

# Gas Transport Properties of CoAlPO<sub>4</sub>-5/PC Membranes

SHIH-HSIUNG CHEN, SHIOW-SHYUNG LIN, DONG-JANG CHANG, JING-SONG CHANG

Department of Environmental Engineering and Health, Chia-Nan College of Pharmacy and Science, Tainan, 717, Taiwan, Republic of China

Received 24 May 1999; accepted 28 September 1999

**ABSTRACT:** The transport phenomena of oxygen and nitrogen across a pure polycarbonate (PC) and CoAlPO<sub>4</sub>-5/PC membranes were studied. Various CoAlPO<sub>4</sub>-5 membranes with different cobalt content were added to polycarbonate membranes to improve the gas transport performance. Oxygen and nitrogen isotherms were studied. Solubility of oxygen and nitrogen was greatly increased by adding CoAlPO<sub>4</sub>-5 to the membranes, which also resulted in a higher solubility ratio of oxygen to nitrogen. It might be that a pinhole of the membrane caused the increase in diffusivity and a decrease in selectivity when excess CoAlPO<sub>4</sub>-5 was added. The results also showed that CoAlPO<sub>4</sub>-5 with a higher cobalt content would more effectively increase the gas solubility, but make only a minor change in the solubility ratio of oxygen to nitrogen. It was found that the increase in oxygen to nitrogen selectivity was mainly due to an increased diffusivity ratio of oxygen to nitrogen when CoAlPO<sub>4</sub>-5 was added into the membranes.  
© 2000 John Wiley & Sons, Inc. *J Appl Polym Sci* 77: 89–95, 2000

**Key words:** CoAlPO<sub>4</sub>-5; polycarbonate (PC); membrane; solubility; diffusivity

## INTRODUCTION

There are two ways to improve the selectivity of polymeric gas separation membranes. One is to synthesize polymers made from monomers with high affinity to oxygen. The other way to improve the membrane selectivity is via oxygen carrier addition. Unfortunately, an increase in gas selectivity usually decreases the gas permeability. Recently, there have been many investigations focused on zeolite–polymer composite membranes because the size and shape of zeolite may enhance the gas transport performance. Battle et al.<sup>1</sup> prepared polyestersulfone–zeolite composite membranes for gas separation. From the SEM study, the microporous cavity and channel were found in the composite membranes. Incompatibility was

found when excess zeolite was added in the membrane. It was concluded that the gas selectivity was increased by inducing appropriate zeolite in the membranes. Goldman and his coworkers<sup>2</sup> prepared NaA–zeolite type composite membranes for pervaporation. It was found that the composite behaved as the pure polymer membrane when the zeolite contents were below a certain limit. They concluded that the mass transport barrier was dependent on the interaction of polymer and zeolite.

A zeolite-blended membrane can be easily prepared by adding different sizes and shapes of zeolites in the membranes.<sup>3–9</sup> However, the effect of adding zeolite on gas separation performance depended on the zeolite shape, zeolite–polymer compatibility, pore size of zeolite, and membrane formation. Some of the studies reported that gas separation performance of the composite membrane was improved by adding an acceptable amount of zeolite.<sup>4,5</sup> However, the other reports found that the key point of modifying the zeolite/

Correspondence to: S.-H. Chen.

Contract grant sponsor: National Science Council of R.O.C.; contract grant number: NSC 86-2216-E-041-004-T.

*Journal of Applied Polymer Science*, Vol. 77, 89–95 (2000)  
© 2000 John Wiley & Sons, Inc.

polymer composite membrane was to control the micromorphology<sup>5</sup> of the composite membrane.

It was found that an increase of oxygen selectivity can be achieved by adding a limited amount of zeolite. However, while the oxygen selectivity was increased, the oxygen permeability was decreased. It appeared that the zeolite added to the membrane affected the gas diffusion and sorption properties of composite membranes.

In the previous investigations,<sup>11–15</sup> gas transport properties with salt-added membranes were studied. An important point is that the solubility ratio of oxygen to nitrogen was found to contribute only a small part of the O<sub>2</sub>/N<sub>2</sub> selectivity. It was indicated that the enhanced improvement of a salt-added membrane was due to the diffusion contribution attributable to the pore size of a membrane.

With the study work of Lin and Weng<sup>15–17</sup> and that of Iton et al.,<sup>18</sup> it was found that the CoAlPO<sub>4-5</sub> (aluminophosphate molecular sieved with some aluminum substituted by cobalt species) also had the oxidizing ability. Thus, it prompted us to prepare the CoAlPO<sub>4-5</sub>-blended membrane to improve the O<sub>2</sub>/N<sub>2</sub> selectivity via the higher affinity of CoAlPO<sub>4-5</sub> to oxygen. The main purpose of this study was to determine the relationship between gas transport properties and CoAlPO<sub>4-5</sub> content in the membranes. Oxygen and nitrogen permeability was measured, and the oxygen and nitrogen isotherms were also developed and modeled by the dual sorption model. In addition, effects of upstream pressure on membrane permeability and selectivity were also determined.

## EXPERIMENTAL

### Materials

Polycarbonate (Uplion S-2000) ( $M_w = 28,000$ ) was supplied by Mitsubishi Gas Chemical Co. Chloroform, triethylamine, and cobalt acetate, etc., were purchased from Merck Co. All the above-mentioned chemicals are of reagent grade, and without further purification.

### CoAlPO<sub>4-5</sub> Preparation

Three types of CoAlPO<sub>4-5</sub> with various cobalt content were synthesized according to the procedures described by Messina et al.<sup>19</sup> The CoAlPO<sub>4-5</sub> precursors were calcined (550°C), treated with

NH<sub>4</sub>NO<sub>3</sub> (1 N) solution, and then calcined again. Thus, acquired CoAlPO<sub>4-5</sub> was characterized by XRD to confirm the right crystal pattern. In addition, with UV/VIS spectrometer measurements, the cobalt species was proven to be in a tetrahedral crystal field. In preparing the membrane, the CoAlPO<sub>4-5</sub> added was the sieved particles with sizes below 100 mesh.

Of the following work, Co-3 will be used to represent CoAlPO<sub>4-5</sub> with mol ratio of Co : Al : P in the mother liquor equals 0.03 : 1 : 1, while Co-5 and Co-13 represent Co : Al : P equals 0.05 : 1 : 1 and 0.13 : 1 : 1, respectively.

### Membrane Preparation

PC and CoAlPO<sub>4-5</sub>/PC membranes were prepared from a casting solution of polycarbonate in chloroform. Casting the solution onto a glass plate to a predetermined thickness using a Gardner knife at room temperature formed the membranes required for the study. The membranes were dried in vacuum for 24 h before gas sorption and permeation measurements.

### Gas Permeability Measurements

The apparatus (Yanaco Gas Permeability Analyzer, model GTR-10) used for measuring the gas permeability was shown in our previous report.<sup>20</sup> Permeability of gas was measured by the following equation:

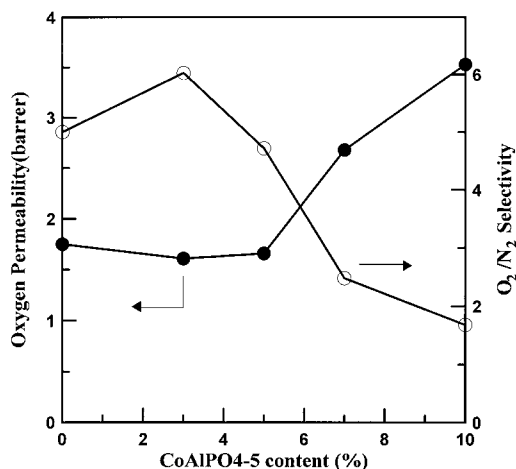
$$P = \frac{l}{(p_1 - p_2)} \frac{q/t}{A}$$

where  $P$  is the gas permeability [cm<sup>3</sup>(STP)-cm/cm<sup>2</sup>-s-cmHg];  $q/t$  is the volume flow rate of gas permeate [cm<sup>3</sup>(STP)/s];  $l$  is the thickness [cm],  $p_1$  and  $p_2$  are the pressures (cmHg) on the high-pressure and low-pressure side of the membrane, respectively; and  $A$  is the effective membrane area [cm<sup>2</sup>]. The gas selectivity is calculated by the following expression:

$$\text{Selectivity} = \frac{P_{O_2}}{P_{N_2}}$$

### Gas Sorption Measurements

The experimental setup for gas sorption measurements was shown in our previous report.<sup>13</sup> Pure oxygen and nitrogen sorptions were measured by a microbalance. The microbalance (Cahn Model



**Figure 1** The effect of CoAlPO<sub>4-5</sub> (Co-3) content on gas permeability and selectivity of polycarbonate membranes.

D-202 Electrobalance) was enclosed in a stainless steel chamber that was placed in a constant temperature box. The system pressure was then vacuumed to about  $4 \times 10^{-3}$  Torr before the gas sorption measurement. The sorption equilibrium measurement were carried out under pressure from 3 to 25 atm. The gas sorption was measured by an microbalance that was enclosed in a stainless chamber. The amount of gas sorption were recorded when sorption equilibrium was reached.

## RESULTS AND DISCUSSION

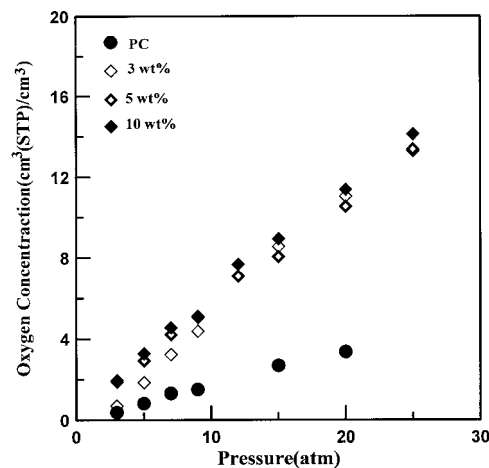
### Effect of CoAlPO<sub>4-5</sub> Content on Gas Selectivity and Permeability

The effects of CoAlPO<sub>4-5</sub> (Co-3) content in polycarbonate membranes on gas permeability and selectivity are shown in Figure 1. As can be seen, oxygen permeability first decreased with increasing the CoAlPO<sub>4-5</sub> content in the membranes, and then increased when more than 3 wt % of CoAlPO<sub>4-5</sub> was added. On the other hand, the O<sub>2</sub>/N<sub>2</sub> selectivity first increased and then decreased when more than 3 wt % of CoAlPO<sub>4-5</sub> was added in the membrane. According to our previous studies,<sup>8,9</sup> the decrease in gas selectivity with increasing additive content could be caused by the exist of a pinhole when excessive additive was added. The polycarbonate (PC) membrane exhibited an oxygen/nitrogen selectivity of 5.0. Compared with a PC membrane, the membranes containing 3 wt % of CoAlPO<sub>4-5</sub> would improve the

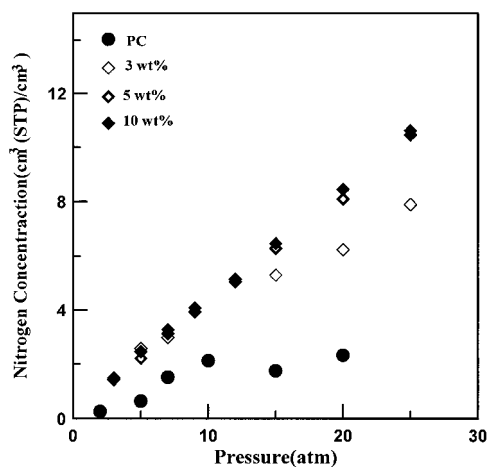
oxygen selectivity from 5.0 to 6.1, with a minor loss of permeability. To understand the gas transport behavior in the membranes, gas solubility and diffusivity were measured.

### Effect of Gas Solubility and Diffusivity on Gas Transport through the Membranes

According to the solution-diffusion model, the O<sub>2</sub>/N<sub>2</sub> selectivity may be attributed to the solubility ratio or the diffusivity ratio of oxygen to nitrogen in the membranes. Gas solubility was determined by the gas sorption measurement. The sorption isotherms of oxygen and nitrogen with various amounts of CoAlPO<sub>4-5</sub> (Co-3) in the membrane are shown in Figures 2 and 3. The amount of oxygen and nitrogen sorption increased with increasing CoAlPO<sub>4-5</sub> content in the membranes. With the amount of gas sorption, gas solubility and diffusivity could be calculated. Gas solubility of the membrane at 3 atm was calculated by the relationship of  $S = C/p$ , where  $S$  is the gas solubility,  $C$  is the amount of gas sorption, and  $p$  is the operating pressure. Besides, gas diffusivity of the membrane is calculated by the relationship of  $P = D \times S$ , where  $P$  is the gas permeability,  $S$  is the gas solubility, and  $D$  is the gas diffusivity of the membranes. The effect of CoAlPO<sub>4-5</sub> content on the gas solubility and diffusivity is shown in Figures 4 and 5. As can be seen, the oxygen and nitrogen solubility increased with CoAlPO<sub>4-5</sub> content, but the oxygen and nitrogen diffusivity initially decreased from 0 to 5 wt % and increased thereafter. But in Figure 5, the diffusivity ratio of

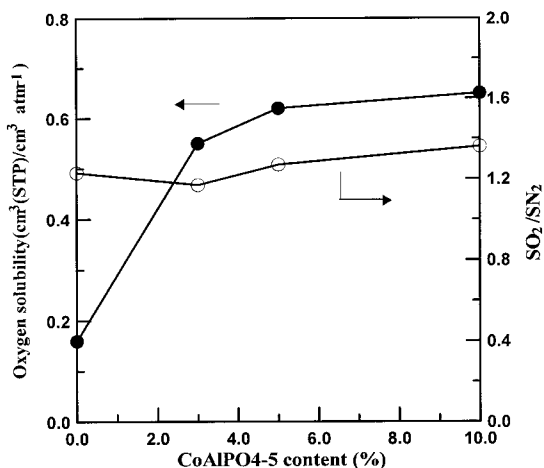


**Figure 2** The sorption isotherm of oxygen with various amount of CoAlPO<sub>4-5</sub> (Co-3) in CoAlPO<sub>4-5</sub>/PC membranes.

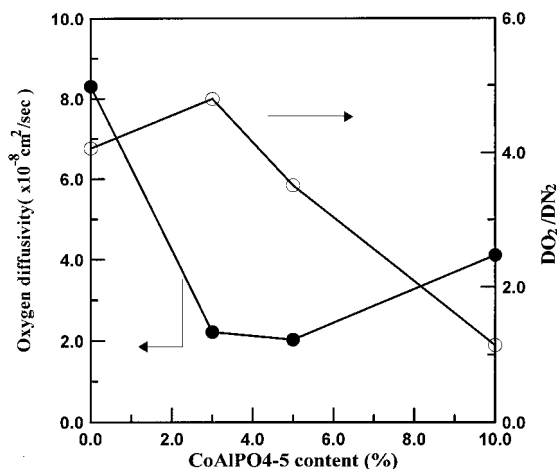


**Figure 3** Sorption isotherm of nitrogen with various amount of  $\text{CoAlPO}_4\text{-5}$  (Co-3) in  $\text{CoAlPO}_4\text{-5/PC}$  membranes.

oxygen to nitrogen reached the maximum value at 3 wt %  $\text{CoAlPO}_4\text{-5}$  content. Moreover, as shown in Figure 4, the solubility ratio of oxygen to nitrogen hardly decreased when  $\text{CoAlPO}_4\text{-5}$  was increased to 3 wt % in the membrane. The permeability, solubility, and diffusivity results were also listed in Table I. Because the increase in oxygen and nitrogen solubility was largely due to the addition of  $\text{CoAlPO}_4\text{-5}$  but the solubility ratio of oxygen to nitrogen only slightly increased from 1.23 to 1.38, it may be concluded that the addition of  $\text{CoAlPO}_4\text{-5}$  into the membrane had a greater impact on gas solubility but a less effect on the solubility ratio of oxygen to nitrogen. From the



**Figure 4** Effect of  $\text{CoAlPO}_4\text{-5}$  (Co-3) content on gas solubility and solubility selectivity of  $\text{CoAlPO}_4\text{-5/PC}$  membranes.



**Figure 5** Effect of  $\text{CoAlPO}_4\text{-5}$  (Co-3) content on gas diffusivity and diffusivity selectivity of  $\text{CoAlPO}_4\text{-5/PC}$  membranes.

above result it can be concluded that the diffusivity ratio of oxygen to nitrogen should be the dominant factor of  $\text{O}_2/\text{N}_2$  selectivity.

It is interesting to note that the gas diffusivity first decreased and then increased. The increase in diffusivity may be caused by pinholes in the membrane, which were induced by the excess addition of  $\text{CoAlPO}_4\text{-5}$ . This assumption was similar to the report of Tsujita.<sup>21</sup> In the work of Tsujita, effects of additive filler on gas transport properties of membranes had been studied, and it showed that polymer–filler systems usually raised the glassy transition temperature due to a higher interface force between polymer and filler. The rise in glassy transition temperature indicated a restricted polymer chain motion because of the strong polymer–filler interaction. Usually, a stronger interaction force between the polymer and filler would result in a higher gas selectivity and lower gas diffusivity. On the contrary, a lower glassy transition temperature indicated that the poor compatibility of polymer and filler existed in the membrane. The poor compatibility usually resulted in the formation of pinholes in the membrane. This, in turn, would lead to lower gas selectivity and higher permeability of polymer–filler membranes when the filler was over the acceptable content. It was indicated that compatibility between polycarbonate and  $\text{CoAlPO}_4\text{-5}$  could be justified by the glassy transition temperatures of membranes.

The effect of  $\text{CoAlPO}_4\text{-5}$  (Co-3) content on glassy transition temperatures of the membranes is shown in Figure 6. In the  $\text{CoAlPO}_4\text{-5/PC}$  sys-

**Table I** Permeability Coefficient, Solubility Coefficient, and Diffusivity Coefficient of CoAlPO<sub>4-5</sub> (Co-3)/PC Membrane

Membrane CoAlPO <sub>4-5</sub> Content	<sup>a</sup> P <sub>O<sub>2</sub></sub> (Barrers)	<sup>a</sup> P <sub>N<sub>2</sub></sub> (Barrers)	<sup>b</sup> S <sub>O<sub>2</sub></sub> (cm <sup>3</sup> cm <sup>-3</sup> atm <sup>-1</sup> )	<sup>b</sup> S <sub>N<sub>2</sub></sub> (cm <sup>3</sup> cm <sup>-3</sup> atm <sup>-1</sup> )	D <sub>O<sub>2</sub></sub> (×10 <sup>-8</sup> cm <sup>2</sup> s <sup>-1</sup> )	D <sub>N<sub>2</sub></sub> (×10 <sup>-8</sup> cm <sup>2</sup> s <sup>-1</sup> )	S <sub>O<sub>2</sub>/S<sub>N<sub>2</sub></sub> Solubility Selectivity</sub>	D <sub>O<sub>2</sub>/D<sub>N<sub>2</sub></sub> Diffusivity Selectivity</sub>
0 wt %	1.75	0.38	0.16	0.13	8.31	2.22	1.23	4.06
3 wt %	1.61	0.28	0.55	0.47	2.22	0.45	1.17	5.15
5 wt %	2.68	0.56	0.62	0.49	3.28	0.87	1.26	3.75
10 wt %	3.53	2.23	0.65	0.47	3.80	3.61	1.38	1.05

<sup>a</sup> Barrer = ×10<sup>-10</sup> cm<sup>3</sup> cm cm<sup>-2</sup> cmHg<sup>-1</sup> s<sup>-1</sup>.

<sup>b</sup> Solubility measured at 3 atm.

tem, the glassy transition temperature decreased with an increase in CoAlPO<sub>4-5</sub> content in the polycarbonate membranes. The decrease in glassy transition temperature indicated that pinholes may exist in the polymer-CoAlPO<sub>4-5</sub> system. It was found that the increase in diffusivity and the decreased in the diffusivity ratio was due to the existence of a pinhole in the membrane when more than 3 wt % of CoAlPO<sub>4-5</sub> was added. Thus, it can be concluded that more pinholes existed when excessive CoAlPO<sub>4-5</sub> was added in the membrane.

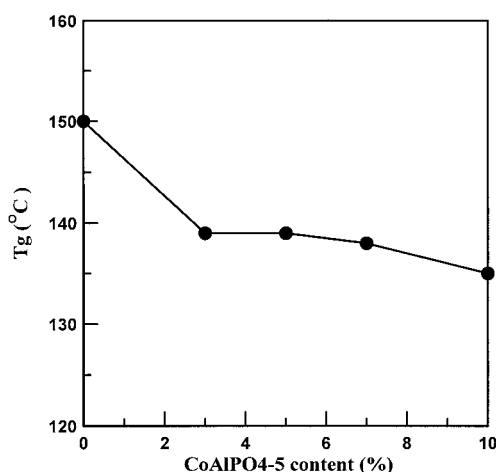
### Effect of Operating Pressure

The effect of operating pressure on the gas separation performance is shown in Figure 7. It was found that the oxygen and nitrogen permeability was independent of the operating pressure. Usually it is the solubility and diffusivity that con-

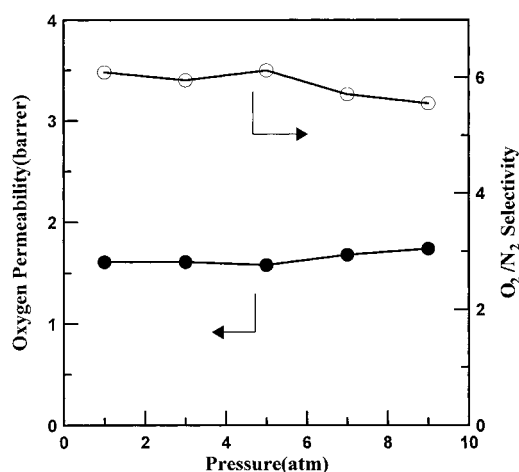
tribute to the gas permeability of a membrane. Figures 8 and 9 show the effect of operating pressure on the gas solubility and effective diffusivity. The solubility of oxygen decreased with an increase in the operating pressure. This result was well described by the dual-sorption model, which indicated that the solubility of Langmuire's type would decrease as the operating pressure increased. The effective diffusivity of oxygen increased with increasing the operating pressure. The increase in effective diffusivity may be attributed to the increase in pore flow that was induced by adding CoAlPO<sub>4-5</sub> into membranes. This result was similar to that of a previous investigation in Co(SalPr)/PC membranes.<sup>13</sup>

### Effect of Various Cobalt Content of CoAlPO<sub>4-5</sub> Additive on Gas Transport Properties

The gas permeability, solubility, and diffusivity of various cobalt contents of CoAlPO<sub>4-5</sub> additive

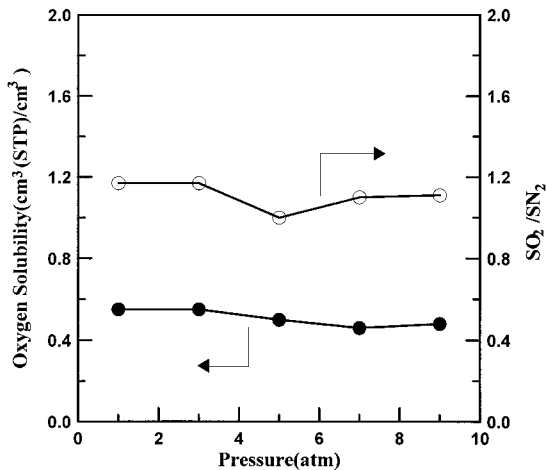


**Figure 6** Effect of CoAlPO<sub>4-5</sub> (Co-3) content on  $T_g$  of CoAlPO<sub>4-5</sub>/PC membranes.



**Figure 7** Effect of operating pressure on gas permeability and selectivity of CoAlPO<sub>4-5</sub>/PC membranes.





**Figure 8** Effect of operating pressure on gas solubility and solubility selectivity of CoAlPO<sub>4</sub>-5/PC membranes.

membranes are listed in Table II. An increase in cobalt content of CoAlPO<sub>4</sub>-5 increased the gas solubility and the solubility ratio of oxygen to nitrogen, but decreased the gas diffusivity and diffusivity ratio of oxygen to nitrogen. The decrease in oxygen diffusivity and diffusivity ratio of oxygen to nitrogen at higher cobalt content CoAlPO<sub>4</sub>-5 may be due to the existence of pinholes when excessive CoAlPO<sub>4</sub>-5 was added to membrane. It has been indicated that the gas solubility increased as the CoAlPO<sub>4</sub>-5 content increased. It can be concluded that the cobalt content in CoAlPO<sub>4</sub>-5 did enhance the gas solubility, but to a lesser extent increased the solubility ratio of oxygen to nitrogen.

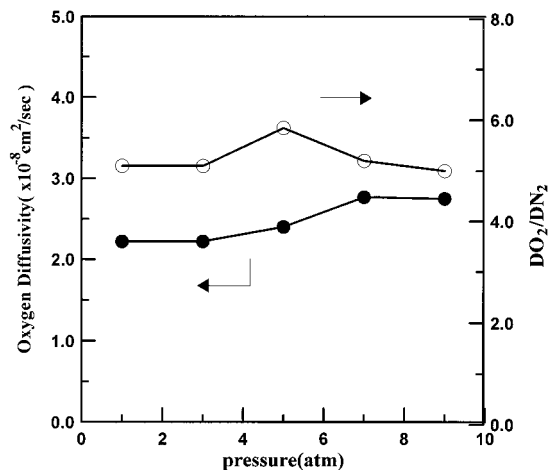
The gas separation factor of the CoAlPO<sub>4</sub>-5/PC membrane was contributed by the diffusivity ratio more than solubility of oxygen to nitrogen. It was found that the solubility ratio of oxygen to nitrogen would increase with increasing cobalt content; nevertheless, the dominant factor of selectivity was the diffusivity ratio rather than the solubility ratio.

An interesting result was that adding filler in membranes generally decreased the diffusivity ratio due to a higher packing density in the polymer and additive system. However, the same percentage of CoAlPO<sub>4</sub> with various cobalt contents was added in PC membranes, and the decreased ratio of the gas diffusivity were different from each other. It was indicated that the compatibility of CoAlPO<sub>4</sub> and PC was dependent on the cobalt content in CoAlPO<sub>4</sub>-5. The CoAlPO<sub>4</sub>-5 membranes with a higher cobalt content would even

more effectively increase the gas solubility, but the ratio of O<sub>2</sub>/N<sub>2</sub> remained unchanged. From the above results it was found that the gas diffusivity ratio played a more important role than the gas solubility ratio for the separation factor of CoAlPO<sub>4</sub>-5 membranes. It can be concluded that the diffusivity ratio of oxygen to nitrogen was the dominated factor.

## CONCLUSION

The CoAlPO<sub>4</sub>-5-blended membrane was prepared by adding CoAlPO<sub>4</sub>-5 into the membranes. It was found that the CoAlPO<sub>4</sub>-5 (Co-3) addition (3 wt %) enhanced oxygen/nitrogen selectivity with a minor decrease in gas permeability. The result of glassy transition temperature and permeability data of CoAlPO<sub>4</sub>-5 (Co-3) membranes showed that pinhole existed in the polymer-CoAlPO<sub>4</sub>-5 system. The pinhole could be induced by adding excessive CoAlPO<sub>4</sub>-5 in the membrane. It was found that CoAlPO<sub>4</sub>-5 with higher cobalt contents would more effectively increase the gas solubility, but to a lesser extent enhanced the solubility ratio of oxygen to nitrogen. It was also found that the dominant factor of gas selectivity was the diffusivity ratio, but not the solubility ratio, when the membrane was blended with various amounts of cobalt in CoAlPO<sub>4</sub>-5. The gas separation factor of CoAlPO<sub>4</sub>-5/PC membrane was contributed by the diffusivity ratio more than the solubility of oxygen to nitrogen.



**Figure 9** Effect of operating pressure on gas diffusivity and diffusivity selectivity of CoAlPO<sub>4</sub>-5/PC membranes.

**Table II Permeability Coefficient, Solubility Coefficient, and Diffusivity Coefficient of Different Type of PC/CoAlPO<sub>4</sub>-5 Membranes**

Membrane CoAlPO <sub>4</sub> -5 Content	<sup>a</sup> Po <sub>2</sub> (Barrers)	<sup>a</sup> Pn <sub>2</sub> (Barrers)	<sup>b</sup> So <sub>2</sub> (cm <sup>3</sup> cm <sup>-3</sup> atm <sup>-1</sup> )	<sup>b</sup> Sn <sub>2</sub> (cm <sup>3</sup> cm <sup>-3</sup> atm <sup>-1</sup> )	Do <sub>2</sub> (×10 <sup>-8</sup> cm <sup>2</sup> s <sup>-1</sup> )	Dn <sub>2</sub> (×10 <sup>-8</sup> cm <sup>2</sup> s <sup>-1</sup> )	So <sub>2</sub> /Sn <sub>2</sub> Solubility Selectivity	Do <sub>2</sub> /Dn <sub>2</sub> Diffusivity Selectivity
PC	1.75	0.38	0.16	0.13	8.31	2.22	1.23	4.06
Co-3 (3 wt %)	1.61	0.28	0.55	0.47	2.22	0.45	1.17	5.15
Co-5 (3 wt %)	1.69	0.32	0.69	0.44	1.86	0.547	1.56	3.64
Co-13 (3 wt %)	1.72	0.30	0.73	0.37	1.80	0.61	1.96	2.87

<sup>a</sup> Barrer = ×10<sup>-10</sup> cm<sup>3</sup> cm cm<sup>-2</sup> cmHg<sup>-1</sup> s<sup>-1</sup>.

<sup>b</sup> Solubility measured at 3 atm.

The authors wish to thank the National Science Council of R.O.C. (NSC 86-2216-E-041-004-T) for financial support.

## REFERENCES

- Battle, T.; Bac, N.; Yilmaz, L. *Sep Sci Technol* 1995, 30, 2365.
- Goldman, M.; Fraenkel, D.; Levin, G. *J Appl Polym Sci* 1989, 37, 1791.
- Kemp, D. R.; Paul, D. R. *J Polym Sci Phys Ed* 1974, 12, 485.
- Duval, J. M.; Folkers, B.; Mulder, M. V.; Desgrandchamps, G.; Somlders, C. A. *J Membr Sci* 1993, 80, 189.
- Suer, M. G.; Bac, N.; Yilmaz, L. *J Membr Sci* 1994, 91, 77.
- Dotremont, C.; Brabants, B.; Geeroms, K.; Mewis, J.; Vandecasteele, C. *J Memb Sci* 1995, 104, 109.
- Jia, M.; Peinemann, K. V.; Behling, R. D. *J Membr Sci* 1991, 57, 289.
- Duval, J. M.; Kemperman, A. J. B.; Folkers, B.; Mulder, M. H. V.; Desgradchamps, G. *J Membr Sci* 1994, 54, 409.
- Gur, T. M. *J Memb Sci* 1994, 93, 283.
- Chen, S. H.; Ruaan, R. C.; Lai, J. Y. *Sep Sci Technol* 1997, 32, 925.
- Chen, S. H.; Lai, J. Y.; Ruaan, R. C.; Wang, A. A. *J Membr Sci* 1997, 123, 197.
- Chen, S. H.; Lai, J. Y. *J Appl Polym Sci* 1996, 59, 1129.
- Ruaan, R. C.; Chen, S. H.; Lai, J. Y. *J Membr Sci* 1997, 135, 9.
- Chen, S. H.; Ruaan, R. C.; Lai, J. Y. *J Membr Sci* 1997, 134, 143.
- Lin, S. S.; Weng, H. S. *Appl Catal* 1993, 105, 289.
- Lin, S. S.; Weng, H. S. *J Chem Eng Jpn* 1994, 27, 211.
- Lin, S. S.; Weng, H. S. *Appl Catal* 1994, 1118, 21.
- Iton, L. E.; Choi, I.; Desjardins, J. A.; Maroni, V. A. *Zeolites* 1989, 9, 535.
- Messina, C. A.; Lok, B. M.; Flanigen, E. M. U.S. Pat. 4,544,143 (1985) to Union Carbide.
- Lai, J. Y.; Chen, S. H.; Lee, M. S.; Shyu, S. S. *J Appl Polym Sci* 1993, 47, 1513.
- Tsujita, Y. *Membrane Science and Technology*; Osada, Y.; Nakagawo, T., Eds.; Dekker: New York, 1992, p. 3.