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Fabrication of carbon nanotubes incorporated double-skinned thin film nanocomposite membranes for enhanced separation performance and antifouling capability in forward osmosis process

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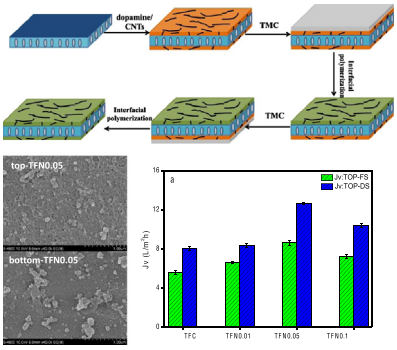
HIGHLIGHTS

- Double-skinned FO membrane was prepared by interfacial polymerization of dopamine/CNTs with TMC on PSf.
- Double-skinned membranes exhibited excellent water flux without sacrificing solute rejection.
- The TFN0.05 membrane demonstrated remarkable antifouling capacity because of CNT addition.

GRAPHICAL ABSTRACT



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ABSTRACT

Novel carbon nanotubes (CNTs) incorporated double-skinned thin film nanocomposite (TFN) membranes were fabricated by interfacial polymerization of polydopamine/CNTs and trimesoylchloride (TMC) on polysulfone (PSf) substrate. As controls, thin film composite (TFC) membrane without CNTs and FO membranes with single-skinned structures (top-skinned or bottom-skinned) was also fabricated. The prepared membranes were characterized and evaluated in terms of membrane morphology, structure, surface property, separation performance and antifouling capacity. The effect of membrane orientation, composition and concentration of draw solutions on FO performance was studied as well. It was found that CNTs had significant influence on the properties and the performances of the synthesized FO membranes. The double-skinned membranes owned excellent solute rejection without sacrificing water flux. By incorporation of CNTs, TFN membranes exhibited higher FO water flux than TFC membranes. The double-skinned TFN0.05 membrane, the optimal FO membrane, showed a 54% enhancement in water flux than double-skinned TFC membrane at TOP-FS orientation by using $MgCl_2$ as draw solution and DI water as feed solution. Moreover, the double-skinned TFN0.05 membrane demonstrated remarkable antifouling capacity because of the prominent foulant resistance induced by CNT addition. This work paved a new way to fabricate high performance FO membrane by the utilization of double-skinned structure and incorporation of CNTs.

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1. Introduction

Global water crisis and clean water shortage have become a serious challenge faced by humanity today. Membrane separation technology is a green and effective way to regenerate clean water [1]. In recent years, forward osmosis (FO) has been identified as an emerging technology because of its potential advantages of less energy input, lower costs, lower fouling propensity or easier fouling removal compared to the currently dominant desalination technology—reverse osmosis (RO) [2–4]. FO has attracted worldwide attention in a variety of applications, including power generation [5,6], seawater desalination [7], waste water treatment [8], liquid food processing [9], biological process [10], and pharmaceutical applications [11].

Generally, FO membrane is asymmetric membrane and possesses a thin selective layer on top of a porous substrate. These conventional asymmetric single-skinned FO membranes experience a major drawback of FO process, i.e. internal concentration polarization (ICP) [12]. ICP is a phenomenon of osmosis driven membrane process as a result of the dilution of draw solutes or the accumulation of feed solutes in the support layer depending on the membrane orientation [13]. When the active layer faces the feed solution (AL-FS orientation), dilutive ICP will occur within the membrane support layer as water permeates across the membrane from the FS to the DS. When the active layer is placed against the draw solution (AL-DS orientation), concentrative ICP occurs as the solute in the FS accumulates within the membrane support layer. Both dilutive and concentrative ICP can lower the effective osmotic driving force for water transport and cause a severe decline in water flux [3,14]. Compared to AL-FS orientation, AL-DS orientation tends to have higher FO water flux induced by the attenuated concentrative ICP [15]. However, AL-DS orientation has greater fouling propensity due to the easy entering of foulants from the feed solution into the porous support [15,16]. Therefore, traditional single-skinned FO membranes face a problem of either experiencing much more severe dilutive ICP in AL-FS mode or having much higher fouling propensity in AL-DS mode [13]. In order to solve this problem, high performance double-skinned FO membranes have been designed and fabricated [13]. These membranes have a typical sandwiched structure (i.e., a porous support between two selective layers), with a primary selective layer to reject the draw solute and a relatively loose secondary selective layer to reject feed foulants [13,17].

To further improve the performances of membranes, many nanomaterials (such as zeolite [18], mesoporous silica [19], metal or metal oxide nanoparticles [4], CNTs [3], and graphene oxide [20]) have been used to fabricate membranes. Among these nanomaterials, carbon nanotubes (CNTs) have gained much attention because they show great potential in enhancing both the permeability and antifouling capacity of membranes [3,21]. The permeability improvement of CNT incorporated membranes may be attributed to the extra water channels caused by CNTs and the nanocorridors between CNTs and polymers [3]. CNTs can damage the cell membranes of microorganisms and disrupt the metabolic pathways accompanied by the oxidative stress, then inactivation and/or death of microorganisms can occur [22].

To date, applications of CNTs in the preparation of FO membranes can be investigated from two directions. The first one is incorporating CNTs in a polymer matrix [23], and the second one is adding CNTs in the active layer to fabricate thin film nanocomposite (TFN) membranes [3,24]. Xu et al. [23] fabricated CNTs embedded PES substrate supported polyamide TFC FO membrane, which exhibited higher performance than pristine PES supported TFC membrane. Wang et al. [24] fabricated CNT immobilized PEI-PAI hollow fiber membrane. Compared to the membranes without CNTs, this membrane showed 44% enhancement in water permeability without significant compromise on the salt permeability. Amini et al. [3] prepared CNT incorporated polyamide TFN membranes via interfacial polymerization method for FO application. The resultant TFN membranes exhibited nearly 160% higher water flux than TFC membrane.

The objective of the current study was to minimize the ICP and increase antifouling property of FO membranes by incorporating CNTs in the double-skinned FO membrane through interfacial polymerization. Dopamine (DA) and trimesoyl chloride (TMC) were used as the monomers to induce interfacial polymerization [1,25]. As shown in Fig. 1, CNTs were dispersed in DA solutions and were immobilized in the active layer of the FO membranes because of the formation of a polydopamine (PDA) layer by DA self-polymerization and a dense active layer formed by PDA and TMC via interfacial polymerization.

2. Experimental

2.1. Materials

CNTs (20 nm in diameter, 0.5–2 μm in length, purity > 95 wt.%) were purchased from Nanjing XFNANO Materials Tech Co., Ltd. Polysulfone (PSf-3500P) was purchased from Solvay Advanced Polymers. 1-Methyl-2-pyrrolidinone (NMP, 99%), n-hexane (99.0%), and polyvinyl pyrrolidone (PVP, 99%, 25,000 g/mol) were obtained from Merck. Dopamine hydrochloride (DA, 98%) and trimesoyl chloride (TMC, 98%) were supplied by Aladdin. Other chemicals were obtained from Sinopharm Chemical Reagent Co., Ltd. All chemicals were used as received. Milli-Q water (Barnstead, compact ultrapure water system) with a resistivity of 18.2 MΩ·cm was used.

2.2. Preparation of polysulfone (PSf) substrates

The PSf substrates were prepared by a phase inversion method [26]. The casting solution was prepared by dissolving 15.5 wt.% PSf in NMP

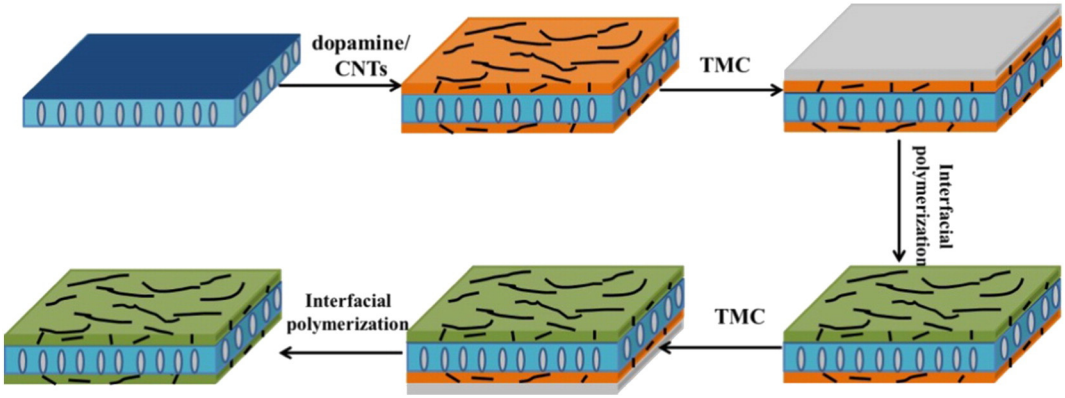

Fig. 1. Schematic of the fabrication procedure of the CNTs incorporated double-skinned FO membrane.

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