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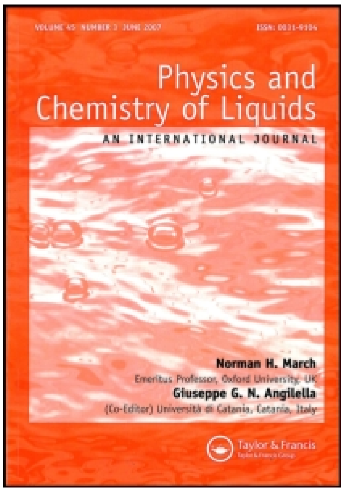

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Densities and viscosities of propylene carbonate with aromatic hydrocarbons (benzene, 1,4-dimethylbenzene and ethylbenzene) at 288.15, 298.15 and 308.15 K

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Densities and viscosities of propylene carbonate with aromatic hydrocarbons (benzene, 1,4-dimethylbenzene and ethylbenzene) at 288.15, 298.15 and 308.15 K

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Densities and viscosities of the binary mixtures of propylene carbonate with benzene, 1,4-dimethylbenzene and ethylbenzene were measured over the entire mole fraction range at 288.15, 298.15 and 308.15 K. Using the experimental values of densities and viscosities the excess molar volumes and viscosity deviations were calculated. The results were fitted by Redlich–Kister equation. The results were discussed in terms of intermolecular interactions.

Keywords: Densities; Viscosities; Excess molar volumes; viscosity deviations; Redlich–Kister equation

1. Introduction

In continuation to our study on thermodynamic properties of liquid mixtures containing propylene carbonate (PC), as one of the binaries [1–2], we report here the densities and viscosities of binary mixtures of (PC) with benzene, 1,4-dimethylbenzene and ethylbenzene at 288.15, 298.15 and 308.15 K and at atmospheric pressure. Densities and viscosities of binary mixtures are important from the theoretical point of view to understand the liquid theory. Knowledge of densities and viscosities are required for the designing of new processes and the latter for evaluation of mass-transfer phenomena and designing equipment.

In this work, we report densities, viscosities, excess molar volumes and viscosity deviations for the aforementioned systems at 288.15, 298.15 and 308.15 K and at atmospheric pressure. The results obtained were fitted to Redlich–Kister equation and are discussed in the light of molecular interactions going on among the components of mixtures.

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Table 1. Values of densities (ρ) and viscosities (η) of various pure liquids at 298.15 K.

	Density ρ (g cm^{-3})		Viscosity η (mPa s)	
Liquid	Obs.	Liter.	Obs.	Liter.
Propylene carbonate	1.19758	1.1970 [3]	2.4997	2.5009 [3]
Benzene	0.87355	0.8735 [4]	0.6126	0.6026 [4]
1,4-dimethyl benzene	0.85676	0.85670 [4]	0.6050	0.6078 [4]
Ethylbenzene	0.86248	0.86253 [5]	0.6367	0.6373 [5]

2. Experimental

Propylene carbonate (Merck, >99) was refluxed over anhydrous calcium carbonate and distilled at atmospheric pressure. Benzene, 1,4-dimethylbenzene and ethylbenzene (all of S.D. Fine Chem, Pvt. Ltd.) were distilled at atmospheric pressure. All the liquids were double distilled. The middle fraction collected of all the liquids was stored over 4Å molecular sieve. All the liquids were degassed before use. The purity of the purified liquids was checked by comparing the measured densities and viscosities with those reported in the literature [3–5]. The experimental and literature values are as shown in table 1. These values agreed well within the precision of experimental error.

The binary liquid mixtures were prepared by mixing known masses of pure liquids in airtight-stoppered bottles in order to minimize the evaporation losses. All measurements of mass were performed on a Mettler one pan balance (E-Mettler, Zurich), which can read up to five decimal places, with an accuracy of ±0.05 mg.

The densities of the pure liquids and their binary mixtures were measured using the single arm capillary pyknometer. The accuracy in the density measurements was found to be of $\pm 5 \times 10^{-5}\,\mathrm{g\,cm^{-3}}$.

Viscosity measurements were performed by using Schott Gerate (AVS 350) viscosity measuring equipment with a series of Ubbelhode 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. Several viscometers were used, with capillary diameters from 0.36 to 1.13 mm, for kinematic ranges of 0.3 to $10\,\mathrm{mm^2\,s^{-1}}$, respectively. In all the measurements, kinetic energy corrections have been taken into account. The experimental repeatability of the viscosity measurements of each solution was ±0.2%. The details of the density and viscosity measurements are as per our previous articles [1,2,6].

For all the measurements, temperature was controlled by circulating the water through an ultra thermostat JULABO F-25 (made in Germany) which has an accuracy of ±0.02°C.

3. Results and discussion

The experimental values of densities (ρ) and viscosities (η) of pure PC with aromatic hydrocarbons and their binary mixtures at 288.15, 298.15 and 308.15 K over the whole composition range are listed in table 2.

Table 2. Densities (ρ), visceosities (η), excess molar volumes (V^E) and viscosity deviations ($\Delta\eta$) of binary liquid mixtures at 288.15, 298.15 and 308.15 K.

x_1	ρ (g cm^{-3})	η (mPa s)	V^E (cm^3 mol^{-1})	$\Delta\eta$ (mPa s)
Benzene				
$T = 288.15$ K				
0.0000	0.88377	0.6510	0.000	0.000
0.0191	0.89016	0.6638	−0.045	−0.034
0.0416	0.89765	0.6793	−0.092	−0.073
0.0585	0.90327	0.6915	−0.125	−0.102
0.0771	0.90945	0.7054	−0.160	−0.133
0.1030	0.91804	0.7255	−0.205	−0.176
0.1982	0.94951	0.8111	−0.338	−0.323
0.2949	0.98134	0.9211	−0.426	−0.448
0.3972	1.01482	1.0702	−0.471	−0.548
0.4938	1.04627	1.2490	−0.474	−0.605
0.5987	1.08024	1.4930	−0.438	−0.616
0.7002	1.11290	1.7861	−0.367	−0.570
0.7907	1.14189	2.1013	−0.276	−0.475
0.9024	1.17748	2.5689	−0.133	−0.280
1.0000	1.20867	3.0865	0.000	0.000
$T = 298.15$ K				
0.0000	0.87355	0.6126	0.000	0.000
0.0191	0.88012	0.6193	−0.068	−0.028
0.0416	0.88779	0.6341	−0.137	−0.055
0.0585	0.89354	0.6458	−0.186	−0.074
0.0771	0.89985	0.6595	−0.237	−0.095
0.1030	0.90861	0.6758	−0.303	−0.126
0.1982	0.94062	0.7426	−0.497	−0.234
0.2949	0.97282	0.8478	−0.625	−0.307
0.3972	1.00649	0.9876	−0.691	−0.355
0.4938	1.03795	1.1450	−0.695	−0.375
0.5987	1.07172	1.3458	−0.641	−0.367
0.7002	1.10399	1.5711	−0.537	−0.328
0.7907	1.13247	1.7995	−0.404	−0.266
0.9024	1.16721	2.1188	−0.195	−0.152
1.0000	1.19758	2.4997	0.000	0.000
$T = 308.15$ K				
0.0000	0.86348	0.5237	0.000	0.000
0.0191	0.87074	0.5377	−0.144	−0.016
0.0416	0.87861	0.5545	−0.236	−0.034
0.0585	0.88462	0.5677	−0.315	−0.047
0.0771	0.89100	0.5824	−0.374	−0.061
0.1030	0.89977	0.6037	−0.444	−0.080
0.1982	0.93213	0.6908	−0.679	−0.140
0.2949	0.96453	0.7937	−0.828	−0.187
0.3972	0.99833	0.9193	−0.905	−0.220
0.4938	1.02992	1.0546	−0.917	−0.234
0.5987	1.06366	1.2211	−0.853	−0.230
0.7002	1.09590	1.4024	−0.739	−0.206
0.7907	1.12432	1.5818	−0.593	−0.167
0.9024	1.15822	1.8262	−0.309	−0.096
1.0000	1.18716	2.0731	0.000	0.000
1,4-Dimethylbenzene				
$T = 288.15$ K				
0.0000	0.86530	0.6787	0.000	0.000
0.0227	0.87726	0.6847	−0.910	−0.049
0.0397	0.88608	0.6902	−1.542	−0.084
0.0573	0.89508	0.6968	−2.153	−0.120
0.0816	0.90735	0.7077	−2.932	−0.168

(*Continued*)

Table 2. Continued.

x_1	ρ (g cm^{-3})	η (mPa s)	V^E (cm^3 mol^{-1})	$\Delta\eta$ (mPa s)
0.0980	0.91545	0.7160	−3.412	−0.199
0.2090	0.96776	0.7985	−5.880	−0.383
0.3029	1.00822	0.9063	−7.038	−0.502
0.3993	1.04620	1.0574	−7.506	−0.583
0.5028	1.08292	1.2680	−7.328	−0.621
0.5978	1.11294	1.5079	−6.638	−0.610
0.7068	1.14307	1.8398	−5.315	−0.541
0.8012	1.16540	2.1763	−3.768	−0.432
0.8996	1.18496	2.5751	−1.804	−0.270
1.0000	1.20867	3.0865	0.000	0.000
$T = 298.15$ K				
0.0000	0.85676	0.6050	0.000	0.000
0.0227	0.86868	0.6155	−0.928	−0.033
0.0397	0.87747	0.6246	−1.573	−0.056
0.0573	0.88643	0.6333	−2.990	−0.115
0.0980	0.90672	0.6527	−3.480	−0.138
0.2090	0.95879	0.7280	−5.993	−0.273
0.3029	0.99904	0.8062	−7.171	−0.373
0.3993	1.03679	0.9317	−7.646	−0.430
0.5028	1.07324	1.1162	−7.462	−0.441
0.5978	1.10301	1.3161	−6.759	−0.422
0.7068	1.13283	1.5840	−5.409	−0.360
0.8012	1.15487	1.8558	−3.832	−0.267
0.8996	1.17412	2.1490	−1.832	−0.160
1.0000	1.19758	2.4997	0.000	0.000
$T = 308.15$ K				
0.0000	0.84792	0.5330	0.000	0.000
0.0227	0.85980	0.5443	−0.947	−0.024
0.0397	0.86856	0.5534	−1.605	−0.041
0.0573	0.87750	0.5634	−2.240	−0.058
0.0816	0.88968	0.5779	−3.049	−0.081
0.0980	0.89772	0.5884	−3.549	−0.096
0.2090	0.94961	0.6727	−6.109	−0.182
0.3029	0.98970	0.7639	−7.307	−0.236
0.3993	1.02728	0.8783	−7.788	−0.270
0.5028	1.06356	1.0257	−7.598	−0.282
0.5978	1.09316	1.1840	−6.878	−0.270
0.7068	1.12278	1.3935	−5.501	−0.228
0.8012	1.14466	1.5986	−3.893	−0.168
0.8996	1.16373	1.8353	−1.855	−0.083
1.0000	1.18716	2.0731	0.000	0.000
Ethylbenzene				
$T = 288.15$ K				
0.0000	0.87136	0.7116	0.000	0.000
0.0294	0.88303	0.7222	−0.648	−0.059
0.0420	0.88801	0.7274	−0.908	−0.084
0.0729	0.90011	0.7417	−1.498	−0.143
0.0821	0.90371	0.7464	−1.663	−0.160
0.1013	0.91113	0.7569	−1.987	−0.195
0.2023	0.94952	0.8299	−3.336	−0.362
0.3008	0.98570	0.9333	−4.133	−0.493
0.4012	1.02129	1.0771	−4.493	−0.587
0.5001	1.05511	1.2624	−4.458	−0.637
0.6001	1.08801	1.4998	−4.082	−0.637
0.7001	1.11967	1.7936	−3.404	−0.581
0.8009	1.15029	2.1528	−2.450	−0.461
0.8992	1.17892	2.5699	−1.286	−0.277
1.0000	1.20867	3.0865	0.000	0.000

(*Continued*)

Table 2. Continued.

x_1	ρ (g cm^{-3})	η (mPa s)	V^E (cm^3 mol^{-1})	$\Delta\eta$ (mPa s)
$T = 298.15$ K				
0.0000	0.86248	0.6367	0.000	0.000
0.0294	0.87412	0.6462	−0.665	−0.045
0.0420	0.87909	0.6512	−0.932	−0.064
0.0729	0.89117	0.6609	−1.539	−0.112
0.0821	0.89476	0.6655	−1.708	−0.124
0.1013	0.90216	0.6735	−2.040	−0.152
0.2023	0.94045	0.7164	−3.425	−0.297
0.3008	0.97650	0.8062	−4.243	−0.391
0.4012	1.01194	0.9344	−4.612	−0.450
0.5001	1.04558	1.0980	−4.577	−0.470
0.6001	1.07829	1.3015	−4.194	−0.453
0.7001	1.10972	1.5438	−3.501	−0.397
0.8009	1.14010	1.8268	−2.526	−0.302
0.8992	1.16846	2.1397	−1.338	−0.172
1.0000	1.19758	2.4997	0.000	0.000
$T = 308.15$ K				
0.0000	0.85348	0.5484	0.000	0.000
0.0294	0.86510	0.5585	−0.682	−0.035
0.0420	0.87006	0.5634	−0.955	−0.049
0.0729	0.88210	0.5767	−1.575	−0.083
0.0821	0.88569	0.5811	−1.748	−0.092
0.1013	0.89306	0.5908	−2.088	−0.112
0.2023	0.93124	0.6552	−3.504	−0.202
0.3008	0.96716	0.7411	−4.337	−0.266
0.4012	1.00245	0.8533	−4.711	−0.307
0.5001	1.03592	0.9894	−4.672	−0.322
0.6001	1.06843	1.1536	−4.275	−0.310
0.7001	1.09966	1.3454	−3.562	−0.270
0.8009	1.12981	1.5669	−2.562	−0.203
0.8992	1.15794	1.8106	−1.344	−0.109
1.0000	1.18716	2.0731	0.000	0.000

Excess molar volume V^E was calculated [7,8] from the measured densities (ρ) by using equation (1),

$$V^E = \frac{x_1 M_1 + x_2 M_2}{(\rho)} - \left(\frac{x_1 M_1}{(\rho_1)} + \frac{x_2 M_2}{(\rho_2)}\right) \tag{1}$$

where ρ is the density of the mixture and M_1, x_1, (ρ_1) and M_2, x_2, (ρ_2) are the molecular mass, mole fractions, and densities of pure PC (1) and aromatic hydrocarbons (2), respectively. The uncertainty in the measurement of V^E was found to be ± 0.003.

Dynamic viscosities (η) of binary mixtures of PC (1) + aromatic hydrocarbons (2) were calculated by using densities and flow times by equation (2),

$$\eta = K \cdot \rho \cdot (t - \mathrm{HC}) \tag{2}$$

where K and ρ are viscometer constant and density of the mixture, respectively. $(t - \mathrm{HC})$ is the flow time adjusted by the Hagenbach correction factor and,

$$\mathrm{HC} = \frac{E}{K \cdot t^2}.$$

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. The viscometers with $t > 200\,\mathrm{s}$ were selected for measurements.

The viscosity deviations of binary mixtures were calculated [9,10] by using equation (3),

$$\Delta\eta = \eta - (x_1\eta_1 + x_2\eta_2) \tag{3}$$

where η is the viscosity of the mixture, η_1, η_2, x_1 and x_2 is the viscosity of pure components 1 and 2, mole fraction of components 1 and 2, respectively.

The calculated excess molar volumes (V^{E}) and viscosity deviations ($\Delta\eta$) were correlated by Redlich–Kister polynomial [11] as shown in equation (4),

$$\Delta Y = x_1 x_2 \sum a_i (x_1 - x_2)^i. \tag{4}$$

The coefficients in equation (4) were estimated by the least squares fit method and the standard deviation was calculated by equation (5),

$$\sigma = \left[\sum \frac{(\Delta Y_{\mathrm{Experimental}} - \Delta Y_{\mathrm{Calculated}})^2}{D - N}\right]^{0.5} \tag{5}$$

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

Regression results for excess molar volumes and viscosity deviations of binary mixtures of PC (1) and aromatic hydrocarbons (2) are as shown in table 3.

Figure 1 shows the graphical variation of the excess molar volumes V^{E} with mole fractions x_1 of PC at 298.15 K (figures at 288.15 and 308.15 K are not shown). In the present study, the V^{E} values are found to be negative over the whole composition range, for all the systems studied and at the said temperatures. The negative values increase as the temperature increases for all the studied systems. The graphical variations are found to be symmetric in nature and the minima occurs at 0.4 M fraction for all the systems. The minima of V^{E} values at 298.15 K vary from $-0.695\,\mathrm{cm^3\,mol^{-1}}$ for benzene to $-7.646\,\mathrm{cm^3\,mol^{-1}}$ for 1,4-dimethylbenzene. The V^{E} values follow the trend as benzene < ethylbenzene < 1,4-dimethylbenzene.

It has been reported [12] earlier that V^{E} values are resultant of the chemical, physical and structural characteristics of liquids. The difference in size and shape of the component molecules and loss of dipolar association lead to expansion whereas physical interaction between unlike molecules such as donor–acceptor and dipole–dipole interactions lead to contraction in volume. In the present study, the negative V^{E} values indicate existence of specific interaction between the mixing components. The introduction of methyl or ethyl group in the ring increases the electron donor–acceptor interactions, hence 1,4-dimethylbenzene and ethylbenzene have the more negative excess molar volumes than benzene.

Figure 2 shows the graphical variations of $\Delta\eta$ with mole fractions x_1 of PC with aromatic hydrocarbons at 298.15 K. The viscosity deviations are also negative over the entire composition range, for all the systems studied and at the said temperatures. As the temperature increases the negative values of $\Delta\eta$ decreases. The viscosity deviation values show no specific trend regarding aromatic hydrocarbons at 288.15 K (figure not shown) and the values are little bit mixed. But as the temperature increases, a specific trend is observed at 298.15 and 308.15 K (figure at 308.15 K is not shown). The minima

Table 3. Coefficients of equation (5) and standard deviations σ (ΔY) for the binary mixtures.

T	a_0	a_1	a_2	σ
PC + Benzene				
V^E (cm^3 mol^{-1})				
288.15	−1.894	0.397	0.000	0.002
298.15	−2.773	0.597	−2.741	0.004
308.15	−3.598	0.707	−1.152	0.023
$\Delta\eta$ (mPa s)				
288.15	−2.447	−0.780	0.104	0.025
298.15	−1.508	−0.148	−0.044	0.004
308.15	−0.936	−0.114	−0.039	0.001
PC + 1,4-dimethylbenzene				
V^E (cm^3 mol^{-1})				
288.15	−29.396	10.297	−0.811	0.061
298.15	−29.937	10.590	−0.832	0.063
308.15	−30.481	10.818	−0.828	0.065
$\Delta\eta$ (mPa s)				
288.15	−2.478	−0.348	−0.128	0.005
298.15	−1.780	−0.029	0.265	0.006
308.15	−1.131	0.048	0.157	0.003
PC + Ethylbenzene				
V^E (cm^3 mol^{-1})				
288.15	−17.843	4.489	−0.505	0.014
298.15	−18.317	4.541	−0.630	0.012
308.15	−18.697	4.571	−0.552	0.015
$\Delta\eta$ (mPa s)				
288.15	−18.126	3.589	2.577	0.145
298.15	−1.893	−0.080	−0.163	0.004
308.15	−1.289	−0.012	0.087	0.001

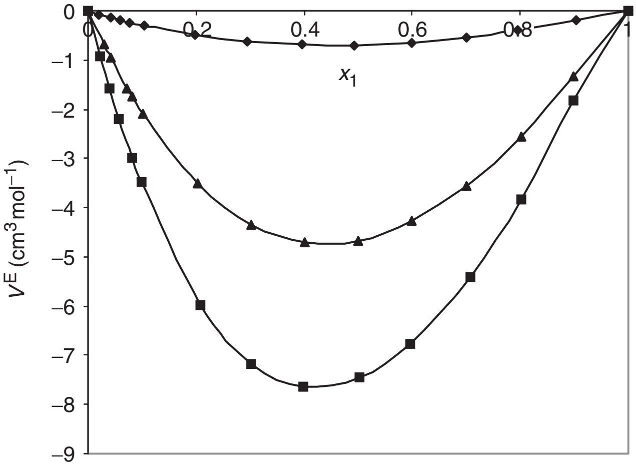

Figure 1. Variation of excess molar volume, V^E for binary mixtures of propylene carbonate with benzene (◆), 1,4-dimethylbenzene (■) and ethylbenzene (▲) at 298.15 K.

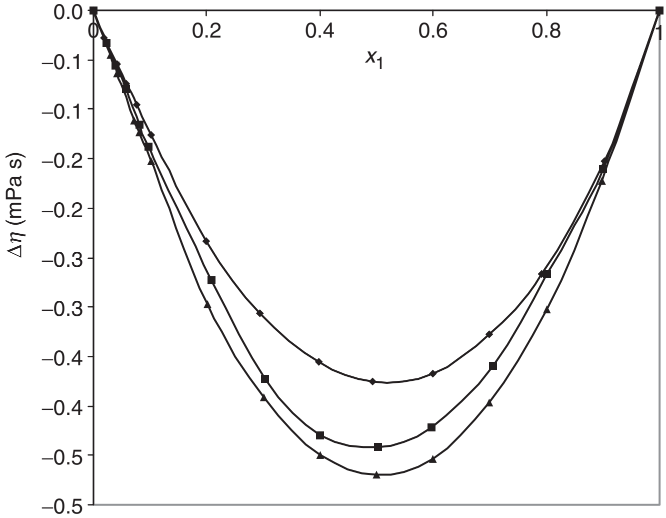

Figure 2. Variation of viscosity deviations, $\Delta\eta$ for binary mixtures of propylene carbonate with benzene (◆), 1,4-dimethylbenzene (■) and ethylbenzene (▲) at 298.15 K.

of $\Delta\eta$ values at 298.15 K vary from −0.375 mPa s for benzene to −0.470 mPa s for ethylbenzene. The plot of $\Delta\eta$ values at equimolar concentration follows the order benzene < 1,4-dimethylbenzene < ethylbenzene at 298.15 K.

For the systems where dispersion, induction and dipolar forces are operating, the values of viscosity deviations are found to be negative, whereas the existence of specific interactions between the mixing components of the various binary systems tends to make viscosity deviations positive. Furthermore, the strength of intermolecular electron donor–acceptor interaction, the molecular size and shape and average degree of association of the mixture are contributing to the viscosity deviations [13]. The negative viscosity deviations can thus be attributed to interstitial accommodation of one component into other due to differences in molecular size of mixing components.

4. Conclusion

New experimental values of density and viscosity for the systems of binary mixtures of PC + benzene, 1,4-dimethylbenzene, ethylbenzene at 288.15, 298.15 and 308.15 K and different compositions are measured. The excess molar volumes and viscosity deviations were correlated using Redlich–Kister polynomial equation. Excellent agreements were observed. The excess molar volumes were negative for all binary mixtures at all temperatures and over the entire range of compositions and negative values increased with rising temperature from 288.15 to 308.15 K. The viscosity deviations are also negative and negative values decrease with increasing temperature. The results are discussed in terms of molecular interactions.

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