Zeolite-polyamide thin film nanocomposite membranes: Towards enhanced performance for forward osmosis

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ABSTRACT

Zeolite-polyamide thin film nanocomposite (TFN) membranes were prepared on a polysulfone (PSf) porous substrate tailored for forward osmosis (thin thickness, high porosity, and straight needle-like pores). The TFN membranes were characterized and evaluated in comparison with a thin film composite (TFC) membrane. The incorporation of NaY zeolite nanoparticles in the polyamide rejection layer significantly changed its separation properties. In the range of 0.02–0.1 wt./v% zeolite loading, the incorporation of zeolite-polyamide exhibited enhanced water permeability of membrane likely due to the porous nature of zeolite. However, further increase in zeolite loading led to a reduction in water permeability, possibly as a result of the formation of a thicker polyamide layer. The most permeable TFN membrane (TFN0.1, with 0.1 wt./v% zeolite loading) had a water permeability approximately 80% higher compared to the baseline TFC membrane. The FO water flux followed a similar trend to that of the membrane water permeability. Under all cases evaluated in the current study (0.5–2.0 NaCl draw solution, DI water and 10 mM NaCl feed solution, and both membrane orientations), the membrane TFN0.1 exhibited highest water flux (up to 50% improvement over the TFC membrane). To the best knowledge of the authors, this is the first report on zeolite-polyamide based TFN membranes for FO applications.

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1. Introduction

Forward osmosis (FO) is an emerging membrane separation technology that utilizes the difference of osmotic pressure between a feed solution (FS) and a concentrated draw solution (DS) to drive water across a semi-permeable membrane [1,2]. Due to its low hydraulic pressure during operation, FO process has been proposed and reported with several advantages, mainly including (1) simplicity and high rejection of a wide range of contaminants, (2) no requirement of high pressure or high temperature, and (3) potentially lower energy consumption and lower fouling propensity in several given cases [3–5]. Therefore, FO can be employed in a number of areas such as seawater desalination, water treatment, wastewater reclamation and food processing [1,6–9].

The development of high performance FO membranes is one of the priority research topics in the FO area [10–18]. Performance of existing commercial FO membranes (asymmetric cellulose triacetate (CTA) membranes from Hydration Technology Inc. (HTI)) are generally limited by their relatively low water permeability and salt rejection [15]. In comparison, thin film composite (TFC) FO membranes have demonstrated superior FO water flux and better solute rejection [11–15]. These high-performance TFC FO membranes consist of a top thin polyamide (PA) rejection layer and a porous membrane support. In order to minimize the internal concentration polarization (ICP), a small structural parameter (S, thickness × tortuosity/porosity) is preferred for the support layer [15]. In parallel, the relatively high water permeability of the PA rejection layer ensures minimized membrane resistance loss [15]. In addition, recent studies also reveals that a high water permeability of the rejection layer helps to reduce the ICP level in the active-layer-facing-feed-solution (AL-FS) orientation [18,19].

Recent studies have demonstrated that mixed matrix membranes formed by embedding porous materials in a polymeric matrix may significantly enhance membrane properties such as permeability, selectivity, stability, surface area, or catalytic activity in various membrane separation processes [20–22]. For example, thin film nanocomposite (TFN) reverse osmosis (RO) membranes have been developed by incorporating pure metal, metal oxide and zeolite nanoparticles into the PA rejection layer. In particular, the incorporation of zeolite in a PA layer has demonstrated to improve its water permeability without significant loss of salt rejection under high pressure during RO process [23–26]. This has
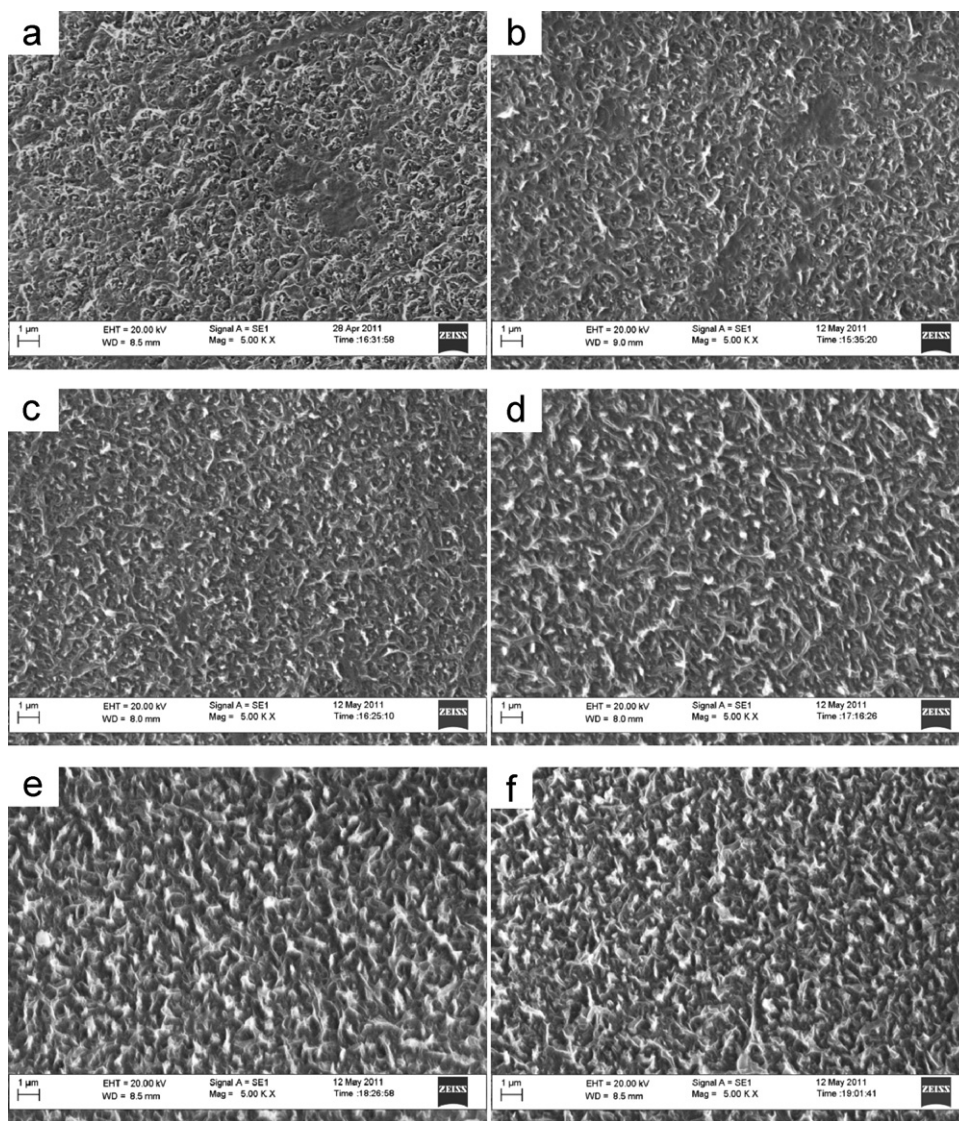
Fig. 1. SEM micrographs of top view of TFC and TFN membranes with different zeolite loadings. (a) TFC membrane, (b) TFN0.02 membrane, (c) TFN0.05 membrane, (d) TFN0.1 membrane, (e) TFN0.2 membrane and (f) TFN0.4 membrane.

been attributed to the well-defined sub-nanometer pores in zeolite nanoparticles that behave as preferential flow channels for water molecules while they are too small for the solutes such as hydrated sodium ions to pass through (i.e., the molecular sieving mechanism). Whereas the zeolite-PA based TFN membranes were originally formulated for RO applications, their enhanced water permeability of active layer may make them ideal candidates for FO.

The objectives of the current study were to synthesize zeolite-PA based TFN FO membranes by incorporating zeolite nanoparticles into a polyamide rejection layer and to characterize these membranes in terms of membrane properties and FO performance. To the best knowledge of the authors, this is the first study reporting the development and application of TFN membranes for FO process.

2. Experimental

2.1. Chemicals and reagents

Unless specified otherwise, all reagents were of analytical grade and were used as received without further purification. Deionized water (DI) was obtained from a Milli-Q system (Millipore,

Billerica, MA). Polysulfone (PSf) beads (Mn:75,000–81,000 Da, Solvay Advanced Polymers, LLC, GA), 1-methyl-2-pyrrolidinone (NMP, Merck Schuchardt OHG, Hohenbrunn), polyvinyl pyrrolidone (PVP, average molecular weight 1,300,000 Da, Alfa Aesar, MA) and lithium chloride (LiCl, Sinopharm Chemical Reagent Co. Ltd., China) were used for the preparation of membrane substrate. 1,3-Phenylendiamine (MPD, >99%, Fisher Scientific), n-hexane (ACS reagent, Fisher Scientific) and 1,3,5-benzenetricarbonyl trichloride (TMC, >98%, Sigma–Aldrich) were used for the synthesis of polyamide rejection layer of thin film composite membranes (TFC). Zeolite nanoparticles (NaY, catalyst support, Sigma–Aldrich) were used for TFN membrane preparation due to its commercial availability and well characterized properties [27]. Based on electron microscopic characterization, the particle size of zeolite ranged from 40 to 150 nm (Appendix B). Sodium chloride (NaCl) used for both FO and RO tests was obtained from Merck Chemicals.

2.2. Preparation of TFC and TFN membranes

PSf substrates were hand-casted following our previous method [15]. Briefly, PSf beads (15.5 wt.%), PVP (0.5 wt.%) and LiCl (3.0 wt.%) were dissolved in NMP, and stirred at 60 °C until homogeneous,