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CO_2 facilitated transport through an acrylamide and maleic anhydride copolymer membrane

Li-guang Wu[a*], Jiang-nan Shen[b], Huan-lin Chen[b], Cong-jie Gao[a]

[a]*The Center for Development of Water Treatment Technology, State Ocean Administration, Hangzhou 310012, China*
Fax: +86 (571) 8886 8427; email: wu_liguang@hotmail.com
[b]*College of Materials and Chemical Engineering, Zhejiang University, Hangzhou 310027, China*

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Abstract

A copolymeric membrane material containing an amine group for CO_2 facilitated transport was synthesized by radical polymerization of acrylamide and maleic anhydride. The relative molecular weight and chemical structure of the copolymer were analyzed by viscometric measurement, elemental analysis, and FTIR. The copolymeric membranes were prepared. The sorption behavior as well as the permeabilities of the membranes for pure CO_2 and CH_4 were investigated. The results show that the copolymeric membrane possesses a higher permeability of CO_2 and a lower permeability of CH_4. The membrane displays a CO_2 permeability of 5×10^{-12} cm^3(STP)/cm^2.s.pa, and a CH_4 permeability of 2×10^{-13} cm^3(STP)/cm^2.s.pa. CO_2 sorption behavior of the copolymeric membrane, which can be classified as a dual-mode sorption model, and CH_4 sorption behavior of the copolymeric membrane accord with the Fickian diffusion model.

Keywords: Facilitated transport membrane; Sorption behavior; Permeability; CO_2; CH_4

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1. Introduction

CO_2 removal from gas streams is required in many industrial applications among which natural gas processing is probably the most important one. Polymeric membranes are used to remove CO_2 from natural gas, but few commercially available polymeric membranes have both high permeability and high selectivity [1]. Facilitated transport membranes exhibit fairly high selectivity as well as high permeability because of carrier-mediated transportation.

Facilitated transport membranes involve carrier-mediated transportation in addition to

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*Corresponding author.

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permeant physical dissolution and diffusion. The existence of a carrier that can react reversibly with the permeant brings about high selectivity and high permeability. There are two types of facilitated transport membranes: one is a mobile carrier membrane (liquid membrane) where the carrier can diffuse in the membrane, and the other is a fixed carrier membrane where the carrier is immobilized in the membrane matrix and cannot move.

Although there is a remarkably high selectivity for liquid membranes, the disadvantage of this type of membrane is that it has poor stability. The fixed carrier membrane, in which the carrier is chemically bonded, is more favorable in comparison with the liquid membrane because there is little chance of carrier leakage. The fixed carrier membrane, which removes acid gas from gas streams and separates organic–organic mixtures, has been investigated extensively in recent years [2–7].

In this paper, the membranes containing amine groups, which act as fixed carriers for CO_2 facilitated transport, were prepared, and their chemical structure analyzed. In addition,the CO_2, CH_4 sorption behavior of the membranes and their CO_2, CH_4 permeability were investigated.

2. Experimental

2.1. Materials

Acrylamide (AM. m.p. 83-84) and maleic anhydride (MA. m.p. 56) were recrystallized from reagent-grade benzene and dried in a vacuum at room temperature. Azobisisobutyronitrile (AIBN) was recrystallized from reagent-grade methanol prior to use. Deionized water was used.

2.2. Synthesis of AM–MA copolymer

AM (7.108 g), MA (9.806 g) and benzene (250 ml) were introduced into a 300-ml three-necked flask equipped with a stirrer, a condenser and a thermometer. The contents were stirred at 80 for complete dissolution. AIBN (0.20 g in four steps) was added to the dissolution as an initiator, and was stirred for 3 h at the same temperature in a nitrogen atmosphere. The polymer precipitated from the solution as polymerization continued, and was then completed. The precipitated product was washed with acetone several times and a hygroscopic white solid was obtained. The yield of copolymers was more than 60 wt%. The products were placed into a mixture of methanol/water (1/1), which was a solvent for these two monomers and the AM homopolymer. The insoluble copolymer was removed by filtration and dried under vacuum to a constant weight at room temperature. A large volume of methanol (more than 1/10) was added to the filtrate to check that the AM homopolymer was absent.

2.3. Membrane preparation

A known concentration of aqueous AM–MA copolymer solution was cast onto a glass plate and the PSF ultrafiltration membrane to form a homogeneous membrane and a composite membrane. The membranes were dried at room temperature for a day. The thickness of the membrane strips was measured on a micrometer with an accuracy of 0.1 μm. The thickness of the homogeneous membranes was 10 μm, and the top layer thickness of the composite membranes was 2 μm.

2.4. Viscometric measurements

Viscometric measurements were carried out with an Ubbelohde viscometer. Viscosity data were calculated according to the Mark-Huggins equations:

$$\eta_{sp}/C = [\eta] + k' [\eta]^2 C$$

where η_{sp} is the specific viscosity, C is the concentration of copolymer; $[\eta]$ is the intrinsic

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