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Excess Molar Volumes and Viscosities of Binary Mixtures of Sulfolane with Benzene, Toluene, Ethylbenzene, *p*-Xylene, *o*-Xylene, and *m*-Xylene at 303.15 and 323.15 K and Atmospheric Pressure

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Experimental data on density and viscosity at 303.15 and 323.15 K are presented for the binary mixtures of sulfolane + benzene, toluene, ethylbenzene, *p*-xylene, *o*-xylene, and *m*-xylene. From these data, excess molar volumes and deviations in viscosity have been calculated. The computed quantities have been fitted to the Redlich−Kister equation to derive the coefficients and estimate the standard error values. The results are discussed in terms of the intermolecular interactions.

Introduction

Solvent extraction is one of the most important methods used to produce high-purity aromatic extracts from catalytic reformats. Sulfolane is one of the most widely used solvents in this process. Densities are significant for the design of new processes, and viscosities are the transport properties needed to evaluate mass-transfer phenomena and to design equipment. The density and viscosity of a binary mixture are important from the theoretical viewpoint to understand the liquid theory.

A survey of the literature shows that very few measurements have been made on the densities and viscosities of binary mixtures for sulfolane + aromatic hydrocarbons. Yu et al.[1] report densities of these binary systems at 298.15 K. Chen and Knapp[2] measured the densities for sulfolane + ethylbenzene at 283.15 K and 313.15 K. Nevertheless, to our knowledge, no viscosity data on these mixtures were previously reported in the literature.

In this work, densities and viscosities for sulfolane + benzene, ethylbenzene, toluene, *p*-xylene, *o*-xylene, and *m*-xylene at 303.15 K and 323.15 K were studied. The present density and viscosity values for pure substances were compared with the literature values. The Redlich−Kister equation was used to correct and fit the experimental excess molar volumes and the viscosity deviations.

Experimental Section

Materials. All of the chemicals used in this study were obtained from Tianjin Reagent Company. All of the chemicals were purified by distillation, and their middle fraction was collected. The liquids were dried over 0.4-nm molecular sieves and partially degassed by ultrasound prior to their experimental use. The mass-fraction purities tested by gas chromatography were as follows: benzene (>0.995), toluene (>0.993), ethylbenzene (0.993), *p*-xylene (>0.998), *o*-xylene (0.996), and *m*-xylene (0.992). The purity of the sulfolane was confirmed by comparing the densities and viscosities with those reported in the literature. In Table 1, we show the densities and viscosities determined in this study and available from the literature.

Table 1. Comparison of Experimental and Literature Values of Density, ρ, and Viscosity, η, for Pure Compounds

		ρ/g·cm^{-3}		η/m·Pas	
liquid	T/K	exp	lit	exp	lit
sulfolane	298.15	1.26540	1.26654[2]		
			1.2640[3]		
	303.15	1.26080	1.26224[4]	10.0742	10.356[5]
			1.26184[5]		10.284[6]
	323.15	1.24336		6.1936	
benzene	298.15	0.87357	0.8737[7]	0.6038	0.6044[13]
			0.8683[8]		
			0.8734[13]		
	303.15	0.86821	0.8683[7]	0.5612	0.5643[8]
			0.8683[14]		0.5627[13]
			0.8681[13]		
	323.15	0.84665	0.8501[8]	0.4362	0.4422[8]
			0.8468[14]		0.4381[14]
toluene	298.15	0.86301	0.86217[10]	0.5540	0.5541[13]
			0.8609[11]		0.5542[11]
			0.8623[13]		0.5541[8]
	303.15	0.85832	0.8565[10]	0.5251	0.5171[9]
			0.8576[13]		0.5226[13]
			0.8578[14]		0.5227[14]
	323.15	0.83945	0.8385[9]	0.4229	0.4219[9]
			0.8387[11]		0.4216[11]
			0.8390[14]		0.4216[14]
ethylbenzene	298.15	0.86433	0.86289[2]	0.6299	0.6293[15]
			0.8625[19]		
	303.15	0.85995	0.86454[16]	0.5981	0.5923[15]
					0.5976[16]
	323.15	0.84227	0.85471[16]	0.4829	0.4797[15]
					0.4852[16]
p-xylene	298.15	0.85670	0.85648[1]	0.6078	0.6045[13]
			0.8565[13]		0.6031[18]
			0.85672[12]		0.6108[12]
			0.8567[19]		
	303.15	0.85235	0.85227[17]	0.5714	0.567[17]
			0.8522[13]		0.5694[13]
			0.85239[12]		0.5762[12]
	323.15	0.83490		0.4599	0.4580[18]
o-xylene	298.15	0.87487	0.87558[12]	0.7540	0.7574[12]
			0.8750[19]		
	303.15	0.87066	0.87136[12]	0.7602	0.7093[12]
	323.15	0.85379		0.5554	
m-xylene	298.15	0.85980	0.86006[12]	0.5892	0.5900[12]
			0.8600[19]		
	303.15	0.85551	0.85587[12]	0.5471	0.5563[12]
			0.8561[14]		0.5494[14]
	323.15	0.83825		0.4428	0.4444[14]

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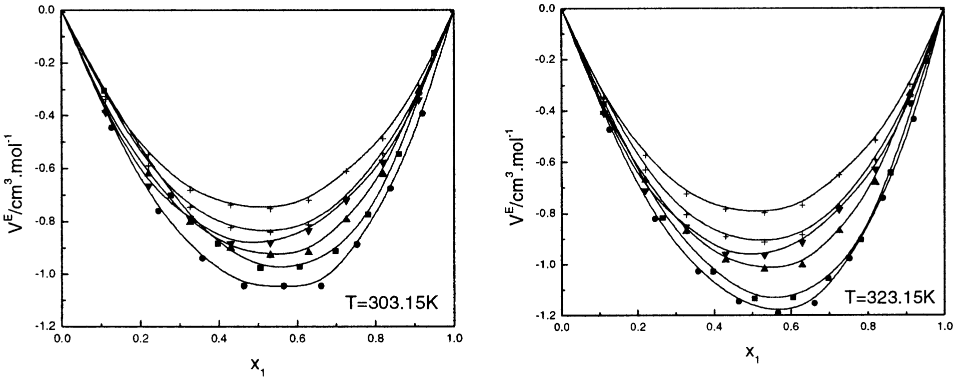

Figure 1. Excess volumes, V^E, of the aromatic hydrocarbons (1) + sulfolane (2) systems at 303.15 and 323.15 K as a function of mole fraction x_1: ■, benzene; ●, toluene; ▲, ethylbenzene; ▼, *p*-xylene; +, *o*-xylene; ×, *m*-xylene. The solid curves are calculated with the Redlich−Kister equation; the symbols represent experimental values.

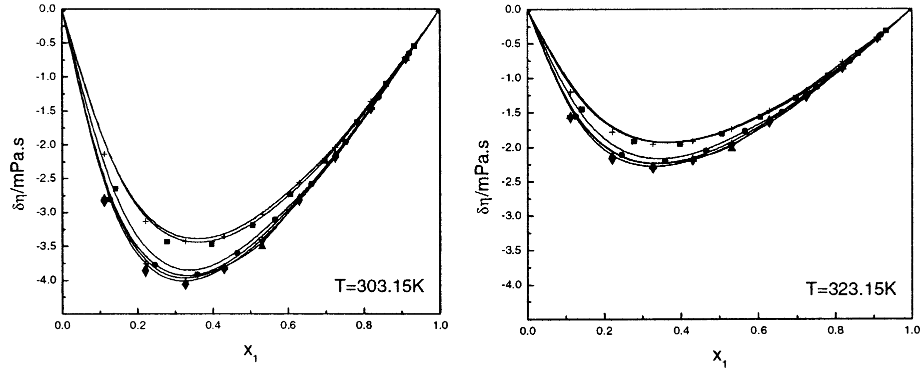

Figure 2. Deviation of viscosity, $\Delta\eta$, of the aromatic hydrocarbons (1) + sulfolane (2) systems at 303.15 and 323.15 K as a function of mole fraction x_1: ■, benzene; ●, toluene; ▲, ethylbenzene; ▼, *p*-xylene; +, *o*-xylene; ×, *m*-xylene. The solid curves are calculated with the Redlich−Kister equation; the symbols represent experimental values.

Apparatus and Procedure. The densities of the pure components and their mixtures were measured with a high-precision vibrating tube digital densimeter (Density/Specific Gravity Meter DA 505, KEM, Japan), and the measurement cell was controlled automatically at 0.01 K. Before each series of measurements the instrument was calibrated at atmospheric pressure with double-distilled water and dry air. Densities in both water and dry air at the various working temperatures were given by the manufacturer. The uncertainty in density measurements was $\pm 5 \times 10^{-5}$ g·cm^{-3}

The liquid mixtures were prepared by weight using a BP210s balance accurate to within ±0.01 mg. The average accuracy in the compositions (mole fraction) of the mixtures was estimated to less than ±0.0001. The precision of excess molar volumes was estimated better than ± 0.002 cm^3·mol^{-1}. All molar quantities are based on the IUPAC relative atomic mass table.

Viscosity was measured using an Ubbelohde suspended-level viscometer, calibrated with double-distilled water, whose viscosity was reported in the literature.[8] An electronic digital stopwatch with a readability of ±0.01 s was used for flow-time measurement. Experiments were repeated a minimum of 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

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

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

The overall uncertainty of the 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 uncertain to within the range ±0.003 mPa·s. All measurements were carried out in a well-stirred water bath with temperature controlled to within ±0.01 K.

Result and Discussion

Excess volumes and excess viscosities were calculated from our measurements according to the following equations

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

Where x_1 and x_2 are mole fractions, M_1 and M_2 are the molar masses, and ρ_1 and ρ_2 are the densities of pure components 1 and 2, respectively. Quantities without subscripts refer to the mixture.

The viscosity derivations were calculated from the following relation

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

Where η is the viscosity of mixtures and η_1 and η_2 are the viscosity of components 1and 2, respectively.

Table 2. Densities (ρ), Viscosities (η), Excess Molar Volumes (V^E), and Viscosity Deviations ($\Delta\eta$) for the Binary Mixtures at 303.15 K and 323.15 K

x	ρ/g·cm^{-3}	η/mPa·s	V^E/cm^3·mol^{-1}	$\Delta\eta$/mPa·s	x	ρ/g·cm^{-3}	η/mPa·s	V^E/cm^3·mol^{-1}	$\Delta\eta$/mPa·s
(x) Benzene + (1 − x) Sulfolane									
T = 303.15 K									
0.0000	1.26080	10.0742	0.0000	0.0000	0.6976	1.00176	1.1999	−0.9139	−2.2380
0.1417	1.21121	6.0747	−0.2608	−2.6515	0.7823	0.96581	0.9625	−0.7748	−1.6697
0.2777	1.16497	4.0025	−0.7012	−3.4299	0.8602	0.93148	0.7860	−0.5457	−1.1051
0.3972	1.12090	2.8261	−0.8850	−3.4695	0.9327	0.89762	0.6520	−0.1540	−0.5494
0.5062	1.07914	2.0659	−0.9787	−3.1928	1.0000	0.86821	0.5612	0.0000	0.0000
0.6063	1.03920	1.5715	−0.9734	−2.7349					
T = 323.15K									
0.0000	1.24336	6.1936	0.0000	0.0000	0.6976	0.98164	0.8791	−1.0580	−1.2981
0.1417	1.19323	3.9276	−0.3120	−1.4502	0.7823	0.94538	0.7170	−0.9054	−0.9726
0.2777	1.14660	2.6755	−0.8110	−1.9192	0.8602	0.91067	0.5933	−0.6429	−0.6478
0.3972	1.10213	1.9499	−1.0288	−1.9569	0.9327	0.87647	0.5029	−0.2048	−0.3208
0.5062	1.05991	1.4685	−1.1361	−1.8107	1.0000	0.84665	0.4362	0.0000	0.0000
0.6063	1.01954	1.1367	−1.1321	−1.5662					
(x) Toluene + (1 − x) Sulfolane									
T = 303.15 K									
0.0000	1.26080	10.0742	0.0000	0.0000	0.6617	0.99405	1.1728	−1.0464	−2.5828
0.1267	1.20984	6.0515	−0.4458	−2.8128	0.7527	0.95736	0.9289	−0.8882	−1.9577
0.2458	1.16172	3.9501	−0.7603	−3.7769	0.8392	0.92276	0.7548	−0.6766	−1.3058
0.3587	1.11578	2.7316	−0.9407	−3.9173	0.9206	0.89024	0.6265	−0.3934	−0.6568
0.4651	1.07279	2.0290	−1.0467	−3.6039	1.0000	0.85832	0.5251	0.0000	0.0000
0.5662	1.03184	1.5569	−1.0467	−3.1106					
T = 323.15 K									
0.0000	1.24336	6.1936	0.0000	0.0000	0.6617	0.97551	0.8828	−1.1525	−1.4924
0.1267	1.19198	3.9064	−0.4748	−1.5561	0.7527	0.93870	0.7151	−0.9780	−1.1349
0.2458	1.14365	2.6621	−0.8211	−2.1131	0.8392	0.90399	0.5893	−0.7420	−0.7616
0.3587	1.09760	1.9191	−1.0290	−2.2046	0.9206	0.87146	0.4967	−0.4367	−0.3843
0.4651	1.05445	1.4580	−1.1472	−2.0517	1.0000	0.83945	0.4229	0.0000	0.0000
0.5662	1.01337	1.1506	−1.1506	−1.7756					
(x) Ethylbenzene + (1 − x) Sulfolane									
T = 303.15 K									
0.0000	1.26080	10.0742	0.0000	0.0000	0.6293	0.99333	1.3237	−0.9176	−2.7872
0.1118	1.20927	6.2242	−0.3794	−2.7906	0.7254	0.95717	1.0567	−0.7932	−2.1436
0.2205	1.16040	4.1551	−0.6197	−3.8296	0.8194	0.92309	0.8613	−0.6215	−1.4482
0.3267	1.11466	2.9424	−0.8023	−4.0360	0.9106	0.89045	0.7155	−0.3064	−0.7298
0.4301	1.07165	2.1852	−0.9014	−3.8133	1.0000	0.85995	0.5981	0.0000	0.0000
0.5309	1.03119	1.5364	−0.9279	−3.5069					
T = 323.15 K									
0.0000	1.24336	6.1936	0.0000	0.0000	0.6293	0.97558	0.9927	−1.0043	−1.6071
0.1118	1.19164	4.0068	−0.4055	−1.5483	0.7254	0.93943	0.8125	−0.8697	−1.2385
0.2205	1.14270	2.8007	−0.6703	−2.1337	0.8194	0.90536	0.6653	−0.6804	−0.8490
0.3267	1.09692	2.0479	−0.8719	−2.2801	0.9106	0.87272	0.5639	−0.3350	−0.4295
0.4301	1.05390	1.5611	−0.9839	−2.1763	1.0000	0.84227	0.4829	0.0000	0.0000
0.5309	1.01347	1.1363	−1.0193	−2.0255					
(x) *p*-Xylene + (1 − x) Sulfolane									
T = 303.15 K									
0.0000	1.26080	10.0742	0.0000	0.0000	0.6293	0.98650	1.2525	−0.8342	−2.8416
0.1118	1.20785	6.1562	−0.3872	−2.8556	0.7252	0.95004	0.9893	−0.7203	−2.1936
0.2206	1.15809	4.0927	−0.6653	−3.8852	0.8191	0.91582	0.8122	−0.5754	−1.4784
0.3266	1.11070	2.8874	−0.7880	−4.0832	0.9106	0.88335	0.6727	−0.3392	−0.7483
0.4301	1.06673	2.1432	−0.8835	−3.8439	1.0000	0.85235	0.5715	0.0000	0.0000
0.5310	1.02522	1.6081	−0.8799	−3.4201					
T = 323.15 K									
0.0000	1.24336	6.1936	0.0000	0.0000	0.6293	0.96888	0.9254	−0.9152	−1.6600
0.1118	1.19021	3.9640	−0.4103	−1.5886	0.7252	0.93240	0.7473	−0.7848	−1.2883
0.2206	1.14037	2.7395	−0.7106	−2.1892	0.8191	0.89826	0.6269	−0.6294	−0.8702
0.3266	1.09299	2.0001	−0.8521	−2.3208	0.9106	0.86585	0.5300	−0.3715	−0.4425
0.4301	1.04903	1.5260	−0.9596	−2.2016	1.0000	0.83490	0.4599	0.0000	0.0000
0.5310	1.00758	1.1659	−0.9642	−1.9831					
(x) *o*-Xylene + (1 − x) Sulfolane									
T = 303.15 K									
0.0000	1.26080	10.0742	0.0000	0.0000	0.6293	1.00009	1.6180	−0.7197	−2.5610
0.1118	1.21086	6.8903	−0.3384	−2.1366	0.7254	0.96488	1.2434	−0.6100	−2.0353
0.2206	1.16338	4.8749	−0.5472	−3.1328	0.8191	0.93197	1.0398	−0.4866	−1.3611
0.3267	1.11872	3.5819	−0.6825	−3.4318	0.9105	0.90064	0.8453	−0.2842	−0.6994
0.4301	1.07659	2.6915	−0.7370	−3.3535	1.0000	0.87066	0.7062	0.0000	0.0000
0.5311	1.03709	2.0657	−0.7516	−3.0332					
T = 323.15 K									
0.0000	1.24336	6.1936	0.0000	0.0000	0.6293	0.98276	1.1727	−0.7680	−1.4727
0.1118	1.19329	4.3582	−0.3530	−1.2051	0.7254	0.94764	0.9211	−0.6506	−1.1826
0.2206	1.14577	3.1690	−0.5739	−1.7808	0.8191	0.91483	0.8044	−0.5168	−0.7709
0.3267	1.10118	2.3966	−0.7239	−1.9550	0.9105	0.88361	0.6503	−0.2989	−0.4097
0.4301	1.05909	1.8537	−0.7832	−1.9149	1.0000	0.85379	0.5554	0.0000	0.0000
0.5311	1.01964	1.4562	−0.7978	−1.7429					

Table 2 (Continued)

x	ρ/g·cm^{-3}	η/mPa·s	V^E/cm^3·mol^{-1}	$\Delta\eta$/mPa·s	x	ρ/g·cm^{-3}	η/mPa·s	V^E/cm^3·mol^{-1}	$\Delta\eta$/mPa·s
				(x) m-Xylene + (1 − x) Sulfolane					
				T = 303.15 K					
0.0000	1.26080	10.0742	0.0000	0.0000	0.6294	0.98881	1.2753	−0.8126	−2.8025
0.1117	1.20779	6.2312	−0.3268	−2.7789	0.7254	0.95264	1.0073	−0.7114	−2.1559
0.2206	1.15839	4.2144	−0.5900	−3.7581	0.8191	0.91849	0.7887	−0.5436	−1.4818
0.3267	1.11181	2.9606	−0.7458	−4.0012	0.9106	0.88627	0.6551	−0.3209	−0.7437
0.4301	1.06811	2.1708	−0.8239	−3.8058	1.0000	0.85551	0.5471	0.0000	0.0000
0.5310	1.02714	1.6499	−0.8412	−3.3654					
				T = 323.15 K					
0.0000	1.24336	6.1936	0.0000	0.0000	0.6294	0.97129	0.9507	−0.8850	−1.6233
0.1117	1.19018	4.0140	−0.3468	−1.5373	0.7254	0.93515	0.7638	−0.7722	−1.2582
0.2206	1.14073	2.7981	−0.6309	−2.1269	0.8191	0.90108	0.6203	−0.5918	−0.8628
0.3267	1.09418	2.0418	−0.8055	−2.2730	0.9106	0.86892	0.5221	−0.3471	−0.4348
0.4301	1.05048	1.5431	−0.8912	−2.1771	1.0000	0.83825	0.4428	0.0000	0.0000
0.5310	1.00956	1.2037	−0.9138	−1.9362					

Table 3. Coefficients of the Redlich−Kister Equation and Standard Deviation of Excess Function for Mixtures

	T = 303.15 K					T = 323.15 K				
function	A_0	A_1	A_2	A_3	s	A_0	A_1	A_2	A_3	s
					(x) Benzene + (1 − x) Sulfolane					
V^E/cm^3·mol^{-1}	−4.1336	−2.0899	2.0415	2.1586	0.0553	−4.7699	−2.2715	2.1009	2.0319	0.0574
$\Delta\eta$/mPa·s	−12.747	7.1561	−5.3916	2.9731	0.0115	−7.2649	3.7542	−2.7280	1.5034	0.0036
					(x) Toluene + (1 − x) Sulfolane					
V^E/cm^3·mol^{-1}	−4.2537	−0.6905	−0.6469	−0.1231	0.0078	−4.6686	−0.7906	−0.6188	−0.2838	0.0092
$\Delta\eta$/mPa·s	−13.821	8.9958	−6.4221	3.0754	0.0071	−7.8784	4.8275	−3.4004	1.6314	0.0043
					(x) Ethylbenzene + (1 − x) Sulfolane					
V^E/cm^3·mol^{-1}	−3.7657	−0.7559	−0.1804	1.1819	0.0109	−4.1236	−0.8674	−0.1115	1.2610	0.0117
$\Delta\eta$/mPa·s	−14.118	9.4601	−7.3385	4.4524	0.0291	−8.0921	5.1174	−3.8671	2.3475	0.0197
					(x) p-Xylene + (1 − x) Sulfolane					
V^E/cm^3·mol^{-1}	−3.5369	0.1791	−0.7713	−0.4733	0.0059	−3.8578	0.1279	−0.7468	−0.5553	0.0065
$\Delta\eta$/mPa·s	−14.140	9.4076	−8.0719	5.0055	0.0150	−8.1748	5.0892	−4.2559	2.6893	0.0078
					(x) o-Xylene + (1 − x) Sulfolane					
V^E/cm^3·mol^{-1}	−3.0001	0.0382	−0.6763	−0.0966	0.0042	−3.1958	−0.0062	−0.6287	−0.0668	0.0045
$\Delta\eta$/mPa·s	−12.601	7.0512	−4.1923	1.9677	0.0081	−7.2236	3.9839	−2.2883	0.9493	0.0100
					(x) m-Xylene + (1 − x) Sulfolane					
V^E/cm^3·mol^{-1}	−3.4008	−0.0714	−0.3204	−0.4724	0.0041	−3.6921	−0.1315	−0.2737	−0.5014	0.0042
$\Delta\eta$/mPa·s	−13.921	9.0023	−7.7187	4.8500	0.0218	−8.0276	4.9976	−4.0027	2.3771	0.0075

The values of V^E and $\Delta\eta$ for each mixture were fitted to the Redlich−Kister polynominal equation

$$Y = x_1(1 - x_1)\sum_{i=0}^{n} A_i(2x_1 - 1)^i \qquad (4)$$

Where $Y = V^E$ or $\Delta\eta$, A_i are adjustable parameters, and x_1 is the fraction of components 1.

In each case, the optimum number of coefficients A_i was determined from an examination of the variation of the standard derivation

$$\sigma(Y) = [\sum(Y_{cal} - Y_{exp})^2/(n - m)]^{(1/2)} \qquad (5)$$

Where n is the total number of experimental values and m is the number of parameters.

The excess molar volume data and the viscosity derivations are presented in Table 2, and they are shown graphically in Figures 1 and 2. Table 3 lists the values of the parameters A_i together with the standard deviations.

The excess volumes are negative over the entire composition range for all the binary mixtures studied and increase in absolute value in the sequence o-xylene < m-xylene < p-xylene < ethylbenzene < benzene < toluene. From Figures 1 and 2, it can be seen the observed minimum in V^E vs x_1 curves tend to shift slightly toward aromatic hydrocarbon rich region of the mixtures.

It has been reported[20] 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 the loss of dipolar association leads to expansion in volume. The physical interaction between unlike molecules such as donor−acceptor and dipole−dipole interactions results in contraction in volume.

The introduction of methyl (or ethyl) groups in the ring will increase the electric donor−acceptor interaction. Hence toluene has a more negative excess volume than benzene. But the bulky methyl or ethyl groups handle the proper orientation of sulfolane to interact with xylene and ethylbenzene and result in a little negtive excess volume than toluene. Therefore, the V^E values for xylene and ethylbenzene should more than that for the toluene mixture. For xylene and ethylbenzene, the specific interaction energy between the two unlike molecules is affected by the positive relation of two ethyl groups in the ring.

The viscosity derivations are negative and decrease in absolute values as the temperature is increased. $\Delta\eta$ values increase in absolute value in the sequence: o-xylene < benzene < toluene < m-xylene < ethylbenzene < p-xylene, although the values for m-xylene, ethlybenzene, and p-xylene are similar. It is known[21] that the strength of the intermolecular electric donor−acceptor interaction is not the only factor that influences the viscosity derivation in liquid mixtures. The molecular size and shape of the components and average degree of association of the mixture are equally important factors. Apart from o-xylene, viscosity derivation increases in absolute with the length of the alkyl chain increase or number of aliphatic groups.

Conclusion

New experimental values of density and viscosity for the system of for the binary mixtures of sulfolane + benzene, toluene, ethylbenzene, *p*-xylene, *o*-xylene, and *m*-xylene at 303.15 and 323.15 K and different compositions are measured. The excess molar volume and viscosity deviations were correlated using the Redlich−Kister polynomial equation. Excellent agreements were observed. The excess molar volumes V^E were negative for all binary mixtures at all temperatures and over the entire range of compositions and increase in absolute value with rising temperature from 293.15 K to 323.15 K. The viscosity deviations are negative too and less negative with increasing temperature. The results are discussed in terms of the intermolecular interactions.

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