

The potential of thin film nanocomposite membrane in reducing organic fouling in forward osmosis process



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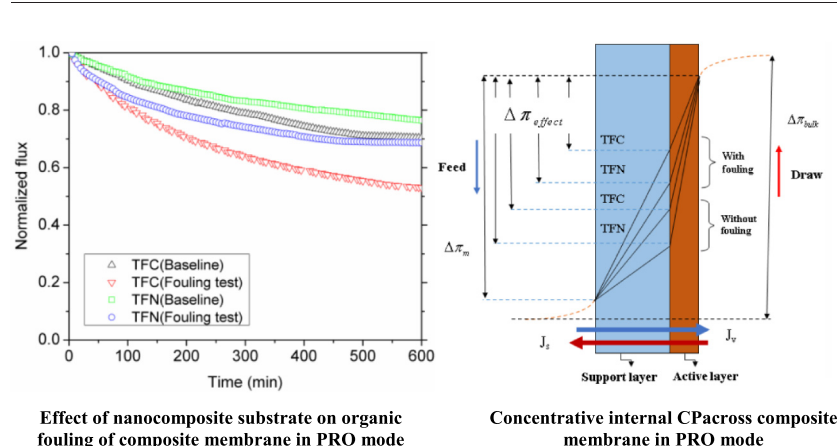
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HIGHLIGHTS

- Synthesis of a novel TFNFO membrane with small S value and minimum ICP.
- Better performance of the TFN membrane compared to a typical TFC membrane.
- The TFN FO membrane prepared has demonstrated superior fouling resistance.

GRAPHICAL ABSTRACT



Effect of nanocomposite substrate on organic fouling of composite membrane in PRO mode

Concentrative internal CP across composite membrane in PRO mode

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ABSTRACT

A major limiting factor of forward osmosis (FO) membrane, particularly in pressure retarded osmosis (PRO) mode, is fouling by natural organic matters. In this work, we investigated the effect of the nanocomposite substrate on the fouling of a thin film nanocomposite (TFN) membrane due to organic foulants in PRO mode. The TFN membrane was synthesized by coating a polyamide film over the surface of substrate made of polysulfone–titanium dioxide. The TFN membrane always showed much higher FO water flux than the typical thin film composite TFC membrane prepared from the pristine polysulfone substrate. Reduced internal concentration polarization following a significant decrease of the structural parameter in the nanocomposite substrate causes the mass transfer coefficient of the substrate to increase. In the PRO mode, BSA removal in the presence of Ca²⁺ confirmed that the TFN FO membrane could significantly mitigate fouling tendency compared to a typical TFC membrane. Results also showed that fouling in TFN FO is highly reversible, recovering >92% permeate flux after a simple water rinse process. A complete study of the membrane fouling was reported with detailed scientific discussion. To the best of our knowledge, this is the first report on the effect of the nanocomposite membrane on membrane fouling in PRO mode.

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1. Introduction

Membrane fouling is the main problem to affect the efficiency of membranes in their applications like brackish water, seawater desalination and water treatment [1–5]. Forward osmosis (FO) membrane shows water flux decline, because of the internal concentration polarization (ICP) and membrane fouling [6–8]. According to Tang's research group [9], membrane orientation is an important factor to the membrane fouling. The performance of the FO membrane was studied using latex particles as model foulant. Furthermore, they found that the boron flux was greater in pressure retarded osmosis (PRO) mode as compared to FO mode [10]. Both PRO and FO modes have been used in various FO applications [11]. According to Arkhangelsky et al. [12], FO mode is favorable, when feed solutions of higher fouling and scaling tendencies (e.g. wastewater treatment) [13], or solutions of high salinity (e.g. seawater desalination) are treated [14–16]. On the other hand, PRO orientation is preferred when using solutions with lower fouling, ICP and scaling tendencies (e.g. brackish water desalination) or where intensive concentration is unnecessary (e.g. power generation) [17,18]. Among the numerous research efforts on PRO mode, the effect of using a nanocomposite substrate on the composite membrane fouling has not been investigated. Therefore, there is a need to understand the intrinsic cause and effect of organic fouling in the PRO mode, which is of primary importance for the development of fouling control in PRO mode.

Recently in year 2013, Rajaeian et al. [19] explored the possibility of embedding aminosilanized titanium dioxide (TiO₂) nanoparticles into the polyamide (PA) layer of the composite membrane in an attempt to prepare a thin film nanocomposite (TFN) nanofiltration (NF) membrane for salt water purification. In comparison to the typical TFC membrane, they experienced that both water permeability and salt selectivity of the TFN membrane were improved. Nevertheless, it must be pointed out that good dispersion of hydrophilic nanoparticles in a non-polar organic solvent/aqueous solution is very difficult to achieve during interfacial polymerization process, and might therefore impede the TFN membrane to realize its full potential. Previous studies have shown the potential of using TiO₂ nanoparticles as filler for the preparation of the ultrafiltration (UF) membrane with improved performance and anti-fouling, particularly in water permeation rate [20–22]. According to Teow et al. [23], the polyvinylidene difluoride (PVDF)/TiO₂ composite UF membrane shows significantly high fouling resistance compared with the neat PVDF membrane during the filtration process of humic acid (HA) in the presence of Ca²⁺. The increase in water permeability and antifouling properties of composite UF could be attributed to the enhanced membrane hydrophilicity and the increase in membrane porosity upon TiO₂ addition. Despite the positive impact of TiO₂ nanoparticles on the performance of the microporous membrane, overloading of nanoparticles should be avoided as it could result in poor membrane performance in which both water flux and solute rejection tend to decrease [24]. The phenomenon is most likely due to the agglomeration of TiO₂ nanoparticles, which reduces the contact area of hydroxyl groups carried by TiO₂ nanoparticles.

In this work, an attempt is made to fabricate a novel TFN membrane for FO process by modifying the polysulfone (PSf) substrate using hydrophilic TiO₂ nanoparticles. The effect of TiO₂ nanoparticles on the physical and chemical interactions between organic foulants and TFN membranes was investigated. The TFN membrane fouling behavior in FO process was studied to understand the fouling mechanisms at the molecular level. The influence of other physical and chemical factors, such as membrane orientation and flux recovery was also investigated.

2. Experimental

2.1. Materials and method

Polysulfone beads (Udel P-1700, Solvay Advanced Polymers), 1-methyl-2-pyrrolidinone (NMP, ~99.5%, Merck) and polyvinyl pyrrolidone

(PVP K30, Sigma-Aldrich) were used to make the substrate without any further purification. Titanium dioxide nanoparticles (TiO₂, Degussa P25, Evonik) were added to the polymeric dope solution to modify substrate properties. Two active monomers, i.e. 1,3-phenylenediamine (MPD, ~99%, Merck) and 1,3,5-benzenetricarbonyl trichloride (TMC, ~98%, Merck) were used to synthesize the PA selective layer for the TFC and TFN membranes. TMC was dissolved in n-hexane (~99%, Merck) while MPD was dissolved in distilled water. For both the FO and RO experiments, sodium chloride (NaCl, ~99.5%, Merck) was used to prepare solutions of different salt concentrations. Bovine serum albumin (BSA, 66 kDa, Sigma-Aldrich) and calcium chloride (CaCl₂, ~99.5%, Merck) were used for the foulants. The test solution contained 200 mg/L as organic foulant and 10 mM NaCl solution. Mixing of the stock solution was performed for over 24 h to ensure complete dissolution of the solutes. The stock solution was stored in sterilized glass bottle at 4 °C until use.

2.2. FO membranes

2.2.1. Preparation of substrate and thin film composite membrane

PSf substrates were prepared using dope solutions containing 17.5 wt.% PSf, 82.0 wt.% NMP and 0.5 wt.% PVP. In order to enhance the hydrophilicity and structure of the substrate, additional 0.5 wt.% TiO₂ nanoparticles was added to one of the dope solutions. As reported in our previous research work [25], it is proved that 0.5 wt.% TiO₂ nanoparticles is the optimum concentration in the preparation of the composite substrate owing to the high water permeability and low reverse solute flux shown by the TFN membrane prepared from this composite substrate. The active rejection layer of TFC and TFN membrane was formed by interfacial polymerization on the top surface of the pre-cast PSf substrate and PSf-TiO₂ nanocomposite substrate, respectively. At first, 100 mL of 2% (w/v) MPD aqueous solution was poured onto the top surface of the PSf substrate, which was held horizontally for 2 min to ensure the penetration of MPD solution into the pores of the substrate. The excess MPD solution was then drained off from the substrate surface and a rubber roller was employed to remove the residual droplets of MPD solution. Then, 100 mL of 0.1% (w/v) TMC solution in hexane was poured onto the substrate surface. The TMC solution was drained off from the surface after 1 min contact time. In order to remove unreacted monomers from the TFN membrane surface, the membrane was rinsed with pure n-hexane and dried at ambient condition for 1 min and further in an oven at 60 °C for 8 min. The prepared TFC and TFN membranes were stored in a deionized water container until it was tested [25].

2.3. Evaluation of membrane performance

The reverse osmosis (RO) experimental setup with cross-flow filtration mode was used for determining permeability and selectivity of TFC/TFN membrane. The permeation cell was designed to have a total effective membrane area of 14.62 cm². In this RO experiments, water flux (*J*) and water permeability (*A*) [26] of membranes were evaluated using Eqs. (1) and (2) as follows:

$$J = \frac{\Delta V}{A_m \cdot \Delta t} \quad (1)$$

$$A = \frac{J}{\Delta P} \quad (2)$$

where *A_m* is the effective membrane area and Δ*V*, Δ*t* and Δ*P* are permeate volume, time and transmembrane pressure difference, respectively.

Feed NaCl concentration was 20 mM and the cross membrane pressure difference was kept at 2.5 bar. In order to minimize external concentration polarization, a cross-flow rate of 32.72 cm/s was employed.

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