



Improved separation performance and durability of polyamide reverse osmosis membrane in tertiary treatment of textile effluent through grafting monomethoxy-poly(ethylene glycol) brushes

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ABSTRACT

To improve the separation performance and durability of the aromatic polyamide thin-film composite reverse osmosis membrane in tertiary treatment of textile effluent, grafting of hydrophilic polymer brushes was performed in this work through sequential surface treatment using acidic aqueous glutaraldehyde and monomethoxy-poly(ethylene glycol) (MPEG) solutions. ATR-FTIR and XPS analyses confirmed that MPEG brushes were grafted on membrane surface at the sites of N–H group of amide linkages and end amino groups in the polyamide active layer. The modification was found to enhance membrane fouling resistance, water permeability and rejections to both COD and conductivity in tertiary treatment of textile effluent. The steady-state flux of the modified membrane to secondary textile effluent was higher by 24.0% compared with the virgin membrane and comparable with that of the commercial membrane BW30FR. The grafting of MPEG brushes was also found to make the membrane less susceptible to chlorine. After ten times of intensified chlorine exposure, i.e. total chlorine exposure of 42,000 ppmh NaClO of 35.0 °C and pH 9.0, the membrane grafted with MPEG brushes maintained COD and conductivity rejections of 97.9% and 98.5%, respectively, to textile effluent, showing better durability against chlorine than the virgin membrane and compared membrane BW30FR.

1. Introduction

Aromatic polyamide-based thin-film composite (TFC) reverse osmosis (RO) membranes have been widely used in the fields of seawater desalination and wastewater treatment due to their relatively good chemical stability and high rejection abilities to the impurities in aqueous like salts and dissolved organic compounds [1–3]. However, the polyamide-based membranes are prone to fouling in treatment of wastewater due to the inherent physico-chemical properties of the polyamide active layer [4–6]. Membrane fouling usually deteriorates membrane separation performance, increases the cost of running and shortens the service life of membrane. Additionally, in tertiary treatment of industrial effluents using reverse osmosis process with polyamide-based TFC membrane, disinfection through chlorination is often performed prior to RO membrane to mitigate the growth of micro-organism [7,8]. Any operational failures of dechlorination will allow for a direct contact of chlorine with the polyamide-based TFC RO

membranes and thus leading to performance degradation, since the polyamide-based RO membranes are readily degraded by chlorine [9,10]. Therefore, the improvements of membrane antifouling property and durability against chlorine are of great importance for the extensive applications of the polyamide-based reverse osmosis membranes in the treatment and reuse of industrial effluents.

Surface modification has been regarded as the most feasible route to enhance the fouling resistance of the polyamide-based RO membranes while maintaining their separation performance [11,12]. Methods of surface coating [13,14] and grafting [15–17] have been proved effective in mitigating the deposition of foulants on membrane surface through tuning membrane surface properties including chemical composition, hydrophilicity, charge as well as roughness. Physical coating is relatively simple while chemical grafting is desirable for the long-term stability of the modified membrane. Of the chemical modification approaches, the grafting of hydrophilic polymer brushes have showed good prospect in improving membrane antifouling ability [18–20].

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However, the reported works on the grafting of polymer brushes were all performed through consuming the carboxylic acid groups on membrane surface, which usually leads to a declined salt rejection.

In recent years, surface modification has also been explored to protect the polyamide-based RO membranes from chlorine degradation. The coating of a protective and/or sacrificial layer on membrane surface can retard or prevent the hydrogen atoms on the site of amide nitrogen (N–H) and end amino groups ($-\text{NH}_2$) of the polyamide backbone from being attacked by free chlorine [21–23], while the grafting at the sites of N–H bonds of amide linkages ($-\text{CONH}-$) of the polyamide active layer can reduce the chlorine susceptible sites in the polyamide layer and thus the N-chlorination to form N-chloroamide [24–26]. The modification of grafting is preferred for the robustness of the modified membrane, while the use coupling agents like glutaraldehyde are relatively simple and easy to be industrialized [27,28].

On the base of above analysis, in order to improve the performance and durability of the aromatic polyamide thin-film composite RO membrane in tertiary treatment of industrial effluent, a novel idea was proposed in this study to improve the fouling resistance and chlorine stability of the polyamide TFC RO membrane through grafting hydrophilic polymer brushes on membrane surface at the sites of chlorine-susceptible N–H bonds of amide linkages in the polyamide active layer. The idea was implemented simply through treating the polyamide TFC membrane sequentially with acidic aqueous solutions of glutaraldehyde (GA) and monomethoxy-poly(ethylene glycol) (MPEG) as schematically illustrated in Fig. 1. Unlike the modification using polyvinyl alcohol (PVA) that formed a dense coating layer on membrane surface [28], the monomethoxy-poly(ethylene glycol) (MPEG) molecules having one end hydroxyl group will form brushes and loosely deposit on membrane surface and thus endow the membrane with improved antifouling property and water permeability in tertiary treatment of industrial wastewater. Additionally, unlike the grafting of polymer brushes at the sites of carboxylic acid groups on membrane surface, the modification performed in this work will not consume the carboxylic acid groups on membrane surface. Membrane surface physico-chemical properties were characterized using ATR-FTIR, XPS, AFM, SEM, water contact angle and streaming potential analyzers. The antifouling property, chlorine stability and separation performance in tertiary treatment of industrial effluent of the polyamide TFC RO membrane grafted with MPEG brushes were evaluated through cross-flow permeation tests and compared with the virgin membrane and commercial antifouling membrane BW30FR of Dow.

2. Experimental

2.1. Materials and reagents

Flat-sheet aromatic polyamide TFC RO membrane for modification was supplied by Hangzhou Tianchuang Environmental Technology Co., Ltd. (Hangzhou, China). Commercial antifouling polyamide TFC RO membrane BW30FR for comparison was provided by DOW Chemical Co. Ltd. (Minneapolis, MN, USA). Monomethoxy-poly(ethylene glycol) (MPEG) with a molecular mass of about 1000 g/mol was purchased from Aldrich. Glutaraldehyde (GA) was purchased from Tianjin Kemiou Chemical Reagent Co., Ltd. (Tianjin, China). Chlorination agent sodium

Table 1

Properties of the textile effluent used in the experiment.

Parameter	Value
Conductivity ($\mu\text{S}/\text{cm}$)	2780 ± 25
Chemical oxygen demand (COD, mg/l)	108.5 ± 2.2
pH	7.2 ± 0.2

hypochlorite solution (NaClO , 10.0 wt% effective chlorine) was purchased from Gaojing Fine Chemical Co., Ltd. (Hangzhou, China). De-ionized water with the conductivity less than $5.0 \mu\text{S}/\text{cm}$ was used to prepare aqueous solutions and to rinse membrane samples. The textile effluent for tertiary treatment has been treated through biological treatment followed by ultrafiltration and its typical properties were tabulated in Table 1. All other reagents were of analytical grade and used as received.

2.2. Membrane modification

Membrane surface modification was conducted in an assembly clean room. All the virgin membrane samples for modification were rinsed thoroughly to remove all preservative materials through soaking with de-ionized water of 50.0°C for at least 10.0 h. Modification was implemented through the following five steps of (1) Contacting the surface of the virgin membrane with 0.1 wt% GA aqueous solution of room temperature and pH 3.0 adjusted with H_2SO_4 for 5 min; (2) Draining off the excess solution and rinsing the GA-coated membrane thoroughly with de-ionized water to remove all the un-reacted GA molecules (the membrane thus obtained was designated as PA-GA); (3) Pouring an aqueous MPEG solution of room temperature, pH 3.0 adjusted with H_2SO_4 and a concentration varying from 0.5 to 2.0 wt% on membrane surface for a residence time of 5 min; (4) Draining off the excess solution and holding the membrane in a hot air dryer of 50.0°C for 5 min; (5) Soaking and rinsing the membrane sample with de-ionized water of 50.0°C for at least 10.0 h. Membranes modified with 0.5, 1.0, 1.5 and 2.0 wt% MPEG aqueous solutions were accordingly designated as PA-GA-MPEG0.5, PA-GA-MPEG1.0, PA-GA-MPEG1.5 and PA-GA-MPEG2.0, respectively.

In order to differentiate the physical adsorption of MPEG on membrane surface, modification was also performed with the virgin membrane through the above steps of (3), (4) and (5) with an aqueous MPEG solution of 2.0 wt%. The membrane thus obtained was designated as PA-MPEG.

2.3. Membrane characterization

Membrane surface chemical structure and composition was analyzed by using Fourier transform infrared spectroscopy (Nicolet Ararat 370 FTIR spectrometer) with an attenuated total reflectance (ATR) unit (ZnSe crystal, 45°) and X-ray photoelectron spectroscopy (XPS, PerkinElmer PHI 5000C ESCA System with Mg/Al Dual Anode Hel/Hell ultraviolet source, USA). Membrane surface morphology was characterized by using a field emission scanning electron microscopy (FE-SEM, Hitachi S-4800) at the magnification of $10,000\times$ and an atomic force microscopy (AFM, Park Instrument Auto Probe CT) under the

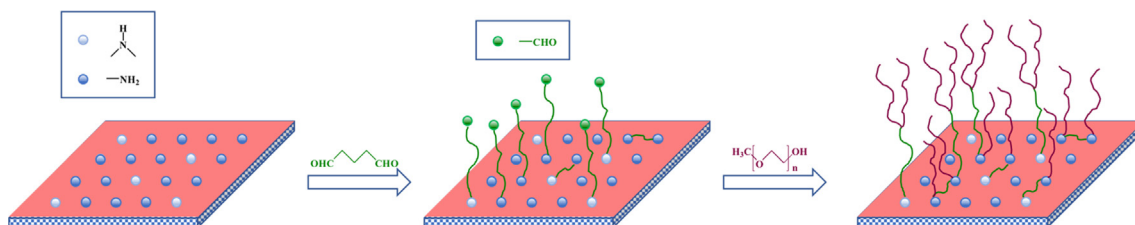


Fig. 1. Schematic for the grafting of polymer brushes onto the surface of polyamide membrane via using GA and MPEG.

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