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Operational Model for Evaluating the Permeation of Mixed Gas Through Poly(dimethylsiloxane) Membrane*

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Abstract An operational model is developed to evaluate and predict the permeation performance of mixed gas through poly(dimethylsiloxane) (PDMS) membranes by combining the ideal gas permeation model with the experimental analysis of the mixed gas transport character. This model is tested using the binary and ternary mixed gas with various compositions through the PDMS membranes, and the predicted data of the permeation flux and the compositions of the permeated gas are in good agreement with the experimental ones, which indicates that the operational model is applicable for the evaluation of the permeation performance of mixed gas through PDMS membranes.

Keywords operational model, poly (dimethylsiloxane) membrane, mixed gas, permeation performance

1 INTRODUCTION

Measurement and analysis of gas permeability in polymeric membranes is an essential element in proper design and selection of operating conditions for gas separation systems[1]. In the permeation of gas mixture through membranes, one component exhibits different transport parameters from the pure state owing to the presence of other components with different characters. Therefore, using the permeation data of pure gas to evaluate the separation performance of gas mixture may lead to erroneous results, particularly in the case of condensable components or one component interacting with the polymer matrix[2]. Extensive studies regarding the difference in permeation behavior between the components in the pure state and that in the mixed state have been made in literatures[1—12]. Of them, the coupling effect including solution coupling and diffusion coupling is thought to be an important factor responsible for the deviation[2,6,8,9], and the deviation is more prominent in glassy membranes than in rubbery membranes[1,6,8]. In addition, the plasticization effect greatly influences the transport process, particularly in the case of the mixtures containing some components such as carbon dioxide and organic vapors. These condensable penetrants interact strongly with membrane matrix, which results in the swelling of matrix and the enhancement of the permeability[9—11]. Moreover, under some operation modes, the concentration polarization has significant influence on the permselectivity at the lower feed flowrate[12].

To acquire a better understanding of permeation behavior in mixed state and to achieve ideal separation efficiency, it is necessary to establish a practical model to guide the estimation of the separation capability under conditions beyond the available experimental ones. However, to the extent of our knowledge, there are scarce models applied to predict practical performance of membranes in terms of the controllable parameters, such as pressure, temperature, *etc*. Ettouney and Majeed[1] developed permeability functions to describe the permeation behavior for pure and mixtures of N_2, O_2, CH_4 and CO_2 through polysulfone and silicone rubber membranes, in which the permeability of one component is expressed as a linear relationship of the partial pressure of the species. Prabhakar *et al*.[13] established a self-consistent model to describe the dependence of gas and vapor permeability on concentration and temperature in rubber polymers, by which the variation of the propane permeability with the permeate pressure was accurately predicted. Conesa *et al*.[14] derived a mathematical model on the basis of mass balance to calculate the composition of penetrants as a function of the different experimental parameters by investigating the transport of H_2-N_2 binary gas mixture across ceramic membranes.

The objective of this work was to develop an operational model to evaluate the separation performance of a given membrane-based gas separation system, and further optimize the membrane system design. In this study, poly(dimethylsiloxane) (PDMS) was chosen as the model polymer because it is used commercially as a gas separation membrane material. The permeation flux and the permeated gas composition through PDMS membrane was investigated at various operation conditions using binary and ternary gas mixtures of O_2, N_2 and CO_2 with various compositions, and the experimental data were compared with the predicted values by the operational model.

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2 BASIC PRINCIPLES AND ASSUMPTIONS

In general, the transport behavior of gas through dense polymer membrane is typically described by the solution-diffusion model. In other words, the permeability is determined by the solubility and the diffusivity of the gas in the membranes[15]. For ideal permeation of binary mixture through membranes where there are no interactions between the gases, as well as no gas-polymer special interactions because of the presence of the second gas, the permeation parameters of the ideal gas mixture could be calculated as follows[2,6,16]:

$$p_i = \frac{Q_i \cdot l}{\Delta P_i} = D_i \cdot S_i \tag{1}$$

$$Q_i = \frac{V_i}{A \cdot t} = \frac{p_i}{l} \cdot \Delta P_i \tag{2}$$

$$Q_t = Q_1 + Q_2 \tag{3}$$

$$Y_i = \frac{Q_i}{Q_t} \tag{4}$$

$$\Delta P_i = (X_i \cdot P_f - Y_i \cdot P_p) \tag{5}$$

where p_i, D_i and S_i are permeability coefficient, diffusivity coefficient and solubility coefficient of the ith gas in the polymer, respectively; l is the thickness of the membrane; ΔP_i is the partial pressure difference of the ith gas through the membrane; V_i is the volume of the ith gas; t is the permeation time and A is the membrane area; Q_t is the steady-state total permeation flux of the mixed gases; Q_i is the steady-state permeation flux of the ith component, and the subscripts 1 and 2 denote component gases, respectively; P_f and P_p are the feed pressure and the permeate pressure, respectively; X_i and Y_i are the molar fractions of the ith component in the feed and permeated gases, respectively.

As mentioned above, because of the existence of coupling effect, plasticization effect and concentration polarization during the transport process of mixed gas through membranes, the permeability of one component in mixed gas is different from that in pure gas state, and the permeability also varies with the operation conditions[13]. This makes the determination of the actual permeability and further evaluating of the separation efficiency difficult. Therefore, an attempt is made to establish a model to evaluate the actual separation performance using the operational parameters.

In the case of binary gas mixture transport through membrane, when the operation parameters such as temperature and permeate pressure are constant, the total permeation flux Q_t and the molar fraction Y_i of one component in the permeated gas are considered to be the function of feed pressure and composition,

$$Q_t = F(P_f, X_i) \tag{6}$$

$$Y_i = G(P_f, X_i) \tag{7}$$

As already known, the total flux is linearly dependent on the pressure difference[17,18]. When X_i is invariable, the total flux Q_t can be expressed as follows,

$$Q_t = F(P_f, X_i) = K \cdot (P_f - P_p) \tag{8}$$

It is also well known that the molar fraction Y_i of one component in permeated gas has nonlinear relationship with the pressure difference. Here it is assumed that the relationship between the Y_i and pressure could be expressed as exponential dependence on the basis of the nonlinear trend in literatures[17,18],

$$Y_i = G(P_f, X_i) = M \cdot \left(\frac{P_f - P_p}{P_0} \right)^N \tag{9}$$

where M and N are constants, P_0 is the standard pressure.

Based on Eq.(4), the permeation flux Q_i of ith component could be expressed as below,

$$Q_i = \frac{K \cdot M}{P_0^N} \cdot (P_f - P_p)^{N+1} = \Phi \cdot (P_f - P_p)^{\Psi} \tag{10}$$

where

$$\Phi = \frac{K \cdot M}{P_0^N} \tag{11}$$

$$\Psi = N + 1 \tag{12}$$

By constructing the curves of Q_t and Y_i with P_f as variable, the parameters of M, N and K can be obtained by fitting Q_t and Y_i to P_f, respectively, then the values of Φ and Ψ can be calculated. In the same way, the corresponding values of Φ and Ψ can be obtained for various X_i. Thereby, Φ and Ψ can be considered as the function of X_i,

$$\Phi = \phi(X_i) \tag{13}$$

$$\Psi = \psi(X_i) \tag{14}$$

By constructing the curves of Φ and Ψ with the X_i as variable, the expressions of $\phi(X_i)$ and $\psi(X_i)$ can be obtained by fitting Φ and Ψ to X_i.

Then the expressions of actual Q_i, Q_t and Y_i can be deduced as below:

$$Q_i = \phi(X_i)(P_f - P_p)^{\psi(X_i)} \quad (i=1,2) \tag{15}$$

$$Q_t = \Sigma\phi(X_i)(P_f - P_p)^{\psi(X_i)} \quad (i=1,2) \tag{16}$$

$$Y_i = \frac{Q_i}{Q_t} = \frac{\phi(X_i)(P_f - P_p)^{\psi(X_i)}}{\Sigma\phi(X_i)(P_f - P_p)^{\psi(X_i)}} \quad (i=1,2) \tag{17}$$

Based on Eqs.(2) and (15), the actual permeability p_i of the ith component in the mixed gas was deduced.

$$p_i = \frac{l\phi(X_i)(P_f - P_p)^{\psi(X_i)}}{X_i P_f - \dfrac{\phi(X_i)(P_f - P_p)^{\psi(X_i)}}{\Sigma\phi(X_i)(P_f - P_p)^{\psi(X_i)}} P_p} \tag{18}$$

3 EXPERIMENTAL

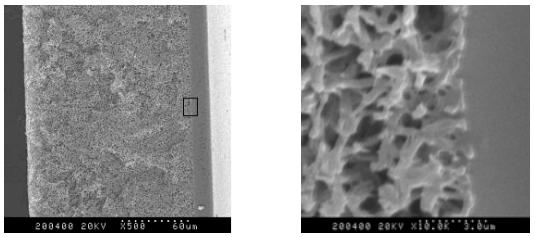
3.1 Materials

Poly (dimethylsiboxane) with an average

molecular weight of 5000 was provided by Shanghai Synthetic Resin Company, China and used without any further purification. *N*-heptane solvent, tetraethyl orthosilicate (TAOS) curing agent and dibutyltin dilaurate (DBTDL) catalyst were obtained from Shanghai Chemical Agents Company, China. Cellulose acetate (CA) porous membrane was purchased from the Shanghai Xinya Purification Company, China. Nitrogen, oxygen and carbon dioxide used in the permeation experiments with a purity of over 99% were purchased from Nanjing Tongguang Special Gas Company, China.

3.2 Membrane preparation

PDMS was dissolved in *n*-heptane to give a 5% (by mass) solution, in which the TAOS and DBTDL were added with mass ratio of 5:1. The ready casting solution was cast on the surface of CA porous membrane which was previously put on the surface of water in a basin, and the solvent was evaporated at ambient temperature for 6 hours. Then the initial PDMS film with support membrane was extracted from the basin and put into a vacuum oven at 60℃ for 6 hours to complete the cross-linking. As shown in Fig.1, the thickness of the PDMS skin layer was approximately 14 microns and the support layer was a microporous structure, and generally, the effect of the support layer on the gas transport across membranes is negligible.



(a) Whole cross-section (b) Local enlargement at the interface

Figure 1 SEM micrograph of the PDMS composite membrane

3.3 Permeation measurements

A laboratory-scale gas permeation set-up is shown in Fig.2. The permeation properties of gases across the prepared PDMS membranes were determined by the constant pressure/variable volume method. The PDMS membranes were fixed into a membrane cell to offer an effective membrane area of 64cm^2. The feed gas with various compositions was obtained by matching the pure gas in a match cell. The feed gas pressure was adjusted by the pressure regulator-6 and retentate valve-10 together, and the permeate pressure was atmospheric. The temperature was maintained at 25℃ using a temperature control system. The same PDMS composite membrane was used for all permeation measurements. The flowrate of feed and permeate gas were measured by soap-film bubble rotameters and flowmeters was as the volume flux in the calculation without being converted into standard volume. The ratio of permeate flow rate to feed flow rate was always less than 0.5%. Under these conditions, the retentate composition was essentially equal to the feed composition and concentration polarization effects were negligible.

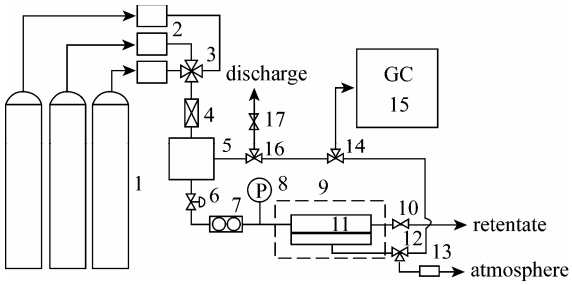


Figure 2 Schematic diagram of experimental setup

1—gas cylinder; 2—flowrate regulator; 3—four-way valve; 4—cleaner; 5—match gas cell; 6—pressure regulator; 7, 13—flowmeter; 8—pressure gauge; 9—temperature control system; 10, 17—valve; 11—membrane cell; 12, 14, 16—three-way valve; 15—GC system

The compositions of feed and permeated gases were determined by Agilent 6890N gas chromatograph (GC) equipped with a thermal conductivity detector (TCD). The GC column was 2m long with 0.32cm inside diameter having TDX-01 as fixing phase, which was provided by Lanzhou Institute of Chemistry and Physics, China. The GC temperature profile was 70℃ (oven), 70℃ (injector), 180℃ (detector). The compositions were measured quickly and accurately with the help of the attached GC chemstation.

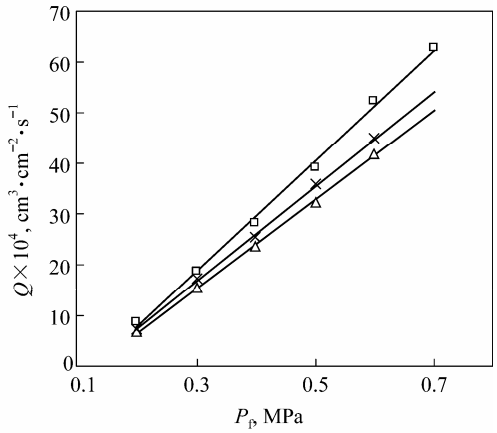
The permeation data used in the article were the average values of the measurements under the same condition and the experimental error was estimated to be about ±5%.

4 RESULTS AND DISCUSSION

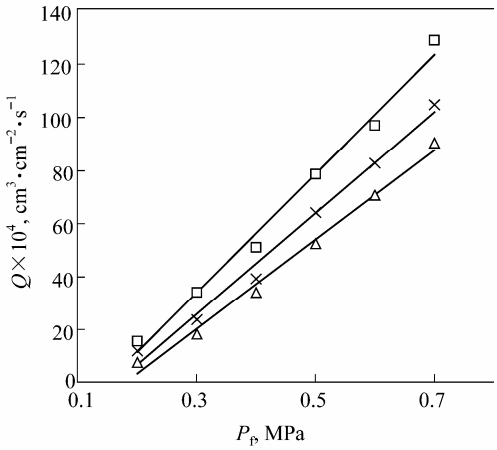
4.1 Permeation of binary gas mixture

It is shown in Fig.3 that the permeation fluxes of O_2-N_2, CO_2-N_2 and O_2-CO_2 binary gas mixtures vary with the feed pressure and the feed molar composition. The total flux of binary mixed gas increases with the increase of feed pressure and the increase in the percentage of the faster gas in the feed. And in the same proportion, the total flux of gas mixture containing faster gas (CO_2) is more than that containing slower ones (N_2) with O_2 as an example. The fitted curves in Fig.3 show that the fluxes are linearly dependent on the feed pressure.

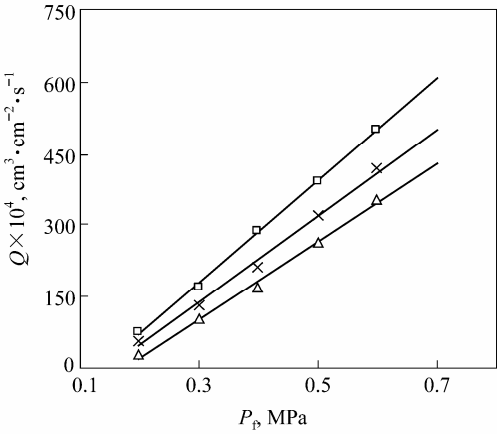
The variations of the molar fraction of one component in the permeated gas with the feed pressure and the feed composition are shown in Fig.4. With the increase of feed pressure, the molar fraction of O_2 in the permeated gas ascends for O_2-N_2 mixed gas and descends for O_2-CO_2 mixed gases. However, the



(a) △ 11% O_2, 89%N_2; × 21%O_2, 79%N_2; □ 31%O_2, 69%N_2; — fitted line



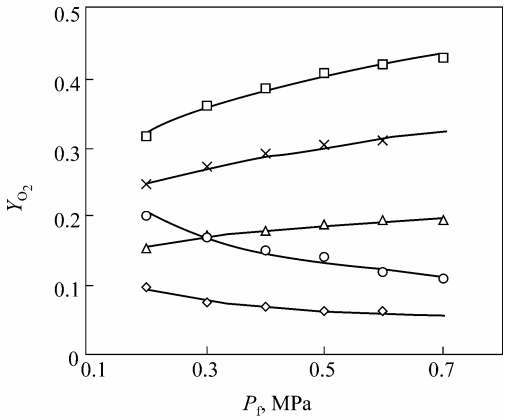
(b) △ 11%CO_2, 89%N_2; × 21%CO_2, 79%N_2; □ 31%CO_2, 69%N_2; — fitted line



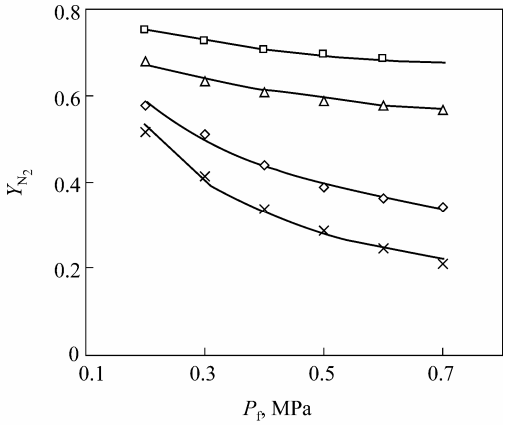
(c) △ 31%O_2, 69%CO_2; × 21%O_2, 79%CO_2; □ 11%O_2, 89%CO_2; — fitted line

Figure 3 Variation of the permeation flux with the feed pressure and the feed molar composition

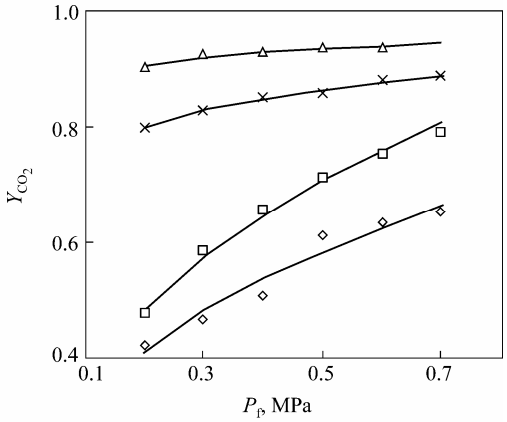
molar fractions of N_2 in the permeated gas descend for both O_2-N_2 and N_2-CO_2 mixed gas, and CO_2 is just the reverse. The variation indicates that the higher permeable component (CO_2 to O_2 or O_2 to N_2) dominates the permeation process[2]. And the increase of feed pressure is favorable to the enrichment of faster component in the permeated gases. The fitted curves in the Fig.4 indicate that there is exponential relationship between the molar fraction of one component in the permeated gas and the feed pressure.



(a) ◇ 21%O_2, 79% CO_2; ○ 31%O_2, 69%CO_2; △ 11%O_2, 89%N_2; × 21%O_2, 79%N_2; □ 31%O_2, 69%N_2; — fitted curve



(b) ◇ 21%CO_2, 79% N_2; × 31%CO_2, 69%N_2; △ 31%O_2, 69%N_2; □ 21%O_2, 79%N_2; — fitted curve



(c) ◇ 21%CO_2, 79% N_2; □ 31%CO_2, 69%N_2; × 31%O_2, 69%CO_2; △ 21%O_2, 79%CO_2; — fitted curve

Figure 4 Variation of the permeated gas composition with the feed pressure and the feed molar composition

Table 1 Fitting parameters for binary gas mixtures

	Component	α	β	γ	δ	Average error of the model prediction, %	
						Total flux	Molar fraction
1	O_2	149.57	0.16	0.23	1.1	3.41	3.77
	N_2	57.85	14.03	0.41	0.62		
2	O_2	105.07	24.6	0.99	0.28	7.22	6.72
	CO_2	626.89	55.52	0.05	0.98		
3	N_2	55.28	4.81	0.87	0.02	5.19	7.07
	CO_2	477.85	29.73	0.01	1.29		

The fitted curves of flux and composition illustrated in Figs.3—4 give the best-fitting parameters of Φ and Ψ, By constructing the curves of the Φ and Ψ with X_i as variable, the expressions of $\phi(X_i)$ and $\psi(X_i)$ are fitted as below,

$$\phi(X_i) = \alpha_i X_i + \beta_i \quad (i=1,2) \tag{19}$$

$$\psi(X_i) = \gamma_i X_i + \delta_i \quad (i=1,2) \tag{20}$$

then

$$Q_i = (\alpha_i X_i + \beta_i)(P_f - P_p)^{(\gamma_i X_i + \delta_i)} \quad (i=1,2) \tag{21}$$

$$Q_t = \Sigma(\alpha_i X_i + \beta_i)(P_f - P_p)^{(\gamma_i X_i + \delta_i)} \quad (i=1,2) \tag{22}$$

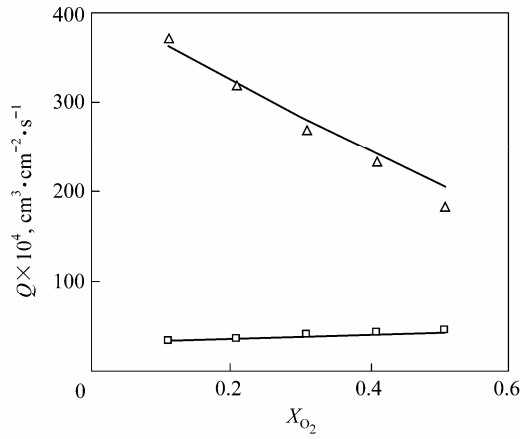
$$Y_i = \frac{(\alpha_i X_i + \beta_i)(P_f - P_p)^{(\gamma_i X_i + \delta_i)}}{\Sigma(\alpha_i X_i + \beta_i)(P_f - P_p)^{(\gamma_i X_i + \delta_i)}} \quad (i=1,2) \tag{23}$$

$$p_i = \frac{lQ_i}{X_i P_f - Y_i P_p} \quad (i=1,2) \tag{24}$$

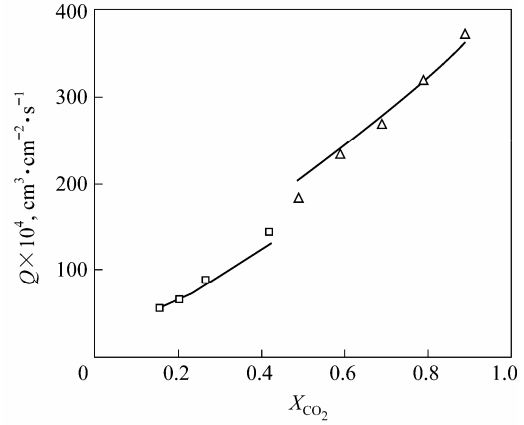
The fitted parameters α, β, γ and δ are the constants for a certain binary gas and their values are listed in Table 1.

A series of binary mixed gas with different feed composition were used to verify the permeation functions at the same feed pressure of 0.5MPa. Figs.5—6 show the comparison of the calculated results (plotted as curves) with the experimental data (plotted as points). It is obvious that the calculated results are in good agreement with experimental data, and the corresponding average errors of the model prediction are listed in Table 1. Fig.5 also reveals the dependence of the permeation flux of binary gas mixtures on the feed compositions, and the flux rises with the increase in the percentage of the faster gas (CO_2 to O_2 and O_2 to N_2) in the feed mixture. Fig.6 shows the variation in the molar fractions of components in the permeated gases with their percentages in the feed gases. The molar fraction of the faster components in permeated gas is higher than that in the feed gas, and the reverse is found for the slower components, which indicates that the faster gas is dominant in the permeation competition[2].

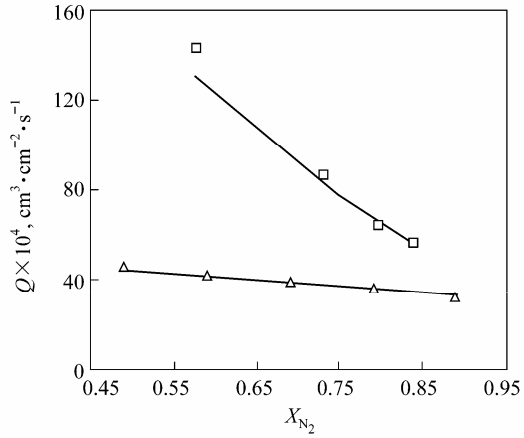
Figure 7 illustrates the variation of the permeability coefficient of O_2 with the composition of binary mixture. It proves that the permeability coefficient of one component in mixture is not invariable, and it depends on the character of the other component.



(a) □ O_2-N_2; △ O_2-CO_2; — calculated values

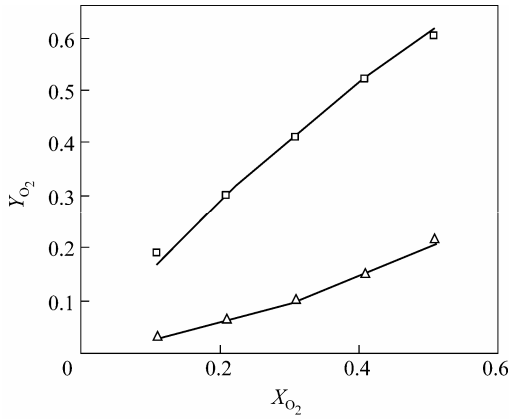


(b) □ N_2-CO_2; △ O_2-CO_2; — calculated values

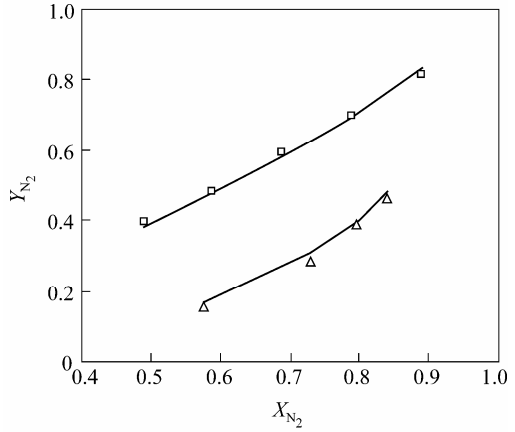


(c) □ CO_2-N_2; △ O_2-N_2; — calculated values

Figure 5 Variation of the permeation flux with the feed molar composition at the feed pressure of 0.5MPa



(a) □ O_2-N_2; △ O_2-CO_2; — calculated values



(b) □ O_2-N_2; △ CO_2-N_2; — calculated values

Figure 6 Variation of the permeated gas composition with the feed molar composition at the feed pressure of 0.5MPa

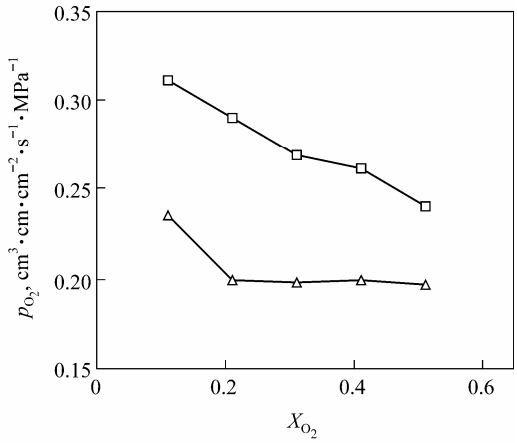


Figure 7 Variation of the permeability of O_2 with the feed composition at the feed pressure of 0.5MPa

□ O_2-CO_2; △ O_2-N_2

4.2 Application of the model to ternary gas mixture

To verify the application of the operational model deduced from binary gas mixture, the model was extended to the ternary gas mixture for test. For the ternary gas mixture, there also exists complex effect among penetrants, and the others influence the permeation behavior of one component together. On the basis of the expression of the model in binary gas mixtures, the corresponding permeation functions of ternary gas mixture have the following expressions,

$$Q_i = (\alpha_i X_i + \beta_i)(P_f - P_p)^{(\gamma_i X_i + \delta_i)} \quad (i=1,2,3) \quad (25)$$

$$Q_t = \Sigma(\alpha_i X_i + \beta_i)(P_f - P_p)^{(\gamma_i X_i + \delta_i)} \quad (i=1,2,3) \quad (26)$$

$$Y_i = \frac{(\alpha_i X_i + \beta_i)(P_f - P_p)^{(\gamma_i X_i + \delta_i)}}{\Sigma(\alpha_i X_i + \beta_i)(P_f - P_p)^{(\gamma_i X_i + \delta_i)}} \quad (i=1,2,3) \quad (27)$$

$$p_i = \frac{lQ_i}{X_i P_f - Y_i P_p} \quad (i=1,2,3) \quad (28)$$

where subscript 1, 2, 3 denote the species, α, β, γ and δ are the constants for a certain ternary gas. In the case of the ternary gas mixture, respective permeation behavior could be strengthened or weakened owing to the complex interactions among the components. And from the corresponding experimental data of binary gas mixtures, the influence from the others can be evaluated approximately. To simplify the calculation, it is assumed that the values of α, β, γ and δ are determined by the geometry mean of the values of two corresponding binary mixtures listed in Table 1, and the calculated values of four constants are given in Table 2.

In order to verify the permeation function, a series of permeation experiments of O_2, N_2 and CO_2 ternary mixtures with various feed compositions were carried out under the same condition as binary gas mixture. Comparison of calculated values and experimental data in Figs.8—9 reveals that there is a good agreement between them and the average errors of the model prediction are listed in Table 2. Therefore, it is proven that the assumption about the model parameters, α, β, γ and δ, are correct and the permeation model is applicable to the ternary mixtures.

Figures 8 and 9 also show the effect of the feed pressure on the flux and the permeated gas compositions of ternary mixed gas. With the increase of the feed pressure, the total flux rises, and the molar fractions of the slower components O_2 and N_2 in

Table 2 Fitting parameters for ternary mixtures

Component	α	β	γ	δ	Average error of model prediction, %	
					Total flux	Molar fraction
O_2	125.36	1.98	0.48	0.55		
N_2	56.55	8.21	0.60	0.11	9.26	8.46
CO_2	547.32	40.63	0.02	1.12		

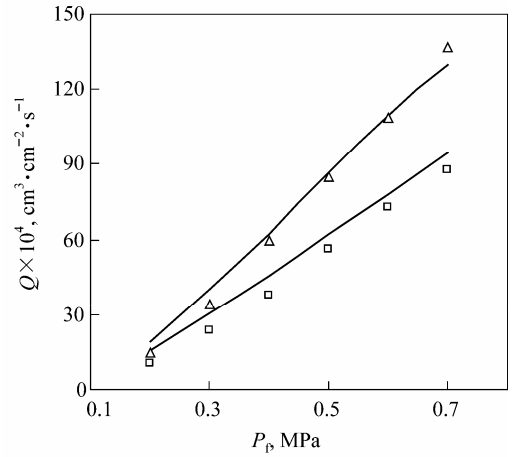
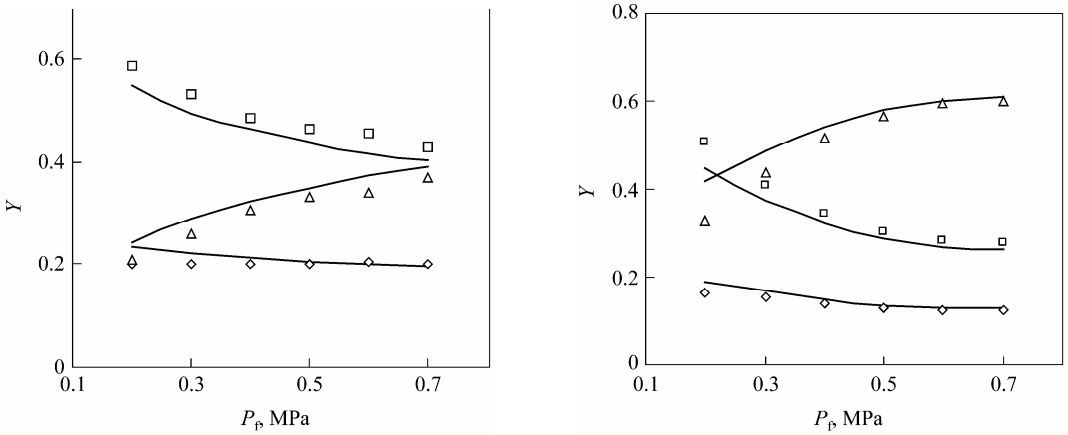


Figure 8 Variation of the permeation flux with the feed pressure and the feed molar composition
△ 15.5%O_2, 62.9%N_2, 21.6%CO_2; □ 17.7%O_2, 70%N_2, 12.3%CO_2; — calculated values



(a) Feed gas (17.7%O_2, 70%N_2, 12.3%CO_2) (b) Feed gas (15.3%O_2, 62.1%N_2, 22.7%CO_2)

Figure 9 Variation of the permeated gas composition with the feed pressure and the feed molar composition
◇ O_2; □ N_2; △ CO_2; — calculated values

Table 3 The variation of the permeated gas composition with feed gases at the feed pressure of 0.5 MPa

Feed molar fraction, %			Experimental data, %			Calculated results, %		
O_2	N_2	CO_2	O_2	N_2	CO_2	O_2	N_2	CO_2
7.5	32.8	59.7	3.0	8.0	89.0	2.8	7.3	89.9
10.9	46.7	42.4	6.0	14.5	79.5	5.9	13.2	80.9
15.3	62.1	22.7	13.0	30.3	56.6	13.6	27.3	59.1
17.7	70.5	11.6	20.8	49.4	29.8	21.9	45.6	32.4
19.1	78.3	2.6	30.1	55.5	14.4	32.4	54.8	12.8

permeated gas descend and that of the faster gas CO_2 ascends. Table 3 illustrates the dependence of the permeated gas composition on the feed composition of ternary gases at constant feed pressure of 0.5MPa. With the variation in the proportion of three species in feed gas, the molar fraction of N_2 in permeated gas is always lower than that in feed gas and the reverse is found for CO_2. However, the variation in the molar fraction of O_2 in permeated gas is uncertain, which depends on the proportion of the much slower gas N_2 and the faster gas CO_2. When the effect of N_2 on the transport of O_2 exceeds the effect of CO_2, then O_2 is enriched in the permeated gas and has a higher molar fraction than that in feed gas.

The above experimental results indicate that the operational model established in this study could be used to predict the flux and the permeated gas composition of one gas mixture through PDMS membranes at some operation conditions accurately and quickly, and further to evaluate the separation capacity of a

membrane system and provide data for the design of gas membrane separation system efficiently.

5 CONCLUSIONS

An operational model is presented to evaluate and predict quantitatively the permeation flux and the permeated gas composition of mixed gas though PDMS membranes by combining the ideal gas permeation model with the experimental analysis of the mixed gas transport character. The permeation behaviors of binary and ternary mixtures of O_2, N_2 and CO_2 through PDMS membrane were investigated at various operation conditions, and the calculated values of the operational model show good agreement with the experimental results. In addition, the model is established mainly based on two adjustable operation parameters, i.e., feed pressure and feed gas composition. Therefore, it is more convenient to acquire the data of the flux and the permeated gas composition, which are the two most important indexes to evaluate the separation capability of the membrane-based gas separation systems. Although the model is presented on the basis of the permeation behaviors of O_2, N_2 and CO_2 through the PDMS membrane, the research method can be employed to study the transport properties of other gas membrane-separation systems macroscopically.

NOMENCLATURE

A	membrane area, cm^2
D	diffusivity coefficient, $cm^2 \cdot s^{-1}$
l	thickness of membrane, cm
P	pressure, MPa
ΔP	pressure difference, MPa
P_0	standard pressure
p	permeability coefficient, $cm^3 \cdot cm \cdot cm^{-2} \cdot s^{-1} \cdot MPa^{-1}$
Q	permeation flux, $cm^3 \cdot cm^{-2} \cdot s^{-1}$
S	solubility constant, $cm^3 \cdot cm^{-3} \cdot MPa^{-1}$
t	permeation time interval, s
V	volume of permeation gas, cm^3
X	molar fraction of feed gas
Y	molar fraction of permeated gas

Subscripts

f	feed
i	component
p	permeated

REFERENCES

1 Ettouney, H., Majeed, U., "Permeability functions for pure and mixture gases in silicone rubber and polysulfone membranes: Dependence on pressure and composition", *J. Membr. Sci.*, **135**, 251—261(1997).

2 Dhingra, S.S., Marand, E., "Mixed gas transport through polymeric membranes", *J. Membr. Sci.*, **141**, 45—63(1998).

3 Jordan, S.M., Koros, W.J., "Permeability of pure and mixed gases in silicone rubber at elevated pressure", *J. Polym. Sci. Part B*: *Polym. Phys.*, **28**, 795—809(1990).

4 Hughes, R., Jiang, B.Q., "The permeabilities of carbon dioxide, nitrous oxide and oxygen and their mixtures through silicone rubber and cellulose acetate membranes", *Gas. Sep. Purif.*, **9**, 27—30(1995).

5 Stern, S.A., "Polymers for gas separation: The next decade", *J. Membr. Sci.*, **94**, 1—65(1994).

6 Yeom, C.K., Lee, S.H., Lee, J.M., "Study of transport of pure and mixed CO_2/N_2 gases through polymeric membranes", *J. Appl. Polym. Sci.*, **78**, 179—189(2000).

7 Merkel, T.C., Bondar, V.I., Nagai, K., Freeman, B.D., Pinnau, I., "Gas sorption, diffusion, and permeation in poly (dimethylsiloxane)", *J. Polym. Sci. B*: *Polym. Phys.*, **38**, 415—434(2000) .

8 Pandey, P., Chanhan, R.S., "Membranes for gas separation", *Prog. Polym. Sci.*, **26**, 853—893(2001).

9 Pinnau, I., He, Z.J., "Pure- and mixed-gas permeation properties of polydimethylsiloxane for hydrocarbon/methane and hydrocarbon/hydrogen separation", *J. Membr. Sci.*, **244**, 227—233(2004).

10 Wang, R., Liu, S.L., Lin, T.T., Chung, T.S., "Characterization of hollow fiber membranes in a permeator using binary gas mixtures", *Chem. Eng. Sci.*, **57**, 967—976(2002).

11 Jordan, S.M., Koros, W.J., Fleming, G.K., "The effects of CO_2 exposure on pure and mixed gas permeation behavior: Comparison of glassy polycarbonate and silicone rubber", *J. Membr. Sci.*, **30**, 191—212(1987).

12 Peng, F.B., Liu, J.Q., Li, J.T., "Analysis of the gas transport performance through PDMS/PS composite membranes using the resistances-in-series model", *J. Membr. Sci.*, **222**, 225—234(2003).

13 Prabhakar, R.S., Raharjo, R., Toy, L., Lin, H.Q., Freeman, B.D., "Self-consistent model of concentration and temperature dependence of permeability in rubbery polymers", *Ind. Eng. Chem. Res.*, **44**, 1547—1556(2005).

14 Conesa, A., Fernandez, A., Pitarch, J.A., Vicente-Mingarro, I., Rodriguez, M.A., "Separation of binary gas mixtures by means of sol-gel modified cermic membranes: Prediction of membrane performance", *J. Membr. Sci.*, **155**, 123—131(1999).

15 Wijmans, J.G., Baken, R.W., "The solution-diffusion model: A review", *J. Membr. Sci.*, **107**, 1—21(1995).

16 Mulder, M., Basic Principles of Membrane Technology, Kluwer Academic Publishers (1996).

17 Huang, M.R., Li, S.X., Dong, Z.Q., Feng, W., Wang, X.Y., Gu, S.Y., Wu, Y.H., Huang, X.A., "Oxygen enrichment from air through multiplayer thin low density polyethylene films", *J. Appl. Polym. Sci.*, **83**, 3013—3021(2002).

18 Huang,M.R., ,Xu,Q.W., Gu, S.Y., Wu, Y.H., Dong, Z.Q., Wang, J., Shao, H.T., Wang, X.S., "Oxygen enrichment across blend membranes of bipyridine and ethyl cellulose", *J. Appl. Polym. Sci.*, **87**, 1371—1381(2003).