

# Compatibility of PTET-60/CA Blends and Separation Performance of Their Membranes for Benzene/Cyclohexane Mixture by Pervaporation

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Received 8 November 2005; accepted 24 March 2006

DOI 10.1002/app.24557

Published online in Wiley InterScience (www.interscience.wiley.com).

**ABSTRACT:** Aromatic copolyester of poly(trimethylene-*co*-ethylene terephthalate) (PTET) with different composition was synthesized and the PTET sample with 60% weight fraction of poly(trimethylene) (PTET-60) was amorphous. The compatibility of PTET-60/cellose acetate (CA) blends and the pervaporation of their membranes for separation of benzene/cyclohexane mixtures were investigated. It was found that PTET-60 is compatible with CA when the weight fraction of PTET-60 in PTET-60/CA blends ( $W_{\text{PTET-60}}$ ) is lower than

0.35 and more than 0.5. Both the degree of swelling (DS) and the permeation flux ( $J$ ) of these blend membranes increased with increasing  $W_{\text{PTET-60}}$  from 0 to 0.35, and a maximum value of the separation factor ( $\alpha$ ) displayed at  $W_{\text{PTET-60}} = 0.25$ . © 2006 Wiley Periodicals, Inc. *J Appl Polym Sci* 102: 2832–2838, 2006

**Key words:** aromatic copolyester; blends; compatibility; pervaporation membrane; benzene/cyclohexane mixture

## INTRODUCTION

Pervaporation is being recognized as an energy-efficient alternative to other traditional liquid mixtures separation methods like distillation, especially in case of the separation of azeotropic mixtures, isomeric components, close boiling point systems.

Separation of benzene from cyclohexane is a very tough target in membrane separation process. Cyclohexane, a very important bulk chemical, is seldom found in crude oil and therefore mainly manufactured through benzene hydrogenation. But benzene/cyclohexane effluent mixtures are difficult to separate because both have very close boiling point and approximately equal molecular volumes. Many researchers have searched many polymer materials,<sup>1–30</sup> including new synthesized materials, modified membranes, blend membranes and facilitate transport membranes.

Aromatic polyesters of poly(trimethylene terephthalate) (PTT) and poly(ethylene terephthalate) (PET) are commonly used as fibers and engineering thermoplastics for their outstanding characteristics such as chemical resistance, resilience, and thermal stability.<sup>31,32</sup> Polyesters such as PET, polyethylene terephthalate/cyclohexane-dimethanol terephthalate (PETG), poly-

arylate, and crosslinked unsaturated polyesters have been used as membranes to separate aromatic/alicyclic mixtures like toluene/isooctane or toluene/*n*-octane by pervaporation, because they show preferential selectivity for aromatic hydrocarbons to alicyclic hydrocarbons.<sup>33–35</sup> But polyesters have not been reported to separate benzene/cyclohexane mixtures. We have tried to use PET and PTT membranes to separate benzene/cyclohexane mixtures. But both of them absorb little amount of benzene because they are semicrystalline polymers. Copolymerization with several amounts of a second glycol or diacid comonomer is an approach frequently used to attain low crystalline polyester, which guarantees large free volume necessary for diffusion. In this article, copolyester poly(trimethylene-*co*-ethylene terephthalate) (PTET) was synthesized from different molar ratios of 1,3-propylene glycol (PG) to ethylene glycol (EG). To modify the brittleness of PTET membranes, we prepared blend membranes of PTET-60 (the molar fraction of PET segments is 40%, i.e., the ratio of EG to PG is 40 : 60) and CA by solution blending. The compatibility of PTET-60/CA blends was studied. Then the swelling behavior and pervaporation performance of PTET-60/CA membranes for separating benzene/cyclohexane mixture were examined.

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Contract grant sponsor: NNSFC; contract grant number: 20376068.

Contract grant sponsor: Major State Basic Research Program of China; contract grant number: 9732003C8615700.

## EXPERIMENTAL

### Materials

1,3-Propylene glycol (PG), ethylene glycol (EG), tetrabutyl titanate and terephthalic acid were supplied by

Aldrich, USA. CA with 40.0 wt % acetylating content (intrinsic viscosity in dioxane at 30°C is 117.4) was purchased from Shanghai Chemical Station (Shanghai, China). The solvent 1,1,2,2-tetrachloroethane and phenol were of AR grade; benzene and cyclohexane were of CP grade. All the products were used directly without further purification.

### Preparation of PTET

Different molar ratios of PG to EG and equivalence of terephthalic acid were mechanically stirred and the mixture was kept at the temperature of 200°C catalyzed by tetrabutyl titanate for 3 h, then the mixture was heated up to 270°C and kept for 3.5 h under reduced pressure. Thus, polyesters of PTET with different molar ratios of PTT to PET could be prepared. The intrinsic viscosity of PTET was measured in the 1 : 1 (w/w) 1,1,2,2-tetrachloroethane/phenol solvent mixture at 25°C, the values are located at the range of 0.677–0.823 dL/g.

### WAXD of PTET

The specimens of PTET with a thickness of 300 μm for wide-angle X-ray diffraction (WAXD) measurements were prepared by melt-pressing. The melts were cooled rapidly to 130°C and crystallized at that temperature for 60 min and cooled to room temperature. WAXD measurements were carried out with a Rigaku D/max- $\alpha$  diffractometer using graphite-monochromatized Cu K $\alpha$  radiation ( $\lambda = 0.1542$  nm; 40 kV; 80 mA; 2K cps).

### Preparation of PTET-60/CA blend membranes

The casting solutions of CA and PTET-60/CA blends of different PTET-60 weight fraction in the blend ( $W_{\text{PTET-60}}$ ) with concentration of 10 g/100 mL were prepared by dissolving the polymers in the 2 : 3 (w/w) phenol/1,1,2,2-tetrachloroethane solvent mixture for a week at 45°C. The casting solutions were then poured onto clean glass plates. After drying at 160°C at vacuum for 24 h, the membranes were peeled off. The membranes obtained were about 20 μm in thickness for pervaporation experiment and 150 μm for DS experiments.

### Intrinsic viscosity measurement

Solutions of PTET-60, CA, and their blends in the 2 : 3 (w/w) phenol/1,1,2,2-tetrachloroethane solvent mixture were prepared at 50°C for 24 h, and then filtered through G2 sintered glass filters. Viscosity measurement was made with an Ubbelohde dilution viscometer at (30.00 ± 0.02)°C. The flux time was recorded with an accuracy of ±0.02 s. Extrapolation procedure from data obtained for five concentrations of solutions was

used to evaluate intrinsic viscosity,  $[\eta]$ .  $[\eta]$  was determined by plotting the  $\eta_{\text{sp}}/C$  against the solution concentration,  $C$ , for each blend composition according to the well known eq. (1):

$$\eta_{\text{sp}}/C = [\eta] + K_H[\eta]^2C \quad (1)$$

where  $K_H$  is the Huggins constant.

### Swelling measurements of PTET-60/CA blend membranes

The DS of the membrane at a given time is defined by the following equation:

$$DS = \frac{m_t - m_0}{m_0} \times 100\% \quad (2)$$

where  $m_0$  and  $m_t$  are the weights of dry and swollen membrane at the given time, respectively. The swelling experiments were carried out at a constant temperature of 40°C, and the membranes were weighted at room temperature by analyzing balance. The free liquid on the surface of the swollen membrane was removed carefully by using filter paper before weighting.

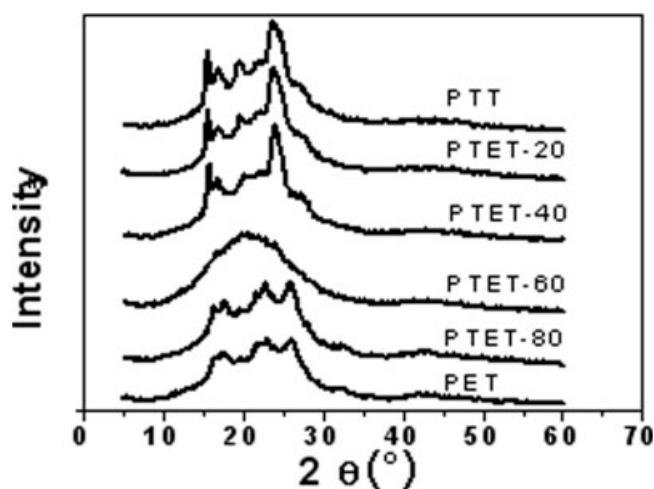
### Pervaporation measurements of PTET-60/CA blend membranes

The pervaporation experiments were conducted with a pervaporation apparatus as reported previously.<sup>36</sup> The permeation flux,  $J$ , and separation factor,  $\alpha$ , for all membranes were calculated according to the following equations:

$$J = \frac{\Delta g}{S \Delta t} \quad (3)$$

$$\alpha = \frac{y_{\text{ben}}/y_{\text{cyh}}}{x_{\text{ben}}/x_{\text{cyh}}} \quad (4)$$

where  $\Delta g$  is the permeation weight during the operation time  $\Delta t$ ,  $S$  is the membrane area,  $x$  and  $y$  are the weight fraction in the feed and permeation, respectively. The membrane area in contact with the feed was 18.1 cm<sup>2</sup>, the temperature of the cell was thermostatically controlled and the vacuum at down streamside was maintained at about 160 Pa by a vacuum pump. All pervaporation experiments were carried out in a continuous steady state and operated at constant temperature of 40°C for benzene/cyclohexane mixture. The pervaporation vapor was condensed by liquid nitrogen (N<sub>2</sub>). After running 2 h of the pervaporation apparatus, the composition of the



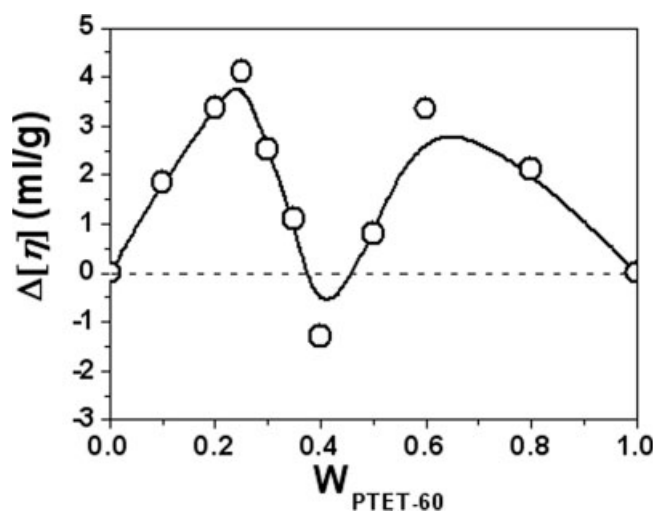
**Figure 1** WAXD patterns for PTET copolyesters with different composition.

permeation flux was analyzed by gas chromatograph equipped with a thermal conductivity detector.

## RESULTS AND DISCUSSION

### WAXD of PTET

Figure 1 shows WAXD patterns of copolyesters crystallized from melt. It is easily seen that only PTET-60 alone does not show crystalline peaks, which means that PTET-60 is amorphous copolyester, whereas other PTET membranes are semicrystalline polymers. This is because copolymerization can destroy ordered structures of both crystalline PET and crystalline PTT, and depress their crystallinity. The breakage will be serious when the two segments in the copolymer are nearly equal.



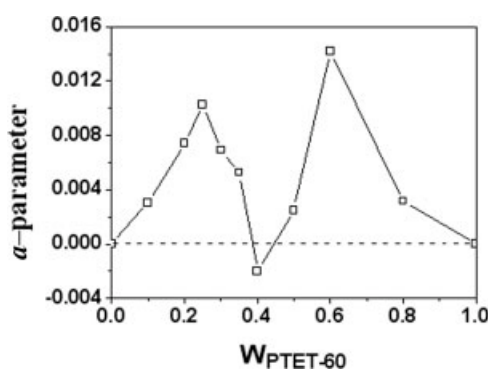
**Figure 2** The variation of  $\Delta[\eta]$  with  $W_{\text{PTET-60}}$  of PTET-60/CA blends at 40°C in the 2:3 (w/w) phenol/1,1,2,2-tetrachloroethane mixture.

**TABLE I**  
The Values of Intrinsic Viscosity  $[\eta]$  and  $b$  for PTET-60/CA Blends in the 2 : 3 (w/w) Phenol/1,1,2,2-Tetrachloroethane Mixture at 40°C

$W_{\text{PTET-60}}$	$[\eta]$ (mL/g)	$b$ ( $10^{-3}$ ) (mL/g) <sup>2</sup>
0	153.45	3.47
0.1	146.59	3.62
0.2	139.42	3.78
0.25	135.81	3.87
0.3	129.88	3.71
0.35	124.12	3.60
0.4	117.38	3.35
0.5	110.78	3.53
0.6	104.62	3.83
0.8	86	3.63
1.0	66.5	3.42

### Compatibility of PTET-60/CA blends

Many experiments and theoretical methods have been used to investigate polymer compatibility.<sup>37–39</sup> Viscometry is an attractive method because of its simplicity and reliability.<sup>39–42</sup> Theoretically, attractive interaction between the chains of two different polymers in solution may cause expanding of the polymer coils and result in increasing of the intrinsic viscosity because the intrinsic viscosity,  $[\eta]$ , represents the effective hydrodynamic volume of a polymer molecule in solution,<sup>43</sup> i.e., it is larger than that calculated according to the additivity law, and *vice versa* for the case of repulsive interaction. Figure 2 shows the  $\Delta[\eta]$  values ( $\Delta[\eta] = [\eta]_{\text{exp}} - [\eta]_{\text{calc}}$ , where  $[\eta]_{\text{calc}}$  was calculated according to the additivity law) of PTET-60/CA blends with various  $W_{\text{PTET-60}}$  in the 2:3 (w/w) phenol/1,1,2,2-tetrachloroethane solvent mixture at 40°C. It can be observed that the  $\Delta[\eta]$  values of the most PTET-60/CA blends are not equal to zero, i.e., deviating from the additivity law. Positive deviation is demonstrated at  $W_{\text{PTET-60}} \leq 0.35$  and  $W_{\text{PTET-60}} \geq 0.5$ , which reflects the expanding of both PTET-60 and CA coil dimensions due to strong intermolecular interaction or compatibility between them, and the negative deviation is dis-



**Figure 3** The  $a$ -parameter as a function of  $W_{\text{PTET-60}}$  for PTET-60/CA blends at 40°C in the 2:3 (w/w) phenol/1,1,2,2-tetrachloroethane mixture.

**TABLE II**  
**Comparison of the Experimental Equilibrium Degree of Swelling ( $DS_E$ ) and the Calculated Additive Equilibrium Degree of Swelling ( $DS_C$ )**

PTET-60 content	0	0.1	0.2	0.25	0.35	0.4	1.0
$DS_E$	12	13.5	15.4	15.6	18.2	24.6	45
$DS_C$	12	15.3	18.6	20.6	23.5	25.2	45
$\Delta DS$	0	1.8	3.2	5.0	5.3	0.6	0

played in the region of  $W_{PTET-60}$  from 0.35 to 0.5, which means the shrinkage of both PTET-60 and CA coil dimensions occurred due to the incompatibility between them.

Crispim et al.<sup>39</sup> used “ $a$ -parameter,” calculated from the eq. (5), to evaluate the interaction and miscibility. If  $a$ -parameter  $\geq 0$ , attractive forces occur between poly-

mers, showing miscibility. If  $a$ -parameter  $< 0$ , repulsive forces prevail and immiscibility is expected.  $K_{BLEND}$  is the Huggins constant of the blend and  $K_1$  is calculated from eq. (6). The values of  $[\eta]$  and  $b$  (in this case,  $b = [d(\eta_{sp}/C)]/dC$  of the eq. (1) or more properly,  $b = K_H[\eta]^2$ ) for pure polymers and their blends, obtained in the 2:3 (w/w) phenol/1,1,2,2-tetrachloroethane solvent mixture at 40°C, are shown in Table I. In Figure 3, the curve of the  $a$ -parameter against  $W_{PTET-60}$  was presented. Again when  $W_{PTET-60} = 0.35-0.5$ , the  $a$ -parameter was low or negative. This suggested that the two polymers of PTET-60 and CA showed immiscible behavior, which is similar to the result presented by the method of  $\Delta[\eta]$  above.

$$a - \text{parameter} = K_{BLEND} - K_1 \quad (5)$$

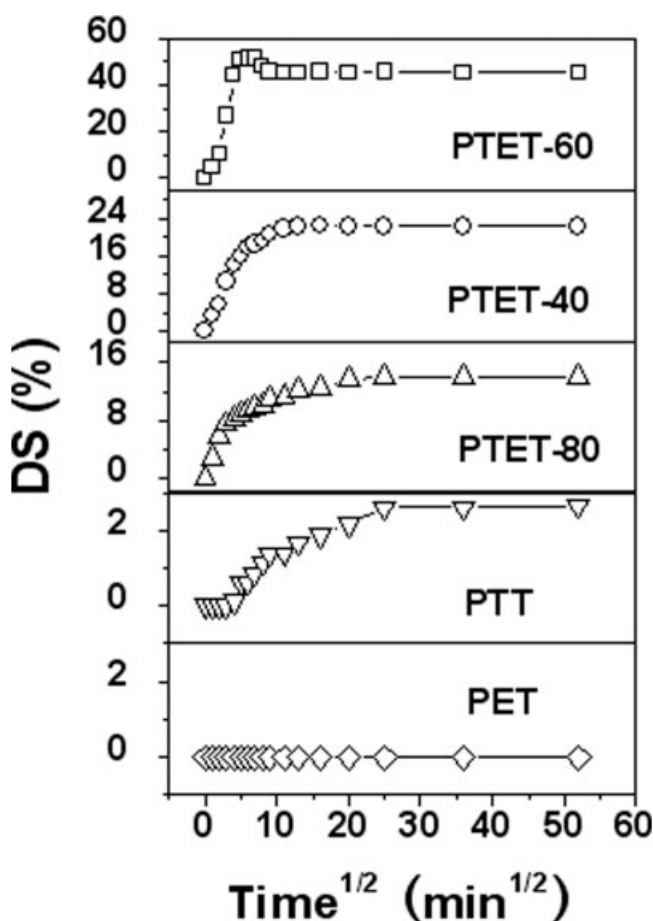
$$K_1 = \frac{b_{PTET-60}W_{PTET-60}^2 + b_{CA}W_{CA}^2 + 2\sqrt{b_{PTET-60}b_{CA}}W_{PTET-60}W_{CA}}{([\eta]_{PTET-60}W_{PTET-60} + [\eta]_{CA}W_{CA})^2} \quad (6)$$

This can also be approved by the swelling experiment as seen in Figure 5 and the difference between the experimental equilibrium degree of swelling ( $DS_E$ ) and the calculated degree of swelling ( $DS_C$ ) in Table II, which is similar to the additivity law of  $[\eta]_{calc}$ . We can see that  $\Delta DS$  ( $DS_C - DS_E$ ) nearly equals to zero when  $W_{PTET-60} = 0.4$  but the values of  $DS_E$  is apparently not equal to  $DS_C$  when  $W_{PTET-60}$  is from 0.1 to 0.35. Because of the incompatibility of CA and PTET-60 with  $W_{PTET-60} = 0.4$ , the absorbance of benzene in CA and PTET-60 were not influenced by the interaction of each other. When the interaction of CA and PTET-60 became larger, physical crosslink will occur in the blend membranes and the experimental equilibrium swelling  $DS_E$  will be much less than  $DS_C$ .

#### Swelling of PTET and PTET-60/CA blend membranes

Swelling of PTET membranes

Figure 4 shows the variation of the DS of PTET membranes with the swelling time in benzene at 40°C. From Figure 4, it can be seen that the DS of PTET-60 membrane in benzene is the highest compared with other PTET membranes and homopolymer membranes of PET and PTT. This can be ascribed to that PTET-60 membrane is amorphous polymer, which guarantees large free volume necessary for diffusion, whereas other PTET membranes are semicrystalline polymers and it is difficult for solvent to penetrate in crystalline phase. So, we prepared blend membranes of PTET-60



**Figure 4** DS of PTET membranes of different composition with swelling time in benzene at 40°C.

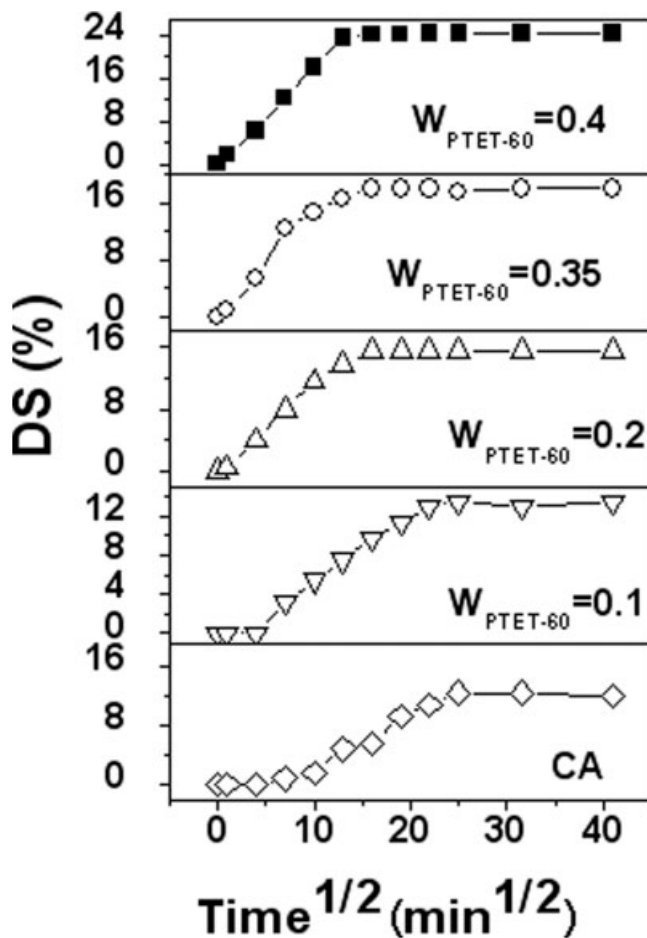


Figure 5 DS of PTET-60/CA membranes of different  $W_{\text{PTET-60}}$  with swelling time in benzene at 40°C.

and CA to separate benzene from benzene/cyclohexane mixture.

#### Swelling of PTET-60/CA membranes

Figures 5–6 give the variation of the DS of PTET-60/CA blend membranes of different  $W_{\text{PTET-60}}$  in benzene and benzene/cyclohexane mixtures with the swelling time at 40°C. As seen in Figure 5, the equilibrium DS of CA and its blend membranes of PTET-60/CA in benzene increased with the increasing of  $W_{\text{PTET-60}}$ , and the equilibrium swelling time decreased from 625 min for CA membrane to 225 min for blend membrane with  $W_{\text{PTET-60}} = 0.4$ . So the addition of PTET-60 can enhance the absorbance of benzene.

Figure 6 shows that the equilibrium DS values of all blend membranes in benzene/cyclohexane (10/90) mixture were lower than that in benzene, and the pure CA membrane almost had no swelling in this case. Therefore, it indicates that CA is good against cyclohexane, and that the addition of PTET-60 to CA could raise the equilibrium DS of PTET-60/CA blend membrane in benzene/cyclohexane mixture. Figure 7 shows the DS of PTET-60/CA membrane ( $W_{\text{PTET-60}} = 0.2$ )

with swelling time in different benzene/cyclohexane mixtures at 40°C. From Figure 7, the equilibrium DS values of the blend membrane increased with the increasing of concentration of benzene in benzene/cyclohexane mixture. That is to say, the affinity of the blend membranes for benzene is greater than that for cyclohexane and this difference can be ascribed to the stronger interaction between aromatic polyester and benzene than that between aromatic polyester and cyclohexane, due to larger density of electron cloud being in benzene than in cyclohexane. Therefore, these blend membranes are expected to preferentially extract benzene from benzene/cyclohexane mixtures.

#### Pervaporation of PTET-60/CA blend membranes

Figure 8 shows the variation of permeation flux,  $J$ , and separation factor,  $\alpha$ , of PTET-60/CA blend membranes with different  $W_{\text{PTET-60}}$  for benzene/cyclohexane (10/90) mixture. It is very similar to their swelling results, the value of  $J$  increases with the increasing of  $W_{\text{PTET-60}}$  in blends, whereas a maximum value of  $\alpha$  exists at  $W_{\text{PTET-60}} = 0.25$ . It leads us to consider that the separation mechanism of PTET-60/CA blend membranes for

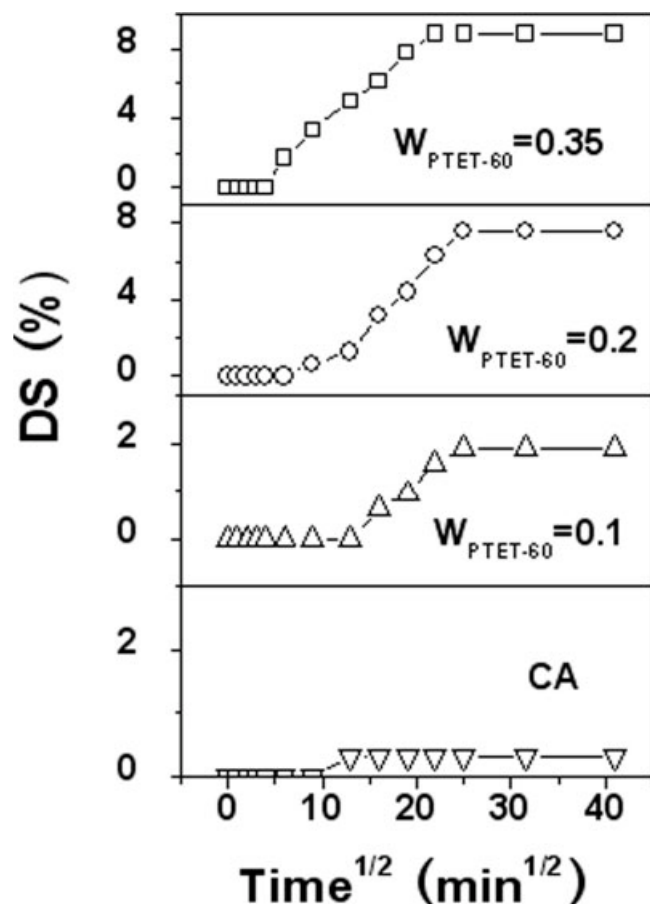


Figure 6 DS of PTET-60/CA membranes of different  $W_{\text{PTET-60}}$  with swelling time in benzene/cyclohexane (10/90 in weight) mixture at 40°C.

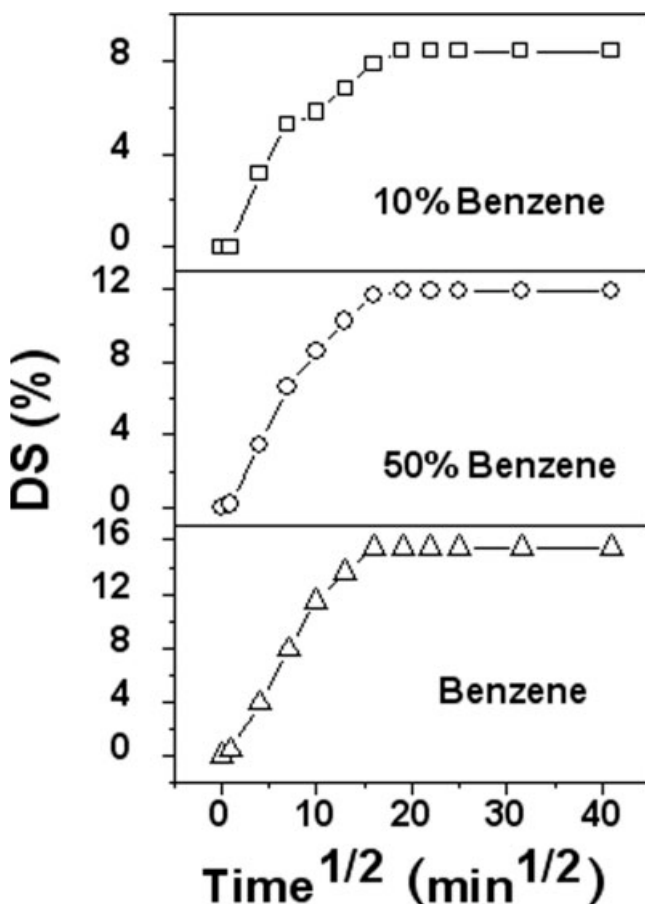


the benzene and cyclohexane mixtures is determined by both solution and diffusion. When  $W_{\text{PTET-60}}$  is smaller than 0.25, the separation process is probably governed mainly by solution. Membranes with more PTET-60 can absorb more amount of benzene, so  $\alpha$  and  $J$  increase with the increasing of  $W_{\text{PTET-60}}$  in the blends, and *vice versa* when  $W_{\text{PTET-60}}$  is larger than 0.25. But in the case of higher PTET-60 content, the separation process is governed mainly by diffusion and the blend membranes are so swollen that benzene and cyclohexane will diffuse more quickly simultaneously, so  $\alpha$  falls down, whereas  $J$  increases continually with the increasing of  $W_{\text{PTET-60}}$ .

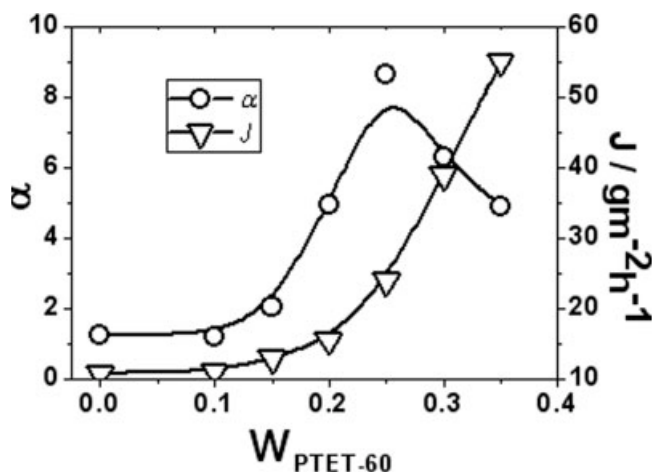
### CONCLUSIONS

The following conclusions can be drawn from the present study:

1. PTET-60 is amorphous copolyester and the DS of PTET-60 membrane in benzene is the highest compared with other PTET membranes and homopolymer membranes of PET and PTT.



**Figure 7** DS of PTET-60/CA membrane ( $W_{\text{PTET-60}} = 0.2$ ) with swelling time in different benzene/cyclohexane mixtures at 40°C.



**Figure 8** The variation of  $J$  and  $\alpha$  of the PTET-60/CA membranes with  $W_{\text{PTET-60}}$  for benzene/cyclohexane (10/90 in weight) mixture at 40°C.

2. PTET-60/CA blends are compatible except for  $W_{\text{PTET-60}}$  from 0.35 to 0.5.
3. The variation of the DS of PTET-60/CA blend membranes of different  $W_{\text{PTET-60}}$  ( $\leq 0.35$ ) in benzene/cyclohexane mixtures with the swelling time at 40°C suggests that these membranes are expected to preferentially extract benzene from benzene/cyclohexane mixtures.
4. The separation process was probably governed mainly by the solution step when  $W_{\text{PTET-60}}$  was smaller than 0.25 and by the diffusion step when  $W_{\text{PTET-60}}$  was larger than 0.25. So a maximum value of  $\alpha$  existed at  $W_{\text{PTET-60}} = 0.25$  and the value of  $J$  increased with the increasing of  $W_{\text{PTET-60}}$ .

### References

1. Cabasso, J.; Jagur-Grodzinski, J.; Vofsi, D. *J Appl Polym Sci* 1974, 18, 2137.
2. Cabasso, J. *Ind Eng Chem Prod Res Dev* 1983, 22, 313.
3. Acharya, H. R.; Stern, S. A.; Lin, Z. Z.; Cabasso, J. *J Membr Sci* 1988, 37, 205.
4. Park, C. K.; Oh, B. K.; Choi, M. J.; Lee, Y. M. *Polym Bull* 1994, 33, 591.
5. Sun, F.; Ruckenstein, E. *J Membr Sci* 1995, 99, 273.
6. Yoshikawa, M.; Kitao, T. *Eur Polym J* 1997, 33, 25.
7. Yoshikawa, M.; Takeuchi, S.; Kitao, T. *Angew Makromol Chem* 1997, 245, 193.
8. Inui, K.; Miyata, T.; Uragami, T. *J Polym Sci Part B: Polym Phys* 1997, 35, 699.
9. Yamasaki, A.; Shinbo, T.; Mizoguchi, K. *J Appl Polym Sci* 1997, 64, 1061.
10. Inui, K.; Okumura, H.; Miyata, T.; Uragami, T. *J Membr Sci* 1997, 132, 193.
11. Inui, K.; Okumura, H.; Miyata, T.; Uragami, T. *J Membr Sci* 1998, 143, 93.
12. Uragami, T.; Tsukamoto, K.; Miyata, T.; Inui, K. *Macromol Chem Phys* 1998, 199, 49.
13. Inui, K.; Tsukamoto, K.; Miyata, T.; Uragami, T. *J Membr Sci* 1998, 138, 67.

14. Kusakabe, K.; Yoneshige, S.; Morooka, S. *J Membr Sci* 1998, 14, 929.
15. Yoshikawa, M.; Tsubouchi, K. *J Membr Sci* 1999, 158, 269.
16. Wang, H.; Tanaka, K.; Kita, H.; Okamoto, K. *J Membr Sci* 1999, 154, 221.
17. Uragami, T.; Tsukamoto, K.; Miyata, T.; Heinze, T. *Macromol Chem Phys* 1999, 200, 1985.
18. Uragami, T.; Tsukamoto, K.; Miyata, T.; Heinze, T. *Cellulose* 1999, 6, 221.
19. Yoshikawa, M.; Tsubouchi, K. *Sep Sci Technol* 2000, 35, 1683.
20. Yildirim, A. E.; Hilmioglu, N. D.; Tulbentci, S. *Chem Eng Technol* 2001, 24, 275.
21. Wang, Y. C.; Li, C. L.; Huang, J.; Lin, C.; Lee, K. R.; Liaw, D. J.; Lai, J. Y. *J Membr Sci* 2001, 185, 193.
22. Ren, J.; Staudtbickel, C.; Lichtenthaler, R. N. *Sep Purif Technol* 2001, 22/23, 31.
23. Kao, S. T.; Wang, F. J.; Lue, S. J. *Desalination* 2002, 149, 35.
24. Wang, Y.; Hirakawa, S.; Wang, H.; Tanaka, K.; Kita, H.; Okamoto, K. *J Membr Sci* 2002, 199, 13.
25. Lue, S. J.; Peng, S. H. *J Membr Sci* 2003, 222, 203.
26. An, Q. F.; Qian, J. W.; Sun, H. B.; Wang, L. N.; Zhang, L.; Chen, H. L. *J Membr Sci* 2003, 222, 113.
27. Pandey, L. K.; Saxena, C.; Dubey, V. *J Membr Sci* 2003, 227, 173.
28. Uragami, T.; Tsukamoto, K.; Miyata, T. *Biomacromolecules* 2004, 5, 2116.
29. Yanagishita, H.; Arai, J.; Sandoh, T.; Negishi, H.; Kitamoto, D.; Ikegami, T.; Haraya, K.; Idemoto, Y.; Koura, N. *J Membr Sci* 2004, 232, 93.
30. Lue, S. J.; Wang, F. J.; Hsiaw, S.-Y. *J Membr Sci* 2004, 240, 149.
31. Jajewats, R.; Ward, I. M.; Wilding, M. A.; Hal, I. H.; Desborough, I. J.; Pass, M. G. *J Polym Sci Polym Phys Ed* 1975, 13, 799.
32. Ward, I. M.; Wilding, M. A.; Brody, H. *J Polym Sci Polym Phys Ed* 1976, 14, 263.
33. Sartori, G. (to Exxon Research and Engineering Company). U.S. Pat. 4,976,868 (1990).
34. Sartori, G. (to Exxon Research and Engineering Company) U.S. Pat. 5,012,036 (1991).
35. Sartori, G. (to Exxon Research and Engineering Company) U.S. Pat. 5,138,023 (1991).
36. Zhu, C. L.; Liu, M. E.; Xu, W.; Ji, W. C. *Desalination* 1989, 71, 1.
37. Beaucauge, G.; Stein, R. S.; Koningsveld, R. *Macromolecules* 1993, 26, 1603.
38. Campos, A.; Gomez, C. M.; Garcia, R.; Figueruelu, J. E.; Soria, V. *Polymer* 1996, 37, 3361.
39. Crispim, E. G.; Rubira, A. F.; Muniz, E. C. *Polymer* 1999, 40, 5129.
40. Kulshreshtha, A. K.; Singh, B. P.; Sharma, Y. N. *Eur Polym J* 1988, 24, 29.
41. Qian, J. W.; Qi, R.; Ding, X. Z.; Yang, S. L. *Chin J Polym Sci* 1997, 15, 35.
42. Yang, H. Y.; Zhu, P. P.; Wang, S. Q.; Guo, Q. P. *Eur Polym J* 1998, 34, 463.
43. Bohclanechy, M.; Kovar, J. *Viscosity of Polymer Solutions*; Elsevier: Amsterdam, 1982.