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Journal of Membrane Science

journal homepage: www.elsevier.com/locate/memsci

Enhancing the performance of aromatic polyamide reverse osmosis membrane by surface modification via covalent attachment of polyvinyl alcohol (PVA)

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ARTICLE INFO

Article history:

Received 18 September 2015

Received in revised form

26 October 2015

Accepted 1 December 2015

Available online 4 December 2015

Keywords:

Reverse osmosis membrane
Aromatic polyamide thin-film composite membrane
Membrane surface modification
Antifouling property
Chlorine stability

ABSTRACT

Surface modification is a promising way to improve membrane performance. In this study, a commercial aromatic polyamide thin-film composite reverse osmosis membrane was modified through sequential surface treatment with glutaraldehyde aqueous solution followed by polyvinyl alcohol (PVA) aqueous solution. ATR-FTIR spectroscopy, SEM, AFM, measurements of streaming potential and contact angle and cross-flow permeation tests were employed to investigate the influence of modification on membrane performance. It was illustrated that PVA molecules were covalently attached on the surface of the pristine membrane and both membrane surface physico-chemical and permeation properties could be tuned through changing the PVA content. The covalent attachment of PVA resulted in an improved surface hydrophilicity, a declined surface negative charge and a slightly increased surface roughness, and could enhance membrane salt rejection and water flux simultaneously. The modification was proved to be effective in improving membrane antifouling property to the foulants of bovine serum albumin, sodium dodecyl sulfate and dodecyltrimethyl ammonium bromide through increasing anti-adsorption capability and in enhancing membrane chlorine stability through reducing chlorination sites and preventing the underlying polyamide backbones from chlorine attack. Furthermore, the modification could effectively enhance the membrane rejection performance and antifouling property in tertiary treatment of industrial effluent.

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1. Introduction

Reverse osmosis (RO) membrane process, which uses polymeric semi-permeable membrane to achieve molecular separation, has become the most promising technology for transforming saline, brackish and contaminated water into a useable and/or potable product [1–3]. Aromatic polyamide (PA) thin-film composite (TFC) membranes are the state-of-the-art polymeric membranes for the process of reverse osmosis because of their high salt rejection and water permeability as well as their excellent chemical, thermal, and mechanical stability [4,5]. However, the major hindrance to the effective application of reverse osmosis technology in the reclamation of wastewater is membrane fouling,

which decreases water productivity, deteriorates permeate quality, and shortens membrane lifespan [6,7]. Approaches of pretreatment of feed solution and chemical cleaning of the fouled membranes that are currently used to cope with membrane fouling can significantly increase the operational cost [8–11]. Furthermore, the chlorine disinfectants that are commonly used to control membrane biofouling can also react with the PA active layer of the membrane and result in material degradation and associated performance deterioration [12,13]. Therefore, it is essential to design membranes that possess optimized surface properties and are less prone to fouling and susceptible to chlorine disinfectants.

Many efforts have been devoted to improve both antifouling property and chlorine stability of the PA-based RO membranes through appropriate surface modifications including physical and chemical treatments that can alleviate the deposition of foulants and the attack of chlorine on the polyamide chains simultaneously. For example, physical modifications of surface layer-by-layer (LBL) deposition of oppositely charged GO nanosheets [14] and surface

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coating of the terpolymer of poly(methylacryloxy ethyldimethyl benzyl ammonium chloride-*r*-acrylamide-*r*-2-hydroxyethyl methacrylate) (P(MDBAC-*r*-Am-*r*-HEMA)) [15] have been reported to be effective in improving membrane resistance to both fouling and chlorine. Chemical surface modifications of free-radical graft polymerization of 3-allyl-5,5-dimethylhydantoin (ADMH) and N,N'-Methylenebis (acrylamide) [16], controlled redox-initiated graft polymerization of N-isopropylacrylamide (NIPAm) followed by acrylic acid (AA) [17], Ce(IV)/Polyvinyl alcohol (PVA) redox-mediated graft copolymerization of 3-sulphopropyl methacrylate (SPMA) and methylene-bis-acrylamide (MBA) [18], and thermal-initiated graft of polyvinyl alcohol (PVA) [19] have been demonstrated to effectively improve the fouling resistance and chlorine stability of the PA-based RO membranes. However, to our knowledge, it is still very challenging to improve the chlorine stability and antifouling property of the PA-based TFC RO membrane simultaneously while having no negative effect on membrane flux and/or salt rejection.

Therefore, in this study, we attempted to explore a novel and simple modification approach of good controllability to improve the resistance of the PA-based TFC RO membrane to both fouling and chlorine while enhancing or maintaining membrane separation performance. Method of sequential surface treatment with glutaraldehyde (GA) aqueous solution followed by polyvinyl alcohol (PVA) aqueous solution was adopted to covalently attach the neutral hydrophilic polymer molecules onto the surface of the PA TFC membrane. As schematically depicted in Fig. 1, GA molecules were firstly bonded on membrane surface through the reaction between the aldehyde group of GA and the amide linkages or unreacted end amino groups of polyamide backbones, then PVA macromolecules were bonded onto the membrane surface through the reaction between the unreacted aldehyde group of the bonded GA molecule and the hydroxyl groups of the PVA molecules. The covalent attachment of PVA molecules would form a durable protective layer to impede the deposition of foulants on membrane surface and to prevent the attack of chlorine on the polyamide chains.

In the experiments, membrane surface property and performance were tuned through varying the PVA content in aqueous solution. The physico-chemical properties of the pristine and modified membranes such as surface chemical structure, morphological structure, hydrophilicity, and charge were characterized by ATR–FTIR spectroscopy, SEM, AFM, and measurements of water contact angle and streaming potential. Membrane permeation properties were evaluated through cross-flow permeation tests. The fouling behaviors of the pristine and modified membranes were investigated and compared through cross-flow fouling

experiments with model foulants of bovine serum albumin (BSA), sodium dodecyl sulfate (SDS) and dodecyltrimethyl ammonium bromide (DTAB) by measuring the time-dependant water flux. The reverse osmosis performance and antifouling property of the modified membrane were also evaluated in tertiary treatment of industrial effluent and compared with the widely used antifouling membrane BW30FR of Dow. Finally, membrane chlorine stability was evaluated through soaking tests using sodium hypochlorite aqueous solution.

2. Experimental

2.1. Materials

A commercially available ultralow-pressure flat-sheet aromatic polyamide (PA) thin-film composite (TFC) reverse osmosis (RO) membrane supplied by Hangzhou Tianchuang Environmental Technology Co., LTD. (China) was used as the pristine membrane (M0) for modification in this work. The neutral hydrophilic polymer polyvinyl alcohol (PVA) with an average molecular weight of 22,000 g/mol and an alcoholysis degree of 88% was purchased from Acros Organic. Glutaraldehyde (GA) purchased from Tianjin Kemiou Chemical Reagent Co., Ltd. (China) was used as the linkage agent. Bovine serum albumin (BSA), sodium dodecyl sulfate (SDS) and dodecyltrimethyl ammonium bromide (DTAB) purchased from Sigma-Aldrich were used as the model foulants. The industrial effluent for tertiary treatment was taken from a textile factory located in Zhejiang province of China and has been subjected to biological treatment and ultrafiltration. The typical characteristics of this effluent are shown in Table 1. Sodium hypochlorite solution (NaClO, 10 wt% free chlorine) for chlorine exposure experiments was purchased from Hangzhou Gaojing Fine Chemical Co., Ltd. (China). De-ionized water with conductivity less than 5×10^{-4} S/m was used to prepare the aqueous solutions and to soak and rinse membrane samples. Other reagents such as sulfuric acid, sodium chloride (NaCl) and potassium chloride (KCl) were of analytical grade and used without further purification.

2.2. Modification of the pristine PA TFC membrane

Modification of the pristine PA TFC membrane was conducted through sequential treatment with GA aqueous solution followed by PVA aqueous solution in an assembly clean room. Prior to modification, all the membrane samples of length 20 cm and width 12 cm were soaked in de-ionized water of 40.0 °C for at least 15.0 h, replacing the water every hour, and then rinsed thoroughly

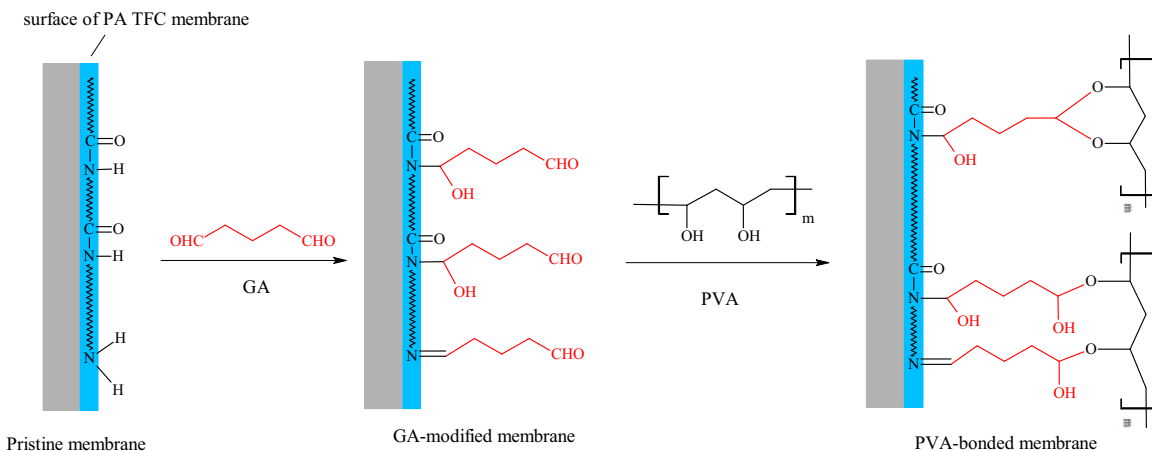


Fig. 1. Schematic diagram for the covalent attachment of PVA molecules onto the surface of polyamide membrane with GA as a linkage agent.

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