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Effective removal of humic acid using xanthan gum incorporated polyethersulfone membranes

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ABSTRACT

In this study, xanthan gum (XA) was used as a hydrophilic biopolymer additive for the modification of polyethersulfone (PES) membrane to removal of humic acid (HA). The membranes are prepared using phase inversion technique and the concentration of XA was varied from 0.5 to 1.5 wt%. The prepared membranes are characterized as a function of hydrophilicity, equilibrium water content (EWC), porosity studies and functional group analysis. Membrane surface and cross-sectional morphology was studied using scanning electron microscope. The lower contact angle value 64.2° was exhibited, when 1.5 wt% of XA incorporated in PES membrane and this ensures that increase of hydrophilicity in pristine PES membrane. Further, higher water permeability (PWP) of 68.9^{-9} m/s kPa was observed for 1.5 wt% of XA/PES membrane. The effect of pH on HA removal was studied for neat PES and XA/PES membranes. The rejection performance of XA incorporated in PES membranes were compared with commercial available PES membrane.

1. Introduction

Surface water is being intensively polluted by various industrial activities, modern agricultural practices, and a natural accumulation of dead leaves, fish and bird faeces, and anthropogenic activities. Moreover, dead leaves degradation and in situ anaerobic digestion of algal or microbial biomass generate humic substances in the surface water streams. Humic acid (HA) is an organic matter. Presence of HA in surface water, can cause the following adverse effects. There are, (a) Impart unpleasant the odour, (b) Changes the colour, (c) Absorbs the metals and (d) Increase the microbial population (Kilduff et al., 1996; Chen et al., 2003; Thurman, 1985). Hence, HA has to be properly treated to provide the portable water to the Public. It is difficult to treat by the conventional water treatment methods. Because, during the conventional water treatment process like coagulation and clarification, it may react with coagulants, forms by-products and hinders the treatment process. Moreover, during the disinfection process, phenolic hydroxyl groups of HA reacts with chlorine and forms trihalomethanes (THMs) as disinfection by-product (Krasner et al., 2006; Zularisam et al., 2006).

Membrane treatment are widely recognized alternative water treatment process to conventional treatment methods for removal the pollutants and meet the stringent environmental regulations (Katsoufidou et al., 2005; Susanto and Ulbricht, 2008; Shao et al., 2011; You et al., 2013). The selection of suitable membrane materials for removal the pollutants with desired properties is important task. Polyethersulfone (PES) is widely used membrane materials because it has good mechanical strength, high thermal stability, better chemical resistance and film-forming property. However, the major bottleneck of usage of PES is hydrophobic property. Hydrophobic surfaces are more prone to fouling (Wang et al., 2011; Wang et al., 2006; Rahimpour et al., 2010; Rahimpour, et al., 2012). Fouling hinders productivity, affects permeation of ions, transport of pollutants and also the lifetime of the membrane. Hydrophobic polymeric membranes have been modified by using additives to enhance permeability and increase their hydrophilicity to prevent the fouling. The work reported the influence of hydrophilic polymeric additives such as Polyethylene glycol (PEG) (Xu et al., 1999b), Polyvinylpyrrolidone (PVP) (Xu et al., 1999a; Marchese et al., 2003) and poly sulfoxide amide (Rahimpour et al., 2009) that increase the hydrophilic property of the modified membrane. Hydrophilic biopolymers are also a potential material for modification of hydrophobic polymers. There had also been reports on novel membranes synthesised by blending cellulose acetate (CA) with hydrophilic biopolymer carboxymethyl cellulose acetate (CMC) which shows enhanced hydrophilic characteristics and higher pure water flux (PWF) (Han et al., 2013). Lakra et al. (2013) reported Chitosan blend membranes such as PES and PES/CA with improved flux and without compromising

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the rejection efficiency.

The increase in HA rejection rates was observed when 1.25 wt% ZnO added in PES membrane (Ahmad et al., 2014). Further, HA flux was also enhanced by incorporation ZnO in PES polymer matrix. The accumulation of humic elements on hydrophobic PES surfaces favoured and reduce the flux due to the prevalent hydrophobic property of these compounds (Peeva et al., 2011). In our previous work (Lukka et al., 2014), removal of HA was studied using zirconia (ZrO_2) embedded PES membranes. Hydrophilicity enhancement was reported due to addition of zirconia nanopartciels in PES membranes. The modification of PES membranes with hydrophilic biopolymer for removal humic acid is limited.

Xanthan gum (XA) is hydrophilic biopolymer and is produced by aerobic fermentation of sugar by a gram-negative bacterium *Xanthomonas campestris* (Fink, 2013; Altay and Gunasekaran, 2013; Jaipal et al., 2013). It has received a great deal of attention as stabilizing agent in food industries and slow drug release applications in pharmaceutical industries due to its non-toxic, biocompatible property, biodegradable property, excellent mechanical property, better thermal stability and pseudoplastic rheological property (Psomas et al., 2007; Jiangyang et al., 2008; Bhattacharya et al., 2013). In the present study, interaction between XA and PES is studied by various membrane characterization techniques like FTIR, contact angle and scanning electron microscope (SEM). The pure water flux and removal of humic acid are extensively studied using low-pressure cross flow UF module. In addition, separation performance of as prepared membranes at different feed pH are carefully investigated and compared with commercially available PES membrane.

2. Materials and methods

2.1. Materials

Polyethersulfone (PES) (Veradale 3000p) is procured from Solvay Chemicals India Ltd, Mumbai, India. Dimethyl acetamide (DMAc) is obtained from Alfa Aesar, U.S.A. Sodium Lauryl Sulphate (SLS) is purchased from Avantor Performance Materials, U.S.A. Xanthum gum and humic acid are supplied by Loba Chemie Pvt. Ltd., Mumbai, India. The commercial PES membrane at 30 kDa was purchase from Orelis Environnement SAS, France. All reagents were analytical reagent grade without further purification. Double distilled (DD) water is used throughout the study.

2.2. Preparation of biopolymer blend PES membranes

Base polymer as PES and solvent as DMAc were chosen for the preparation of membrane using phase inversion method. Base polymer PES was dried in hot air oven at a temperature of 80 °C before use. Initially casting dope solution was prepared by dissolving PES in a solvent DMAc using magnetic stirrer at a temperature of 60 °C. And then homogeneous mixture is cast on glass plates using casting knife with a thickness of 250 µm. Then, cast membranes are allowed to evaporate in air for 30 s and then glass plates was immersed in a non-solvent water bath containing 2 wt% SLS at 10 °C for 3 h. The resultant membrane was stored in container containing water for 24 h. In order to improve the membrane performance, XA was used as an additive. Similarly, the above procedure was followed for preparation of XA incorporated membranes and its composition are provided in Table 1.

2.3. Membrane characterizations

Chemical structure of neat and modified PES membranes are studied at 4 cm^{-1} resolution using ATR mode of Thermo Fisher Scientific Nicolet i5S FTIR analyser (Thermo Nicolet Corporation, USA). To further understand the relationship between surface morphology and biopolymer loading, cross section and top surface layer images are taken using SEM (TESCAN VEGA 3 SEM, USA). The hydrophilicity of the neat and modified PES membranes, contact analysis is performed by the sessile drop method using goniometer (Model 250-F1 Rame Hart Instruments, Succasunna, NJ).

Table 1
Compositions of PES and PES/XA membranes

Membrane type	Composition of dope solution in solvent DMAc (82.5 wt%)	
	PES (wt%)	XA (wt%)
M-I	100	–
M-II	99.5	0.5
M-III	98.5	1.0
M-IV	97.5	1.5

2.4. Equilibrium water content (EWC) and porosity studies

EWC is an indirect method to analysis the hydrophilicity or hydrophobicity of the membrane. The membrane samples are soaked in DD water for 24 h at room temperature. Weight of the soaked membrane is measured (W_1) using an electronic weighing balance immediately after mopping with tissue paper. These wet samples are placed in a vacuum oven for 24 h at 60 °C and then dry weights of membranes (W_2) are determined. The percentage water content was calculated as follows. (Chakrabarty et al., 2008) and values are tabulated in Table 2.

$$\mathrm{EWC}(\%) = \frac{w_1 - w_2}{w_1} \times 100 \quad (1)$$

Porosity of membranes are calculated by the formula (Raguime et al., 2007) given below and depicted in Table 2.

$$\mathrm{Porosity}\% = \frac{w_1 - w_2}{\rho_w x V_T} \times 100 \quad (2)$$

Where

$$V_T = \frac{w_1 - w_2}{\rho_w} + \frac{w_2}{\rho_{Mo}} \quad (3)$$

where V_T is the membrane volume (m^3) in the wet state and ρ_w is the density of water (1 g/cm^3), ρ_{Mo} is the density of the membrane in the dry state.

2.5. Pure water permeability

Commercial PES membrane and PES/XA membranes were precompressed with DD water at 200 KPa for 1 h. After that, water flux measurements are conducted at 200 KPa pressure by using cross flow module (Model: PLEIADE Rayflow®, Orelis Environnement SAS, France). Pure water flux (PWF) of the membranes was calculated using the following expression:

$$J_w = \frac{Q}{TA} \quad (4)$$

where J_w is the pure water flux (L m^{-2} h^{-1}); Q is the amount of permeate collected (L); T is the sampling PO time (h); and A is the membrane area (m^2).

Pure water permeability (PWP) is the slope plotted between pure water flux and transmembrane pressure (ΔP).

$$\mathrm{PWP} = \frac{J_w}{\Delta P} \quad (5)$$