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Enhanced desalination of polyamide thin film nanocomposite incorporated with acid treated multiwalled carbon nanotube-titania nanotube hybrid

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HIGHLIGHTS

- Multiwalled carbon nanotubes-titania nanotube hybrid was synthesized by hydrothermal method.
- Surface charge of as-synthesized MWCNT-TNT was modified by acid treatment.
- TFN was fabricated by incorporating MWCNT-TNT hybrid and applied for RO desalination.
- TFN composite membrane exhibited higher flux and salt rejection.

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ABSTRACT

Polyamide (PA) thin film nanocomposite (TFN) membrane incorporated with multiwalled carbon nanotubes-titania nanotube (MWCNT-TNT) hybrid was successfully fabricated. The hybrid was introduced to the PA selective layer during the interfacial polymerization (IP) of trimesoyl chloride (TMC) and m-phenylenediamine (MPD) monomers over porous commercial polysulfone (PS) ultrafiltration support. The resultant TFN was characterized and applied for desalination. The results revealed that the acid treated MWCNT-TNT, which act as filler in the PA membrane, improved the surface properties of the membrane in term of surface charge, surface roughness and contact angle. Consequently, the water permeability increased significantly without compromising the salt rejection performance. The highest water permeability of 0.74 L/m^2 h bar was achieved for the TFN membrane containing 0.05 wt% acid treated MWCNT-TNT, which is approximately 57.45% than that of the neat PA membrane. The NaCl and Na_2SO_4 rejection of this membrane was 97.97% and 98.07%, respectively that is almost similar to the neat membrane.

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1. Introduction

Currently, providing clean water from limited water resources with only 0.8% fresh water of the total earth's water [1] for human consumption, agriculture, energy production and industries is one of the most important concerns faced by human society. Desalination has been identified as one of the key pillars of current and future water systems to ensure sustainable supplies of safe and clean drinking water [2]. Thin film composite (TFC) reverse osmosis (RO) membrane with excellent water flux and rejection has been commonly fabricated for desalination [3–5]. Recently, the advancement in nanotechnology opens the doors to fabricate thin film nanocomposite (TFN) membranes. The incorporation of nanomaterials in the support membrane and/or dense thin polyamide (PA) layer has been evidenced to heighten the performance of the RO membranes and consequently reduced the desalination cost and environmental impacts [6]. Several studies have revealed that the proper selection of these fillers can significantly increase the flux, stability and rejection of the TFN membranes [7,8].

Carbon nanotube (CNTs) and titanate nanotube (TNTs) are some commonly used nanomaterials for the fabrication of TFN membrane desalination. CNTs is one of the fullerene-based nanomaterials which has been widely applied to strengthen and modify membrane surface chemistry [9]. CNTs in PA layer provide an additional water path for water molecules to flow through the nanochannel while the formation of ordered hydrogen bonds increases the rate of water transport [10,11]. The functionalization of CNTs with –COOH and –OH groups can further improve their surface hydrophilicity [12]. The capability of CNTs to reject certain dissolved ions based on their structural properties is highly desired for membrane desalination application [13]. Work conducted by Amini et al. (2013) [14] showed that TFN membrane embedded with 0.05 wt% functionalized MWCNTs exhibited nearly 14% water

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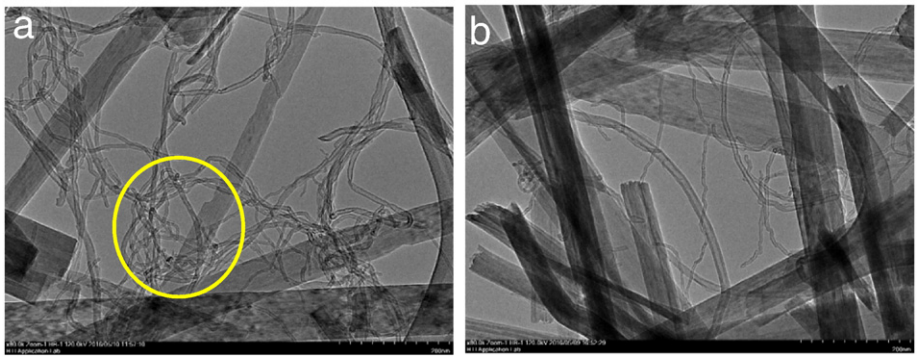

Fig. 1. TEM images of MWCNT-TNT hybrid (a) and a-MWCNT-TNT hybrid (b). Agglomeration of MWCNTs in a-MWCNT-TNT hybrid is reduced (yellow circle).

permeability improvement (3.6 L/m^2 h) and 28% increase in NaCl rejection (89.3%) compared to that of neat TFC membrane

On the other hand, TNTs with high hydrophilicity, the abundant surface functional group as well as high pore volume and the specific surface area has also offers unique properties for desalination application [8,15]. Moreover, the tubular structure of TNTs is found to be less adhesive compared to CNTs, resulted in lower agglomeration [16,17]. Emadzadeh et al. (2015) [18] observed two times improvement in water flux without sacrificing salt rejection when 0.05 wt% of functionalized TNTs was embedded into the PA layer of TFN membrane.

Inspired by the desired properties demonstrated by these nanomaterials, the objective of this study is to develop a highly permeable and selective membrane comprising a thin film layer containing a hybrid of MWCNT-TNT. The MWCNT-TNT hybrid was modified with acid to facilitate the nanomaterial dispersion and alter the surface charge of the resultant TFN membrane. As a single nanomaterial is incapable of offering all beneficial features at a time, it is anticipated that the hybrid can render desired functionalities in terms of surface hydrophilicity, charges and roughness. Additionally, the existence of pore channels of both fillers in this hybrid would also provide additional water channels to enhance the water permeability [19]. The physicochemical characteristics of the hybrid and TFN membrane were characterized, followed by performance evaluation in term of pure water permeability and NaCl and Na_2SO_4 salt rejection. To the best of our knowledge, this is the first attempt to prepare and incorporate modified MWCNT-TNT hybrid nanoparticles into TFN RO membranes for desalination. Hence, this study can shed insights into the possible potential of the surface modified hybrid fillers to fulfill the requirements of the desalination process.

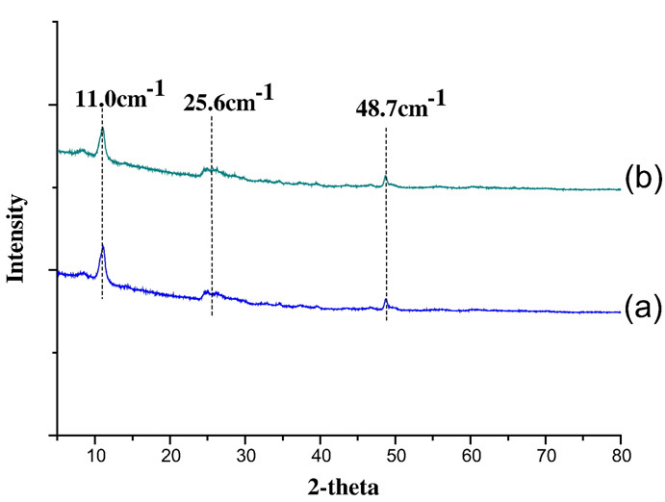

Fig. 2. XRD patterns for MWCNT-TNT (a) and a-MWCNT-TNT (b).

2. Experimental

2.1. Materials

A commercial ultrafiltration PS support membrane (PS35, Nanostone Flat Sheet Membrane) was purchased from Sterlitech Corporation. 1,3,5-benzenetricarboxylic acid chloride (TMC) was obtained from Acros Organic. N-hexane was supplied from Fisher Chemical. 1,3-phenylenediamine (MPD), hydrochloric acid (HCl, fuming 37%), sulfuric acid (H_2SO_4, 95–97%), nitric acid (HNO_3, 65%) sodium hydroxide (NaOH) and sodium chloride (NaCl) were purchased by Merck. Titanium dioxide nanoparticles (TiO_2) was obtained from Degussa P25 Evonik, multiwalled carbon nanotube (MWCNTs, 98% carbon basis, OD × ID × L: 10 ± 1 nm × 4.5 ± 0.5 nm × 3–6 μm) was obtained from Aldrich. Sodium sulphate (Na_2SO_4) was obtained from Riedel-de Haën. TNTs was self-synthesized from the TiO_2 nanoparticles.

2.2. Synthesis of MWCNT-TNT and a-MWCNT-TNT hybrid

MWCNT-TNT hybrid was synthesized by hydrothermal method. 1 g of MWCNTs was added into 100 ml of H_2SO_4:HNO_3 (each 3 M) mixed acids' solution with volume ratio of 3:1 (v/v) [20] at 80 °C for 24 h to introduce carboxylic acid functional groups onto the MWCNTs surface. The dispersion solution was collected by filtration and was rinsed with RO water until the result reached neutral before left to dry overnight at 60 °C. Next, 3 g of oxidized MWCNTs and TiO_2 in the ratio of 1:5 (w/w) were stirred in 100 ml aqueous solutions of 10 M NaOH and the suspension was transferred into an autoclave coated by Teflon and heated in an oven at 120 °C for 24 h. After the heating process, the autoclave was allowed to cool down to room temperature before dark grey

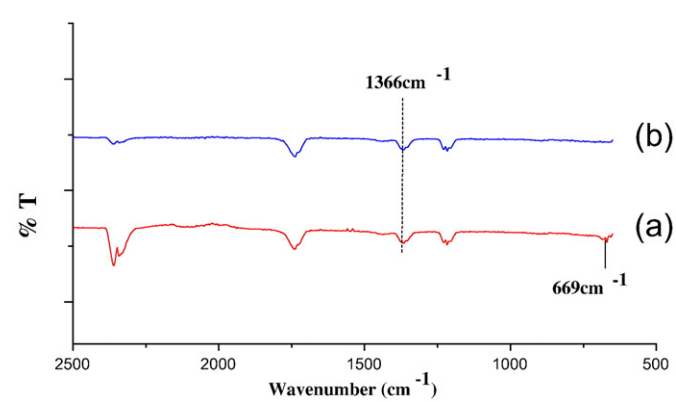

Fig. 3. ATR-FTIR spectra for MWCNT-TNT (a) and a-MWCNT-TNT (b). It was observed that the peak at 669 cm^{-1} disappeared for a-MWCNT-TNT due to less entangled nanotube structure.

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