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Novel reverse osmosis membranes composed of modified PVA/Gum Arabic conjugates: Biofouling mitigation and chlorine resistance enhancement

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ABSTRACT

A novel crosslinked Poly (vinyl alcohol) (PVA) reverse osmosis (RO) thin film membrane conjugated with Gum Arabic (GA) with superb performance and features was synthesized for water desalination. RO membrane desalination parameters, such as hydrophilicity, surface roughness, water permeability, salt rejection, Chlorine resistance and biofouling resistance were evaluated using a dead end RO filtration unit. The incorporation of Pluronic F127 and the conjugation of Gum Arabic improved the overall RO performance of the membranes. This study has shown that the membrane PVA-GA-5 that contains 0.9 wt% Gum Arabic provided excellent permeation, salt rejection, Chlorine and biofouling resistance and mechanical strength. The most remarkable result to arise from this research is that the overall RO performance enhancement has been achieved while utilizing PVA/Gum Arabic as a separation layer without the use of a substrate, which eliminates negative effects associated with the use of a substrate like internal concentration polarization.

1. Introduction

Desalination and purification of seawater and brackish water utilizing reverse osmosis (RO) technology has been progressively crucial in an effort to tackle the issue of the calamitous global fresh water resources scarcity (Drioli & Giorno, 2010; Greenlee, Lawler, Freeman, Marrot, & Moulin, 2009; Lee, Arnot, & Mattia, 2011; Perera et al., 2014). Due to the up-to-date advances in RO membrane technologies and their reduced cost compared to thermal desalination methods, RO became the predominant desalination technology nowadays (Quevedo et al., 2011).

One of the challenges that affect the RO process is membrane biofouling that causes a significant decline in performance. Due to membrane fouling, RO modules need to be cleaned and chemically treated frequently. Cleaning and pretreatment ultimately shorten the membrane life, and that increases the cost of the overall process (Baker & Dudley, 1998; Herzberg & Elimelech, 2007). Membrane biofouling is the buildup of microbial layers on the surface or within the pores of the membrane. Micro-organisms such as bacteria, algae and fungi are pseudo particles, which means that they can grow, multiply or even relocate. Hence, biofouling occurs even after feed water pretreatment and after the application of disinfectants (Abd El Aleem, Al-Sugair, & Alahmad, 1998; Baker & Dudley, 1998; Flemming, Schaule, Griebe, Schmitt, & Tamachkiarowa, 1997; Flemming, 2002; Goosen et al., 2004; Kim, Jung, Sohn, Kim, & Lee, 2009; Matin, Khan, Zaidi, & Boyce, 2011). The buildup of a biofilm on the surface of the membrane causes several adverse effects including, operating pressure increase, loss of salt

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rejection, membrane bio-degradation and flux decline (Ridgway, 1988; Ridgway & Safarik, 1990).

Poly (vinyl alcohol) (PVA) is a highly hydrophilic water-soluble biodegradable polymer with good film-forming properties and outstanding thermal, mechanical and chemical stability. It is considered as one of the best polymers to fabricate membranes with high chemical stability, good Chlorine tolerance and excellent fouling resistance. Subsequently, PVA has been utilized by many researchers to fabricate membranes for separation and pressure driven processes such as microfiltration, ultrafiltration, reverse osmosis and pervaporation (Bezuidenhout, Hurndall, Sanderson, & van Reenen, 1998; Bolto, Tran, Hoang, & Xie, 2009; Hu, Zhang, Lawless, & Feng, 2012; Liu, Wang, Ma, Hsiao, & Chu, 2013; Liu, Zhou et al., 2014; Yee, Ong, Mohamed, & Tan, 2014; You et al., 2012). One of the drawbacks that are associated with the use of PVA in aqueous media is swelling, which occurs due to its very high hydrophilicity that makes it an open structure. Swelling affects the membrane performance, primarily the membrane solute rejection. Therefore, crosslinking of PVA is needed to balance the hydrophilic-hydrophobic properties of such membranes (Gohil & Ray, 2009; Huang, Moreira, Notarfonzo, & Xu, 1998; Katz & Wydeven, 1982). PVA could be crosslinked using multifunctional compounds like dicarboxylic acids, dialdehydes and dianhydrides, which could react with the −OH groups of PVA (Cha, Hyon, & Ikada, 1993; Giménez, Mantecón, & Cádiz, 1996; Gebben, van den Berg, Bargeman, & Smolders, 1985; Huang & Rhim, 1993; Korsmeyer & Peppas, 1981; Macho, Fabíni, Rusina, Bobula, & Harustiak, 1994).

Gum Arabic (GA) is a well-known hydrophilic, negatively charged, non-toxic natural composite polysaccharide derived from the excretions of Acacia Senegal and Vachellia Seyal trees (Roque, Bicho, Batalha, Cardoso, & Hussain, 2009; Tsai et al., 2014). It is an arabinogalactan polysaccharide that consists of three distinctive segments and is composed of more than 97% carbohydrates and less than 3% proteins (Shahgholian & Rajabzadeh, 2015). It is one of the widely accepted constituents in the pharmaceutical, cosmetic and food industries (Kong et al., 2014; Shahgholian & Rajabzadeh, 2015). Gum Arabic is proven to have excellent antibacterial properties, which play an important role in membranes biofouling mitigation (Chang, McLandsborough, & McClements, 2014; Hu, Gerhard, Upadhyaya, Venkitanarayanan, & Luo, 2016; Juby et al., 2012). Furthermore, Gum Arabic has shown superb surface activity and viscoelastic film forming capabilities (Cai et al., 2014; Li et al., 2015; Tan, Xie, Zhang, Cai, & Xia, 2016).

Polyethylene oxide–polypropylene oxide–polyethylene oxide (PEO–PPO–PEO) triblock copolymers, known as Poloxamers or Pluronics are amphiphilic copolymers comprising hydrophilic PEO segments and hydrophobic PPO segments. Compared to other Pluronics, Pluronic F127 has high extractability into aqueous phase, high molecular weight (Mw 12,600) and good hydrophilic/lipophilic balance value (HLB = 22) (Kim, Chung, & Park, 2006; Lv et al., 2007). The incorporation of Pluronic F127 into PVA thin films enhances the separation performance of membranes, as it has been reported in literature (Amanda, Kulprathipanja, Toennesen, & Mallapragada, 2000; Lv et al., 2007). Additionally, It has been reported that amphiphilic copolymers can alter the diffusion rate of water through membranes, which increases the permeate flux through the membrane (Iwasaki, Yamasaki, & Ishihara, 2003; Ishihara, Hanyuda, & Nakabayashi, 1995; Lv et al., 2007; Yajima, Sonoyama, Suzuki, & Kimura, 2002).

Many researchers have used PVA as a hydrophilic modifier or as a coating on polymeric or ceramic surfaces for many separation applications (Bano, Mahmood, Kim, & Lee, 2014; Billard & Kind, 2003; Guo, Ma, Hu, Jiang, 2007; Kang et al., 2012; Liu, Chen, Wang, Yu, & Gao, 2015; Li, Chen, Hu, Zhang, & Hu, 2014; Liu, Zhou et al., 2014; Pourjafar, Rahimpour, & Jahanshahi, 2012; Peters, Benes, Buijs, Vercauteren, & Keurentjes, 2006; Shang & Peng, 2007; Yang, 2007; Ying, 1983; Zhang, Wang, Wang, & Xiang, 2008). Nonetheless, to the best of our knowledge, no research has been presented on the use of crosslinked PVA incorporated with Pluronic F127 and conjugated with Gum Arabic as an active layer in reverse osmosis application without the use of polymeric or ceramic substrates. The use of the polymer as an active layer without a substrate reduces some negative consequences, such as internal concentration polarization that may cause an increase in the applied pressure. In this research, crosslinked PVA membranes incorporated with Pluronic F127 and conjugated with Gum Arabic for RO were fabricated. The newly synthesized membranes were then characterized and analyzed using various techniques like attenuated total reflection Fourier transform infrared spectroscopy (ATR-FTIR), contact angle measurements, X-ray diffraction (XRD), scanning electron microscopy (SEM), atomic force microscopy (AFM) and mechanical testing. The actual reverse osmosis performance of the membranes, including permeation testing, salt rejection and Chlorine resistance, was analyzed using a dead-end RO permeation unit.

2. Experimental procedure

2.1. Materials

Analytical grade PVA (Mw = 89000 Da), Gum Arabic (Mw = 250,000 Da), bisphenol A diglycidyl ether (DGEBA) (crosslinker), Pluronic F127 (average molecular weight: 12.6 kDa), dimethyl sulphoxide (DMSO) and sodium hypochlorite (NaClO) were acquired from Sigma Aldrich (St Louis, MO, USA). All chemicals were used without further purification.

2.2. Membrane casting

2.2.1. Crosslinking of PVA with DGEBA

Various weight percentages of DGEBA crosslinker, as shown in Table 1, were mixed into PVA solutions, where DMSO was used as a solvent with a 17:83 solute/solvent weight percent ratio. The mixing was performed at 70 °C with continuous stirring for 2 h until a homogenous, transparent solution was produced. Utilizing the dissolution casting method, the solution was then transferred slowly into identical Petri dishes with identical amounts of 10 ml to insure uniformity. Petri dishes were then heated to 65 °C in a controlled evaporation environment to ensure uniform film thicknesses of 0.1 ± 0.012 mm, measured by a screw gauge. After complete evaporation, the thin film membranes were removed from the Petri dishes with the aid of sharp blades. Five membranes were synthesized from each concentration for testing. The resultant dense membranes were examined for swelling, permeation and salt rejection to come up with the optimal crosslinker weight percent. As will be demonstrated in the results section, the 0.16 wt% of DGEBA provided the optimum membrane properties, thus, the weight percent of 0.16 was used for further modifications.

2.2.2. Incorporation of Pluronic F127 into the crosslinked PVA membranes

Various weight percentages of Pluronic F127, as shown in Table 1, were blended into the solutions of PVA and 0.16 wt% of DGEBA. The solution preparation and the film casting method followed the procedure mentioned in the previous section. The resultant composite membranes with different weight percentages of Pluronic F127 were characterized and evaluated. The overall evaluation of the Pluronic F127 membranes as will be detailed later on, showed that 6 wt% of Pluronic F127 was the optimal percentage and was used for further modifications. Fig. 1 shows a scheme of the chemical reaction.