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Influence of concentration polarization on the permeation behavior of VOCs/N_2 mixtures through PDMS membrane

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1. Introduction

About ten year ago, the first membrane unit for organic vapor recovery from a gas stream using rubbery membranes was installed. Since then about 100 units have been installed commercially. The recovery of volatile organic compounds (VOCs) from waste or vent gases can be motivated in terms of environmental protection and economical aspect. Usually VOCs cause a serious environmental pollution when they release out into atmosphere without any treatment. VOCs are also known as a main cause of cancers as well as of destroying the ozone layer. From the economical point of view, expensive VOCs lost into air have to be recovered for reuse.

For the recovery of VOCs, silicone rubbery membranes are preferred because they are much permeable to VOCs. Especially, when VOCs vapors are the minor components of feed gas, the rubbery membranes could be used favorably because less feed gas has to permeate the membrane to remove the bulk of the vapor, requiring relatively small membrane areas. Among the rubbery membranes, poly(dimethylsiloxane) (PDMS) exhibits an excellent membrane performance for the removal of VOCs from noncondensable gase; high permeability and high selectivity for VOCs. However, as a result of very high selectivity of a membrane, concentration polarization may take place badly in a layer of feed adjacent to membrane surface, in which a more permeating component is depleted. The concentration polarization at the layer reduces both permeation rate and selectivity. There have been very few studies on the concentration polarization effect on the permeation of VOCs/gas mixture so far although numerous researches are reported on liquid separations.

In this study, the permeations of chlorinated hydrocarbon/N_2 mixtures through homogeneous PDMS membrane were carried out at various operating conditions, and membrane performances, such as, the permeability, diffusion, and solubility coefficients of respective mixture were determined in on-line mode. The concentration polarization phenomena were characterized through analysis of the membrane performances determined.

2. Experimental

Homogeneous PDMS (GE655) membranes with a thickness of 130-160 μm were prepared by radical crosslinking between PDMS oligomer terminated with vinyl groups and a mixture of Pt catalyst and PDMS oligomer with active hydrogens. Methylene chloride, chloroform, 1,2-dichloroethane, and 1,1,2-trichloroethane were used as VOCs component. Feed mixtures used were the chlorinated hydrocarbon/N_2 mixtures. VOCs content in feed ranged from 0.3 to 1.5 vol.% and operating temperatures were 35-65 °C. Feed flowed at a rate of 0 - 600 cc/min to investigate the concentration polarization effect. The permeability, diffusion, and solubility coefficients of respective mixture were directly determined in on-line mode by using a novel equipment which employed a continuous-flow technique.

3. Results and discussions

The PDMS membrane shows very good membrane performance; permeability coefficients of 370-1100 barrers and selectivities of 115-350, depending on the kind of VOCs and its content in feed. It is observed that selectivity increases with increasing the condensability of VOC component. Also, the permeation of the VOCs/N_2 mixtures through the PDMS membrane could be confirmed to be a sorption selective process, as can be seen in other permeations through rubbery membrane. In all of the permeations of the mixtures, both permeability and selectivity decreased dramatically with decreasing feed flow rate below 200 cc/min as shown in Fig. 1, The decrease of membrane performance could be attributed to concentration polarization occurring in the feed layer (boundary layer) adjacent to membrane surface. At slower feed flow, the boundary layer was larger in thickness. The mass transfer coefficient of the boundary layer, k, which is dependent on the hydraulic dynamic of feed flow, is expressed by

$$k = \frac{D}{\delta}$$

where D is a diffusion coefficient of VOC component in the layer and δ denotes the thickness of the layer. Thus k has a smaller value owing to larger δ at slower feed flow. Reducing permeability is accompanied with reducing the solubility coefficient of the mixture, correspondingly, However, the diffusivity of the mixture was kept almost constant regardless of the concentration polarization occurrence. With increasing the condensability, the decrease is found to be greater. Since the selectivity of VOCs/gas mixtures increases with increasing the condensability of VOC component as mentioned previously, the depletion of VOC component with greater condensability happens more significantly in the layer, resulting in more severe concentration polarization.

Fig. 2 shows the membrane performance of various VOCs/N_2 mixtures with temperature. The effect of operating temperature has on membrane permeability in opposite way; at higher operating temperature the activity of VOC is lower while membrane mobility is enhanced. Looking at the concentration polarization with temperature, concentration polarization is less significant at low temperature. It could be explained in terms of higher selectivity and lower diffusivity due to larger viscosity in the layer.

It is concluded that the concentration polarization at the boundary layer is a function of feed flow, selectivity and permeability; the concentration polarization takes place more severly at slower feed flow, higher selectivity or/and greater permeability.

4. References

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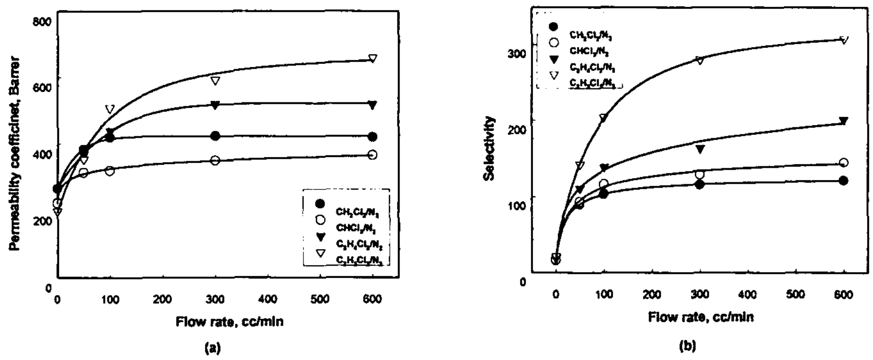

Fig. 1. (a)Permeability coefficient and (b)selectivity with flow rate.
Permeation temp.: 45 ℃, feed VOCs content : 0.9 vol.%

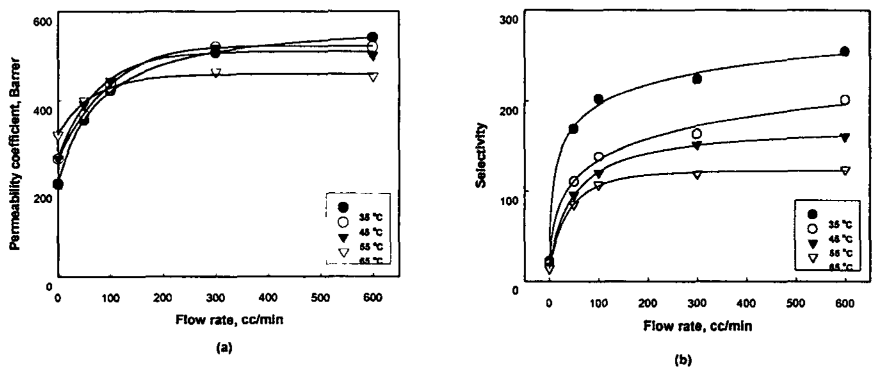

Fig. 2. (a)Permeability coefficient and (b)selectivity with permeation temperature.
Feed VOC : $C_2H_4Cl_2$, feed VOCs content : 0.9 vol.%