CHEMICAL ENGINEERING RESEARCH AND DESIGN XXX (2014) XXX–XXX

Contents lists available at ScienceDirect

Chemical Engineering Research and Design

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

Fabrication of asymmetric polyethersulfone membranes for separation of carbon dioxide from methane using polyetherimide as polymeric additive

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ABSTRACT

Polyetherimide (PEI) was used as a polymeric additive for preparing an asymmetric polyethersulfone (PES) membrane for the separation of CO_2 from CH_4. In pure gas experiments, the higher skin layer thickness and the lower porosity of the sub layer for the membrane prepared from the polymer blend with the composition of 98:2 lead to an increase in CO_2/CH_4 selectivity and a decrease in the CO_2 permeance in contrast with a pristine PES. For higher PEI contents, the higher fractional free volume of the membranes improves the gas permeance and reduces the CO_2/CH_4 selectivity. The incorporation of PEI in PES reduces the CO_2 sorption in PES via decreasing the non-equilibrium free volume and imparts antiplasticization properties to the membrane.

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Keywords: Membrane; Polyethersulfone; Polyetherimide; Carbon dioxide; Methane; Plasticization

1. Introduction

Natural gas sweetening and enhanced oil recovery (EOR) are examples of important industrial processes that demand separation of CO_2 from CH_4. The membrane technology is one of the new processes that competes with such traditional processes as amine absorption and pressure swing adsorption because of its low energy consumption and flexibility and lower toxicity (Simons et al., 2010; Xiao et al., 2009). Appropriate properties of PES such as suitable strength and flexibility are important in making PES a good choice for membrane preparation (Sadrzadeh et al., 2009; Shi et al., 2007). One of the easiest and most common methods to get improved polymers with a better performance is polymer blending. Therefore polymer blending is known as a popular research interest both in industry and among academic groups (Lin et al., 2008). The membranes prepared from PEIs have high CO_2/CH_4 selectivity, high chemical and thermal stability, high strength, and high flux (Hwang et al., 2011; Simons et al., 2010). Hwang et al. (2011) used PEI for preparation of the polyphenylsulfone/polyetherimide blend membranes in order to improve the polyphenylsulfone membrane properties and humic acid removal performance. Swier and Weiss (2006) added PEI to improve the mechanical stability and decrease the water swelling of sulfonated poly (ether ketone ketone) as the proton-conducting membrane. The thermal and mechanical properties, miscibility, morphology, and gas separation performance of the polybenzimidazole/polyetherimide composite hollow fiber membranes were investigated by Chung and Xu (1998). Also PEI has been used to reduce the membrane swelling in sulfonated poly (ether ether ketone) ion-exchange membranes (Cui et al., 1998).

The present study is an attempt to study the effect of PEI as a polymeric additive on the structure and performance of the PES membrane for separation of carbon dioxide from methane.

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Received 3 August 2013; Received in revised form 29 January 2014; Accepted 9 February 2014
http://dx.doi.org/10.1016/j.cherd.2014.02.010
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Please cite this article in press as: Saedi, S., et al., Fabrication of asymmetric polyethersulfone membranes for separation of carbon dioxide from methane using polyetherimide as polymeric additive. Chem. Eng. Res. Des. (2014), http://dx.doi.org/10.1016/j.cherd.2014.02.010

2. Experimental

2.1. Materials

Polyethersulfone (PES Ultrason E6020P with MW = 58,000 g/mol) was procured from BASF company (Germany), the polyether imide (PEI) was purchased from Ultem, polydimethyl siloxane (PDMS) was supplied by LG. Dimethylacetamide (DMAc), and n-hexane was obtained from Merck.

2.2. Membrane preparation

All the used membranes were prepared by the dry/wet phase inversion method. The PES and PES/PEI casting solutions with the concentration of 30 wt.% were prepared in DMAc as the solvent. The casting solutions were prepared as the PES/PEI blend compositions of 100:0, 98:2, 96:4, 94:6 and 92:8. The casting processes were carried out using a self-made casting knife. The prepared polymeric films were kept at ambient condition for 1 min and afterwards immersed in distillated water. The prepared membranes were kept in the distillated water for 24 h and finally were dried at ambient condition for 48 h. For sealing the membrane's defects and for enhancing membrane selectivity the synthesized membranes were coated with the 5 wt.% of PDMS in n-hexane by dip coating method and were placed in an oven at the temperature of 177 °C for 30 min for crosslinking the PDMS layer.

2.3. Viscosity measurements

The viscosity of PES/PEI/DMAc of each casting solution was measured with Bohlin CS Rheometer (Bohlin Instruments Ltd.).

2.4. Differential scanning calorimetry (DCS)

DSC thermograms of the PES and PES/PEI membranes were obtained using a Mettler Toledo DSC calorimeter under nitrogen atmosphere with the heating rate of 10 °C/min.

2.5. X-ray diffraction analysis

The crystal structure of the PES and PES/PEI membranes was investigated with Philips PW 3040. The accelerating voltage and emission current used were 40 kV and 30 mA, respectively.

2.6. Scanning electron microscopy

KYKY-EM3200 scanning electron microscope (SEM) was employed in order to investigate the morphology of the cross section and the top surface of the PES and PES/PEI membranes.

2.7. Porosity measurement

The porosity measurements were carried out as described in our previous work (Saedi et al., 2014).

2.8. Gas permeation measurements

The gas performance of the prepared membranes was evaluated using the experimental set up as explained in our previous work (Saedi et al., 2012). All of the permeation experiments were repeated three times and the mean values were reported. The experimental errors for all the experiments were smaller than 5%.

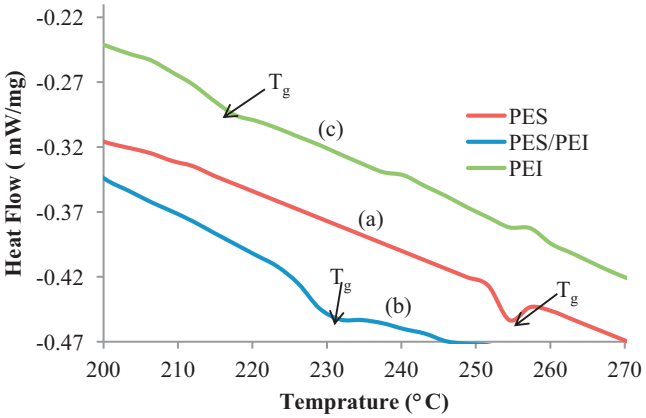

Fig. 1 – DSC thermograms of the membranes prepared from PES (a), PES/PEI with blend composition of 92:8 (b) and PEI (c).

2.9. Sorption measurement

To clarify the effect of PEI on the gas sorption behavior of the PES membranes, the gas sorption experiments were carried out for the dense PES and PES/PEI polymeric films (Chen et al., 2009). The gas sorption experiments were carried out using the method as described in our previous study (Saedi et al., 2013).

3. Results and discussions

3.1. Differential scanning calorimeter

Fig. 1 shows the DSC thermograms of the PES, PES/PEI and PEI. As shown in Fig. 1, there is a single T_g for PES/PEI with the blend composition of 92:8. This observation demonstrated that PES and PEI are miscible in the other blend compositions that were studied in this research. Also as can be seen in Fig. 1 and as predicted by Fox equation, PES/PEI blend has a lower T_g than the pure PES because of the lower T_g of PEI in contrast with PES (Hwang et al., 2012). Moreover, in the presence of PEI, the chain mobility of PES increases and the chain entanglement decreases; so less thermal energy is needed for overcoming the chain–chain interactions of the polymer (Lau and Ismail, 2009). It means that the T_g of PES/PEI blend is less than pure PES.

3.2. XRD analysis

The XRD diffraction patterns of PES and PES/PEI are shown in Fig. 2. We can see the XRD amorphous peak of PES ($2\theta = 18.025°$) (Liang et al., 2012) in both PES and PES/PEI membrane although it slightly shifts to lower values. As can be deduced from Bragg's law, a decline in 2θ implies that the d-spacing of the membrane increases. The main reason for the greater d-spacing is the higher interstitial chain–chain distance and the fractional free volume (FFV) of the membrane (Hosseini et al., 2008; Weng et al., 2008) due to the presence of the PEI in PES structure as a polymeric additive.

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