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TECHNICAL NOTE

Some Characteristics of Pervaporation for Dilute Ethanol–Water Mixtures by Alcohol-Permselective Composite Membrane*

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ABSTRACT

Zeolite-filled polydimethylsiloxane/polysulfone composite membranes were prepared and used to separate ethanol from dilute ethanol–water mixtures through a pervaporation process. During this process the relationship between flux (J) and temperature (T) is $J = 597.95 \exp(-E_0/RT)$ ($g/m^2 \cdot h$), $E_0 = 3.292$ kJ/mol. The experimental results show that the flux decreases with increasing downstream pressure while the selectivity ($\alpha_{EtOH/Water}$) rises, the flux increases, and the selectivity decreases with a rise of feed concentration, and the flux as well as the selectivity increase with increasing bulk velocity on the upstream side of the membrane.

Key Words. Pervaporation; Alcohol-permselective membranes; Composite membranes

INTRODUCTION

Pervaporation is a promising membrane separation method for liquid mixtures. Most researchers have studied the pervaporation of alco-

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hol–water mixtures by using water-permselective membranes; only a few by using alcohol-permselective membranes. As far as we know, in the literature only a few hydrophobic membrane materials such as polydimethylsiloxane (PDMS), nitrile-butadiene rubber, styrene-butadiene, and polysubstituded acetylenes were used. Although PDMS membranes offer low selectivity and flux, they are convenient to use for pervaporation of dilute alcohol–water mixtures. In order to improve PDMS membrane's selectivity and flux in pervaporation for removing alcohol from aqueous solutions, some researchers (1–3) have recently prepared zeolite-filled PDMS membranes and obtained good results.

In this paper an alcohol-permselective composite membrane with a top layer of PDMS-filled zeolite and a support of PS is presented. There were three reasons for us to prepare the membrane: to concentrate ethanol from dilute ethanol–water mixtures by this membrane, to assure both permeation flux and selectivity reach a higher level, and to have a relatively high strength for industrial utilization. By using this kind of membrane, we investigated the effects of different parameters on the pervaporation of dilute ethanol–water mixtures.

EXPERIMENTAL

Materials

Zeolite (Silicalite-I, ZSM-5 Type) with a crystal size of under 40 μm (provided by the Department of Chemistry, Nankai University, People's Republic of China), silicon rubber, *n*-ethylsilicate (crosslinker), and *n*-heptane were mixed together. The weight ratio of Silicalite-I and silicon rubber was 1.0. A composite membrane was made by coating this suspension on a polysulfone membrane. The coated membrane was dried in air at room temperature for 8 hours to ensure complete curing.

Ethanol–water mixtures were prepared from ultrapure deionized water and absolute ethanol (AR, over 99.9 wt%).

Pervaporation

The steady-state pervaporation experiments were performed in a permeation cell as shown in Figs. 1 and 2. The effective membrane area of cell was 40.2 cm^2. The membrane was supported by a porous titanium disk. The membrane was allowed to swell for 1 hour in the feed liquid before pervaporation. The permeate was collected in liquid nitrogen. The flux was measured by weighing the amount collected over a given period, and the compositions were analyzed by gas-phase chromatography.

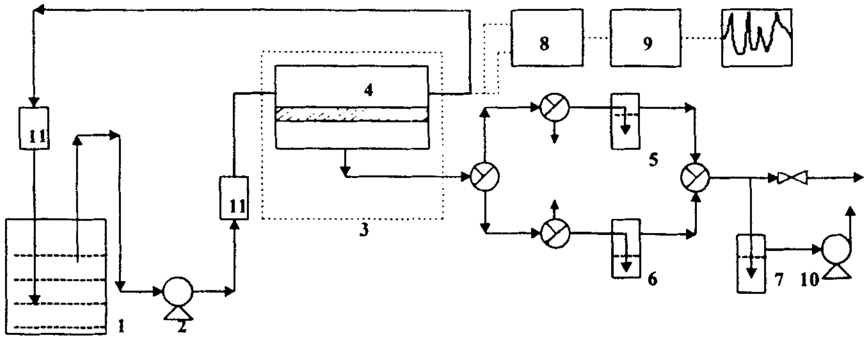

FIG. 1 Pervaporation equipment schematic: (1) feed tank; (2) feed pump; (3) membrane cell; (4) membrane; (5, 6, 7) liquid nitrogen condenser; (8) gas-phase chromatograph (GC); (9) GC recorder; (10) vacuum pump; (11) flowmeter.

We determined the flux (J) and selectivity (α) as follows:

$$J = W/S \cdot t \qquad (\mathrm{kg/m^2 \cdot h})$$

$$\alpha_{\mathrm{EtOH/Water}} = Y_{\mathrm{EtOH}} X_{\mathrm{water}} / Y_{\mathrm{water}} X_{\mathrm{EtOH}}$$

where W refers to the weight of the permeate, S is the membrane area, t

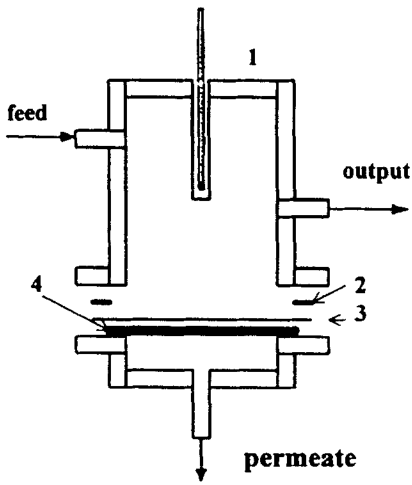

FIG. 2 Pervaporation cell: (1) thermometer; (2) gasket ring; (3) membrane; (4) porous support plate.

is the given pervaporation time, and Y and X denote concentrations of the permeate and the feed, respectively.

RESULTS AND DISCUSSION

1. Effect of Temperature on J and α

Figure 3 shows that the rise in temperature not only intensifies the flux but also the selectivity $\alpha_{EtOH/Water}$. It differs from hydrophilic or water-permselective membranes for pervaporation of water–ethanol mixtures. As regards the latter, a rise in temperature usually intensifies the flux but decreases the selectivity $\alpha_{EtOH/Water}$ because the solubility and diffusivity increase with temperature. It may be assumed that increased swelling of the membrane at higher temperature results in an increase of polymer segmental motions and Brownian movements.

The relationship between flux (J) and temperature (T) for our pervaporation by composite membranes is experimentally established:

$$J = 597.95 \exp(-E_0/RT) \qquad (\text{g/m}^2\cdot\text{h})$$

$$E_0 = 3.292 \text{ kJ/mol}$$

The effect of temperature on selectivity $\alpha_{EtOH/Water}$ is relatively complicated. In general, for a water-permselective membrane, $\alpha_{water/EtOH}$ decreases with rising temperature, but the experimental results in this paper show that $\alpha_{EtOH/Water}$ increases with rising temperature.

2. Effect of Downstream Pressure on J and α

The flux decreases with increasing downstream pressure while $\alpha_{EtOH/Water}$ rises. These results are different from those of some other authors who reported that downstream pressure does not affect flux and selectivity for ethanol–benzene and ethanol–cyclohexane systems (3), but agree well

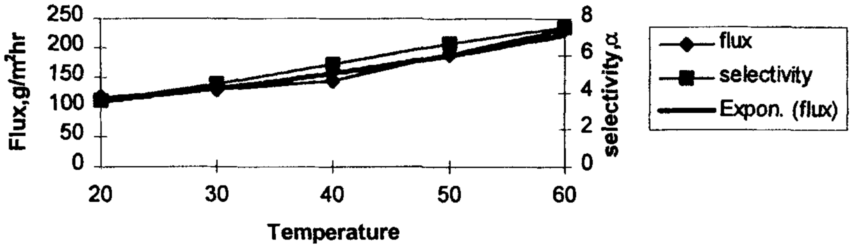

FIG. 3 Relationship between temperature and separation property.

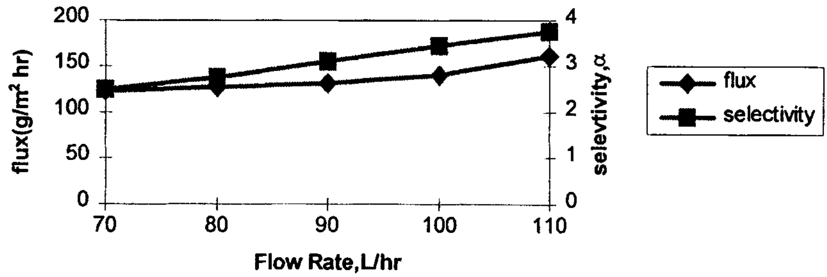

FIG. 4 Relationship between flow rate and separation property.

with the authors of Ref. 4. The reason for our results is probably a rise in the degree of saturation in the downstream chamber, which then tends to swell the membrane. In other words, when the downstream pressure increases, the equilibrium concentration is higher at the downstream surface of the membrane and the desorption rate decreases. Therefore, the surface of membrane remains nearly "wet," the flux decreases, and $\alpha_{EtOH/Water}$ increases. Our results are shown in Fig. 4.

3. Effect of Feed Composition on *J* and α

In Fig. 5 the flux increases and the selectivity decreases as the feed concentration rises. The higher ethanol content in the feed makes the hydrophobic membrane more swollen and destroys the interactive bonding within the PDMS material to some extent. It probably enhances the flux and lowers the selectivity.

4. Effect of Bulk Velocity on *J* and α

Figure 6 shows that the flux as well as $\alpha_{EtOH/Water}$ increase with increasing bulk velocity on the upstream side of the membrane. It is quite evident

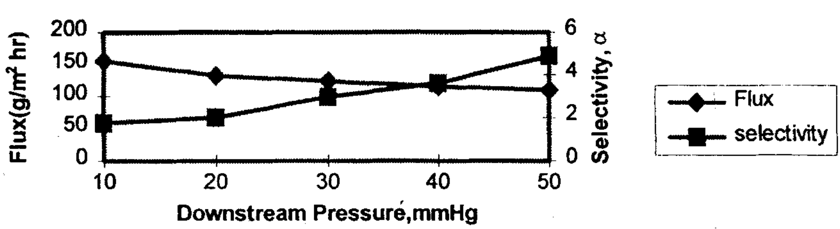

FIG. 5 Relationship between downstream pressure and separation property.

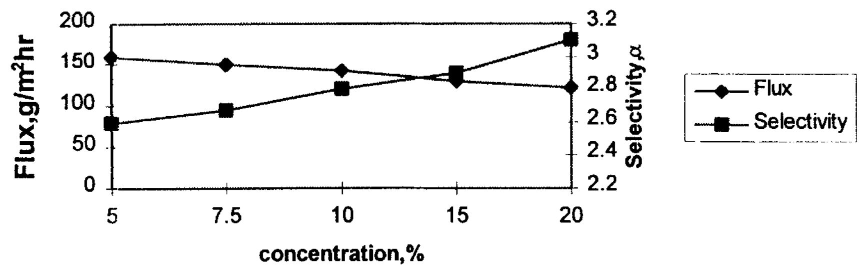

FIG. 6 Relationship between concentration and separation property.

that a higher velocity will promote the flux because it could lessen the concentration polarization on the membrane surface and provide more opportunity for the sorption of ethanol on the hydrophobic membrane.

CONCLUSIONS

Zeolite-filled PDMS/PS composite membranes have been prepared. In the pervaporation of 4.8% (wt) ethanol–water mixtures at a temperature of 60°C and a downstream pressure of 22 mmHg, $\alpha_{EtOH/Water}$ is 7.52 while flux is 231.3 g/m^2·h. The flux as well as $\alpha_{EtOH/Water}$ increase with increasing bulk velocity on the upstream side; a rise in temperature not only intensifies the flux but also the selectivity, and the downstream pressure might promote the selectivity and reduce the flux. An increase of feed concentration increases the flux and decreases the selectivity.

ACKNOWLEDGMENT

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REFERENCES

1. M. Jia, K. V. Peinemann, and R. D. Behling, "Pervaporation and Characterization of Thin-Film Zeolite–PDMS Composite Membranes," *J. Membr. Sci.*, *73*, 119–128 (1992).
2. H. J. C. Te Hennepe, D. Bargeman, M. H. V. Mulder, and C. A. Smolders, "Zeolite-Filled Silicone Rubber Membrane, Part 1. Membrane Pervaporation and Pervaporation Results," *Ibid.*, *35*, 39–55 (1987).
3. X. Li and S. Wang, "A Mass Transfer Model for Pervaporation of Ethanol–Water Mixture by Zeolite-Filled PDMS Membrane," in *Proceeding of International Sympo-*

sium on Membrane and Membrane Processes, Hongzhou, People's Republic of China, 1994, p. 275.
4. J. Neel, P. Aptel, and R. Clement, "Basic Aspect of Pervaporation," *Desalination*, *53*, 297–326 (1985).

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