



Forward osmosis membranes with unprecedented water flux



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ABSTRACT

The internal concentration polarization is recognized as a major obstacle for forward osmosis (FO) membranes to achieve high water flux. Herein, porous substrates with vertical pores are first proposed to be used as supports for fabricating FO membranes in thin film composite structure. The addition of acetone in the aqueous phase facilitates the successful interfacial polymerization on the substrates with vertical pores. Positron annihilation lifetime analyses indicate that the fabricated FO membranes possess thicker and denser selective layer than traditional FO membranes with asymmetric substrates. These novel FO membranes have a low structure parameter, indicating a greatly reduced internal concentration polarization effect. They exhibit an unprecedented water flux up to $93.6 \text{ L m}^{-2} \text{ h}^{-1}$ when the selective layer is oriented towards the deionized water feed solution with 2 mol L^{-1} sodium chloride solution as draw stream. This performance outstands from those FO membranes reported in literatures and available commercially. The results demonstrate that substrates with vertically oriented porous structure are ideal supports for developing FO membranes with minimized or even eliminated internal concentration polarization and ultrahigh water flux.

1. Introduction

Increasing demand but scarcity of potable water and clean energy are the greatest challenges for our world with population growth, industry development, and environment pollution [1–3]. Various materials and technologies have been suggested to overcome these challenges in recent decades. Among them, osmotically-driven membranes and corresponding separation processes are those that can provide a sustainable solution for the global need of drinkable water and clean energy simultaneously [4–6]. Although they have been explored for several decades by using osmotic pressure difference between two streams with high and low salinity across a semipermeable membrane, lack of optimal separation membranes and draw solutions has hindered the competition superiority of forward osmosis/pressure retarded osmosis (FO and PRO) to those existing water purification/power generation technologies [7–9]. Plenty of research has been conducted to develop thin film composite (TFC) membranes in recent years, as the membrane is the core of FO and PRO processes for practical applications. However, typical TFC FO membranes always suffer from limited water flux, leading to low desalination efficiency and insufficient power density. It has been demonstrated that the internal concentration polarization (ICP), caused by the porous sup-

ports, is mainly responsible for this obstacle [10–12]. Qin *et al.* demonstrated that water flux can be reduced by as much as 99.9% in the presence of ICP [13]. Therefore, it is a great challenge to fabricate advanced TFC FO membranes with reduced or even eliminated ICP effect in the porous supports.

Theoretical models using the classical solution-diffusion mechanism have been developed to evaluate the relationship between the support structure and the ICP effect [14–16]. ICP is proportionate to the thickness and tortuosity of the supports in TFC membranes, and varies inversely with the support porosity. Current supports for TFC FO membranes are usually fabricated via phase inversion method and thus have a tortuous sponge-like porous structure. This porous structure is the major bottleneck to increase the water flux of corresponding TFC FO membranes [17]. Therefore, various FO membranes were suggested to overcome this bottleneck. Xiao *et al.* demonstrates a sacrificial-layer approach by co-casting to prepare a polysulfone support with open bottom surface morphology for fabricating TFC FO membranes [18]. The PSf membrane has a markedly low structure parameter value compared with conventional PSf membrane. Li *et al.* used SiO_2 nanoparticles as pore-generating template to generate a highly porous and interconnected-pore structure in the support layer, which results in the reduction of the ICP effect [19]. Chung and his coworkers

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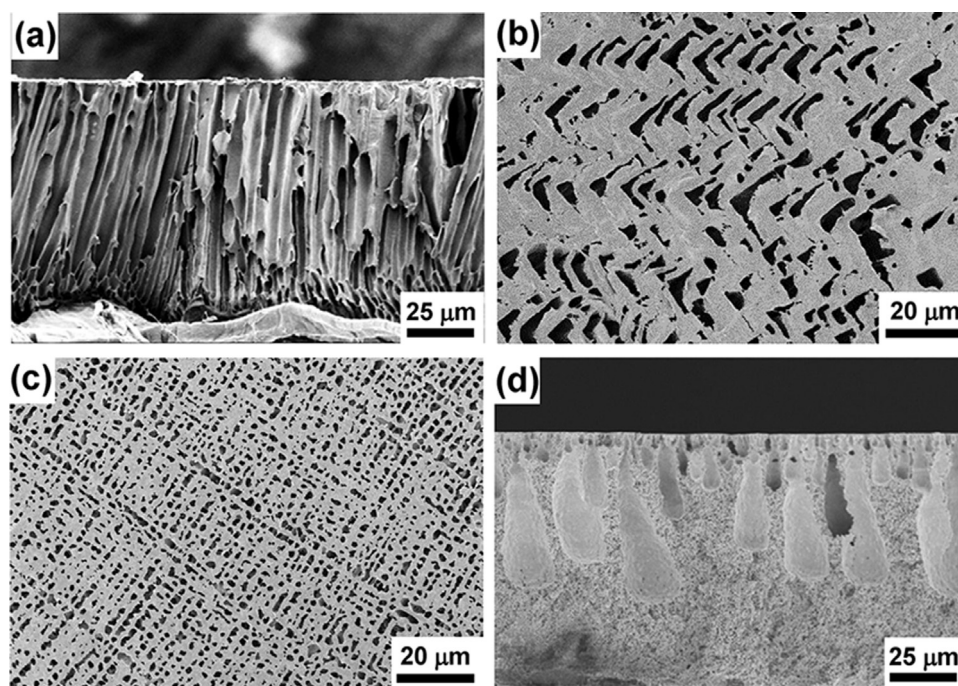


Fig. 1. SEM images of PVDF VOPS-1 (a) cross-section, (b) top surface, and (c) bottom surface. (d) SEM image from the cross-section of a conventional PVDF support prepared by the phase inversion method.

fabricated hydrophilic cellulose ester support with highly bicontinuous porous structure, which was found to reduce the ICP effect and to enhance the water flux effectively [20,21]. Great attentions were also paid to nanofibrous supports which possess low tortuosity, high porosity, and thin thickness [22–27]. Song et al. revealed these unique characteristics provide a good solution for reducing the ICP effect, and also guarantee direct paths for salt diffusion and water permeation [28]. Recently, graphene-based membranes were also studied experimentally and theoretically as potential candidates for FO processes [29–31]. Liu et al. fabricated freestanding ultrathin reduced graphene oxide (rGO) membranes by vacuum filtration method [31]. The results indicated that the ICP effect decreases to almost zero by using rGO membrane directly in a FO process, and the water flux is linearly proportional to the salt concentration. Though great efforts have been made to overcome the mentioned obstacles to some extent, ICP is still an extremely common phenomenon to significantly reduce water flux in FO processes.

We report here for the first time to use vertically oriented porous substrates (VOPSs) as supports for minimizing or eliminating the ICP effect of TFC FO membranes. The VOPSs are prepared by a bidirectional crystallization process of polymer/diluent mixtures at ambient temperature [32]. The vertical pores are constructed with crystallizable diluents as templates in a series of polymer matrices, based on our previous work [33]. This porous structure endows the TFC FO membranes with very low tortuosity, and provides direct paths for salt diffusion and water permeation. It is noteworthy that the TFC FO membranes fabricated on the VOPSs exhibit an unprecedented water flux up to $93.6 \pm 1.4 \text{ L m}^{-2} \text{ h}^{-1}$ in the FO mode using a draw solution of 2 mol L^{-1} NaCl and a feed of deionized water. The results provide an alternative avenue to minimize or even eliminate the ICP effects, which can greatly enhance the advancement of TFC FO membranes for drinkable water and clean energy.

2. Experimental

2.1. Materials

Poly(vinylidene difluoride) (PVDF, $M_n=110,000 \text{ g mol}^{-1}$, Solef

6010) was a commercial product of Solvay Solexis, Belgium. Dimethyl sulfone (DMSO2, 99%) was purchased from Dakang Chemicals Co., China. N-methyl-2-pyrrolidone (NMP) was supplied by Sigma-Aldrich. m-phenylenediamine (MPD, ACROS Organics, Belgium), trimesoyl chloride (TMC, TCI Co. Ltd, Japan), sodium dodecyl sulfate (SDS, Nihon Shiyaku Reagent, Japan), acetone (ECHO Chemical Co. Ltd, Taiwan), glycerol (ECHO Chemical Co. Ltd, Taiwan) and n-hexane (ECHO Chemical Co. Ltd, Taiwan) were used during interfacial polymerization and post-treatments. Sodium chloride (NaCl, Nihon Shiyaku Reagent, Japan) was used to prepare draw solution, and DI water was used as feed solution. Commercial TFC-ES membranes were purchased from Hydration Technology Innovations, USA. PVDF powders were dried at $60 \text{ }^\circ\text{C}$ under vacuum for 6 h before use, and other reagents were used without further purification.

2.2. Fabrication of vertically oriented porous substrates

PVDF VOPSs were prepared via bidirectional freezing of PVDF/DMSO2 solution using oriented DMSO2 crystal as templates, according to our previous work [33]. PVDF powder was first dissolved in DMSO2 at $160 \text{ }^\circ\text{C}$ to form homogeneous solution with a polymer concentration of 25 wt%. After degassing air bubbles, the solution was sealed in a home-made mould which was preheated in an oven at $160 \text{ }^\circ\text{C}$. The mould consists of two plates with thickness of 2 mm, one is made from stainless steel and another is from glass. A Teflon film with a circular opening (diameter=10 cm) was inserted between the two plates to hold the polymer solution and adjust substrate thickness. The two plates were clamped tightly by steel clips, whose surface was covered with Teflon tapes. The mould was then vertically put into a reservoir, and $30 \text{ }^\circ\text{C}$ water was added into the reservoir at a rate of 4 cm min^{-1} . After total solidification, the nascent substrate was taken out of the mould and immersed in deionized water to extract DMSO2. To gain a dry substrate, the wet one was washed with an ethanol–hexane sequence, and then dried in vacuum for 24 h at $60 \text{ }^\circ\text{C}$.

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