



Separation performance of thin-film composite nanofiltration membrane through interfacial polymerization using different amine monomers



Yafei Li, Yanlei Su, Yanan Dong, Xueting Zhao, Zhongyi Jiang*, Runnan Zhang, Jiaojiao Zhao

Key Laboratory for Green Technology of Ministry of Education, School of Chemical Engineering and Technology, Tianjin University, Tianjin 300072, China
Synergetic Innovation Center of Chemical Science and Engineering (Tianjin), Tianjin 300072, China

HIGHLIGHTS

- Four kinds of amine monomers were used for nanofiltration membrane preparation.
- The membranes were prepared through interfacial polymerization.
- Amine solubility in organic phase greatly affects membrane separation performance.

ARTICLE INFO

Article history:

Received 11 July 2013

Received in revised form 21 October 2013

Accepted 19 November 2013

Available online 15 December 2013

Keywords:

Thin-film composite nanofiltration membrane

Interfacial polymerization

Amine monomers

Solubility

Separation performance

ABSTRACT

Four kinds of thin-film composite (TFC) membranes were prepared via interfacial polymerization using diethylenetriamine (DETA), triethylenetetramine (TETA), tetraethylenepentamine (TEPA) and piperazine (PIP) as water-soluble monomer, and trimesoyl chloride (TMC) as organic-soluble monomer. The surface chemical features of the resultant membranes were confirmed by contact angle measurement and Fourier transform infrared spectroscopy (FTIR). The membrane morphology and surface charges were investigated through Scanning electronic microscopy (SEM) and Zeta potential, respectively. Salt rejection was used to evaluate the separation performance of the four kinds of TFC membranes. The results showed that all the four kinds of TFC membranes exhibited typical negatively charged nanofiltration membrane characteristics. The salt rejections followed the sequence: $\text{Na}_2\text{SO}_4 > \text{MgSO}_4 > \text{MgCl}_2$ and the rejection of Na_2SO_4 was all over 80%. It was also found that the solubility of water-soluble monomer in organic solvent played an important role in manipulating the membrane structure, charge properties and thus the separation performance.

© 2013 Elsevier B.V. All rights reserved.

1. Introduction

With the continuing decline of available freshwater supplies and the increasing demand of freshwater due to population booming and worldwide industrialization, membrane technologies have gained great attention in recent years [1]. Nanofiltration, a process intermediate between reverse osmosis and ultrafiltration, enables higher flux and lower operation pressure than reverse osmosis and could afford high rejection of organic molecules with molecular weight cutoff (MWCO) ranging from 100 to 1000 Da [2]. Since the late 1980s, the application range of nanofiltration has extended tremendously from water softening to drinking water production and removal of pesticides, endocrine disruptors and dyes [3–8]. Among the various nanofiltration membrane fabrication techniques, interfacial polymerization (IP) has

been developed as a dominating method for preparing polyamide TFC nanofiltration membranes [9]. Due to the negligible solubility of organic-soluble monomer in water and the fairly good solubility of water-soluble monomer in the organic solvent, the interfacial polymerization reaction usually takes place at the organic phase side near the interface [10].

The interfacial polymerization process is self-inhibiting through a limited supply of reactants to the already formed layer, resulting in an extremely dense thin film of thickness from 50 nm to several hundred nanometers [11]. The thin film formed on the top of porous support will determine the overall solute retention and permeate flux, which can be optimized by varying the type and concentration of monomer in each solution, monomer ratios, and reaction time of the interfacial polymerization [12]. Considerable attempts have been made to improve the comprehensive performance of TFC nanofiltration membrane. Seman et al. [11] prepared polyester TFC nanofiltration membranes with improved antifouling properties by interfacial polymerization using bisphenol A and trimesoyl chloride (TMC) monomers. Zhang et al. [13] selected the interfacial reaction between Tannic acid and

* Corresponding author at: Key Laboratory for Green Technology of Ministry of Education, School of Chemical Engineering and Technology, Tianjin University, Tianjin 300072, China. Tel.: +86 22 27406646; fax: +86 22 23500086.

E-mail address: zhyjiang@tju.edu.cn (Z. Jiang).

TMC and the prepared composite nanofiltration membranes had excellent anti-fouling properties and chlorine resistance. An et al. [14] synthesized a new type of zwitterionic amine (AEPPS) and prepared novel nanofiltration membranes through interfacial polymerization using the blend of piperazine and AEPPS as aqueous phase monomer. The resultant blend membranes had better antifouling properties than the control membrane. Soroush et al. [15] prepared several polyamide composite membranes by interfacial polymerization of *m*-phenylene diamine (MPD) and TMC. They investigated the effect of polymerization time on active layer thickness, morphology, structure, hydrophilicity, roughness and thermal properties.

As the structure of the thin-film was derived from the interplay between the diffusion and reaction of the monomers, the solubility and diffusivity of the monomers during the interfacial reaction might affect the morphology and network structure, and ultimately the separation performance of the membrane. In this study, four kinds of water-soluble monomers, diethylenetriamine (DETA), triethylenetetramine (TETA), tetraethylenepentamine (TEPA) and piperazine (PIP) were selected to react with organic-soluble monomer trimesoyl chloride (TMC) to fabricate the TFC membranes for nanofiltration separation. The separation properties of the composite membranes were evaluated through dead-end nanofiltration of dye aqueous solution and salt aqueous solution and the antifouling properties of the TFC membranes were evaluated using orange GII aqueous solution as a model system. The resultant TFC membranes displayed excellent antifouling properties and separation performance, which had potential applications in dye wastewater treatment.

2. Materials and methods

2.1. Materials

Trimesoyl chloride (TMC, or 1,3,5-benzenetricarbonyl trichloride) was purchased from Alfa Co. (USA). Diethylenetriamine (DETA), triethylenetetramine (TETA), tetraethylenepentamine (TEPA), and piperazine (PIP) were all received from Guangfu Fine Chemical Research Institute (Tianjin, China). Polyethersulfone (PES, Ultrason E6020P) purchased from BASF Co. (Germany) was dried at 110 °C for 12 h before used. Poly(ethylene glycol) (PEG) (MW = 2000 g/mol), *N,N*-dimethyl formamide (DMF), *n*-heptane and the inorganic salts including sodium sulfate and magnesium sulfate were purchased from Kewei Chemical Reagent Co. (Tianjin, China). Organic anionic dyes including tracid brilliant red 10b, Congo red, orange GII and methyl orange were purchased from Dingguo Biological Technology Co. (Beijing, China). The chemical structures of TMC and polyamines are shown in Table 1. The water used was RO water with pH 6.5.

2.2. Preparation of the TFC membrane

PES ultrafiltration membranes were used as the porous support. The detailed fabrication process of PES support had been described in our previous works [16]. The support membrane was firstly immersed into an aqueous solution of amines (DETA, TETA, TEPA and PIP) with amine concentrations of 0.20 wt.% and allowed for 10 min immersion. Then the membrane was taken out and excess solution was drained off with filter papers. The organic phase prepared by dissolving 0.20 wt.% TMC in *n*-hexane was subsequently introduced to cover the surface of the impregnated membrane for 2 min, which resulted in the formation of an ultra-thin film over the surface of PES support. Afterwards, the resultant composite membrane was air dried for 30 min to allow the *n*-heptane to evaporate and subsequently post-treated for attaining the desired stability of the formed structure. Finally, the resultant membrane was rinsed with RO water and kept in RO water before carrying out evaluation studies. To facilitate the discussion, the four composite membranes were designated as DETA/TMC, TETA/TMC, TEPA/TMC and PIP/TMC.

2.3. Characterization of the TFC membranes

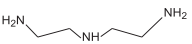

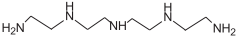

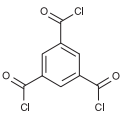
The cross sectional and surface morphologies of the TFC membranes were characterized by a field emission scanning electron microscope (FESEM, Nanosem 430) measurement. The membrane samples frozen in liquid nitrogen were broken and sputter with gold prior to SEM observation.

FTIR spectra of PES support and the four TFC membranes were recorded using an attenuated total reflectance (ATR) technique with a spectrometer (Thermo Nicolet, Nicolet 6700) in the range of 4000–500 cm^{-1} . The experiments run with air as the background. Scans were accumulated with a resolution of 2 cm^{-1} for each spectrum.

The hydrophilicity of the PES support substrate and TFC membrane surface was determined by conducting the static contact angle measurement with a contact angle goniometer (JC2000C Contact Angle Meter, Powereach Co., Shanghai, China) at temperature 25 ± 0.5 °C. At least five contact angles at different locations on one surface were averaged to get a reliable value.

The Zeta potential was determined from streaming potential measurements by an Electro Kinetic Analyzer (Austria/Anton Paar KG, Austria) equipped with a plated sample cell. The measurements were carried out at temperature 25 ± 0.5 °C in KCl solution (0.001 M, pH 6.5). Prior to use, the samples were conditioned in 0.001 M KCl solution for at least 24 h. The resultant Zeta potential was calculated using the Helmholtz–Smoluchowski equation.

Table 1
Molecular structures of the monomers involved.

Abbreviation	Monomers	Molecular structure	Solubility parameters ($\text{MJ}^{1/2} \cdot \text{m}^{-3/2}$)
DETA	Diethylenetriamine		25.8
TETA	Triethylenetetramine		22.7
TEPA	Tetraethylenepentamine		20.7
PIP	piperazine		16.8 ^a
TMC	trimesoyl chloride		-

^a Received and predicted by group contribution method.

Download English Version:

<https://daneshyari.com/en/article/623757>

Download Persian Version:

<https://daneshyari.com/article/623757>

[Daneshyari.com](https://daneshyari.com)