

# Viscosities, Ultrasonic Velocities at (288.15 and 298.15) K, and Refractive Indices at (298.15) K of Binary Mixtures of 2,4,6-Trimethyl-1,3,5-trioxane with Dimethyl Carbonate, Diethyl Carbonate, and Propylene Carbonate

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Viscosities, ultrasonic velocities, and refractive indices (at 298.15 K) of binary mixtures of 2,4,6-trimethyl-1,3,5-trioxane with dimethyl carbonate, diethyl carbonate, and propylene carbonate have been measured at (288.15 and 298.15) K. From these measurements, viscosity deviations ( $\delta\eta$ ), excess molar isentropic compressibilities ( $K_{S,m}^E$ ), and refractive index deviations ( $\Delta n_D$ ) were calculated. These results were fitted to the Redlich–Kister polynomial. The results were discussed in terms of molecular interactions.

## Introduction

Measurements of physicochemical properties such as viscosity, density, ultrasonic velocity, and refractive index of pure components and their binary mixtures are being increasingly used as tools for investigations of the properties of pure components and the nature of intermolecular interactions between the components of liquid mixtures.<sup>1</sup> Moreover, excess thermodynamic properties are used as important parameters for understanding molecular interactions. Earlier we reported<sup>2–4</sup> density and viscosity studies of binaries of paraldehyde + propylene carbonate at (288.15, 293.15, 298.15, 303.15, and 308.15) K, propylene carbonate, + *n*-alkanols at 298.15 K, and 2,4,6-trimethyl-1,3,5-trioxane + ethanol, 1-propanol, and 1-butanol at (298.15, 303.15, and 308.15) K. As part of the experimental investigation of the excess thermodynamic properties of industrially important liquids, we report in the present study, along with previously determined values of densities and viscosities of the system paraldehyde + propylene carbonate, the viscosity deviations ( $\delta\eta$ ), excess molar isentropic compressibility ( $K_{S,m}^E$ ), and refractive index deviations ( $\Delta n_D$ ) for binary mixtures of the above-mentioned systems at (288.15 and 298.15) K.

## Experimental Section

**Materials.** 2,4,6-Trimethyl-1,3,5-trioxane (Merck,  $\geq 98\%$ ) was used as such without further purification. The carbonates were dried by refluxing with anhydrous calcium carbonate and were distilled at atmospheric pressure.<sup>2–5</sup> The middle fraction collected was stored over 4 Å type molecular sieves. All of the solvents used were degassed before measurements. The purity of these solvents was checked by comparing the observed densities and refractive indices with those reported in the literature.<sup>5–10</sup> The measured values are included in Table 1 along with the literature values.

**Measurements.** The binary liquid mixtures were prepared by mixing known masses of pure liquids in airtight-stoppered bottles to minimize evaporation losses. All measurements of mass were performed on a Mettler one-pan balance (E-Mettler, Zurich), which allows reading the fifth decimal digit, with a

precision of  $\pm 0.05$  mg. The uncertainty in the mole fractions of the mixtures was estimated to be  $\pm 5 \times 10^{-5}$ .

Densities of pure components and their mixtures were measured using a single-arm capillary pycnometer having a bulb volume of approximately 5 cm<sup>3</sup> and a capillary bore with an internal diameter of 0.75 mm. At a level of confidence of 99.7 %, the uncertainty in the density measurement was  $\pm 3 \times 10^{-5}$ . Viscosity measurements were performed by using Schott Gerate (AVS 350) viscosity measuring equipment with a series of Ubbelohde viscometers. According to the method suggested by Hagenbach (AVS 350, viscosity measuring system, Introduction Manual Schott Gerate Hofheim, IS Germany 1986), replicate experiments were performed (five times) for each solution, and the results were averaged. At a level of confidence of 99.7 %, the uncertainty in the viscosity measurement was  $\pm 3 \times 10^{-2}$ . The experimental technique for density and viscosity measurements was the same as described in our previous paper.<sup>2,3</sup> The ultrasonic velocities were measured at 1 MHz with a single-crystal variable-path interferometer (F-81 Mittal Enterprises, New Delhi). The uncertainty of the ultrasonic measurement was  $\pm 0.1\%$ . Refractive indices were measured with a thermostated Abbe refractometer (Focus AR-204 85010) using the sodium D line. The refractometer was calibrated by means of a glass test piece of known refractive index supplied by the manufacturer. At a level of confidence of 99.7 %, the uncertainty in the refractive index measurement was  $\pm 3 \times 10^{-4}$ . For all of the above measurements of density, viscosity, ultrasonic velocity, and refractive index, temperature was controlled by circulating water through a JULABO F-25 ultrathermostat (made in Germany), which kept the temperature fluctuations within  $\pm 0.02$  K.

## Results

Table 2 lists the experimental values of densities,  $\rho$ , ultrasonic velocities,  $u$ , and refractive indexes,  $n_D$ , of binary mixtures of 2,4,6-trimethyl-1,3,5-trioxane + dimethyl carbonate, diethyl carbonate, and propylene carbonate.

Dynamic viscosities,  $\eta$ , of binary mixtures at different temperatures were calculated by using densities and flow times using the relation

$$\eta = K\rho(t - HC) \quad (1)$$

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**Table 1. Comparison of Experimental and Literature Values of Density,  $\rho$ , Isobaric Thermal Expansivities,  $\alpha_P$ , Molar Isobaric Heat Capacities,  $C_{P,m}$ , and Refractive Indices,  $n_D$ , of Pure Compounds**

compound	$T/K$	$\rho/g \cdot cm^{-3}$		$\alpha_P/kK^{-1}$	$C_{P,m}/J \cdot K^{-1} \cdot mol^{-1}$	$n_D$	
		expt	lit		lit	expt	lit
2,4,6-trimethyl-1,3,5-trioxane	288.15	0.99969		1.110 <sup>c</sup>	220 <sup>a</sup>		
	298.15	0.98835		1.164 <sup>c</sup>	250 <sup>a</sup>	1.4029	
dimethyl carbonate	288.15	1.07643		1.239 <sup>c</sup>	164.10		
	298.15	1.06310	1.0632 <sup>6</sup>	1.256 <sup>c</sup>	165.80	1.3660	1.3667 <sup>6</sup>
diethyl carbonate	288.15	0.98041		1.036 <sup>c</sup>	217.69 <sup>10</sup>		
	298.15	0.96915	0.96926 <sup>5</sup> 0.9690 <sup>6</sup>	1.317 <sup>c</sup>	220.24 <sup>10</sup>	1.3830	1.3828 <sup>5</sup>
propylene carbonate	288.15	1.20867		0.872 <sup>c</sup>	160.19 <sup>b</sup>		
	298.15	1.19758	1.1988 <sup>7</sup>	0.958 <sup>8</sup>	162.31 <sup>8</sup>	1.4203	1.4199 <sup>5</sup>

<sup>a</sup> Private communication with K. N. Marsh. <sup>b</sup> Calculated from group additivity. <sup>c</sup> Calculated from measured densities of pure components at different temperatures.

where  $K$  and  $\rho$  are the viscometer constant and density, respectively,  $(t - HC)$  is the flow time adjusted by the Hagenbach correction factor, and

$$HC = \frac{E}{Kt^2}$$

The value  $E/K = 70\,500$  is given in the instruction manual provided by the manufacturer. The viscometer constant was calibrated using double-distilled water at 298.15 K. Viscometers with  $t > 200$  s were selected for measurements ( $K = 0.005$  mm<sup>2</sup>·s<sup>-2</sup>, used for the propylene carbonate binary system, and  $K = 0.003$  mm<sup>2</sup>·s<sup>-2</sup>, used for the dimethyl carbonate + diethyl carbonate binary system).

The calculated values of viscosity  $\eta$  are reported in Table 2. The viscosity deviations,  $\delta\eta$ , of binary mixtures were calculated by using the relation

$$\delta\eta = \eta - \sum_{i=1}^2 x_i \eta_i \quad (2)$$

where  $\eta$  is the viscosity of the mixture,  $\eta_i$  is the viscosity of pure component  $i$ , and  $x_i$  is the mole fraction of component  $i$ .

The values of the ultrasonic velocity,  $u$ , and mixture density,  $\rho$ , were used to calculate the isentropic compressibility,  $\kappa_S$ , by using the relation

$$\kappa_S = u^{-2} \rho^{-1} = V(Mu^2)^{-1} \quad (3)$$

The product  $K_{S,m}$  of the molar volume  $V$  and isentropic compressibility  $\kappa_S$  has been calculated from the relation

$$K_{S,m} = -\left(\frac{\partial V}{\partial P}\right)_S = V\kappa_S = V^2(Mu^2) \quad (4)$$

The calculated values of  $K_{S,m}$  are reported in Table 2.

The excess molar isentropic compressibility,  $K_{S,m}^E$ , was obtained from the relation

$$K_{S,m}^E = K_{S,m} - K_{S,m}^{id} \quad (5)$$

where  $K_{S,m}^{id}$ <sup>11,12</sup> is the isentropic compressibility of an ideal mixture of the components.

$$K_{S,m}^{id} = \sum_i x_i \left[ K_{S,i} - T A_{P,i} \left\{ \frac{\sum x_i A_{P,i}}{\sum x_i C_{P,i}} - \frac{A_{P,i}}{C_{P,i}} \right\} \right] \quad (6)$$

where  $A_{P,i}$  (the product of the molar volume  $V_i$  and the isobaric

expansivity  $\alpha_{P,i}$ ),  $C_{P,i}$  (the isobaric molar heat capacity), and  $K_{S,i}$  (the product of the molar volume  $V_i$  and isentropic compressibility  $\kappa_{S,i}$ ) are properties of pure liquid component  $i$ . The values of  $\alpha_{P,i}$  and  $C_{P,i}$  used for these calculations are listed in Table 1 along with their sources.

The refractive index deviations,  $\Delta n_D$ , were obtained from the relation

$$\Delta n_D = n_D - x_1 n_{D1} - x_2 n_{D2} \quad (7)$$

The calculated viscosity deviations ( $\delta\eta$ ), excess molar isentropic compressibility ( $K_{S,m}^E$ ), and refractive index deviations ( $\Delta n_D$ ) were correlated by the Redlich–Kister polynomial<sup>13</sup> by using the relation

$$\Delta Y = x_1 x_2 \sum_{i=0}^2 a_i (x_1 - x_2)^i \quad (8)$$

The coefficients,  $a_i$ , in eq 8 were estimated by the least-squares fitting method, and the standard deviation,  $\sigma$ , was calculated by using the relation

$$\sigma = \left[ \sum_{i=0}^D \frac{(\Delta Y_{\text{expt}} - \Delta Y_{\text{calcd}})^2}{(D - N)} \right]^{0.5} \quad (9)$$

where  $D$  and  $N$  are the number of data points and parameters, respectively.

Regression results for viscosity deviations ( $\delta\eta$ ), excess molar isentropic compressibility ( $K_{S,m}^E$ ), and refractive index deviations ( $\Delta n_D$ ) of binary mixtures of 2,4,6-trimethyl-1,3,5-trioxane + dimethyl carbonate, diethyl carbonate, and propylene carbonate at different temperatures are reported in Table 3.

## Discussion

The values of  $\eta$  and  $n_D$  for the three systems studied are given in Table 2. The dependence of  $\eta$  and  $n_D$  values on the mole fraction of 2,4,6-trimethyl-1,3,5-trioxane follows the trend dimethyl carbonate < diethyl carbonate < propylene carbonate. The values of  $\eta$  and  $n_D$  of the binary liquid mixtures of dimethyl carbonate and diethyl carbonate increase with the mole fraction of 2,4,6-trimethyl-1,3,5-trioxane but decrease for the mixture with propylene carbonate. Similar trends were also obtained by Pal et al.<sup>6,14</sup> for mixtures of trimethylene glycol dimethyl ether with similar carbonates.

Figure 1 shows the graphical variations of  $\delta\eta$  against  $x_1$  for 2,4,6-trimethyl-1,3,5-trioxane at (288.15 and 298.15) K. The values of  $\delta\eta$  are found to be negative over the entire range of

**Table 2. Densities ( $\rho$ ), Viscosities ( $\eta$ ), Ultrasonic Velocities ( $u$ ), the Product ( $K_{S,m}$ ) of Molar Volume and Isentropic Compressibility at (288.15 and 298.15) K, and Refractive Indices ( $n_D$ ) at (298.15) K for Binary Mixtures of 2,4,6-Trimethyl-1,3,5-trioxane (1) + Dimethyl Carbonate (2), + Diethyl Carbonate (2), + and Propylene Carbonate (2)**

$x_1$	$\rho/\text{g}\cdot\text{cm}^{-3}$	$\eta/\text{mPa}\cdot\text{s}$	$u/\text{m}\cdot\text{s}^{-1}$	$K_{S,m}/\text{mm}^3\cdot\text{mol}^{-1}\cdot\text{MPa}^{-1}$	$n_D$	$x_1$	$\rho/\text{g}\cdot\text{cm}^{-3}$	$\eta/\text{mPa}\cdot\text{s}$	$u/\text{m}\cdot\text{s}^{-1}$	$K_{S,m}/\text{mm}^3\cdot\text{mol}^{-1}\cdot\text{MPa}^{-1}$	$n_D$
2,4,6-Trimethyl-1,3,5-trioxane (1) + Dimethyl Carbonate (2)											
$T = 288.15\text{ K}$											
0.0000	1.07643	0.647	1242.0	50.40		0.3994	1.03814	0.844	1235.0	65.03	
0.0204	1.07419	0.660	1242.2	51.07		0.4940	1.03068	0.887	1231.2	68.85	
0.0409	1.07196	0.673	1242.3	51.77		0.5906	1.02362	0.936	1227.2	72.83	
0.0590	1.07003	0.684	1242.4	52.37		0.6960	1.01652	1.002	1223.1	77.21	
0.0845	1.06735	0.699	1242.4	53.25		0.8080	1.00963	1.091	1219.3	81.88	
0.1091	1.06481	0.713	1242.3	54.10		0.9289	1.00287	1.214	1216.7	86.75	
0.2258	1.05337	0.770	1240.4	58.33		1.0000	0.99969	1.374	1217.7	89.18	
0.3012	1.04650	0.803	1238.4	61.18							
$T = 298.15\text{ K}$											
0.0000	1.06310	0.579	1197.4	55.59	1.3660	0.3994	1.02583	0.727	1189.6	71.77	1.3860
0.0204	1.06099	0.590	1197.4	56.34	1.3673	0.4940	1.01835	0.757	1186.0	76.00	1.3894
0.0409	1.05889	0.600	1197.3	57.11	1.3686	0.5906	1.01131	0.793	1182.3	80.39	1.3925
0.0590	1.05707	0.609	1197.3	57.79	1.3696	0.6960	1.00426	0.840	1178.5	85.22	1.3956
0.0845	1.05453	0.620	1197.0	58.77	1.3711	0.8080	0.99747	0.906	1175.3	90.28	1.3985
0.1091	1.05211	0.630	1196.8	59.71	1.3725	0.9289	0.99120	0.998	1173.1	95.53	1.4014
0.2258	1.04101	0.673	1194.6	64.39	1.3786	1.0000	0.98835	1.107	1174.5	98.08	1.4029
0.3012	1.03420	0.697	1192.7	67.53	1.3820						
2,4,6-Trimethyl-1,3,5-trioxane (1) + Diethyl Carbonate (2)											
$T = 288.15\text{ K}$											
0.0000	0.98041	0.868	1225.6	81.82		0.4272	0.98980	0.985	1213.2	86.08	
0.0215	0.98099	0.877	1224.0	82.15		0.5162	0.99135	1.011	1213.8	86.58	
0.0425	0.98154	0.885	1222.6	82.45		0.6080	0.99287	1.047	1214.8	87.06	
0.0615	0.98203	0.892	1221.4	82.71		0.7032	0.99441	1.098	1215.7	87.58	
0.0816	0.98254	0.899	1220.3	82.97		0.7984	0.99594	1.167	1216.2	88.15	
0.1036	0.98308	0.906	1219.1	83.25		0.8957	0.99754	1.260	1216.7	88.72	
0.1911	0.98512	0.931	1215.7	84.23		1.0000	0.99969	1.374	1217.7	89.18	
0.3170	0.98774	0.959	1213.4	85.33							
$T = 298.15\text{ K}$											
0.0000	0.96915	0.749	1173.4	91.35	1.3830	0.4272	0.97757	0.836	1174.4	94.18	1.3926
0.0215	0.96957	0.756	1173.8	91.44	1.3835	0.5162	0.97928	0.854	1173.5	94.93	1.3945
0.0425	0.96999	0.763	1174.2	91.53	1.3840	0.6080	0.98100	0.878	1172.6	95.72	1.3963
0.0615	0.97036	0.768	1174.5	91.61	1.3844	0.7032	0.98274	0.913	1171.9	96.50	1.3982
0.0816	0.97076	0.774	1174.8	91.71	1.3848	0.7984	0.98441	0.962	1171.5	97.25	1.3999
0.1036	0.97119	0.779	1175.0	91.83	1.3853	0.8957	0.98604	1.028	1171.8	97.89	1.4015
0.1911	0.97292	0.798	1175.6	92.35	1.3873	1.0000	0.98835	1.107	1174.5	98.08	1.4029
0.3170	0.97541	0.819	1175.3	93.27	1.3901						
2,4,6-Trimethyl-1,3,5-trioxane (1) + Propylene Carbonate (2)											
$T = 288.15\text{ K}$											
0.0000	1.20867	3.086	1478.4	31.97		0.6022	1.06471	2.155	1314.0	61.45	
0.0910	1.18168	2.966	1451.8	35.61		0.7006	1.04698	1.974	1289.8	67.56	
0.2019	1.15181	2.809	1420.2	40.43		0.8028	1.02973	1.779	1265.5	74.34	
0.3007	1.12763	2.661	1392.9	45.07		0.9027	1.01400	1.581	1242.1	81.46	
0.4055	1.10407	2.494	1364.7	50.37		1.0000	0.99969	1.374	1217.7	89.18	
0.5011	1.08421	2.333	1339.7	55.57							
$T = 298.15\text{ K}$											
0.0000	1.19758	2.499	1442.3	34.22	1.4203	0.4055	1.09350	1.977	1325.5	54.40	1.4142
0.0910	1.17078	2.386	1415.0	38.20	1.4188	0.5011	1.07363	1.846	1299.9	60.21	1.4127
0.2019	1.14110	2.245	1382.6	43.45	1.4172	0.6022	1.05405	1.705	1273.4	66.72	1.4111
0.3007	1.11702	2.116	1354.5	48.55	1.4157	0.7006	1.03619	1.563	1248.5	73.59	1.4094

composition for systems containing dimethyl carbonate and diethyl carbonate and at the said temperatures. Comelli et al.<sup>15</sup> have also reported a similar trend in  $\delta\eta$  values for poly(ethylene glycols) with dialkyl carbonate mixtures. The values of  $\delta\eta$  for systems containing propylene carbonate were found to be positive.<sup>2</sup>  $\delta\eta$  values at equimolar concentrations follow the order propylene carbonate > diethyl carbonate > dimethyl carbonate. The minima in  $\delta\eta$  values at 298.15 K vary from  $-0.106\text{ mPa}\cdot\text{s}$  to  $0.044\text{ mPa}\cdot\text{s}$ . The maxima in  $\delta\eta$  values for propylene carbonate systems occur at 0.5 mole fraction, whereas for dimethyl carbonate and diethyl carbonate systems the minima occur at 0.7 mole fraction. The difference in the maximum in  $\delta\eta$  values for the said temperatures is almost same for all three systems. The observed trend is in agreement with the dielectric constant values, which are higher for propylene carbonate (64.92 at 298.15 K) and lower for dimethyl carbonate (3.087 at 288.15 K).

The values of ultrasonic velocities  $u$  decrease, and those of product  $K_{S,m}$  increase with increase in  $x_1$  of 2,4,6-trimethyl-1,3,5-trioxane, as evident from Table 2. The graphical variations of  $K_{S,m}^E$  against  $x_1$  for 2,4,6-trimethyl-1,3,5-trioxane at (288.15 and 298.15) K are shown in Figure 2. The values of  $K_{S,m}^E$  are found to be negative for the system 2,4,6-trimethyl-1,3,5-trioxane with propylene carbonate at the said temperatures, whereas positive values are observed for the system 2,4,6-trimethyl-1,3,5-trioxane with diethyl carbonate at 288.15 K. The negative  $K_{S,m}^E$  indicates strong interactions,<sup>16</sup> whereas positive values indicate weak interactions. For the systems 2,4,6-trimethyl-1,3,5-trioxane with dimethyl carbonate at 288.15 and 298.15 K and that with diethyl carbonate at 298.15 K, negative  $K_{S,m}^E$  values are observed up to a  $x_1 = 0.7$  concentration of 2,4,6-trimethyl-1,3,5-trioxane and becomes positive at higher

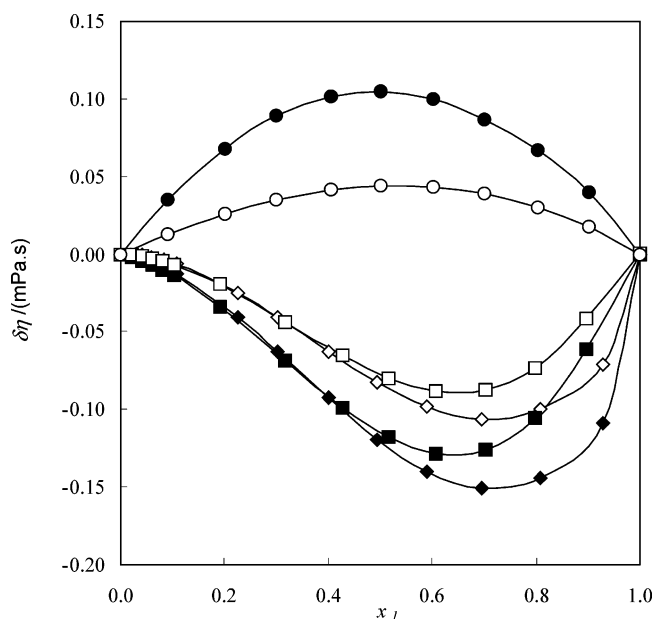
**Table 3.** Value of Coefficients,  $a_i$ , of the Redlich–Kister Polynomial<sup>a</sup> and Standard Deviations ( $\sigma$ )<sup>b</sup> of Binary Mixtures of 2,4,6-Trimethyl-1,3,5-trioxane (1) + Dimethyl Carbonate (2), + Diethyl Carbonate (2), and + Propylene Carbonate (2) at (288.15 and 298.15) K

parameter	$T$	$a_0$	$a_1$	$a_2$	$\sigma$
2,4,6-Trimethyl-1,3,5-trioxane (1) + Dimethyl Carbonate (2)					
$\delta\eta/\text{mPa}\cdot\text{s}$	288.15	-0.471	-0.619	-0.314	0.0095
	298.15	-0.329	-0.443	-0.177	0.0054
$K_{S,m}^E/\text{mm}^3\cdot\text{mol}^{-1}\cdot\text{MPa}^{-1}$	288.15	-2.91	4.89	2.40	0.0427
	298.15	-2.33	5.97	3.17	0.0494
$\Delta n_D$	298.15	0.0207	0.0060	0.0007	0.0001
2,4,6-Trimethyl-1,3,5-trioxane (1) + Diethyl Carbonate (2)					
$\delta\eta/\text{mPa}\cdot\text{s}$	288.15	-0.447	-0.369	3.616	0.0045
	298.15	-0.313	-0.265	0.062	0.0015
$K_{S,m}^E/\text{mm}^3\cdot\text{mol}^{-1}\cdot\text{MPa}^{-1}$	288.15	3.37	-3.02	1.68	0.0294
	298.15	-0.51	4.34	1.52	0.0349
$\Delta n_D$	298.15	0.0047	0.0231	0.0002	0.0001
2,4,6-Trimethyl-1,3,5-trioxane (1) + Propylene Carbonate (2)					
$\delta\eta/\text{mPa}\cdot\text{s}$	288.15	0.418	0.000	0.020	0.0010
	298.15	0.177	0.025	0.000	0.0010
$K_{S,m}^E/\text{mm}^3\cdot\text{mol}^{-1}\cdot\text{MPa}^{-1}$	288.15	-22.61	-2.35	-2.02	0.0369
	298.15	-24.95	-3.16	-1.92	0.0419
$\Delta n_D$	298.15	0.0046	0.0040	0.0000	0.0001

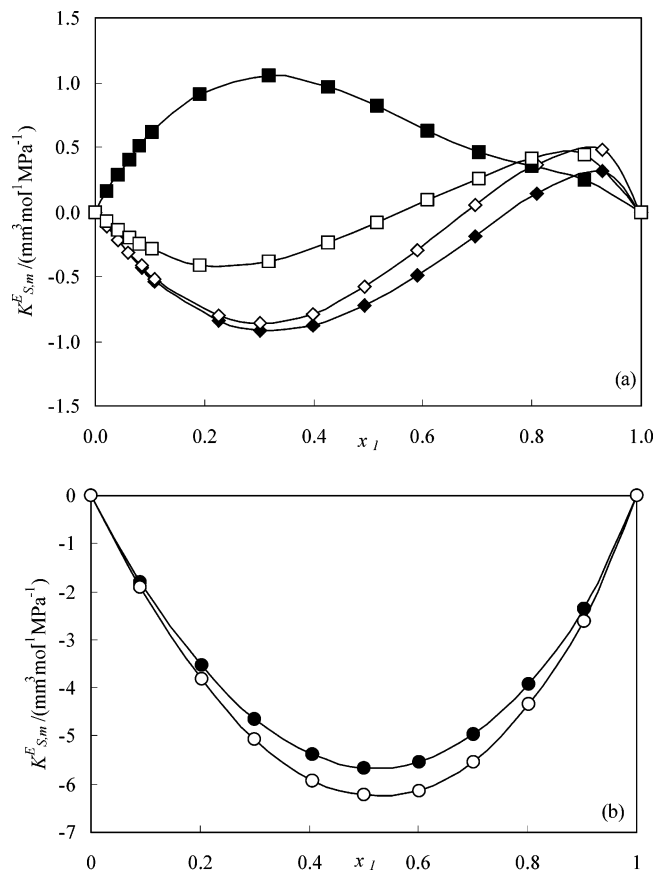
<sup>a</sup> Equation 8. <sup>b</sup> Equation 9.

concentrations of 2,4,6-trimethyl-1,3,5-trioxane. The observed larger negative and symmetrical  $K_{S,m}^E$  variation in the case of propylene carbonate may be attributed to specific dipolar interactions among mixing components, whereas sigmoidal variation observed for dimethyl carbonate and diethyl carbonate mixtures suggest some specific interactions in lower concentrations of 2,4,6-trimethyl-1,3,5-trioxane and dispersive interactions in higher concentrations of 2,4,6-trimethyl-1,3,5-trioxane. The values of  $K_{S,m}^E$  decreases with increasing temperature in the case of diethyl carbonate and propylene carbonate, whereas the reverse is observed in the case of dimethyl carbonate. The  $K_{S,m}^E$  values at equimolar concentration follow the order propylene carbonate < dimethyl carbonate < diethyl carbonate.

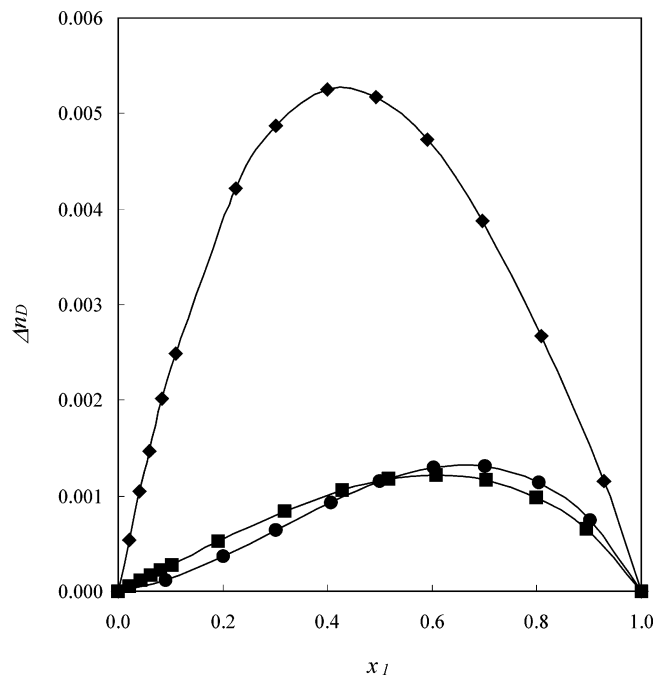
Figure 3 shows the graphical variations of  $\Delta n_D$  against  $x_1$  for 2,4,6-trimethyl-1,3,5-trioxane at (298.15) K. The values of



**Figure 1.** Variation of viscosity deviations ( $\delta\eta$ ) for binary mixtures of 2,4,6-trimethyl-1,3,5-trioxane with dimethyl carbonate ( $\blacklozenge$ ,  $\diamond$ ), diethyl carbonate ( $\blacksquare$ ,  $\square$ ), and propylene carbonate ( $\bullet$ ,  $\circ$ ) at (288.15 (filled symbols) and 298.15 (open symbols)) K.



**Figure 2.** Variation of the excess molar isentropic compressibility ( $K_{S,m}^E$ ) for binary mixtures: (a) 2,4,6-trimethyl-1,3,5-trioxane with dimethyl carbonate ( $\blacklozenge$ ,  $\diamond$ ) and diethyl carbonate ( $\blacksquare$ ,  $\square$ ) and (b) 2,4,6-trimethyl-1,3,5-trioxane with propylene carbonate ( $\bullet$ ,  $\circ$ ) at (288.15 (filled symbols) and 298.15 (open symbols)) K.



**Figure 3.** Variation of refractive index deviations ( $\Delta n_D$ ) for binary mixtures of 2,4,6-trimethyl-1,3,5-trioxane with dimethyl carbonate ( $\blacklozenge$ ), diethyl carbonate ( $\blacksquare$ ), and propylene carbonate ( $\bullet$ ) at (298.15 K).

$\Delta n_D$  are found to be positive over the entire range of composition for all of the studied systems. The  $\Delta n_D$  values for the 2,4,6-trimethyl-1,3,5-trioxane + propylene carbonate mixture are

larger and more positive and symmetric than those observed for 2,4,6-trimethyl-1,3,5-trioxane + dimethyl carbonate and diethyl carbonate. Also, for the 2,4,6-trimethyl-1,3,5-trioxane + dimethyl carbonate or diethyl carbonate mixture, positive  $\Delta n_D$  values are more or less identical as seen in Figure 3. The maxima of  $\Delta n_D$  values for the propylene carbonate system occur at 0.5 mole fraction, whereas that for the dimethyl carbonate and diethyl carbonate system occurs at 0.7 mole fraction.

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