

## CHAPTER-VI

Excess Properties for Binary Mixtures of Propylene Carbonate with 2-Methoxyethanol, 1,2-Dimethoxyethane, Tetrahydrofuran and Methanol at 25, 35 and 45°C

Recent years have witnessed considerable interest<sup>1-5</sup> in the thermodynamic excess properties of binary liquid mixtures. These properties have been very helpful to obtain information on the intermolecular interactions and geometrical effects in these systems<sup>4</sup>. The potentiality of propylene carbonate (PC) as solvent for a variety of applications particularly in high energy batteries has been mentioned earlier (Chapters I and III). Solvents like 2-methoxyethanol (ME; methyl cellosolve), 1,2-dimethoxyethane (DME), tetrahydrofuran (THF) and methanol too find a wide range of applications of technological importance namely, as solvents and solubilizing agents in organic syntheses<sup>6</sup>, reaction kinetics and electrochemical studies<sup>6,7</sup>. However, barring a few<sup>8,9</sup>, the binary liquid systems of propylene carbonate with other non-aqueous solvents have not been studied.

In the present study, densities and viscosities have been measured for four binary liquid mixtures of PC + ME, PC + DME, PC + THF and PC + Methanol at different temperatures. The excess molar volumes ( $V^E$ ), excess viscosities ( $\eta^E$ ) and excess molar free energies of activation of flow ( $G^{*E}$ ) have been determined for these binary systems over the entire range of their compositions at 25, 35 and 45°C. Attempts have been made to explain the behaviour of

the liquid mixtures on the basis of sign and magnitude of these properties. McAllister equation<sup>10</sup> has also been used to calculate the interaction parameters ( $\nu_{ij}$ ) from the kinematic viscosity data.

### Experimental

Propylene carbonate (E. Merck, Germany; > 99% pure) was purified as described earlier (Chapter - II).

2-methoxyethanol [ME] (G.R. E. Merck) was distilled in an all-glass distillation apparatus before use. The physical properties at 25°C of purified ME were : density 0.96002 gcm<sup>-3</sup> and viscosity 1.5414 cp. The viscosity and density values are in reasonable agreement with the reported literature values<sup>11</sup> which are 1.60 cp and 0.96024 gcm<sup>-3</sup> respectively.

1,2-dimethoxyethane [DME] (Fluka, purum) was shaken well with FeSO<sub>4</sub> (AR, BDH) for 1-2 h, decanted and distilled. The distillate was refluxed for 12 h and redistilled over metallic sodium<sup>12</sup>. The boiling point (84.5°C) and density (0.86132 gcm<sup>-3</sup> at 25°C) compared fairly well with the corresponding literature values<sup>13</sup> which are 85°C and 0.86120 gcm<sup>-3</sup> respectively.

Tetrahydrofuran [THF] (E. Merck, India) was kept over KOH, refluxed for 24 h and distilled over  $\text{LiAlH}_4$ <sup>14</sup>. The density ( $0.88072 \text{ gcm}^{-3}$ ) and viscosity ( $0.4630 \text{ cP}$ ) at  $25^\circ\text{C}$  of the purified sample are in good conformity with the literature values<sup>15</sup> ( $\rho = 0.8811 \text{ gcm}^{-3}, \eta = 0.46 \text{ cP}$ ).

Methanol (E. Merck, India, Uvasol grade) was dried over 3A molecular sieves and distilled fractionally. Middle fraction was taken and further distilled<sup>16</sup>. Physical properties of purified methanol at  $25^\circ\text{C}$  ( $\rho = 0.78663 \text{ gcm}^{-3}$  and  $\eta = 0.5437 \text{ cP}$ ) agreed well with the literature values<sup>11</sup> which are :  $\rho = 0.78664 \text{ gcm}^{-3}$  and  $\eta = 0.5445 \text{ cP}$ .

Freshly distilled solvents were always used for the preparation of solvent mixtures. All solutions were prepared by weight and special air-tight bottles were used for mixture preparation. When once the mixtures were made, all measurements were done on the same day to avoid vapour loss, moisture absorption, contamination and oxidation of the mixtures.

Densities were measured with an Ostwald-Sprengel type pycnometer having a bulb volume of  $25 \text{ cm}^3$  and an internal diameter of capillary of about 1 mm. The pycnometer was calibrated at  $25, 35$  and  $45^\circ\text{C}$  with doubly distilled deionized water. The temperature control had an accuracy of  $\pm 0.01^\circ\text{C}$ . The precision of density measurement was  $\pm 3 \times 10^{-5} \text{ gcm}^{-3}$ .

Kinematic viscosities were measured at the desired temperature (accuracy  $\pm 0.01^\circ\text{C}$ ) by using a suspended Ubbelohde-type viscometer. The precision of viscosity measurements was  $\pm 0.05\%$ . The kinematic viscosities were converted into the absolute viscosities by multiplying the former with density.

In all the cases, the experiments were performed in three replicates for each composition and at each temperature and the results were averaged.

### Results and Calculations

The experimental results of density and viscosity measurements at various mole fractions are reported in Table 1.

The kinematic viscosity data have been analyzed by means of McAllister equation<sup>10</sup>:

$$\ln \nu = x_1^3 \ln \nu_1 M_1 + x_2^3 \ln \nu_2 M_2 - \ln(x_1 M_1 + x_2 M_2) + 3x_1^2 x_2 \ln \nu_{12} \left( \frac{2M_1 + M_2}{3} \right) + 3x_2^2 x_1 \ln \nu_{21} \left( \frac{2M_2 + M_1}{3} \right) \quad (1)$$

The interaction parameters  $\nu_{ij}$  along with the kinematic viscosity data at different temperatures are also reported in Table 1.

The excess functions have been calculated by the following equations,

$$V^E = (x_1 M_1 + x_2 M_2) / \rho - x_1 M_1 / \rho_1 - x_2 M_2 / \rho_2 \quad (2)$$

$$\eta^E = \eta - x_1 \eta_1 - x_2 \eta_2 \quad (3)$$

$$G^{*E} = RT [\ln \eta V - (x_1 \ln \eta_1 V_1 + x_2 \ln \eta_2 V_2)] \quad (4)$$

where,  $x_1$  and  $x_2$  are the mole fractions of components 1 and 2 respectively;  $V$ ,  $V_1$  and  $V_2$  are the molar volumes and  $\eta$ ,  $\eta_1$  and  $\eta_2$  are the measured co-efficients of viscosity of the mixture, component 1 and PC respectively. The subscript 1 refers either to ME or DME or THF or  $\text{CH}_3\text{OH}$  in the concerned binary mixture. The molar volume  $V$  is defined by the relationship

$$V = (x_1 M_1 + x_2 M_2) / \rho \quad (5)$$

where,  $M_1$  and  $M_2$  are the molecular weights of component 1 and 2 respectively and  $\rho$  is the density of the mixture. The excess properties of the binary mixtures at different temperatures have been recorded in Table 2.

Graphical representations of  $V^E$  and  $\eta^E$  as functions of the mole fraction of pure component in the binary mixtures are given in

the figures 1A to 1D and 2A to 2D respectively.

The excess functions have been fitted to Redlich-Kister equation<sup>17</sup>,

$$X^E = x_1 x_2 \sum_{j=0}^n A_j (x_1 - x_2)^j \quad (6)$$

where  $X^E$  is  $V^E$  or  $\eta^E$  and  $A_0, A_1, A_2, \dots$  etc. are the adjustable parameters. These parameters were evaluated by the method of least squares. The values of these parameters along with the standard deviations,  $\sigma(X^E)$  of  $X^E$  as defined by

$$\sigma(X^E) = \left[ \frac{\sum (X_{\text{exptl}}^E - X_{\text{calcd}}^E)^2}{N - M} \right]^{0.5} \quad (7)$$

are recorded in Table 3. In equation (7),  $N$  is the total number of experimental points and  $M$  is the number of adjustable parameters. The results obtained from the correlation equation (eq. 6) are quite satisfactory for majority of the systems investigated with the possible exception of  $V^E$  values for PG + ME binary mixture. Owing to the unsymmetrical nature of the variation of  $V^E$  with respect to mole fraction in this mixture, higher  $\sigma(V^E)$ s are obtained.

### Discussion

Kinematic viscosity data for the binary solvent mixtures were correlated with McAllister equation<sup>10</sup> of three body interactions. In all the studied binary mixtures viz. PC + ME, PC + DME, PC + THF and PC + CH<sub>3</sub>OH, the values of  $\nu_{ij}$  are found to change with temperature. From Table 1, it is observed that in PC + ME mixtures, values of  $\nu_{12}$  are slightly greater than the corresponding  $\nu_{21}$  values at each temperature. In PC + ME binary system,  $\nu_{12}$  and  $\nu_{21}$  refer to the interactions of types ME-ME-PC and PC-PC-ME respectively. However, for the other three binary solvent systems, the values of  $\nu_{12}$  are less than the corresponding  $\nu_{21}$  values. It indicates that the interaction parameters resulting from the interactions of PC-PC-DME, PC-PC-THF and PC-PC-CH<sub>3</sub>OH types are greater than the parameters originating from the corresponding DME-DME-PC, THF-THF-PC and CH<sub>3</sub>OH - CH<sub>3</sub>OH - PC interactions in PC + DME, PC + THF and PC + CH<sub>3</sub>OH solvent mixtures respectively. Further, the values of  $\nu_{ij}$  and  $\nu_{ji}$  decrease with increase of temperature in general, for all the systems. Similar results have also been observed in other non-aqueous-non-aqueous solvent mixtures.<sup>18</sup>

It is well known<sup>19</sup> that the sign and magnitude of  $V^E$  give a good estimate of the strength of unlike interactions in the binary mixtures. Large positive  $V^E$  values are taken as indicative of weak intermolecular interactions, whereas, large negative values of  $V^E$  are attributed to different sizes of molecules or to the dipole-



dipole interactions between them<sup>19</sup>.

From Fig. 1A, it is observed that in PC + ME solvent mixtures, the excess molar volume varies with the mole fraction of pure component in an unsymmetrical fashion. IR studies<sup>20,21</sup> on 2-methoxyethanol indicate that the predominant configuration of ME molecule is the gauche form and it favours the intramolecular hydrogen bonding in ME. The unsymmetrical nature of  $V^E$  vs mole fraction curves of PC + ME mixtures, appears to be due to the steric factors arising from a change in the proportion of the different geometric forms of ME molecules with change in its mole fraction. Similar unsymmetrical variation of  $V^E$  has been reported for THF + p-dioxane binary system in the literature<sup>22</sup>.

From Fig. 1B, we see that the  $V^E$  values of PC + DME binary mixtures are negative over the entire composition range and become more negative as the temperature rises. This can be explained in terms of like and unlike intermolecular interactions. Studies on the dielectric and spectroscopic properties of liquid propylene carbonate<sup>23</sup> indicate that it behaves as a normal polar liquid with strong dipole-dipole interactions with little or no specific interaction present. It appears that the intermolecular interactions between PC and DME molecules in the mixture predominate over the intermolecular interactions between like molecules. Another effect which may result for negative values of  $V^E$  in binary solven

mixtures having components of different sizes is the accommodation of one component into the structure of the other. By considering the molar volumes of PC and DME (85.16 and 104.63 cm<sup>3</sup>mol<sup>-1</sup> respectively), the interstitial accommodation of one component into the other seems not to be easy in this mixture. Thus, it appears that the dipolar forces between the different kinds of molecules cause the decrease in volume of this mixture. This model is also in accordance with Prigogine theory<sup>19</sup>, which attributes the negative  $V^E$  values to different sizes of molecules or to the dipole-dipole interactions between them.

In PC + THF solvent mixtures, the excess molar volumes at different temperatures are negative over the whole range of compositions (Fig.1C). The magnitude of  $V^E$  increases with increase in temperature. The minima of  $V^E$  appear at a mole fraction of 0.5. Several effects may contribute to the  $V^E$  values and out of these, three may be considered as being important : (a) break-up of intermolecular dipolar interactions in PC, (b) dipole-dipole interactions between unlike molecules and (c) interstitial accommodation of one component into the other. It appears that on mixing THF with PC, the dipole-dipole interactions between PC molecules become weaker and the dipolar interactions between unlike molecules predominate, as a result of which a sharp decrease in volume occurs. Again, since the molar volumes of PC and THF (85.16 and 81.88 cm<sup>3</sup>mol<sup>-1</sup> respectively) are not far apart from each other, the third

effect i.e. the possibility of accommodation of one molecule into the structure of the other may be neglected for this system.

In PC + CH<sub>3</sub>OH binary system, the excess molar volumes are negative at each temperature. With rise in temperature, the values of  $V^E$  decrease further and the minima of  $V^E$  appear in the vicinity of 0.7 mole fraction of CH<sub>3</sub>OH, as shown in Fig. 1D. In the present system, considerable amount of positive contributions to  $V^E$  values are expected from the break-up of interactions between like molecules viz. the disruption of intermolecular hydrogen bonds in methanol and loosening of the dipolar interactions between PC molecules. Results indicate that the negative contributions predominate in this mixture. The important source for negative contributions is the interactions between unlike molecules. However since methanol (with a donor number 19.1) is a stronger base and thus probably a stronger hydrogen bond acceptor than PC (donor number 15.1) and PC has no such desired hydrogen atom, it appears that the hydrogen bond formation between these two unlike molecules is not favoured in this system. Another possibility of specific interaction lies with the bonding through hydroxyl group of CH<sub>3</sub>OH. But there is a lack of well-developed centre of positive charge<sup>24</sup> in PC and thus the hydrogen bonding between CH<sub>3</sub>OH and PC even if occurs, appears to be weak. Considering these factors, we think that the contributions due to PC-CH<sub>3</sub>OH heteroassociation alone are not sufficient enough to override the positive contributions. Simple dipole-dipole interactions between PC and CH<sub>3</sub>OH molecules may also

contribute to the negative values of  $V^E$ . However, the most important source for negative contributions appears to be the differences in size and shape of the pure components in the mixture. The molar volumes of PC and  $\text{CH}_3\text{OH}$  at  $25^\circ\text{C}$  are 85.16 and  $40.73 \text{ cm}^3 \text{ mol}^{-1}$  respectively and it might allow the components to fit into each other's structure causing the reduction in volume of the mixture.

Values of the quantity,  $\eta^E$ , which refer to the deviations from a rectilinear dependence<sup>25</sup> of the viscosity of the mixture on mole fraction, can be discussed from the view point of intermolecular interactions<sup>9,25</sup>. For systems, where dispersion and dipolar forces are operating, the values of  $\eta^E$  are found to be negative, whereas, the existence of charge-transfer and hydrogen bond interactions leading to the formation of complex species between the two components of the binary system make the values of  $\eta^E$  positive. For systems where, all types of intermolecular forces are operating, the values of  $\eta^E$  will be due to the net effect of individual contributions from all types of interactions.

The system, PC + ME shows negative deviations of  $\eta^E$  from ideality over the entire mole fraction range and over the whole range of temperatures (Fig. 2A). The minima of  $\eta^E$  values correspond to 0.4 mole fraction of ME. As the temperature increases, the magnitude of excess viscosity decreases, showing a tendency towards ideal behaviour. The negative deviation of  $\eta^E$  indicates that mixing of ME with PC tends to loosen the strong dipolar forces

between PC molecules and intermolecular dipolar or dispersion interactions between unlike molecules (PC and ME) predominate in the mixture.

In PC + DME binary system, the negative contributions to  $\eta^E$ , arising from the intermolecular interactions are predominant. Fig. 2B further shows that the minima of  $\eta^E$  lie at 0.4 mole fraction of DME at each temperature. Results indicate that the system lacks heteroassociation or specific interaction between the two different kinds of molecules and the dispersion and dipolar interactions between PC and DME molecules are primarily responsible for negative values of excess viscosity.

The third system i.e. PC + THF, also shows negative excess viscosity values. Like PC + ME and PC + DME binary systems, PC + THF mixtures also have the same trend of  $\eta^E$ , as is evident from Figs. 2A-2C. Negative values of  $\eta^E$  appear to be due to the predominant dispersion and dipolar interactions between the unlike molecules.

Binary mixtures of PC and  $\text{CH}_3\text{OH}$  also show negative values of  $\eta^E$  over the entire composition range and over the whole range of temperatures. The minima of  $\eta^E$  correspond to 0.4 mole fraction of  $\text{CH}_3\text{OH}$  at each temperature (Fig. 2D). As the temperature increases, the magnitude of  $\eta^E$  decreases and approaches towards the ideal behaviour. According to Fort and Moore<sup>25</sup>, negative values of  $\eta^E$

occur for systems of different molecular sizes and the dispersion forces are primarily responsible for the interactions. The molar volume of PC ( $85.16 \text{ cm}^3 \text{ mol}^{-1}$  at  $25^\circ\text{C}$ ) is much greater than that of  $\text{CH}_3\text{OH}$  ( $40.73 \text{ cm}^3 \text{ mol}^{-1}$  at  $25^\circ\text{C}$ ) and it might allow one component to fit into the structure of the other, resulting in a decrease in viscosity of the binary system. However, intermolecular association between PC and  $\text{CH}_3\text{OH}$  molecules, if present, might have some positive contributions to the  $\eta^E$  values. The other possible effect i.e. the intermolecular dipolar interactions between unlike molecules (PC and  $\text{CH}_3\text{OH}$ ) may also have some negative contribution to  $\eta^E$ . However in the present system, the interstitial accommodation of one component into the structure of the other appears to be predominant over the other effects.

Another thermodynamic function, which has also been investigated, is the excess molar free energy of activation of flow ( $G^{*E}$ ). Values of  $G^{*E}$  have been calculated from eqn. (4), which results from the Eyring equation<sup>9</sup> of viscosity flow modified for a binary mixture. According to Reed and Taylor<sup>26</sup> and Meyers et al<sup>27</sup>, the  $G^{*E}$  parameter may be considered as a reliable measure to detect the presence of interactions between molecules. Positive values of  $G^{*E}$  can be seen in binary systems where specific interactions between unlike molecules take place. The binary systems PC + ME, PC + DME and PC + THF have distinct negative values of  $G^{*E}$  at all compositions over the entire range of temperatures (Table 2).

The negative signs of  $G^{*E}$  indicate that the specific interactions between unlike molecules even if present, are not predominant in these systems. In fact, mixtures where a strong specific interaction between unlike molecules is predominant, are characterized by distinct maxima in the viscosity vs composition curves, by positive excess viscosities and positive values of  $G^{*E}$ <sup>25,28-31</sup>. The negative values of  $\eta^E$  and the absence of any maxima in viscosity as a function of mole fraction indicate further that the intermolecular association between unlike molecules is likely to be absent in the binary mixtures of PC + ME, PC + DME and PC + THF. It further supports the view point that the dispersion and dipolar forces between unlike molecules are the controlling forces and play the major role in the stabilization of these systems. However, for PC + CH<sub>3</sub>OH mixtures, signs of  $G^{*E}$  are positive and its magnitudes are small at each temperature. Small positive values of  $G^{*E}$  along with negative  $v^E$  and  $\eta^E$  have also been found for other binary mixtures<sup>9</sup> where a strong dipole-dipole and dispersion interactions are predominant. Analysis of viscosity, excess molar volume and excess viscosity data for PC-CH<sub>3</sub>OH mixtures reveals that the system lacks a strong specific interaction between unlike molecules. Considering all findings, it appears that the interstitial accommodation of one component into the structure of the other play the major role for the stabilization of PC-CH<sub>3</sub>OH binary system.

Table 1. Experimental Density ( $\rho$ )/gcm<sup>-3</sup>, Kinematic Viscosity ( $\nu$ )/St, Absolute Viscosity ( $\eta$ )/cP and Interaction Parameters ( $\nu_{1j}$ ) Data for the Binary Mixtures of Propylene Carbonate 25, 35 and 45°C

$x_1$	$\rho$	$\nu$	$\eta$	$\nu_{12}$	$\nu_{21}$
PC + ME at 25°C					
0.0000	1.19883	2.0613	2.4711		
0.0632	1.18541	1.9384	2.2977		
0.1045	1.17620	1.8648	2.1934		
0.1523	1.16489	1.8060	2.1038		
0.2117	1.14959	1.7430	2.0037		
0.2527	1.14046	1.7097	1.9499		
0.3050	1.13021	1.6725	1.8902	1.5363	1.4861
0.3489	1.12038	1.6446	1.8425		
0.3970	1.10935	1.6170	1.7938		
0.4547	1.09591	1.5923	1.7450		
0.4979	1.08566	1.5814	1.7169		
0.5531	1.07227	1.5668	1.6801		

Contd..



Table 1 (Contd..)

0.5954	1.06174	1.5672	1.6640
0.6954	1.03576	1.5636	1.6195
0.7346	1.02571	1.5535	1.5934
0.7959	1.01037	1.5580	1.5742
0.8414	0.99902	1.5701	1.5686
0.9025	0.98376	1.5706	1.5451
0.9593	0.96985	1.5941	1.5460
1.0000	0.96002	1.6056	1.5414

PC + ME at 35°C

0.0000	1.18970	1.7211	2.0476	
0.0632	1.17626	1.6267	1.9134	
0.1045	1.16722	1.5628	1.8241	
0.1523	1.15630	1.5189	1.7563	
0.2117	1.14207	1.4699	1.6786	
0.2527	1.13256	1.4429	1.6341	
0.3050	1.12061	1.4110	1.5811	
0.3489	1.11068	1.3838	1.5370	
0.3970	1.09980	1.3596	1.4925	1.3027
				1.2591

Contd..

Table 1 (Contd..)

0.4547	1.08661	1.3399	1.4559
0.4979	1.07218	1.3334	1.4296
0.5954	1.05260	1.3159	1.3851
0.6388	1.04182	1.3064	1.3610
0.6954	1.02764	1.3053	1.3413
0.7346	1.01770	1.3017	1.3247
0.7959	1.00234	1.3068	1.3099
0.8414	0.99098	1.3084	1.2966
0.9025	0.97570	1.3078	1.2760
0.9593	0.96228	1.3156	1.2659
1.0000	0.95356	1.3192	1.2579

PC + ME at 45°C

0.0000	1.17796	1.4630	1.7234
0.0632	1.16485	1.3841	1.6123
0.1045	1.15626	1.3326	1.5409
0.1523	1.14614	1.2947	1.4840
0.2117	1.13323	1.2557	1.4230

Contd..

Table 1 (Contd..)

0.2527	1.12441	1.2278	1.3813		
0.3050	1.11252	1.2035	1.3389		
0.3489	1.10261	1.1850	1.3065		
0.3970	1.09156	1.1662	1.2729	1.1087	1.0808
0.4547	1.07799	1.1352	1.2237		
0.4979	1.06769	1.1372	1.2142		
0.5531	1.05423	1.1225	1.1834		
0.5954	1.04369	1.1226	1.1716		
0.6388	1.03295	1.1100	1.1466		
0.6954	1.01878	1.1065	1.1273		
0.7346	1.00883	1.1013	1.1110		
0.7959	0.99359	1.1046	1.0976		
0.8414	0.98236	1.1019	1.0825		
0.9025	0.96743	1.1123	1.0760		
0.9593	0.95418	1.1061	1.0554		
1.000	0.94715	1.1080	1.0494		

Contd..

Table 1 (Contd..)

PC + DME at 25°C					
0.0000	1.19883	2.0613	2.4711		
0.0493	1.18024	1.8869	2.2270		
0.0999	1.16206	1.6863	1.9600		
0.1524	1.14235	1.5335	1.7518		
0.1867	1.13033	1.4479	1.6366		
0.2298	1.11468	1.3524	1.5075		
0.3128	1.08643	1.1864	1.2889		
0.3708	1.06594	1.0898	1.1617		
0.4036	1.05526	1.0400	1.0974	0.7566	0.96687
0.4686	1.04097	0.9318	0.9699		
0.5193	1.01609	0.8742	0.8882		
0.6382	0.97683	0.7444	0.7272		
0.6767	0.96394	0.7142	0.6885		
0.7589	0.93705	0.6453	0.6047		
0.8103	0.92058	0.6071	0.5589		
0.8432	0.91034	0.5963	0.5429		
0.9035	0.89135	0.5466	0.4872		
0.9483	0.87695	0.5204	0.4564		
1.0000	0.86132	0.4918	0.4236		

Contd..

Table 1 (Contd..)

PC + DME at 35°C			
0.0000	1.18970	1.7211	2.0476
0.0493	1.17117	1.5653	1.8332
0.0999	1.15234	1.4396	1.6589
0.1524	1.13349	1.3194	1.4955
0.1867	1.12093	1.2507	1.4019
0.2298	1.10559	1.1727	1.2966
0.2769	1.08843	1.0867	1.1828
0.3708	1.05560	0.9448	0.9973
0.4036	1.04482	0.9412	0.9447
0.5193	1.00622	0.7731	0.7779
0.5704	0.98953	0.7254	0.7178
0.6767	0.95613	0.6385	0.6105
0.8103	0.91468	0.5523	0.5052
0.8432	0.90450	0.5228	0.4728
0.9035	0.88518	0.4949	0.4381
0.9483	0.87025	0.4680	0.4073
1.0000	0.85129	0.4519	0.3847

0.6214

0.9547

Contd..

Table 1 (Contd..)

PC + DME at 45°C			
0.0000	1.17796	1.4631	1.7234
0.0493	1.15999	1.3460	1.5614
0.0999	1.14185	1.2406	1.4166
0.1524	1.12302	1.1368	1.2766
0.2298	1.09638	1.0152	1.1130
0.3128	1.06819	0.9058	0.9676
0.3708	1.04842	0.8403	0.8810
0.4036	1.03767	0.7972	0.8272
0.5193	0.99856	0.6837	0.6827
0.5704	0.98137	0.6510	0.6389
0.6382	0.95858	0.5976	0.5728
0.6767	0.94547	0.5763	0.5449
0.7589	0.91810	0.5209	0.4782
0.8103	0.90116	0.5073	0.4571
0.8432	0.89078	0.4913	0.4376
0.9035	0.87144	0.4555	0.3970
0.9483	0.85667	0.4378	0.3750
1.0000	0.84076	0.4158	0.3496
			0.5700
			0.8230

Contd..

Table 1 (Contd..)

PC + THF at 25°C			
0.0000	1.19883	2.0613	2.4711
0.0524	1.18373	1.8813	2.2270
0.1036	1.16899	1.7301	2.0225
0.1429	1.15743	1.6153	1.8695
0.2064	1.13888	1.4695	1.6736
0.2431	1.12777	1.3891	1.5666
0.3083	1.10786	1.2576	1.3932
0.3478	1.09581	1.1894	1.3034
0.4095	1.07679	1.0877	1.1712
0.6094	1.01221	0.8315	0.8417
0.6479	0.99923	0.7842	0.7836
0.7082	0.97900	0.7324	0.7171
0.7635	0.96036	0.6820	0.6549
0.8062	0.94620	0.6547	0.6195
0.8388	0.93489	0.6237	0.5831
0.9035	0.91352	0.5864	0.5357
0.9631	0.89326	0.5430	0.4850
1.0000	0.88072	0.5257	0.4630

0.7517

1.1677

Contd..

## PC + THF at 35°C

0.0000	1.18970	1.7211	2.0476		
0.0524	1.17480	1.5817	1.8581		
0.1036	1.15960	1.4738	1.7090		
0.1429	1.14834	1.3888	1.5948		
0.2054	1.12941	1.2680	1.4321		
0.2431	1.11855	1.2079	1.3511		
0.3083	1.09884	1.0993	1.2080		
0.3478	1.08681	1.0425	1.1330		
0.4095	1.06788	0.9595	1.0247		
0.5098	1.03638	0.8446	0.8753	0.6745	1.0408
0.5621	1.01955	0.7889	0.8044		
0.6094	1.00412	0.7473	0.7503		
0.6479	0.99130	0.7077	0.7016		
0.7082	0.97123	0.6636	0.6445		
0.7635	0.95253	0.6257	0.5960		
0.8388	0.92667	0.5786	0.5361		
0.9035	0.90441	0.5387	0.4872		
0.9631	0.88035	0.5053	0.4449		
1.0000	0.87033	0.4914	0.4277		

Contd..



Table 1 (Contd..)

## PC + THF at 45°C

0.0000	1.17796	1.4630	1.7234		
0.0524	1.16457	1.3648	1.5894		
0.1036	1.14998	1.2689	1.4592		
0.1429	1.13889	1.2074	1.3751		
0.2064	1.12024	1.1057	1.2386		
0.2431	1.10988	1.0516	1.1672		
0.3083	1.09041	0.9689	1.0565		
0.3478	1.07857	0.9197	0.9920		
0.5098	1.02837	0.7567	0.7782	0.5981	0.9404
0.5621	1.01179	0.7138	0.7222		
0.6094	0.99665	0.6742	0.6719		
0.6479	0.98406	0.6446	0.6343		
0.7082	0.96425	0.6043	0.5827		
0.8062	0.93169	0.5472	0.5098		
0.8388	0.92053	0.5220	0.4805		
0.9035	0.89934	0.4921	0.4425		
0.9631	0.87570	0.4613	0.4040		
1.0000	0.86140	0.4530	0.3902		

Contd..

Table 1 (Contd..)

PC + CH<sub>3</sub>OH at 25°C

0.0000	1.19883	2.0613	2.4711		
0.0512	1.18900	1.9060	2.2662		
0.0983	1.17959	1.1778	2.0971		
0.1406	1.17072	1.6724	1.9579		
0.1883	1.16007	1.5607	1.8106		
0.2587	1.14283	1.4138	1.6157		
0.3430	1.12018	1.2627	1.4144		
0.3986	1.10408	1.1758	1.2982	0.9084	1.2641
0.4723	1.08032	1.0802	1.1670		
0.5124	1.06648	1.0379	1.1069		
0.5819	1.04009	0.9624	1.0009		
0.6762	0.99933	0.8684	0.8678		
0.7580	0.95820	0.8042	0.7707		
0.8297	0.91668	0.7513	0.6887		
0.9256	0.84986	0.7038	0.5981		
0.9495	0.83021	0.6992	0.5805		
1.0000	0.78663	0.6912	0.5437		

Contd..

Table 1 (Contd..)

PC + CH <sub>3</sub> OH at 35°C			
0.0000	1.18970	1.7211	2.0476
0.0512	1.17991	1.6062	1.8952
0.0983	1.17095	1.5007	1.7572
0.1406	1.16178	1.4184	1.6479
0.1883	1.15091	1.3323	1.5334
0.2587	1.13389	1.2147	1.3773
0.3430	1.11119	1.0915	1.2129
0.4723	1.07151	0.9351	1.0020
0.5124	1.05738	0.8966	0.9480
0.5819	1.03107	0.8390	0.8651
0.6762	0.99425	0.7625	0.7581
0.8297	0.90779	0.6647	0.6034
0.8931	0.86530	0.6376	0.5517
0.9256	0.84155	0.6277	0.5282
0.9495	0.82210	0.6225	0.5118
1.0000	0.77718	0.6175	0.4799

0.7847

1.1093

Contd.. .

Table 1 (Contd..)

PC + CH <sub>3</sub> CH at 45°C						
0.0000	1.17796	1.4630	1.7234			
0.0512	1.16875	1.3638	1.5939			
0.0983	1.15927	1.2770	1.4804			
0.1406	1.15085	1.2105	1.3931			
0.1883	1.14050	1.1361	1.2958			
0.2587	1.12410	1.0398	1.1688			
0.3430	1.10199	0.9383	1.0340			
0.3986	1.08592	0.8847	0.9607	0.6933		0.9509
0.4723	1.06278	0.8170	0.8663			
0.5124	1.04879	0.7849	0.8232			
0.5819	1.02251	0.7383	0.7549			
0.8297	0.89905	0.5874	0.5281			
0.8931	0.85658	0.5637	0.4829			
0.9256	0.83204	0.5554	0.4621			
0.9495	0.81301	0.5518	0.4486			
1.000	0.76720	0.5495	0.4216			

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Table 2--Excess Molar Volumes ( $V^E$ )/ $\text{cm}^3 \text{mol}^{-1}$ , Excess Viscosities ( $\eta^E$ )/cP and Excess Molar Free Energies of Activation of Flow ( $G^{*E}$ )/ $\text{J mol}^{-1}$  for the Binary Mixtures of Propylene Carbonate at 25, 35 and 45°C

$x_1$	$V^E$	$\eta^E$	$G^{*E}$
PC + ME at 25°C			
0.0000	0.0000	0.0000	0.000
0.0632	-0.0493	-0.1146	-107.381
0.1045	-0.0553	-0.1806	-174.249
0.1523	-0.0198	-0.2257	-220.357
0.2117	-0.1079	-0.2706	-267.709
0.2527	-0.0882	-0.2863	-287.659
0.3050	-0.0467	-0.2973	-307.290
0.3489	-0.0756	-0.3042	-320.060
0.3970	-0.0942	-0.3082	-330.692
0.4547	-0.1083	-0.3034	-331.990
0.4979	-0.1103	-0.2913	-321.707
0.5531	-0.0978	-0.2768	-310.519
0.5954	-0.0729	-0.2536	-284.172
0.6954	0.0529	-0.2051	-230.762
0.7346	0.0856	-0.1947	-224.200
0.7959	0.0978	-0.1570	-182.515
0.8414	0.0975	-0.1203	-138.324
0.9025	0.0889	-0.0670	-104.823
0.9593	0.0476	-0.0332	-39.408
1.0000	0.0000	0.0000	0.000

Contd..

Table 2 (Contd..)

PC + ME at 35°C			
0.0000	0.0000	0.0000	0.000
0.0632	-0.0365	-0.0843	-95.447
0.1045	-0.0463	-0.1410	-166.393
0.1523	-0.0297	-0.1708	-203.002
0.2117	0.0325	-0.2018	-242.583
0.2527	0.0478	-0.2139	-259.583
0.3050	0.0487	-0.2256	-278.559
0.3489	0.0366	-0.2351	-293.377
0.3970	0.0170	-0.2416	-312.294
0.4547	-0.0037	-0.2326	-304.422
0.4979	-0.0085	-0.2248	-287.010
0.5954	0.0517	-0.1923	-254.865
0.6388	0.0806	-0.1821	-244.766
0.6954	0.1213	-0.1571	-210.324
0.7346	0.1541	-0.1428	-192.472
0.7959	0.1796	-0.1092	-144.069
0.8414	0.1936	-0.0866	-113.333
0.9025	0.2007	-0.0589	-78.091
0.9593	0.1318	-0.0241	-29.982
1.0000	0.0000	0.0000	0.000

Contd..

Table 2 (Contd..)

PC + ME at 45°C

0.0000	0.0000	0.0000	0.000
0.0632	-0.0362	-0.0685	-93.938
0.1045	-0.0627	-0.1121	-160.138
0.1523	-0.0857	-0.1368	-197.410
0.2117	-0.0970	-0.1577	-230.428
0.2527	-0.1161	-0.1718	-255.781
0.3050	-0.0998	-0.1789	-268.863
0.3489	-0.0964	-0.1817	-275.816
0.3970	-0.0836	-0.1829	-281.165
0.4547	-0.0518	-0.1796	-308.707
0.4976	-0.0222	-0.1736	-271.637
0.5531	0.0317	-0.1672	-265.497
0.5954	0.0862	-0.1505	-234.840
0.6388	0.1314	-0.1463	-233.680
0.6954	0.1964	-0.1274	-202.250
0.7246	0.2476	-0.1173	-187.854
0.7959	0.2936	-0.0894	-138.310
0.8414	0.3142	-0.0738	-114.634
0.9025	0.3188	-0.0391	-50.573
0.9593	0.2593	-0.0214	-29.436
1.000	0.0000	0.0000	0.000

Contd..

Table 2 (Contd.)

PC + DME at 25°C

0.0000	0.0000	0.0000	0.000
0.0493	-0.1186	-0.1432	-43.064
0.0999	-0.2794	-0.3070	-141.124
0.1524	-0.3543	-0.4073	-189.342
0.1867	-0.4517	-0.4522	-209.345
0.2298	-0.5135	-0.4931	-224.956
0.3128	-0.7268	-0.5417	-253.969
0.3708	-0.7678	-0.5502	-258.047
0.4036	-0.8514	-0.5473	-257.480
0.4686	-0.9143	-0.5417	-280.171
0.5193	-0.9137	-0.5196	-276.752
0.6382	-0.8936	-0.4372	-252.925
0.6767	-0.8284	-0.3971	-219.147
0.7589	-0.6817	-0.3126	-179.513
0.8103	-0.5743	-0.2531	-148.521
0.8432	-0.5186	-0.2018	-76.653
0.9035	-0.3501	-0.1340	-79.307
0.9483	-0.1528	-0.0731	-42.577
1.000	0.0000	0.0000	0.000

Contd.



Table 2 (Contd..)

PC + DME at 35°C

0.0000	0.0000	0.0000	0.000
0.0493	-0.1346	-0.1324	-73.281
0.0999	-0.2584	-0.2226	-113.560
0.1524	-0.4098	-0.2987	-156.343
0.1867	-0.4723	-0.3352	-175.278
0.2298	-0.5672	-0.3689	-191.986
0.2769	-0.6136	-0.4043	-225.277
0.3708	-0.7386	-0.4337	-261.532
0.4036	-0.8173	-0.4318	-261.649
0.5193	-0.9361	-0.4062	-266.296
0.5704	-0.9783	-0.3813	-254.394
0.6767	-1.0780	-0.3118	-217.417
0.8103	-1.0500	-0.1950	-132.844
0.8432	-1.0086	-0.1726	-161.223
0.9035	-0.8137	-0.1071	-95.956
0.9483	-0.5582	-0.0634	-86.427
1.0000	0.0000	0.0000	0.000

Contd..

Table 2 (Contd..)

PC + DME at 45°C

0.0000	0.0000	0.0000	0.000
0.0493	-0.1785	-0.0943	-55.515
0.0999	-0.3572	-0.1696	-102.016
0.1524	-0.5126	-0.2374	-157.624
0.2298	-0.7765	-0.2947	-198.377
0.3128	-1.0187	-0.3261	-222.836
0.3708	-1.1352	-0.3330	-228.011
0.4036	-1.2213	-0.3417	-257.957
0.5193	-1.3120	-0.3273	-279.069
0.5704	-1.3018	-0.3009	-238.944
0.6382	-1.2319	-0.2738	-239.719
0.6767	-1.1432	-0.2489	-207.942
0.7589	-0.9387	+0.2026	-202.471
0.8103	-0.8172	-0.1531	-102.229
0.8432	-0.6343	-0.1274	-77.878
0.9035	-0.4673	-0.0852	-77.898
0.9483	-0.2076	-0.0456	-34.839
1.000	0.0000	0.0000	0.000

Contd..

Table 2 (Contd..)

PC + THF at 25°C

0.0000	0.0000	0.0000	0.000
0.0524	-0.0687	-0.1389	-42.203
0.1036	-0.1435	-0.2406	-70.544
0.1429	-0.1863	-0.3146	-103.478
0.2064	-0.2731	-0.3830	-116.754
0.2431	-0.2987	-0.4163	-128.896
0.3083	-0.3386	-0.4588	-150.364
0.3478	-0.3682	-0.4693	-152.288
0.4095	-0.4057	-0.4776	-162.359
0.6094	-0.3486	-0.4057	-149.898
0.6479	-0.3023	-0.3865	-166.051
0.7082	-0.2410	-0.3319	-133.778
0.7635	-0.1827	-0.2830	-127.284
0.8062	-0.1613	-0.2327	-87.342
0.8388	-0.1042	-0.2036	-100.271
0.9035	-0.0897	-0.1211	-41.677
0.9631	-0.0325	-0.0521	-39.004
1.000	0.0000	0.0000	0.000

Contd..

Table 2 (Contd..)

## PC + THF at 35°C

0.0000	0.0000	0.0000	0.000
0.0524	-0.0935	-0.1046	-41.244
0.1036	-0.1446	-0.1708	-51.649
0.1429	-0.2174	-0.2213	-73.237
0.2064	-0.2878	-0.2812	-96.340
0.2431	-0.3382	-0.3027	-99.674
0.3083	-0.4040	-0.3402	-126.908
0.3478	-0.4416	-0.3512	-133.739
0.4095	-0.4962	-0.3596	-145.381
0.5098	-0.5447	-0.3465	-148.193
0.5621	-0.5453	-0.3327	-154.896
0.6094	-0.5326	-0.3101	-142.894
0.6479	-0.5039	-0.2965	-159.722
0.7082	-0.4631	-0.2559	-134.031
0.7635	-0.4063	-0.2148	-110.845
0.8388	-0.2987	-0.1527	-76.801
0.9035	-0.2083	-0.0968	-59.613
0.9631	-0.0931	-0.0426	-50.001
1.0000	0.0000	0.0000	0.000

Contd..

Table 2 (Contd..)

PC + THF at 45°C

0.0000	0.0000	0.0000	0.000
0.0524	-0.1983	-0.0641	-14.108
0.1036	-0.2869	-0.1261	-41.758
0.1429	-0.3672	-0.1578	-46.796
0.2064	-0.4489	-0.2096	-76.210
0.2431	-0.5326	-0.2321	-91.692
0.3083	-0.6077	-0.2559	-101.453
0.3478	-0.6541	-0.2677	-114.241
0.5098	-0.7495	-0.2655	-122.773
0.5621	-0.7613	-0.2518	-115.342
0.6094	-0.7649	-0.2390	-120.456
0.6479	-0.7478	-0.2253	-121.133
0.7082	-0.7188	-0.1965	-107.767
0.8062	-0.6524	-0.1387	-74.093
0.8388	-0.6038	-0.1246	-101.388
0.9035	-0.4537	-0.0763	-64.853
0.9631	-0.2132	-0.0354	-59.700
1.0000	0.0000	0.0000	0.000

Cohtd..

Table 2 (Contd.)

PC + CH<sub>3</sub>OH at 25°C

0.0000	0.0000	0.0000	0.000
0.0512	-0.0376	-0.1062	2.953
0.0983	-0.0813	-0.1845	8.838
0.1406	-0.1213	-0.2422	15.007
0.1883	-0.1594	-0.2976	18.532
0.2587	-0.1907	-0.3568	24.782
0.3430	-0.2316	-0.3956	34.290
0.3986	-0.2728	-0.4046	41.120
0.4723	-0.3002	-0.3938	63.392
0.5124	-0.3241	-0.3766	85.104
0.5819	-0.3415	-0.3486	96.339
0.6762	-0.3574	-0.3000	85.373
0.7580	-0.3528	-0.2394	77.899
0.8297	-0.3307	-0.1832	40.973
0.9256	-0.2035	+0.0890	27.436
0.9495	-0.1204	-0.0605	6.449
1.0000	0.0000	0.0000	0.000

Contd..

Table 2 (Contd..)

PC + CH<sub>3</sub>OH at 35°C

0.0000	0.0000	0.0000	0.000
0.0512	-0.0449	-0.0721	17.897
0.0983	+0.1236	-0.1363	19.948
0.1406	-0.1468	-0.1793	31.212
0.1883	-0.1735	-0.2190	43.169
0.2587	-0.2243	-0.2647	53.945
0.3430	-0.2670	-0.2970	64.995
0.4723	-0.3539	-0.3052	76.156
0.5124	-0.3618	-0.2963	86.206
0.5819	-0.3874	-0.2703	109.311
0.6762	-0.4123	-0.2294	100.068
0.8297	-0.3832	-0.1435	46.812
0.8931	-0.3122	-0.0958	21.909
0.9256	-0.2774	-0.0683	11.649
0.9495	-0.2013	-0.0473	6.666
1.0000	0.0000	0.0000	0.000

Contd..

Table 2 (Contd..)

PC + CH<sub>3</sub>OH at 45°C

0.0000	0.0000	0.0000	0.000
0.0512	-0.0876	-0.0628	9.166
0.0983	-0.1294	-0.1150	11.546
0.1406	-0.2035	-0.1473	25.533
0.1883	-0.2637	-0.1825	30.083
0.2587	-0.3527	-0.2178	42.819
0.3430	-0.4268	-0.2429	55.002
0.3986	-0.4683	-0.2438	78.478
0.4723	-0.5299	-0.2403	93.780
0.5124	-0.5412	-0.2332	104.599
0.5819	-0.5592	-0.2110	133.664
0.8297	-0.5032	-0.1152	50.313
0.8931	-0.4159	-0.0779	18.355
0.9256	-0.3318	-0.0563	6.177
0.9495	-0.2699	-0.0387	2.438
1.0000	0.0000	0.0000	0.000

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Table 3. Coefficients of Least-square Fit by Equation (6) for Excess Molar Volumes and Excess Viscosities at 25, 35 and 45°C

Property	Temperature	$A_0$	$A_1$	$A_2$	$A_3$	$A_4$	$\sigma(X^E)$
				PC + ME			
$V^E$	25°C	-0.4593	0.2245	-4.0510	-1.8413	-4.3483	0.039
	35°C	0.0944	0.0327	1.5310	-2.7296	-0.2745	0.020
	45°C	0.0465	-1.1076	-0.2040	-2.9147	4.5610	0.031
$\eta^E$	25°C	-1.1487	-0.5278	-0.4743	-0.1158	0.1315	0.005
	35°C	-0.8865	-0.4349	-0.2059	-0.0730	-0.0269	0.004
	45°C	-0.6958	-0.3135	-0.1750	-0.1116	-0.0516	0.003

Contd..

Table 3 (Contd..)

		PC + DME					
$v^E$	25°C	-3.6259	-0.9344	0.3058	-0.6439	0.5591	0.026
	35°C	-3.6537	1.6564	-2.6306	3.7041	-2.0226	0.022
	45°C	-5.1634	1.0595	0.7986	-0.7305	0.5523	0.026
$\eta^E$	25°C	-2.0639	-1.1171	-0.9847	0.1500	0.8107	0.013
	35°C	-1.6672	-0.6583	0.0695	-0.2123	-0.6540	0.004
	45°C	-1.3088	-0.5272	-0.1560	-0.0979	-0.0543	0.005

Contd..

Table 3 (Contd..)

## PC + THF

$v^E$	25°C	-1.5753	-0.6710	1.0392	0.4804	-0.6790	0.012
	35°C	-2.1799	0.4211	1.0100	-0.0240	-1.2610	0.007
	45°C	-3.0046	0.9241	-0.4780	0.2448	-2.2059	0.017
$\eta^E$	25°C	-1.8582	-0.6661	-0.1397	-0.1191	-0.2310	0.004
	35°C	-1.4198	-0.4252	0.1714	-0.0984	-0.5239	0.004
	45°C	-1.0778	-0.3930	-0.0057	0.2345	-0.0852	0.003

Contd..

Table 3 (Contd..)

PC + CH<sub>3</sub>OH

$\nu^E$	25°C	-1.2160	0.8540	-1.4431	0.2948	0.9537	0.012
	35°C	-1.4192	0.6055	-0.9593	1.3855	-0.7141	0.016
	45°C	-2.1560	0.6085	0.0096	1.8400	-2.2453	0.011
$\eta^E$	25°C	-1.5631	-0.6403	-0.1394	0.1970	-0.0677	0.011
	35°C	-1.1825	-0.4085	-0.1675	0.1499	0.1057	0.003
	45°C	-0.9375	-0.3505	-0.2078	0.1006	0.0737	0.002

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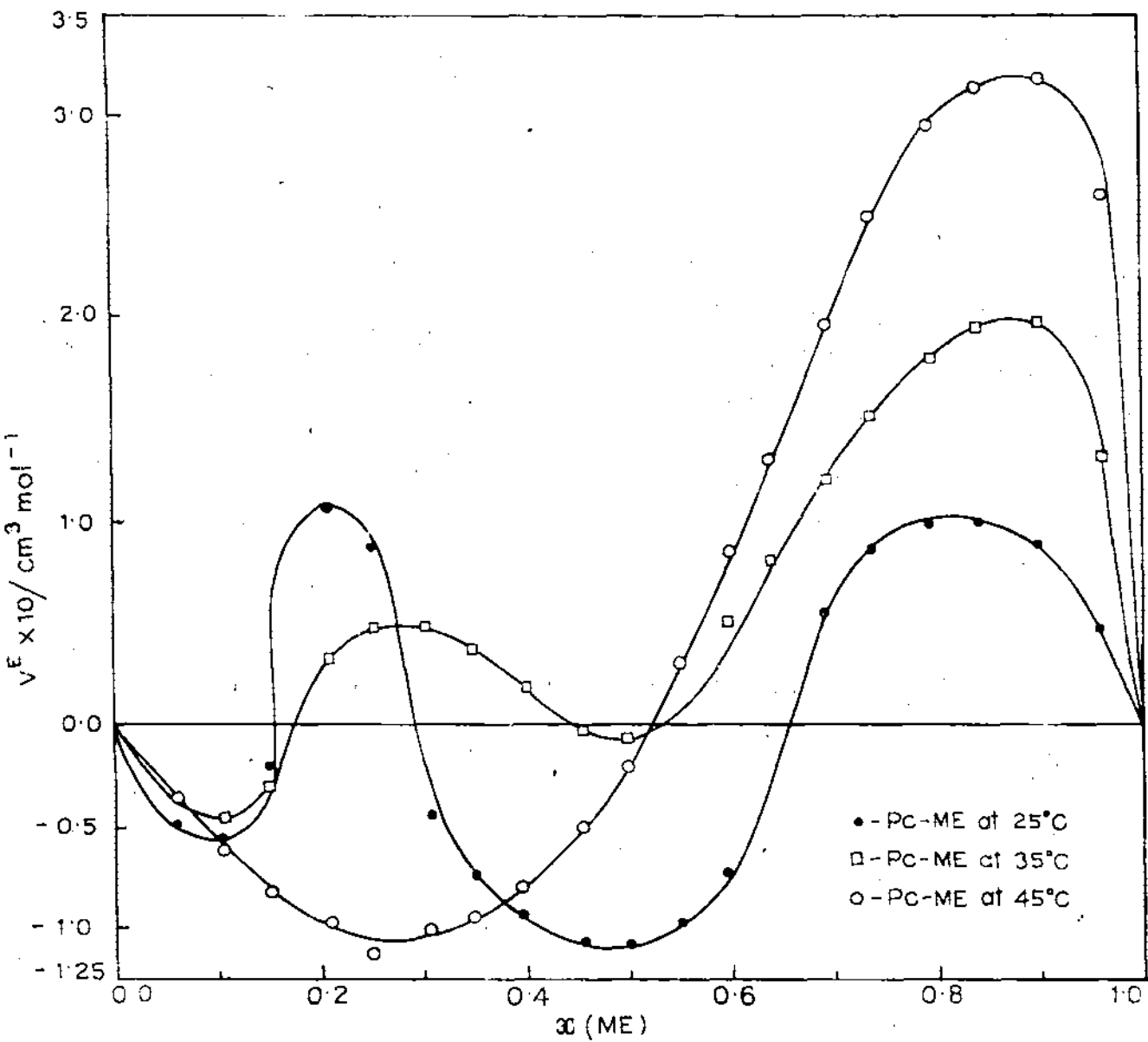


Fig. 1A. Variation of excess molar volume with the mole fraction of ME for PC + ME mixtures.

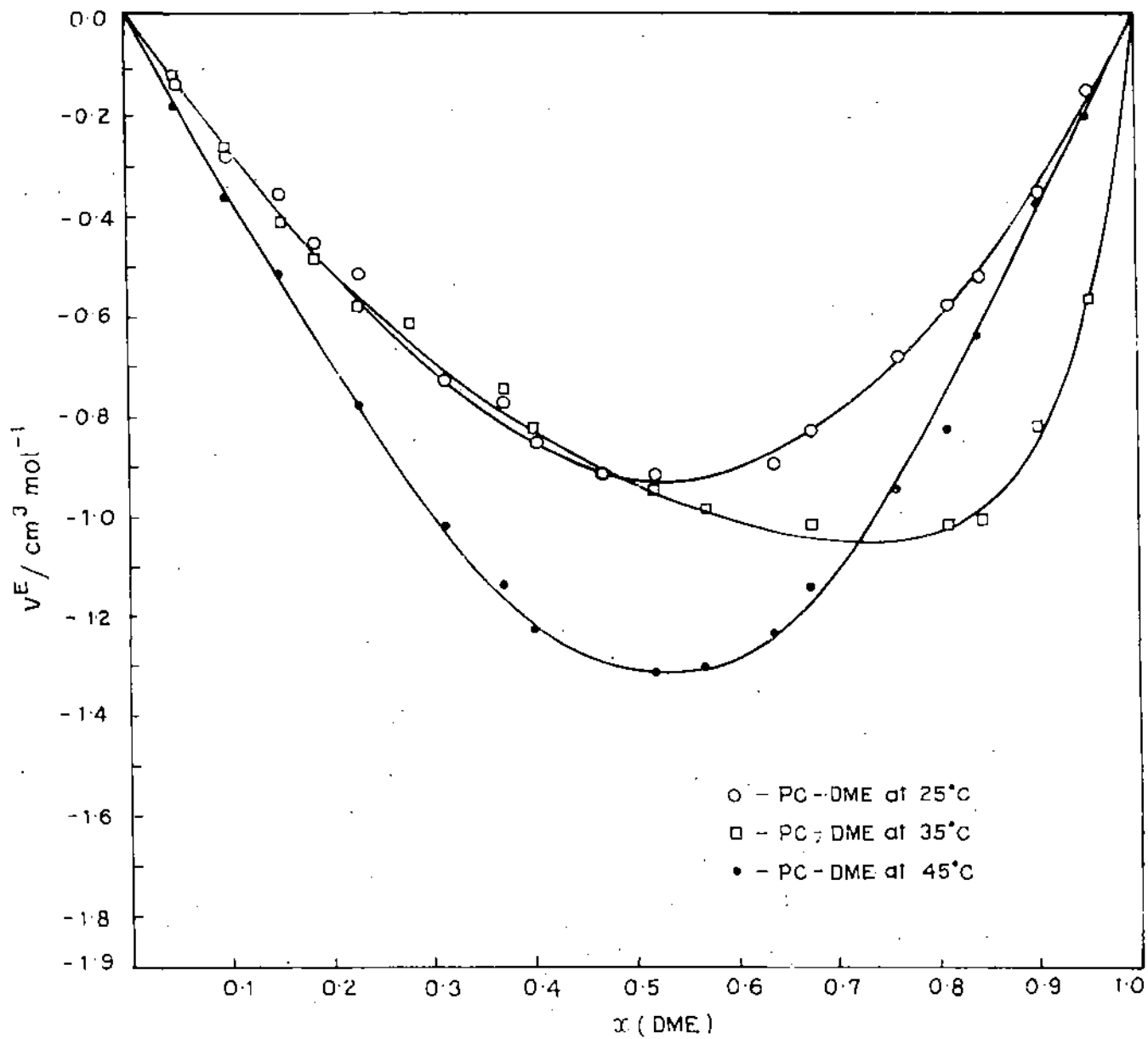


Fig. 1B. Variation of the excess molar volume with the mole fraction of DME for PC + DME mixtures.

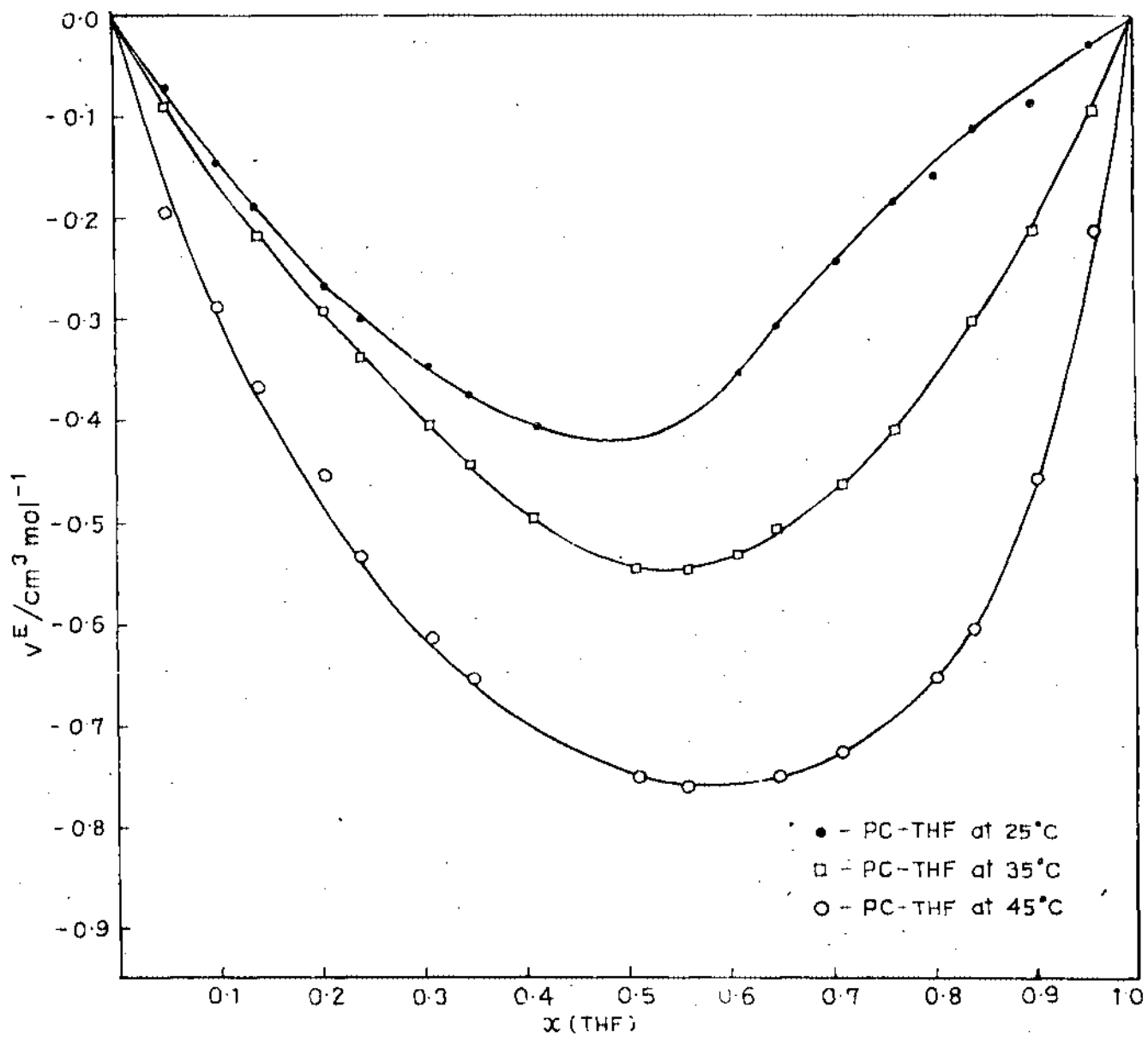


Fig. 1C. Variation of excess molar volume with the mole fraction of THF for PC + THF mixtures.

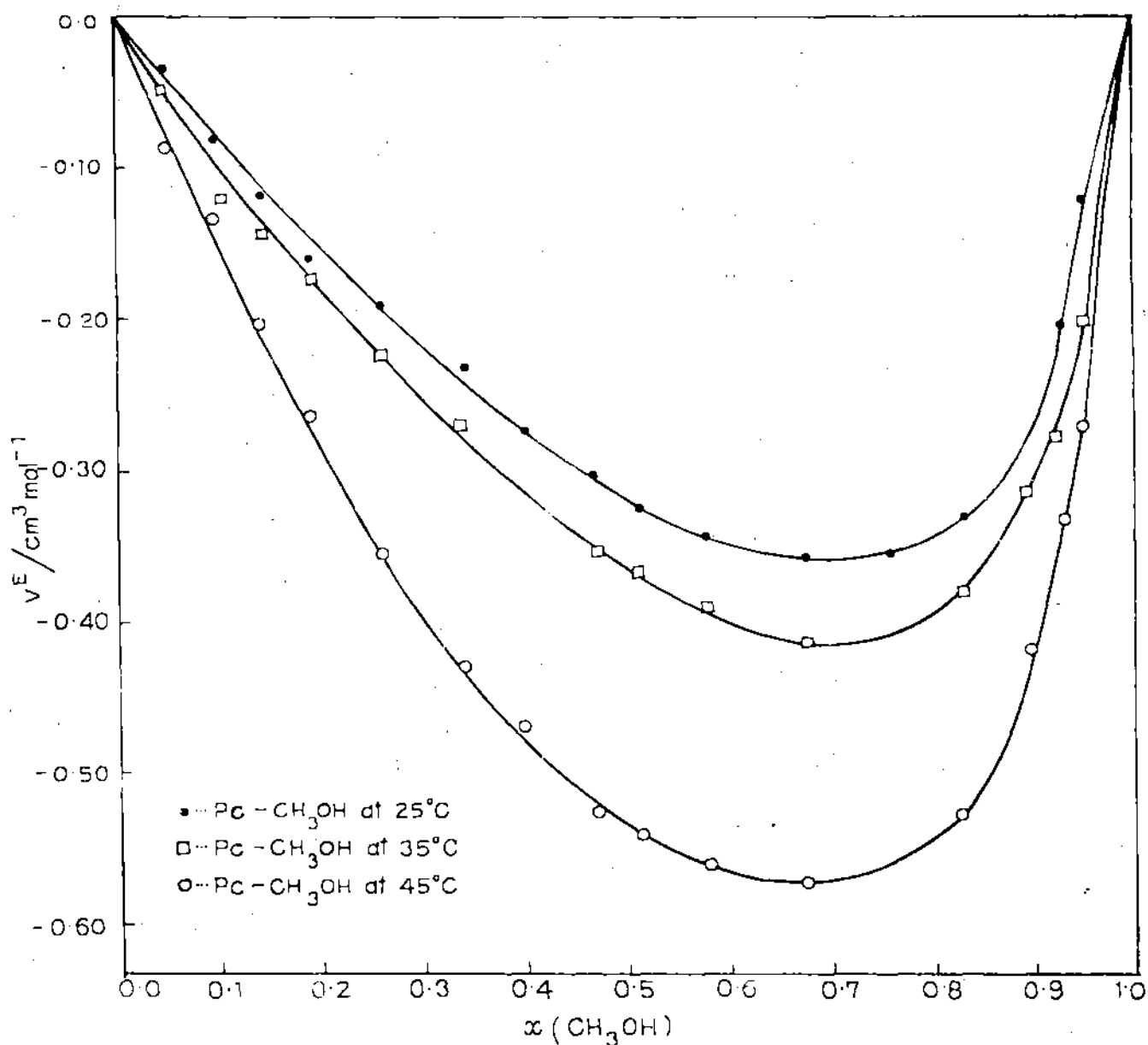


Fig. 1D. Variation of excess molar volume with the mole fraction of  $\text{CH}_3\text{OH}$  for PC +  $\text{CH}_3\text{OH}$  mixtures.



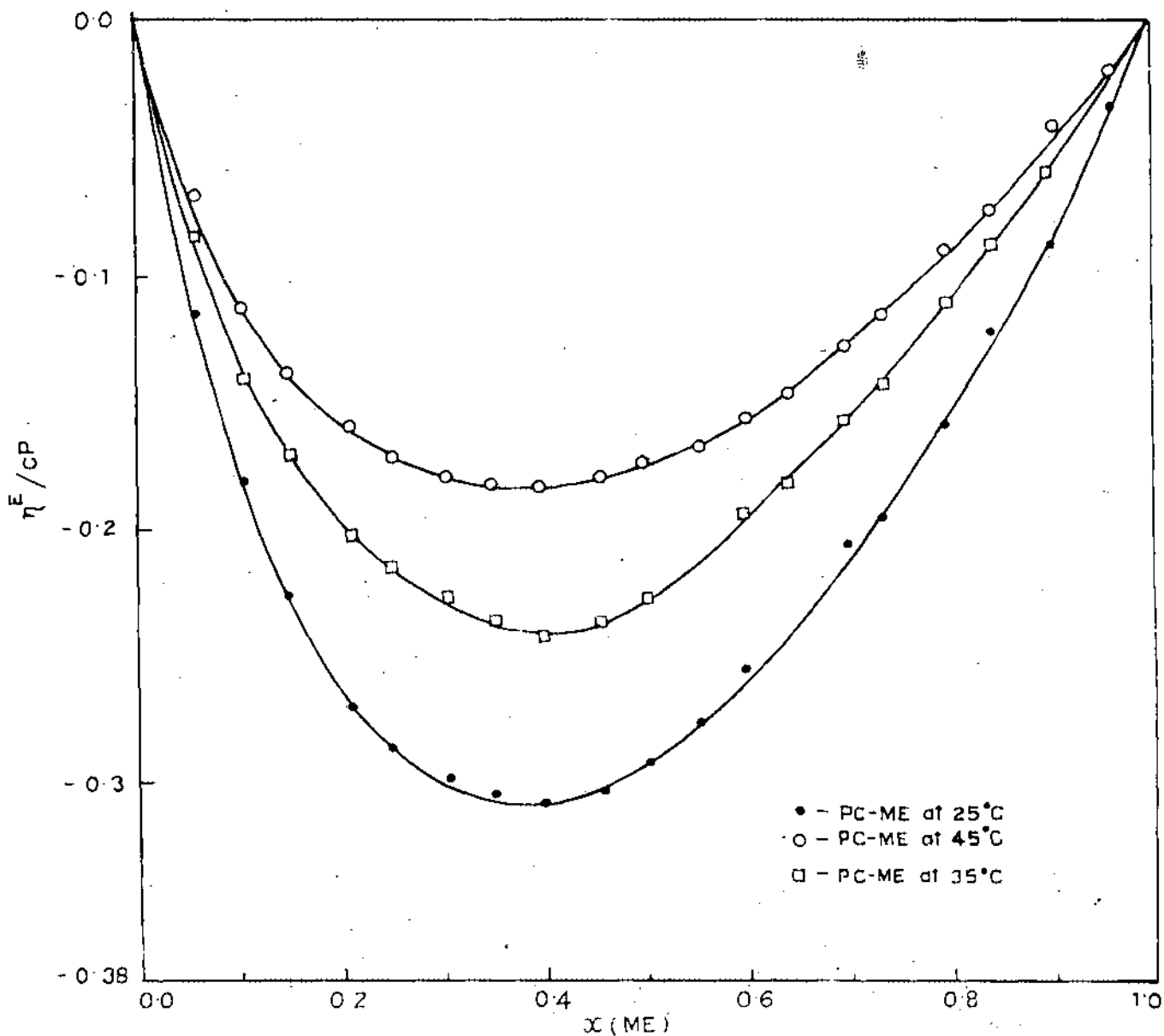


Fig. 2A. Variation of excess viscosity with the mole fraction of ME for PC+ME mixtures.

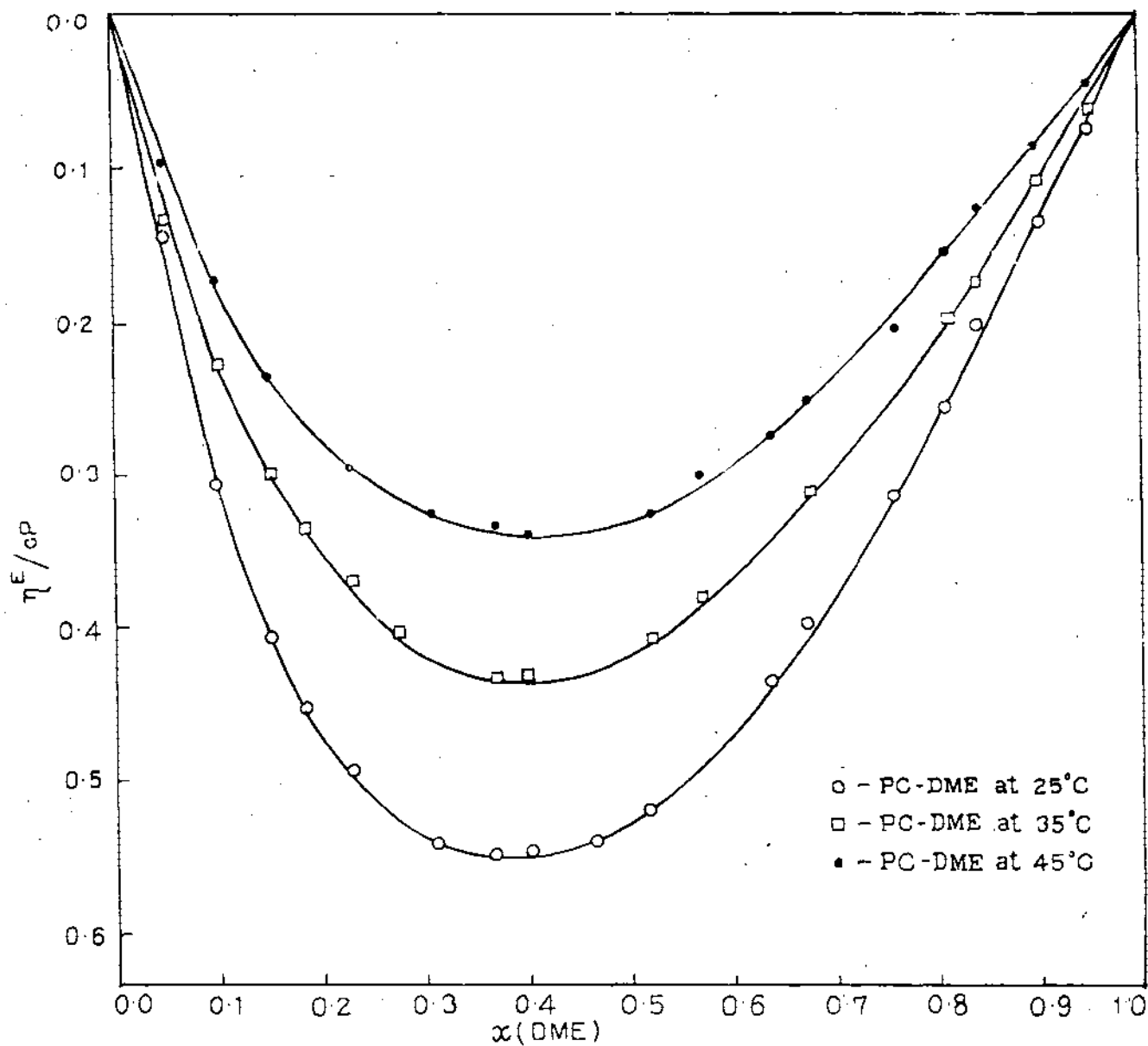


Fig. 2B. Variation of excess viscosity with the mole fraction of DME for PC + DME mixtures.

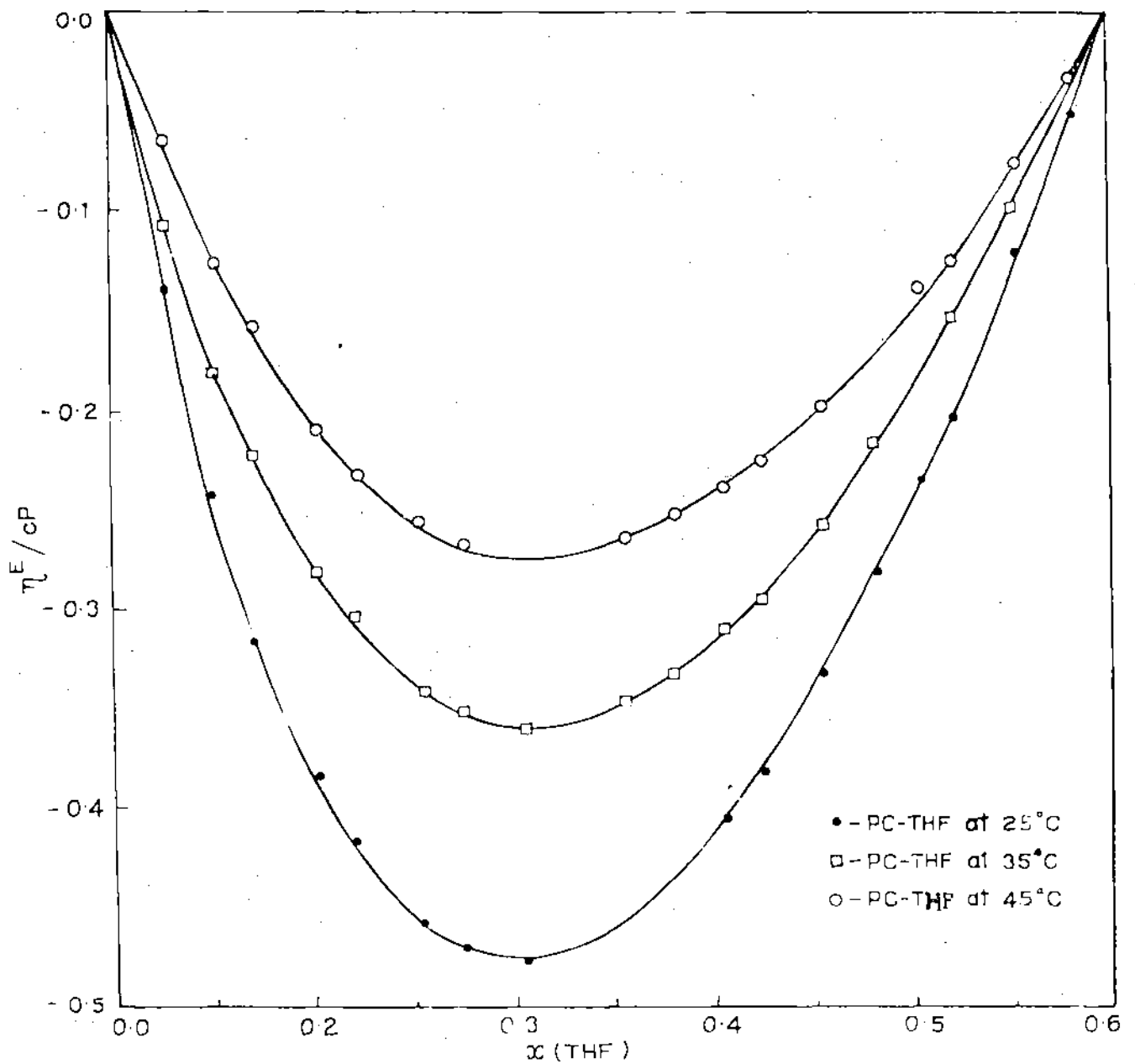


Fig. 2C. Variation of excess viscosity with the mole fraction of THF for PC+THF mixtures.

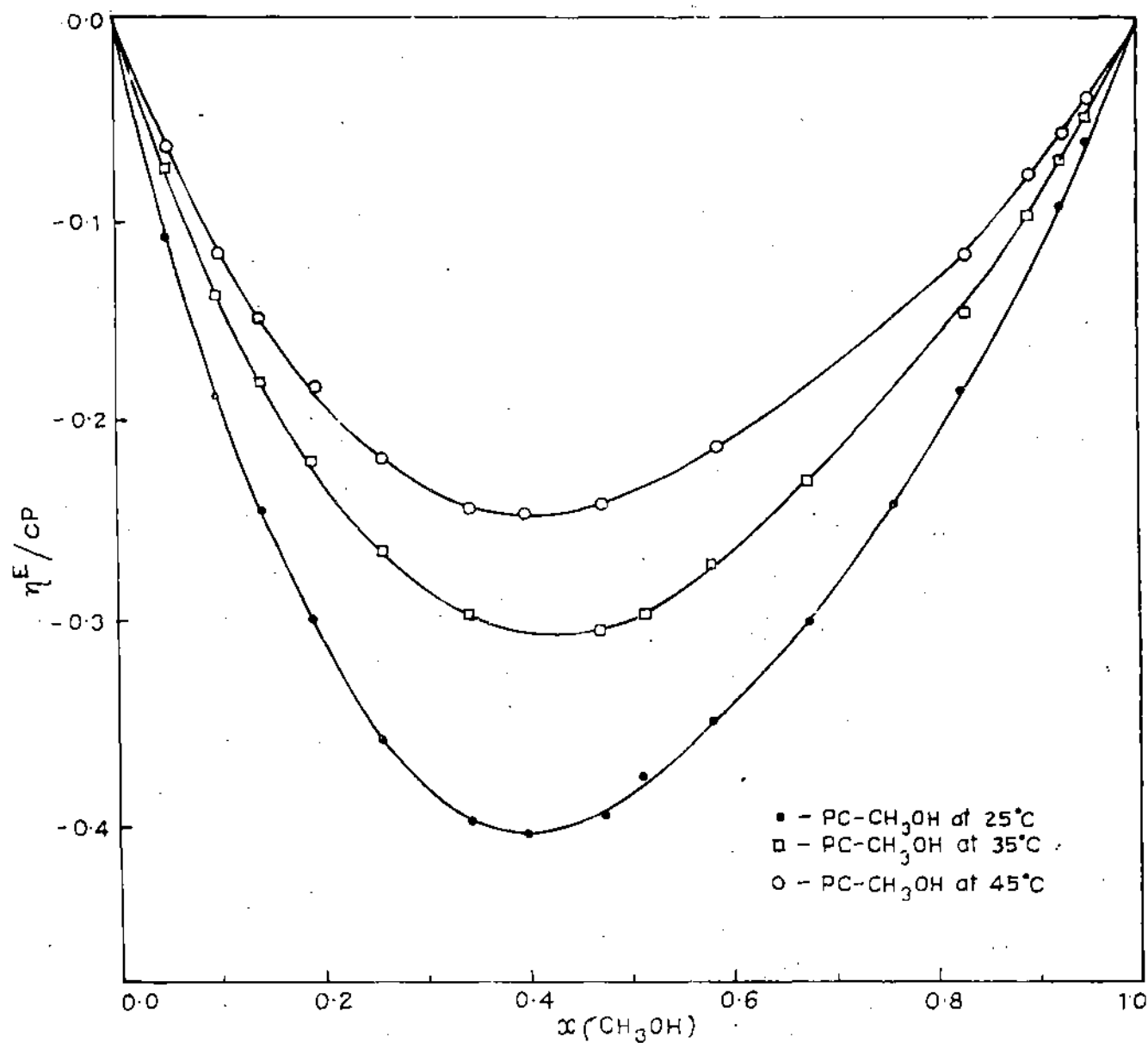


Fig. 2D. Variation of excess viscosity with the mole fraction of  $CH_3OH$  for PC+ $CH_3OH$  mixtures.

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