



## Polyethylene-supported high performance reverse osmosis membranes with enhanced mechanical and chemical durability

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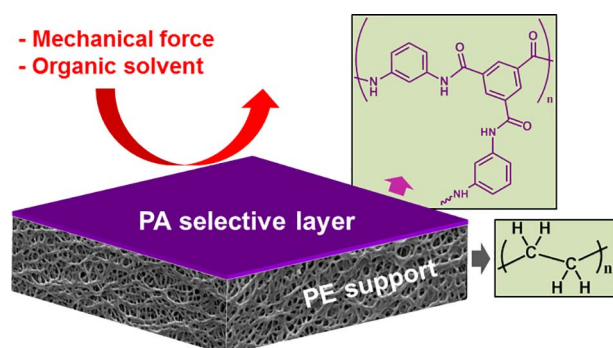
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### GRAPHICAL ABSTRACT



### ARTICLE INFO

#### Keywords:

Reverse osmosis  
Thin film composite membrane  
Polyamide  
Interfacial polymerization  
Polyethylene support

### ABSTRACT

A polyamide (PA) thin film composite (TFC) reverse osmosis (RO) membrane having high permselectivity and excellent mechanical/chemical durability was prepared using a polyethylene (PE) support. Although the PE support's uniform pores and high surface porosity are beneficial for enhancing membrane permselectivity, its intrinsic hydrophobicity makes the fabrication of a PA selective layer challenging. An oxygen plasma treatment on the PE support, combined with using a sodium dodecyl sulfate (SDS) during interfacial polymerization, allowed a PA layer to be formed on the support due to by enhancing its water wettability. The systematic optimization of the membrane fabrication parameters (e.g., plasma pretreatment, monomer and SDS compositions and post-heat treatment) achieved high membrane performance. The fabricated PE-supported membrane (TFC-PE) showed ~30% higher water flux with ~0.4% enhancement in NaCl rejection compared to a commercial RO membrane. Furthermore, the TFC-PE membrane had mechanical properties and organic solvent resistance superior to the commercial membrane, which is attributed to the excellent mechanical and chemical stability of the PE material. The proposed strategy could expand the application of RO membranes to mechanically and chemically harsh operating environments.

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## 1. Introduction

Over the past few decades, polyamide (PA) thin film composite (TFC) membranes, where a topmost PA selective layer is assembled on polymeric supports ( $\sim 100\ \mu\text{m}$  in thickness), have been extensively used as commercialized reverse osmosis (RO) membranes for desalination owing to their high permselectivities [1–3]. PA TFC membranes are fabricated by forming a PA selective layer through interfacial polymerization (IP) between *m*-phenylenediamine (MPD) and trimesoyl chloride (TMC) dissolved in two immiscible solvents on porous supports [4,5]. For the TFC membrane, the support determines its chemical and mechanical stability while the PA selective layer mainly controls its separation performance [6]. Although each membrane component can be independently chosen, the structure and chemistry of the support have a critical influence on the structure and thus separation performance of the formed PA layer, thus posing constraints on the selection of the support material [7,8]. For example, it is known that a support having moderate hydrophobicity is favorable for fabricating a highly permselective PA layer [9], which is consistent with the fact that commercial RO membranes are fabricated mostly using polysulfone (PSF) or polyethersulfone (PES) as a support material [10]. However, PSF or PES-supported TFC membranes have a technical limitation when applied to harsh environments due to their relatively low mechanical strength and poor chemical stability (or organic solvent resistance) [11,12]. To improve the mechanical and chemical durability of TFC membranes, various polymer materials including polypropylene (PP) [13–15], polyacrylonitrile [16,17], polyvinylidene fluoride [12,18], poly(tetrafluoroethylene) [19], polyimide [20] and sulfonated polyphenylsulfone [21] have been explored as supports.

Among them, PP, one of common polyolefins, has been considered a strong candidate for such supports owing to its high mechanical strength and excellent chemical durability. Although its strong hydrophobicity, one of the obstacles in fabricating TFC membranes, was resolved by adopting hydrophilic modifications such as plasma or chemical pretreatments [13–15], its irregular slit-like pore shape (an aspect ratio of  $\sim 2.8$ ), unevenly distributed surface pores and low surface porosity ( $\sim 11\%$ ) stood in the way of attempts to fabricate a defect-less and high flux PA layer. In fact, other researchers have shown that the fabricated PP-supported membrane had very low water flux and NaCl rejection ( $\sim 87\%$ ) far below RO performance level due to its unfavorable support pore structure [13].

Polyethylene (PE) is another class of polyolefins and exhibits remarkably high mechanical and chemical durability, like PP. PE has been manufactured into a porous membrane by sequential processes consisting of melt extrusion, mechanical stretching and diluent extraction, unlike PP membranes whose pore structures are formed by the simple mechanical stretching of an extruded film [22]. As a result, the porous PE membrane has higher porosity with a more regular pore shape than the PP membrane and thus has been successfully commercialized as a lithium ion battery separator [23,24]. This PE membrane also has beneficial structural features as a TFC membrane support together with its inherently strong mechanical and chemical stability. Its

relatively uniform pore structure and high surface porosity could facilitate the formation of a uniform and highly cross-linked PA selective layer via the IP process [25]. In particular, its high surface porosity could improve the mass transport at the PA layer-support interface, thereby enhancing the membrane water flux [8]. In addition, its highly interconnected, open pore structure could further improve membrane permeation [26–28].

Despite many potential advantages, the PE membrane has not been explored as a support material for the RO membrane. In this work, for the first time, we demonstrate that the commercialized, porous PE membrane can be used as a support to fabricate highly performing, mechanically strong and chemically durable RO membranes via conventional IP. The fabrication parameters, including the support pretreatment, monomer and additive compositions and post-heat treatment, were systematically controlled to achieve high separation performance of the PE-supported TFC (TFC-PE) membrane. The fabricated TFC-PE membrane had  $\sim 30\%$  higher water flux with  $\sim 0.4\%$  enhancement in NaCl rejection compared to a commercial RO membrane (SWC4+). In addition, it was demonstrated that the TFC-PE possesses superior mechanical robustness and organic solvent resistance compared to the commercial counterpart by characterizing the mechanical and chemical stability of the membranes. The structures and physicochemical properties of the prepared TFC membranes were comprehensively characterized using various analysis tools including scanning electron microscopy (SEM), atomic force microscopy (AFM), Fourier transform infrared spectroscopy (FT-IR), X-ray photoelectron spectroscopy (XPS) and contact angle measurement and correlated to their separation performance to understand the membrane structure-property-performance relationship.

## 2. Materials and methods

### 2.1. Materials

Trimesoyl chloride (TMC) and *m*-phenylenediamine (MPD) were purchased from Tokyo Chemical. Sodium dodecyl sulfate (SDS) and dodecane were obtained from Sigma-Aldrich. Tetrahydrofuran (THF), toluene and *n*-hexane were purchased from Daejung Chemical. Sodium chloride (NaCl) was obtained from Samchun Chemical. De-ionized (DI,  $18.2\ \text{M}\Omega\ \text{cm}$ ) water was supplied from a water purification apparatus (Millipore Milli-Q). A PE support ( $\sim 20\ \mu\text{m}$  in thickness) and a commercial RO membrane (SWC4+) were provided from SK Innovation and Hydranautics/Nitto Denko, respectively.

### 2.2. Membrane fabrication

The fabrication process of a TFC-PE membrane was depicted in Fig. 1. High hydrophobicity of the pristine PE support evidenced by its high water contact angle ( $\sim 120^\circ$ ) prevents the support pores from being impregnated with an MPD aqueous solution [25]. Additionally, weak chemical attraction of the support with the PA layer due to the absence of polar functional groups on the pristine PE hampers the formation of a

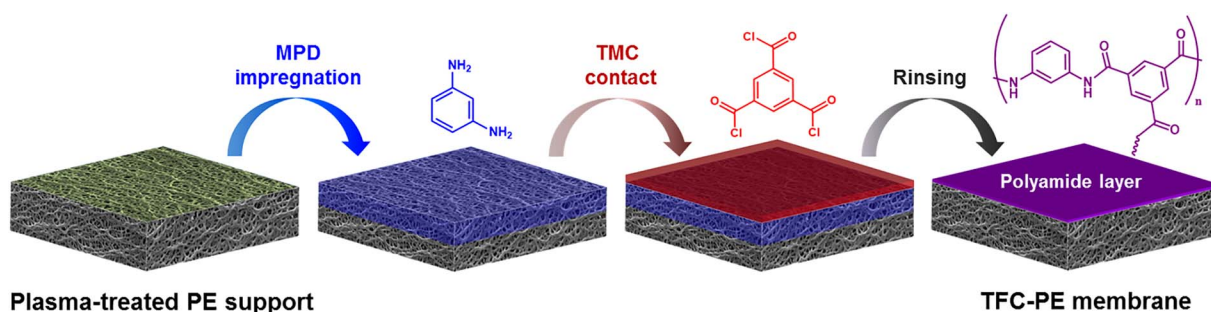


Fig. 1. Fabrication of the TFC-PE membrane via the IP process.

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