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## Enhanced both water flux and salt rejection of reverse osmosis membrane through combining isophthaloyl dichloride with biphenyl tetraacyl chloride as organic phase monomer for seawater desalination

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#### ABSTRACT

The mixed organic monomers of isophthaloyl dichloride (IPC) and 3,3',5,5'-biphenyl tetraacyl chloride (BTEC) were used to prepare polyamide thin film composite (PA TFC) membranes for seawater desalination. IPC and BTEC act as a linear monomer and a cross-linking agent, respectively. As a result, the ratio of linear portion to cross-linking portion of polyamide film can be controlled by adjusting the fraction of monomer. It was found that the chemical composition, chain segment relaxation property, and the surface morphology of polyamide films changed significantly with the ratio of IPC to BTEC increasing. The rougher surface is formed while less pendant carboxylic acid groups present at higher IPC concentrations or lower BTEC concentrations. As the fraction of IPC increases, the length of linear segment between two cross linking point is increased, hence the enlarged polyamide network pore is formed. When IPC weight percentage increases from 0% to 70%, the water flux of the TFC membrane increases from 30 to 43.7 L/m<sup>2</sup> h and the salt rejection increases from 99.3% to 99.7% at 5.5 MPa, 32800 ppm NaCl,  $25 \pm 3$  °C testing condition. We demonstrated that both water flux and salt rejection of the polyamide membrane can be improved by the control of ratio of IPC to BTEC.

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#### 1. Introduction

Reverse osmosis (RO) process, removing salts and most of organics, has been used extensively in many fields such as for the desalination of seawater, ultrapure water production, and wastewater treatment [1,2]. In RO process, a high operating pressure is applied in the saline water feed on one side of the membrane in order to overcome osmotic pressure and achieve desirable water flux. This takes up 65–85% of total energy required in typical seawater RO (SWRO) desalination [3]. Obviously RO membrane plays a highly significant role in determining the economics of the RO process. Therefore, it is desirable to develop RO membranes having superior water flux and high salt rejection.

Most commercial RO membranes are composite and have a partially cross-linked aromatic polyamide active layer prepared by interfacial polymerization of m-phenyleneamine (MPDA) and trimesoyl chloride (TMC) [4]. The active layer is consisted of the crosslinked portion and the linear portion containing free pendant carboxylic acid from the partial hydrolysis of acyl chloride unit of TMC during the interfacial polymerization. It is widely accepted that the chemical structure of the active layer determines its physicochemical properties such as hydrophilicity, surface charge, polymer chain mobility and in turn influences the separation performance of the membrane [5–7]. The polymerization conditions such as monomer concentration [8,9], choice of solvents [10], temperature [11], and catalysts and additives [12] can influence the chemical structure of the active layer and further influence water flux and salt rejection [13]. The water flux can be improved by introducing hydrophilic monomers such as including sulfonic acid or carboxylic acid monomers [4,8] or forming a ultrathin active layer and ridge-andvalley roughness surface structure which increase the contact area between water and membrane [14–16] using additives [15,17–19].

Abbreviations: A, effective membrane area (m<sup>2</sup>); Ac, C=O stretch of carboxylic acid characteristic peaks area integration; A<sub>NH</sub>, amide II band bending characteristic peaks area integration; AFM, atomic force microscopy; ATR-IR, attenuated total reflectance infrared; BTEC, 3,3',5,5'-biphenyl tetraacyl chloride; *Cp*, concentration of the permeation(mg mL<sup>-1</sup>); C<sub>f</sub>, concentration of the feed (mg mL<sup>-1</sup>); CA, contact angle; *F*, water flux (L/m<sup>2</sup> h); IPC, isophthaloyl dichloride; L, liter; MPDA, *m*-phenyleneamine; PA, polyamide; R<sub>(j)</sub>, salt rejection (%); RO, reverse osmosis; SWRO, seawater RO; SEM, scanning electron microscope; T, time(h); T<sub>1p</sub>, spin-lattice relaxation time in rotation frame; TEA, triethyl amine; TMC, trimesoyl chloride; TFC, thin film composite; *V*, volume of permeate water (L); XPS, X-ray photoelectron spectroscopy

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The high salt rejection was achieved by increasing curing temperature [11] and using multiple functional monomers [20,21] to enhance the cross linking degree. However, there is a limit to optimize the membrane property by experimenting with a variety of interfacial polymerization conditions. For this reason, many researchers are seeking to select or design monomers or reactants for optimizing membrane separation performance. Perera et al.[22] introduced 1,3-diamino-2-hydroxypropane (DAHP), a linear hydrophilic amine, into aqueous phase and observed water flux enhanced with a litter reduction of salt rejection because of a more flexible backbone and a thinner active layer compared to membrane produced with MPD. Yu et al. [23] demonstrated that blending diacyl chloride such as isophthaloyl chloride with 5-chloroformyloxyisophthaloyl chloride (CFIC) was able to effectively improved polyamide-urethane TFC permeability while maintaining good rejection. Zhao et al. [24] added the hydrophilic additives, o-aminobenzoic acid-triethylamine(o-ABA-TEA) salt, into the aqueous m-phenylenediamine (MPDA) solution. Compared with MPDA, the cross-linked degree of PA TFC membrane reduces led to water flux increase with a slight decrease in salt rejection. But because the "trade-off" phenomenon [25] exits in the transport process, most studies improve the water flux through decreasing the rigidity and chain cross-linked degree with sacrificing salt rejection or improve the salt rejection with water flux decrease. Therefore, how to keep high salt rejection with excellent water flux is a question deserving to study further.

In the previous paper, we reported the preparation of RO membranes from biphenyl acyl chloride such as 3,3',5,5'-biphenyl tetraacyl chloride (BTEC), 2,3',4,5',6-biphenyl pentaacyl chloride (BPAC) and 2,2',4,4',6,6'-biphenyl hexaacyl chloride (BHAC), and found that the functionality of the acid chloride monomer strongly influenced the properties of the RO membrane [20,21]. As the functionality of the acid chloride monomer increased, the resulting membrane skin layer became more negatively charged, thinner, smoother, and less permeable than the membrane prepared by TMC. The decreased water flux could be contributed to the stiffness of polyamide containing biphenyl moieties and high degree of cross-linking. In this paper, attempting is made to enhance the performance, especially the water flux of biphenyl polyamide RO membrane by blending of 3,3',5,5'-biphenyl tetraacyl chloride (BTEC) and isophthaloyl chloride (IPC). The reaction path way is illustrated in Fig. 1. The introduction of isophthaloyl chloride is to increase the linear portion and decrease the cross-linked portion of the polyamide active layer. We show that the water flux and the salt rejection simultaneously increase with the content of IPC increasing. The effect of blending on the chemical composition, cross-linking, polymer chain segment relaxation property and the morphology of the polyamide active layer are investigated to explain the mechanism.

#### 2. Experimental

#### 2.1. Materials

The BTEC was synthesized as previously described [21]. *m*-phenylenediamine (MPDA, analytical grade) and isophthaloyl dichloride (IPC) were purchased from Tianjin guangfu fine chemical research institution. The preparation of microporous polysulfone support membrane followed the previous literature procedure [20].

#### 2.2. Preparation of TFC membrane

A rubber frame was firstly manufactured to fix the microporous polysulfone support membrane. The aqueous solution (a) including *m*-phenylenediamine (MPDA) (3.5% w/v) and camphor sulfonic acid was adjusted pH to 10.5 using triethyl amine (TEA). Then solution (a) was poured on the surface of polysulfone support membrane and immersed 2 min. Redundant solution was drained from the surface and the membrane was dried in air to no obvious droplets. After that the isopaG solution (b) (the total concentration was 0.2% w/v) of different BTEC and IPC concentration ratio was poured off, and the membrane was kept at 90 °C in an air dry oven for 6 min so that the reaction could continue to form more crosslinked structure. Finally, the TFC membrane was put into 1% Na<sub>2</sub>CO<sub>3</sub> solution for 2.5 min then kept in deionized water 24 h. All operations were finished at 30 °C thermostatic chamber.

#### 2.3. Preparation of polymer

The recipes were same to the preparation of TFC membrane, but interfacial polymerization was occurred in beaker. Then the polymer were washed by ethanol, methanol and deionized water three times to remove residual monomers and dried in vacuum oven at 40 °C, 24 h.

#### 2.4. Evaluation of membrane performance

The membrane samples used rhodamine aqueous solution to stain in order to avoid some obvious flaws before test. If there are defects, the membrane surface becomes pink. All tests for RO performance were conducted at 5.5 MPa using a 32800 ppm NaCl solution at  $25 \pm 3$  °C in cross-flow cells. Testing membrane samples were positioned in the test cell with the polyamide skin layer facing the incoming feed. The valid membrane area of each cell was about 15.79 cm<sup>2</sup>. The membranes sustained a pure water pressure of 4.0 MPa for 4 h before testing.

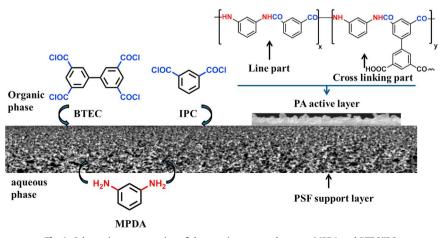


Fig. 1. Schematic representation of the reaction process between MPDA and BTEC/IPC.

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