4 February 2016

Keywords:

Carboxylated MWCNTs

Flux

PTFE

DCMD

Mass transfer coefficient

ABSTRACT

Carbon nanotube (CNT) enhanced direct contact membrane distillation (DCMD) is presented for water desalination. We investigated the immobilization of CNTs on a polytetrafluoroethylene (PTFE) membrane. The presence of CNTs on the hydrophobic membrane favorably altered the water–membrane interactions to promote water vapor transport while preventing salt water penetration into the membrane pores. For a salt concentration of $34,000 \text{ mg l}^{-1}$ and at 70°C , the CNT incorporation led to a 54% enhancement in permeate flux, which was as high as $69 \text{ kg/m}^2 \text{ h}$.

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1. Introduction

As the population increases in many parts of the world, fresh water is becoming a scarce commodity. A promising source of virtually limitless supply of water is the sea; however, desalination technology has been too expensive for extensive utilization. Membrane Distillation (MD) has emerged as an alternative to conventional techniques such as reverse osmosis and thermal distillation [1–3]. MD is a thermally driven process based on a vapor pressure gradient across a hydrophobic membrane [4], and Direct Contact Membrane Distillation (DCMD) is the most commonly used configuration in which a porous hydrophobic membrane is imposed between the aqueous brine solution at a higher temperature and a colder distillate on the permeate side [5,6]. IN DCMD, water vapor diffuses through a porous membrane and condenses into the cold distillate. The main effort in optimal design of such DCMD membrane involves the maximization of solute rejection and flux, with high feed side heat transfer coefficient [7,8] but low thermal conduction by the membrane. A key component in such a process is the membrane itself because it determines both water vapor flux and selectivity.

Recent advances in nanotechnology offers new approaches to address the challenges such as higher flux, low fouling, stability and selectivity. These developments have opened new possibilities for incorporating nanomaterials [9–20] in membranes

thereby enhancing the water vapour permeability and overall flux. Carbon nanotubes (CNTs) possess some interesting properties such as high hydrophobicity, surface area, thermal stability, ease of functionalization [21], which makes them promising candidates for many separations including desalination [11–14]. CNT based membranes have been used in reverse osmosis, forward osmosis, ultrafiltration, nanofiltration, pervaporation and extraction [10–13,19,20,22]. The CNT hollow fiber membranes exhibits enhanced water flux, six times higher than that of polymeric membranes with the similar pore size of $\sim 100 \text{ nm}$ [23]. Synthesis of free-standing and silicon-chip supported vertically aligned CNTs membranes by a complex synthesis method have been reported for RO, and high permeability for such CNTs membranes has been confirmed [24]. Functionalized multi-walled carbon nanotube blended cellulose acetate membranes have been synthesized for forward osmosis (FO) through phase inversion technique that have shown enhanced performance over conventional FO membranes [22]. In the free standing CNT membranes the CNTs serve as pores for selective transport of water while rejecting the salt-water clusters that are slightly bigger in size [22]. Recent efforts have involved the assemblies of CNTs into paper-like structures, called Bucky papers (BP) as self-supporting membranes for DCMD [25]. DCMD performances of membranes embedded with MWCNTs and graphite into the hydrophilic inner layer of a dual layer hollow fiber membrane has been studied and has shown that the MWCNTs formed a random conducting network and improved thermal conductivity of the inner layer, resulting an increase in the water vapor flux [26].

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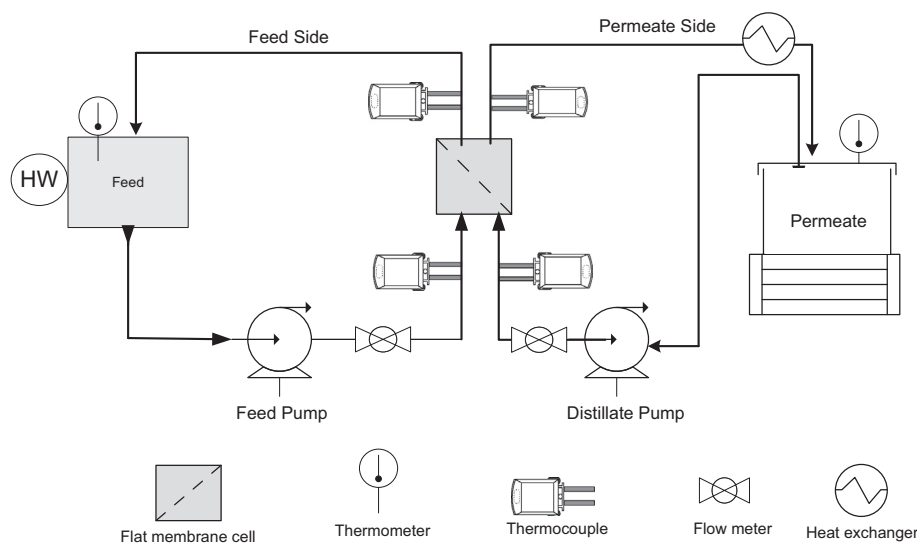


Fig. 1. Schematic representation of experimental setup.

Recently we have demonstrated that by immobilizing CNTs within the membrane pores, the solute-membrane interactions could be altered, which is one of the major physicochemical factors affecting the permeability and selectivity of a membrane. CNTs in such carbon nanotube immobilized membrane (CNIM) provide additional pathways for water vapor transport. So far, these membranes have been used in sweep gas membrane distillation applications and have demonstrated superior performance [19,27]. It was observed that the water vapor flux for SGMD with polypropylene-CNIM remained constant with increasing salt concentration, reaching up to $19.2 \text{ kg/m}^2 \text{ h}$ [27]. However, the SGMD configuration is relatively complex and higher condenser capacity is required to entrap the water vapors compared to direct contact membrane distillation (DCMD) where pure water is used in the permeate side as the condensing medium. The overall water vapor flux in SGMD was also reported lower compared to DCMD [28,29]. Another important consideration that is yet to be fully exploited is whether functionalization of CNTs could further lead to specific interactions with water vapour and if such membranes could be further employed in DCMD configuration. Moreover, the most common membrane materials in MD applications are polypropylene (PP), Polyvinylidene fluoride (PVDF) and Polytetrafluoroethylene (PTFE) and PTFE is known to be the most hydrophobic. The objective of this research is to investigate CNIM for DCMD using carboxylated CNTs. Of particular interest is the immobilization of the CNTs on PTFE backbone. Therefore, the objective of this work was to test the highly hydrophobic Teflon as the base for CNIM using carboxylated CNTs and study the MD desalination in the DCMD mode.

2. Experimental

2.1. Materials and methods

The membrane module used for DCMD was a cylindrical module utilizing a flat membrane with a gasket diameter of 4.3 cm and an effective membrane area of 14.5 cm^2 . The membrane used was that of polytetrafluoroethylene (PTFE) laminated onto a non-woven Polypropylene fabric support for improved mechanical strength and temperature handling (Gore-Tex). The feed used in these experiments ranged from 3000 to 34,000 ppm NaCl solutions (Sigma Aldrich) and permeate was that of deionized water. Both hot and cold sides were circulated through the module using

peristaltic pumps (Cole Parmer, model 7518-60). The preheated hot feed solution was passed through a heat exchanger, which was used to maintain the desired temperature throughout the experiment. The hot feed was recycled to the feed tank and the permeate was collected in the distillate tank. Inlet and outlet temperatures of the feed and distillate were monitored continuously throughout the experiment. Viton and PFA tubings and connectors (Cole Parmer) were used to make connections in the experimental set up. The ionic strength of the original feed solution and permeate were measured using an Electrode Conductivity Meter (Jenway 4310). The schematic of DCMD experimental system is shown in Fig. 1. Each experiment was run for 2 h and was repeated three times to estimate reproducibility.

MWCNT was purchased from Cheap Tubes, Inc., Brattleboro, VT, USA. CNT carboxylation was carried out in a Microwave Accelerated Reaction system (CEM Mars) fitted with internal temperature and pressure controls using a method described before [30].

The functionalized CNT membrane was prepared using PTFE composite membrane with a non-woven fabric support and a pore size of $0.4 \mu\text{m}$. For the preparation of carboxylated CNTs dispersion, 10 mg of MWCNT-COOH were dispersed in a solution containing 0.2 mg of Poly tetrafluoroethylene (PTFE) powder of $1 \mu\text{m}$ particle size (Sigma Aldrich) in 10 ml of Fluorinert FC-40 solvent (Sigma Aldrich) by sonicating for three hours. The PTFE-nanotube dispersion was thereafter sprayed uniformly with a dropper over the membrane held on a flat surface to form the CNIM. The wet CNIM was kept under the hood for overnight drying.

The morphology of CNIM was studied using a scanning electron microscopy (SEM, Model LEO 1530, (Carl Zeiss SMT AG Company, Oberkochen, Germany). This was done by cutting the membranes into 0.5 cm long pieces and coating with carbon films. Confocal Raman imaging and Raman spectra were measured using a Thermo electron Nicolet Raman spectrometer. Furthermore, thermal gravimetric analysis (TGA) was performed using a Perkin Elmer Pyris 7 TGA instrument with a heating rate of 10°C/min under air atmosphere to study the thermal stability of the membrane. To verify the hydrophobic-hydrophilic nature of the unmodified PTFE and CNIM, water droplets ($2 \mu\text{l}$) were deposited using a micro syringe Hamilton ($0\text{--}100 \mu\text{l}$) on the PTFE and the CNIM. The particle positions were recorded and the contact angle was measured using a digital video camera mounted at the top of the stage. Five readings were obtained and the average value together with the standard deviation is reported in this study. The surface roughness of the membranes was measured by atomic force microscopy (Bruker, Dimension ICON

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