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# Poly(tetrafluoroethylene)/polyamide thin-film composite membranes via interfacial polymerization for pervaporation dehydration on an isopropanol aqueous solution

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#### ABSTRACT

Poly(tetrafluoroethylene)/polyamide (PTFE/PA) thin-film composite membranes were prepared via interfacial polymerization using surface-modified PTFE films as substrates. The composite membranes from the amine-functionalized PTFE films exhibit good layer compatibility and membrane stability due to the formation of covalent linkages between the PTFE substrate and PA layer. The composite membranes are applied to pervaporation dehydration processes on a 70 wt% isopropanol aqueous solution. The membranes are stable under the pervaporation dehydration operations and show a high-permeation flux of 1720 g/h m² and a separation factor of 177. These PTFE-based thin-film composite membranes are potentially useful in pervaporation separation for other organic mixtures.

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#### 1. Introduction

Thin-film composite (TFC) membranes are attractive in membrane separations such as reverse osmosis [1–5] and nanofiltration [6–8]. The selective layers of the TFC membranes are dense and thin to warrant their high-permeate fluxes and effective separation performance. One major approach to prepare TFC membranes is interfacial polymerization [1–8], which is usually performed with a polycondensation reaction between two monomers on the surface of a microporous support. The most studied active layers of TFC membranes are polyamides (PA) [4–11], which employ amine and acid chloride compounds as monomers in interfacial polymerization. Many polyamide-based TFC membranes have been prepared. The separation performances of the TFC membranes are highly dependent on the monomers used in interfacial polymerization [9–11].

In addition to the active layer, the microporous supports of the TFC membranes are also of interest. Polysulfone and polyethersulfone are two major materials using as the porous substrates for interfacial polymerization [4–11]. However, other polymeric materials are also utilized to improve the stability of the resulting TFC membranes. Li et al. [12] utilized poly(ether ether ketone) in preparation of TFC membranes due to its good thermal stability and

high-chemical resistance. Peinemann et al. [13] reported a similar work in which polyetherimide was used as the supports of TFC membranes. Polypropylene (PP) was also utilized as the porous support of TFC membranes [14] due to their attractive properties such as high durability, chemical resistance, and sustainability in pH variation. However, surface modification on PP is necessary to provide certain compatibility between the substrates and the reaction solutions in interfacial polymerization. Polyamide-based TFC membranes are also obtained from surface-modified polyacrylonitrile (PAN) porous films, which possess carboxylic acid groups on their surfaces [15]. The carboxylic acid groups on PAN film surface were proposed to involve in the interfacial polymerization and to introduce strong interactions between the substrate and the polyamide layer.

Pervaporation is one important method to separate liquid mixtures in azeotropic compositions, which are hard being separated by conventional distillation processes. Composite membranes are an important class of membranes for pervaporation separations [16–19]. Composite membranes usually possess very thin separation layer. Therefore, the permeation resistance of the membranes is reduced so as to increase the permeation fluxes of the membranes. On the other hand, polyamides and polyimides are both important materials using in preparation of pervaporation separation membranes [20–23]. Therefore, TFC membranes could be potentially applied to pervaporation separation processes. Polysulfone/polyimide TFC membranes, which were prepared from interfacial polymerization of polyimide on a

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polysulfone support, were applied to pervaporation dehydration on ethanol aqueous solutions [24]. PAN/polyamide TFC membranes were also utilized in the pervaporation dehydration on isopropanol (IPA)/water mixtures [25,26]. However, the separation performances of the reported TFC membranes are not satisfied.

Poly(tetrafluoroethylene) (PTFE) shows great applications in industrials basing on its superior chemical resistance, good thermal stability, and high-mechanical strength. Our previous work also demonstrated the attractive performances of PTFE and its composite membranes using in pervaporation separations [19,27]. Therefore, to the best of our knowledge, this work aims on the first preparation of PTFE/polyamide TFC membranes by interfacial polymerization. A proper surface modification on PTFE film could improve the interfacial polymerization behaviors on PTFE films and the properties of the obtained TFC membranes. The performance of

the prepared TFC membranes in pervaporation separation on IPA aqueous solutions is also examined [28–33].

#### 2. Experimental

#### 2.1. Materials

Poly(tetrafluoroethylene) films with a thickness of  $500\,\mu m$  and pore sizes of 0.2– $0.4\,\mu m$  were received from Yu-Min-Tai Co., Ltd., Taiwan. Commercial products of sodium 4-styrenesulfonate (NaSS, Fluka Chemie), acrylic acid (AAc, Acros Chimica), ethylene diamine (EDA, Acros Chimica), trimesoyl chloride (TMC, Aldrich Chemical Co.) were used as received. Other chemicals used in the chemical reactions are reagent grade. Preparation and characterization of poly(styrene sulfuric acid)-grafted PTFE films (PTFE-g-PSSA) and poly(acrylic acid)-grafted PTFE

Fig. 1. Reaction routes for (a) surface modification on PTFE films and (b) preparation of PTFE/PA thin-film composite membranes via interfacial polymerization.

(b) Interfacial polymerization of PA on PTFE surface

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