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Activation promoted ionic liquid modification of reverse osmosis membrane towards enhanced permeability for desalination

Jinglong Zhang^{a,d}, Zhenping Qin^{a,c}, Libin Yang^{a,c}, Hongxia Guo^{b,c,*}, Siyuan Han^a

^a College of Environmental and Energy Engineering, Beijing University of Technology, 100124 Beijing, PR China

^b College of Materials Science and Engineering, Beijing University of Technology, 100124 Beijing, PR China

^c Beijing Key Laboratory for Green Catalysis and Separation, Beijing University of Technology, 100124 Beijing, PR China

^d Beijing Scinor Membrane Technology Company Limited, 100083 Beijing, PR China

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ABSTRACT

The ionic liquid (1,3-dimethylimidazolium dimethyl phosphate ([MMIM][DMP])) synthesized by addition reaction was used to modify a commercial reverse osmosis (RO) membrane thorough activation method. The activation promoted modification process was completed within just 6–12 min under optimal condition of the immersion time of 4 min, activation temperature of 110 °C and activation time of 4 min. The variation of surface morphologies and properties of the RO membrane were characterized by SEM, AFM, water contact angle, FTIR, and the RO performance was evaluated by desalination of 1000 mg·L⁻¹ NaCl aqueous solution. The results showed that the active layer of the modified RO membrane became thinner, smoother, and more hydrophilic with increasing of activation temperature. The IL modified membrane achieved water flux of $33.7 \,\text{Lm}^{-2} \,\text{h}^{-1}$ and rejection of 93.2%, showing 62% higher water flux comparing to the pristine membrane and 2.6% decrease in salt rejection. Moreover, the IL modified membrane showed an enhanced anti-fouling property with 95.5% of normalized water flux recovery. This work provides a facile and efficient way to modify and improve separation performance and anti-fouling property of commercial thin film composite (TFC) RO membranes.

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1. Introduction

The global water scarcity together with the water pollution issues has become the world focus presently [1]. Membrane technology has been proven the most effective and environment-friendly method to purify water [2–5]. Reverse osmosis (RO) has been one of well-known membrane technology extensively used in desalination of abundant seawater and brackish water to produce drinking water and in other applications [6–8]. The polyamide (PA) thinfilm composite (TFC) membrane composed of a nonwoven support layer and a thin polyamide layer [9], has taken up over ninety percent of the RO membrane market due to its satisfactory water flux and salt rejection [10,11]. However, the disadvantages of high energy consumption, high operation pressure and high fouling tendency of RO membrane present challenges for its widespread application [12].

Various researches have been explored to develop better RO membrane for enhancing their energy efficiency and fouling re-

* Corresponding author at: College of Materials Science and Engineering, Beijing University of Technology, 100124, Beijing, PR China.

E-mail address: hxguo@bjut.edu.cn (H. Guo).

sistance [13-17]. For example, amino functionalized titanate nanotubes was embedded in the PA layer to prepare RO membrane, resulting in 93% enhancement in water flux compared to the control RO membrane [15]. The RO membrane incorporated with tannic acid modified graphene oxides nanoparticles exhibited higher water flux and simultaneously enhanced chlorine-resistant property and bio-fouling resistance than the pristine membrane [16]. Moreover, RO membrane embedded with hydrophilic clinoptilolite in the PA layer showed 39% flux improvement and 12% higher flux recovery ratio compared with the unmodified membrane [17]. Furthermore, hydrofluoric acid, critic acid, sulfuric acid and nitric acid can also be used to modify the polyamide layer of RO membrane, resulting in an order of magnitude higher water flux, considerable salt rejection and improved surface hydrophilicity [18]. However, these strategies may cause environmental problem due to the use of highly-volatile, poisonous or highly corrosive organic reagents during modification process.

lonic liquids (ILs) are low-melting salts, comprising of highly asymmetric organic cations paired with bulky inorganic anions. With unique advantages of unflammability, nontoxicity and low volatility [19–22], ILs are considered as an attractive replacement for the conventional environment-unfriendly organic reagents [23–27]. The anion groups of ILs, a strong hydrogen acceptor, can

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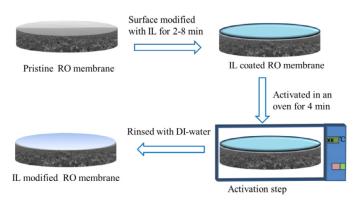
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J. Zhang et al./Journal of the Taiwan Institute of Chemical Engineers 000 (2017) 1-9

Nomenclature	
A Am B C ₀ C _f J J/Jo FRR R R R _{ir} R _r R _t t t ΔP $\Delta \pi$	water permeability coefficient $(Lm^{-2} \cdot h^{-1} \cdot bar^{-1})$; the effective filtration area of the membrane (m^2) ; salt permeability coefficient $(Lm^{-2} \cdot h^{-1})$; the concentration of the feed solution (g/l) ; the concentration of the permeate (g/l) ; the water flux of the RO membrane $(Lm^{-2} \cdot h^{-1})$; normalized water flux; flux recovery ratio (%); salt rejection (%); irreversible fouling resistance (%); reversible fouling resistance (%); total fouling resistance (%); filtration time (h); hydraulic pressure (MPa ⁻¹); the osmotic pressure of the feed (MPa ⁻¹);
riangle V	the volume increment of the permeate (L)

effectively destroy the inter- and intra-molecular hydrogen bonds of polymers [28], thereby being used to modify or dissolve polymeric membranes. Therefore, ILs are used as a kind of green solvent for polymers, *i.e.*, cellulose, chitosan and starch, through the formation of strong hydrogen bonds between polymers and IL anions [28]. As the ionization degree of ILs is greatly depended on the temperature [29], a high temperature promotes the interaction between ILs and polymer, and thus strengths the dissolution ability of ILs towards polymers. Recently, the high dissolution ability of the IL [BMIM]Cl has been also used to modify commercial RO membrane [30]. The modified membrane showed a thinner, more hydrophilic PA selective layer, achieving 20% increment of water flux compared to the pristine membrane [30]. Yet, this modification process is time-consuming due to the long immersion time of 48 h for RO membrane modification in [BMIM]Cl [31]. Furthermore, the long immersion time may induce the degradation of both the IL and the RO membrane [32], thereby hindering the scalability of RO membrane modification in IL.

Herein, we presented a fast and facile method to modify a commercial TFC RO membrane using 1,3-dimethylimidazolium dimethyl phosphate ([MMIM][DMP]) as the IL modification reagent. The modification was carried out through activation promotion, which is based on the fact that the dissolubility of ILs towards polymer is greatly enhanced with increasing temperature [28,29,32]. Thus, the effect of activation temperatures, the concentration of [MMIM][DMP] and immersion time on morphology and the surface property of the membranes was investigated. And the RO performance was evaluated by desalination of 1000 mg·L⁻¹ NaCl aqueous solution and the anti-fouling property was investigated by 500.0 mg·L⁻¹ BSA aqueous solution. The modification method in this work would provide a possibility to reduce the energy consumption of RO process in a facile and environmental friendly way.



[m5G;August 8, 2017;13:5]

Fig. 2. Schematic illustration of the activation promoted IL modification process.

2. Materials and methods

2.1. Materials

N-methylimidazole and trimethyl phosphate were obtained from Aladdin Industrial Corporation (China) and were used as the precursor of [MMIM][DMP]. Absolute ether and sodium chloride (NaCl) were purchased from Beijing Chemical Plant (China). The low-pressure RO membrane (DOW BW30LE) was supplied by Dow Chemical Company in United States. All the chemicals and materials were of analytical degree and used without further purification. Deionized water was produced by an ultrapure water system (Milli-Q, MA 01730, USA).

2.2. Synthesis of [MMIM][DMP] ionic liquid

The [MMIM][DMP] was synthesized using the method in Ref. [33]. Specifically, the equimolar amount of N-methylimidazole and trimethyl phosphate were added in a round bottom flask fitted with a reflux condenser, and stirred for 10 h at 150 °C to synthesize [MMIM][DMP] as Fig. 1. The obtained light brown liquid was thoroughly washed with ether, followed by vacuum evaporation to remove the residual ether. The final product was obtained and kept at ambient condition.

2.3. Surface modification of the RO membrane

The pristine RO membrane was first cut into round pieces and rinsed with deionized (DI) water for 24 h to remove the residual glycerinum before modification. Then IL modification of RO membrane was carried out according to the process shown in Fig. 2. Specifically, the pristine RO membrane with the active layer upward was first immersed in a certain concentration (12.5 –100.0 wt.%) [MMIM][DMP] for 2–8 min at 25 ± 1 °C. Then the [MMIM][DMP]-coated membrane was activated in an oven with a constant temperature of 25–140 °C for 4 min. Finally, the modified membrane was rinsed thoroughly with deionized water and stored in deionized water bath before use.

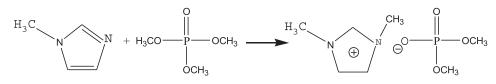


Fig. 1. Chemical reaction for synthesis of [MMIM][DMP].

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