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Densities, Viscosities and Related Properties for Binary Mixtures of Sulfolane + *p*-Xylene, Sulfolane + Ethylbenzene in the Temperature Range from 303.15 K to 353.15 K

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Abstract Densities and viscosities of the binary systems of sulfolane + ethylbenzene, sulfolane + *p*-xylene have been experimentally determined in temperature interval 303.15—353.15 K and at atmospheric pressure for the whole composition range. The excess molar volumes and viscosity deviations were computed. The computed quantities have been fitted to Redlich-Kister equation. Excess molar volumes and viscosity deviation show a systematic change with increasing temperature. Two mixtures exhibit negative excess volumes with a minimum which occurs approximately at $x = 0.5$. The effect of the size, shape and interaction of components on excess molar volumes and viscosity deviations is discussed.
Keywords density, viscosity, excess molar volume, sulfolane

1 INTRODUCTION

Sulfolane is a high-boiling solvent with excellent extraction properties and is widely used in the extraction of aromatics from refinery process streams. Thermodynamic and transport properties provide important information for design of the molecular interactions existing in liquid mixtures.

Chen and Knapp[1] measured the densities and excess molar volumes for sulfolane + ethylbenzene at 283.15 K and 313.15 K. Yu *et al.*[2] reported densities of binary systems of sulfolane with aromatic hydrocarbons at 298.15 K. No report is available in the literature on viscosity of two binary liquid systems to be studied.

In this work, we present the densities, viscosities of systems for sulfolane + *p*-xylene and sulfolane + ethylbenzene in the temperature range from 303.15 K to 358.15 K and of mixture compositions over the whole range of compositions.

From these data, excess molar volume, V^{E}, and viscosity deviation, $\Delta\eta$ have been calculated. These results are fitted to the Redlich-Kister polynomial equation to estimate the adjustable parameter and standard deviations between the calculated and experimental results. The calculated results are discussed in terms of the binary intersection and the effect of alkane chain length in addition to their shapes.

2 EXPERIMENTAL SECTION

2.1 Materials

Ethylbenzene is analytical reagent and sulfolane is chemical reagent (all from Tianjin Reagent Company). *p*-Xylene was supplied by Tianjin Petroleum Engineering Company. All chemicals were purified by distillation and their middle fraction were collected. Then the liquids were dried over 0.4 nm molecular sieves. The purities of *p*-xylene and ethylbenzene were better than 99.5% (by mass), checked by gas chromatograph. The purities of the sulfolane were confirmed by comparing the densities and viscosities with those reported in the literature, as shown in Table 1, aromatic hydrocarbons were further ascertained by the same method. The data agree with the published results.

2.2 Apparatus and procedures

Binary mixtures were prepared by mass on an analytical BP 210 S balance with ±0.01 mg accuracy. The possible error in the mole fraction is estimated to be around ±0.0001.

The densities of the pure components and the binary mixtures were measured by means of vibrating tube densimeter (Density/Specific Gravity Meter DA 505, KEM, Japan) in a thermostat at ±0.01 K. The accuracy of density measurements was $\pm 1 \times 10^{-5}$ g·cm^{-3}. The calibration was carried out with both deionized double-distilled water and dry air.

Viscosity was measured using an Ubbelohde suspended-level viscometer, calibrated with double-distilled water. An electronic digital stopwatch with a readability of ±0.01 s was used for flow time measurement. Experiments were repeated at least four times at each temperature for all compositions, and the results were averaged. The viscosity η of the liquid was then calculated from the following relationship

$$\eta/\rho = k(t - \theta) \tag{1}$$

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Table 1 Comparison of experimental and literature values[2−11] of densities, ρ, and viscosities, η, for pure compounds

Liquid	T, K	ρ, g·cm^{-3}		η, mPa·s	
		exp.	lit.	exp.	lit.
sulfolane	298.15	1.26540	1.26654[2]	10.0742	10.356[5]
			1.2640[3]		10.284[6]
	303.15	1.26080	1.26224[4]		
			1.26184[5]		
	313.15	1.25190	1.2532[1]		
	323.15	1.24336			
	333.15	1.23462	1.2359[1]		
p-xylene	298.15	0.85670	0.85648[1]	0.6078	0.6045[8]
			0.8565[8]		0.6031[9]
			0.85672[11]		0.6083[10]
	303.15	0.85235	0.85227[7]	0.5715	0.567[7]
			0.8522[8]		0.5694[8]
					0.5762[11]
	313.15	0.84364	0.8434[8]	0.5080	0.5064[8]
			0.85239[11]		
	323.15	0.83490		0.4599	0.4580[9]
ethylbenzene	298.15	0.86433	0.86289[2]	0.6298	0.6293[9]
	303.15	0.85995	0.86454[10]	0.5981	0.5923[9]
					0.5976[10]
	313.15	0.85114	0.8496[1]	0.5368	0.5369[10]
			0.85976[10]		
	323.15	0.84227	0.85471[10]	0.4829	0.4797[9]
					0.4852[10]
	333.15	0.83334	0.8318[1]	0.4370	0.4410[10]
			0.84975[10]		

where t is the flow time, η is the kinetic viscosity, k and θ are the viscometer constant and the Hagenbach correction factor respectively.

The overall uncertainty of viscosity measurements is dependent on the equilibrium stability of the viscometer, the time of flow, and the change of concentration, which are in the order of 1×10^{-2}, 1×10^{-2}, and 1×10^{-4}, respectively; viscosity values are accurate to within ±0.0003 mPa·s. All measurements were carried out in a well-stirred water bath with temperature controlled to within ±0.01 K.

3 RESULTS AND DISCUSSION

The densities of the pure components and their mixtures were used to calculate the excess molar volumes V^E, as the following equation[12,13] indicates

$$V^E = \frac{x_1M_1 + x_2M_2}{\rho} = \frac{x_1M_1}{\rho_1} - \frac{x_2M_2}{\rho_2} \quad (2)$$

where ρ is the density of the mixture and x_1, ρ_1, M_1; x_2, ρ_2 and M_2 are the mole fraction, densities, and molecular weight of pure components 1 and 2, respectively.

The viscosity deviation $\Delta\eta$ was obtained from dynamic viscosity and composition data through

$$\Delta\eta = \eta - (x_1\eta_1 + x_2\eta_2) \quad (3)$$

where η_{12}, η_1 and η_2 are viscosity of the mixture and those of pure components 1 and 2, respectively.

Table 2 shows the experimental densities, viscosities, excess molar volumes, and viscosity deviations for each binary mixture at working temperatures.

The V^E and $\Delta\eta$ values were fitted with composition x through least-squares to a Redlich-Kister type equation[14,15].

$$Y = x(1-x)\sum_{k=0}^{N} A_k(2x-1)^k \quad (4)$$

where $Y \equiv (V^E \text{ or } \Delta\eta)$. The fitting parameters A_k were obtained by the unweighted least-squares method. Table 3 shows the fitting parameters and the standard deviations for each of the binary mixtures at different temperatures.

As shown in Table 2, the excess molar volumes were negative over the entire composition range for both systems studied and at temperatures researched. The values of V^E for system of *p*-xylene + sulfolane are plotted in Fig. 1 and for system of ethylbenzene + sulfolane in Fig. 2. They are U-shaped, nearly symmetrical, composition dependence with increase in absolute values with increase of temperature.

Table 2 Densities(ρ), viscosities(η), excess molar volumes(V^E), and viscosities deviations($\Delta\eta$) for the binary mixtures at several temperatures

x_1	ρ, g·cm^{-3}	η, mPa·s	V^E, cm^3·mol^{-1}	$\Delta\eta$, mPa·s	x_1	ρ, g·cm^{-3}	η, mPa·s	V^E, cm^3·mol^{-1}	$\Delta\eta$, mPa·s
(x)*p*-xylene + (1 − x) sulfolane					(x)ethylbenzene+(1 − x) sulfolane				
		T = 303.15 K					T = 303.15 K		
0.0000	1.26080	10.0742	0.0000	0.0000	0.0000	1.26080	10.0742	0.0000	0.0000
0.1118	1.20785	6.1562	−0.3872	−2.8556	0.1118	1.20927	6.2242	−0.3794	−2.7906
0.2206	1.15809	4.0927	−0.6653	−3.8852	0.2205	1.16040	4.1551	−0.6197	−3.8296
0.3266	1.11070	2.8874	−0.7880	−4.0832	0.3267	1.11466	2.9424	−0.8023	−4.0360
0.4301	1.06673	2.1432	−0.8835	−3.8439	0.4301	1.07165	2.1852	−0.9014	−3.8133
0.5310	1.02522	1.6081	−0.8799	−3.4201	0.5309	1.03119	1.5364	−0.9279	−3.5069
0.6293	0.98650	1.2525	−0.8342	−2.8416	0.6293	0.99333	1.3237	−0.9176	−2.7872
0.7252	0.95004	0.9893	−0.7203	−2.1936	0.7254	0.95717	1.0567	−0.7932	−2.1436
0.8191	0.91582	0.8122	−0.5754	−1.4784	0.8194	0.92309	0.8613	−0.6215	−1.4482
0.9106	0.88335	0.6727	−0.3392	−0.7483	0.9106	0.89045	0.7155	−0.3064	−0.7298
1.0000	0.85235	0.5715	0.0000	0.0000	1.0000	0.85995	0.5981	0.0000	0.0000
		T = 313.15 K					T = 313.15 K		
0.0000	1.25190	7.8084	0.0000	0.0000	0.0000	1.25190	7.8084	0.0000	0.0000
0.1118	1.19886	4.8829	−0.3964	−2.1093	0.1118	1.20044	4.9431	−0.4027	−2.0524
0.2206	1.14906	3.3411	−0.6826	−2.8568	0.2205	1.15153	3.3851	−0.6524	−2.8199
0.3266	1.10169	2.3845	−0.8131	−3.0395	0.3267	1.10577	2.4521	−0.8422	−2.9807
0.4301	1.05774	1.7912	−0.9133	−2.8772	0.4301	1.06277	1.8346	−0.9471	−2.8463
0.5310	1.01627	1.3509	−0.9122	−2.5810	0.5309	1.02232	1.3157	−0.9756	−2.6323
0.6293	0.97756	1.0656	−0.8623	−2.1487	0.6293	0.98445	1.1351	−0.9617	−2.0973
0.7252	0.94122	0.8516	−0.7539	−1.6625	0.7254	0.94831	0.9235	−0.8324	−1.6101
0.8191	0.90695	0.7111	−0.5905	−1.1175	0.8194	0.91423	0.7568	−0.6498	−1.0932
0.9106	0.87450	0.5929	−0.3401	−0.5677	0.9106	0.88160	0.6317	−0.3196	−0.5552
1.0000	0.84364	0.5080	0.0000	0.0000	1.0000	0.85114	0.5368	0.0000	0.0000
		T = 323.15 K					T = 323.15 K		
0.0000	1.24336	6.1936	0.0000	0.0000	0.0000	1.24336	6.1936	0.0000	0.0000
0.1118	1.19021	3.9640	−0.4103	−1.5886	0.1118	1.19164	4.0068	−0.4055	−1.5483
0.2206	1.14037	2.7395	−0.7106	−2.1892	0.2205	1.14270	2.8007	−0.6703	−2.1337
0.3266	1.09299	2.0001	−0.8521	−2.3208	0.3267	1.09692	2.0479	−0.8719	−2.2801
0.4301	1.04903	1.5260	−0.9596	−2.2016	0.4301	1.0539	1.5611	−0.9839	−2.1763
0.5310	1.00758	1.1659	−0.9642	−1.9831	0.5309	1.01347	1.1363	−1.0193	−2.0255
0.6293	0.96888	0.9254	−0.9152	−1.6600	0.6293	0.97558	0.9927	−1.0043	−1.6071
0.7252	0.93240	0.7473	−0.7848	−1.2883	0.7254	0.93943	0.8125	−0.8697	−1.2385
0.8191	0.89826	0.6269	−0.6294	−0.8702	0.8194	0.90536	0.6653	−0.6804	−0.8490
0.9106	0.86585	0.5300	−0.3715	−0.4425	0.9106	0.87272	0.5639	−0.3350	−0.4295
1.0000	0.83490	0.4599	0.0000	0.0000	1.0000	0.84227	0.4829	0.0000	0.0000
		T = 333.15 K					T = 333.15 K		
0.0000	1.23462	5.0089	0.0000	0.0000	0.0000	1.23462	5.0089	0.0000	0.0000
0.1118	1.18139	3.2834	−0.4257	−1.2135	0.1118	1.18285	3.3304	−0.4246	−1.1674
0.2206	1.13151	2.3179	−0.7391	−1.6808	0.2205	1.13386	2.3504	−0.7026	−1.6504
0.3266	1.08410	1.7162	−0.8885	−1.7972	0.3267	1.08806	1.7631	−0.9157	−1.7521
0.4301	1.04015	1.3206	−1.0038	−1.7189	0.4301	1.04502	1.3471	−1.0344	−1.6954
0.5310	0.99870	1.0020	−1.0103	−1.5754	0.5309	1.00457	0.9945	−1.0718	−1.5872
0.6293	0.96001	0.8081	−0.9602	−1.3192	0.6293	0.96666	0.8757	−1.0546	−1.2561
0.7252	0.92352	0.6593	−0.8217	−1.0288	0.7254	0.93050	0.7242	−0.9135	−0.9682
0.8191	0.88939	0.5595	−0.6571	−0.6987	0.8194	0.89641	0.5966	−0.7121	−0.6660
0.9106	0.85698	0.4741	−0.3835	−0.3651	0.9106	0.86376	0.5051	−0.3493	−0.3406
1.0000	0.82609	0.4298	0.0000	0.0000	1.0000	0.83334	0.4370	0.0000	0.0000
		T = 343.15 K					T = 343.15 K		
0.0000	1.22592	4.1261	0.0000	0.0000	0.0000	1.22592	4.1261	0.0000	0.0000
0.1118	1.17257	2.7594	−0.4409	−0.9478	0.1118	1.17393	2.7956	−0.4331	−0.9135
0.2206	1.12264	1.9735	−0.7694	−1.3259	0.2205	1.12489	2.0052	−0.7271	−1.2985
0.3266	1.07522	1.4834	−0.9312	−1.4187	0.3267	1.07906	1.5407	−0.9532	−1.3670
0.4301	1.03124	1.1582	−1.0527	−1.3560	0.4301	1.03602	1.1769	−1.0827	−1.3451
0.5310	0.98980	0.8713	−1.0642	−1.2647	0.5309	0.99554	0.8824	−1.1230	−1.2638
0.6293	0.95110	0.7097	−1.0124	−1.0579	0.6293	0.95762	0.7720	−1.1064	−1.0072
0.7252	0.91451	0.5832	−0.8552	−0.8250	0.7254	0.92143	0.6463	−0.9575	−0.7745
0.8191	0.88046	0.4985	−0.6915	−0.5578	0.8194	0.88732	0.5358	−0.7446	−0.5343
0.9106	0.84806	0.4254	−0.4043	−0.2879	0.9106	0.85467	0.4588	−0.3663	−0.2712
1.0000	0.81717	0.3783	0.0000	0.0000	1.0000	0.82426	0.3966	0.0000	0.0000

Table 2(Continued)

x_1	ρ, g·cm^{-3}	η, mPa·s	V^E, cm^3·mol^{-1}	$\Delta\eta$, mPa·s	x_1	ρ, g·cm^{-3}	η, mPa·s	V^E, cm^3·mol^{-1}	$\Delta\eta$, mPa·s
(x)p-xylene + (1 − x) sulfolane					(x)ethylbenzene+(1 − x) sulfolane				
		T = 353.15 K					T = 353.15 K		
0.0000	1.21680	3.4397	0.0000	0.0000	0.0000	1.21680	3.4397	0.0000	0.0000
0.1118	1.16338	2.3518	−0.4553	−0.7412	0.1118	1.16514	2.3707	−0.4839	−0.7254
0.2206	1.11344	1.7069	−0.7976	−1.0486	0.2205	1.11608	1.7358	−0.7946	−1.0262
0.3266	1.06599	1.2945	−0.9649	−1.1322	0.3267	1.07018	1.3190	−1.0276	−1.1165
0.4301	1.02201	1.0262	−1.0905	−1.0795	0.4301	1.02710	1.0392	−1.1616	−1.0786
0.5310	0.98056	0.7690	−1.0994	−1.0237	0.5309	0.98658	0.7850	−1.2009	−1.0229
0.6293	0.94185	0.6261	−1.0403	−0.8618	0.6293	0.94863	0.6923	−1.1795	−0.8132
0.7252	0.90559	0.5194	−0.9155	−0.6710	0.7254	0.91240	0.5830	−1.0183	−0.6271
0.8191	0.87121	0.4475	−0.6920	−0.4517	0.8194	0.87828	0.4841	−0.7920	−0.4371
0.9106	0.83881	0.3843	−0.3833	−0.2312	0.9106	0.84558	0.4166	−0.3873	−0.2243
1.0000	0.80822	0.3382	0.0000	0.0000	1.0000	0.81518	0.3661	0.0000	0.0000

Table 3 Parameters (A_k) of Eq. (4) and standard deviation for V^E (cm^3·mol) and $\Delta\eta$ (mPa·s) for the binary systems p-xylene + sulfolane and ethylbenzene + sulfolane

T, K		A_0	A_1	A_2	A_3	δ
		(x) p-xylene + (1 − x) sulfolane				
303.15	V^E	−3.5369	0.1791	−0.7713	−0.4733	0.0059
	$\Delta\eta$	−14.140	9.4076	−8.0719	5.0055	0.0150
313.15	V^E	−3.6740	0.0938	−0.6640	−0.2703	0.0056
	$\Delta\eta$	−10.617	6.7157	−5.7863	3.8577	0.0161
323.15	V^E	−3.8578	0.1279	−0.7468	−0.5553	0.0065
	$\Delta\eta$	−8.1748	5.0892	−4.2559	2.6893	0.0078
333.15	V^E	−4.0437	0.0840	−0.7010	−0.4845	0.0069
	$\Delta\eta$	−6.4277	3.7558	−3.1808	1.9419	0.0092
343.15	V^E	−4.2461	0.0926	−0.6897	−0.6012	0.0082
	$\Delta\eta$	−5.1289	2.8631	−2.3557	1.5954	0.0077
353.15	V^E	−4.4395	−0.0883	−0.3856	0.0613	0.0073
	$\Delta\eta$	−4.1389	2.1698	−1.7072	1.2819	0.0068
		(x) ethylbenzene + (1 − x) sulfolane				
303.15	V^E	−3.7657	−0.7559	−0.1804	1.1819	0.0109
	$\Delta\eta$	−14.118	9.4601	−7.3385	4.4524	0.0291
313.15	V^E	−3.9522	−0.7944	−0.2055	1.3111	0.0110
	$\Delta\eta$	−10.546	6.8049	−5.2820	3.2852	0.0245
323.15	V^E	−4.1236	−0.8674	−0.1115	1.2610	0.0117
	$\Delta\eta$	−8.0921	5.1174	−3.8671	2.3475	0.0197
333.15	V^E	−4.3356	−0.9150	−0.0757	1.3450	0.0122
	$\Delta\eta$	−6.3237	3.9556	−2.7317	1.4275	0.0154
343.15	V^E	−4.5432	−0.9813	0.0188	1.3220	0.0123
	$\Delta\eta$	−5.0206	2.9786	−2.0767	1.2480	0.0133
353.15	V^E	−4.8554	−0.9838	−0.1152	1.5755	0.0132
	$\Delta\eta$	−4.0576	2.3871	−1.5991	0.8257	0.0105

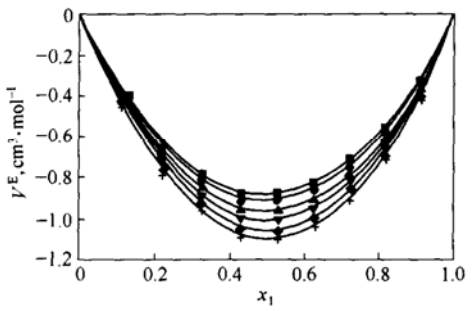

Figure 1 Excess molar volume variation with mole fraction for p-xylene (1) + sulfolane (2)
T, K: ■ 303.15; ● 313.15; ▲ 323.15; ▼ 333.15; ◆ 343.15; + 353.15

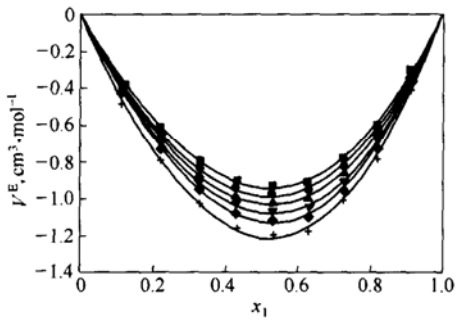

Figure 2 Excess molar volume variation with mole fraction for ethylbenzene (1) + sulfolane (2)
T, K: ■ 303.15; ● 313.15; ▲ 323.15; ▼ 333.15; ◆ 343.15; + 353.15

It has been reported[16] that V^E values of the binary mixture result from the chemical, physical and structural characteristics of liquids. The difference in size and shape of the component molecules and loss of dipolar association leads to expansion in volume. The physical interaction between unlike molecules such as donor-acceptor, dipole to dipole interactions, result to contraction in volume. Sulfolane is self-association according to a hydrogen-bonding scheme similar to self-association in pyridine and quinoline. Kalra *et al.*[17] have proposed that there are intermolecular interactions between quinoline and aromatic hydrocarbon molecules through weak hydrogen bonding. A similar interaction between amine with aliphatic alcohols[18] and aromatic hydrocarbon with aliphatic alcohols[19] through hydrogen bonding has been suggested. The negative values V^E suggest strong interaction between sulfolane and aromatic hydrocarbons. The introduction of $-CH_3$ and $-CH_2CH_3$ groups in aromatic ring would increase the electron density, and hence *p*-xylene and ethylbenzene would have a higher electron donor capacity than benzene, and result in weaker bonding between the H atom associated with the C atom in the aromatic ring (which has less positive character) and S atom of sulfolane. The larger the aliphatic groups in the ring, the weaker the interaction between sulfolane with aromatic hydrocarbons. Therefore, the V^E values for ethylbenzene system are more negative than the values of *p*-xylene system. From Fig. 1 and Fig. 2, it can be seen the mixtures of sulofane + *p*-xylene provide a more compact arrangement.

In Figs. 3 and 4, the viscosity deviation for sulfolane + *p*-xylene and sulfolane + ethylbenzene were plotted against compositions respectively. As may be observed, the $\Delta\eta$ were negative over the entire composition range for two systems, and they were less negative as the temperature increased. The graphs of $\Delta\eta$ *vs.* x are asymmetrical, and the curves are slightly skewed to the left for the two systems. The minima are observed in the rich aromatic hydrocarbon region. Comparing Fig. 3 with Fig. 4, the absolute viscosity deviation decreases with the increase of chain length.

As mentioned above, although the interaction between sulfolane and aromatic hydrocarbon molecules is stronger, however it is weaker than the interaction of self-association by sulfolane. Therefore the viscosity deviations were negative over the entire composition range.

4 CONCLUSIONS

Densities and viscosities of the binary systems of sulfolane + ethylbenzene and sulfolane + *p*-xylene have been experimentally determined in temperature

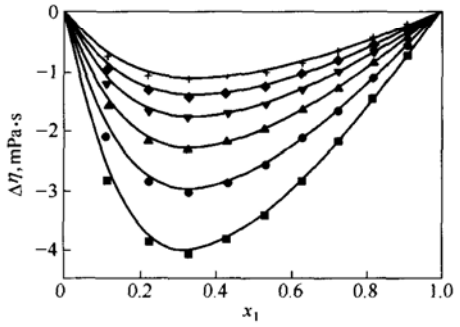

Figure 3 Viscosity deviation with mole fraction for *p*-xylene (1) + sulfolane (2)
T, K: ■ 303.15; ● 313.15; ▲ 323.15; ▼ 333.15; ◆ 343.15; + 353.15

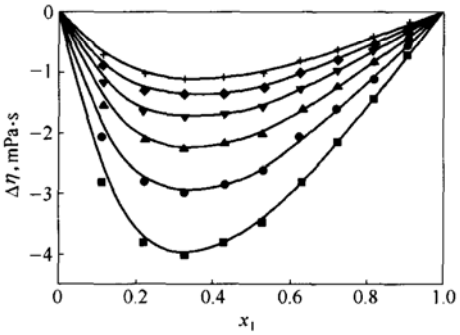

Figure 4 Viscosity deviation with mole fraction for ethylbenzene (1) + sulfolane (2)
T, K: ■ 303.15; ● 313.15; ▲ 323.15; ▼ 333.15; ◆ 343.15; + 353.15

interval 303.15—353.15 K and at atmospheric pressure for the whole composition range. The excess molar volumes and viscosity deviations were computed. The computed quantities have been fitted to Redlich-Kister equation. Excess molar volumes and viscosity deviation show a systematic change with increasing temperature. Two mixtures exhibit negative excess volumes with a minimum which occurs approximately at $x = 0.5$. The effect of the size, shape and interaction of components on excess molar volumes and viscosity deviation is discussed.

NOMENCLATURE

A_k	the coefficients of Redlich-Kister equation
k	viscometer constant
M_i	molar mass of component i, g
N	total number of experimental values
T	temperature, K
t	flow time, s
V^E	excess molar volume, $cm^3 \cdot mol^{-1}$
x_i	molar fraction of component i
Y	V^E or $\Delta\eta$
δ	standard deviation
η	dynamic viscosity of mixtures, mPa·s
η_i	derivation of viscosity for component i, mPa·s

$\Delta\eta$ derivation of viscosity for the mixture, mPa·s
θ Hagenbach correction factor for viscometer
ρ density of the mixture for binary system, $g \cdot cm^{-3}$
ρ_i density of component i, $cm^{-3} \cdot mol^{-1}$, $g \cdot cm^{-3}$

Superscripts

E excess molar property

Subscripts

i component i
1 component 1
2 component 2

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