Density and Viscosity of the Binary Mixtures of Hexan-1-ol with Isomeric Xylenes at $T = (308.15 \text{ and } 318.15)$ K and Atmospheric Pressure

M. Habibullah,*[†] Kamalendra N. Das,*[†] Ismail M. M. Rahman,*[†,‡] M. Ashraf Uddin,[†] Khaled Saifuddin,[†] Koichi Iwakabe,[§] and Hiroshi Hasegawa[‡]

Department of Chemistry, Faculty of Science, University of Chittagong, Chittagong-4331, Bangladesh, Graduate School of Natural Science and Technology, Kanazawa University, Kakuma, Kanazawa 920-1192, Japan, and Prime Polymer Co., Ltd., Ichihara, Chiba 299-0108, Japan

Densities and viscosities of binary liquid mixtures of hexan-1-ol + *o*-xylene, + *m*-xylene, or + *p*-xylene were measured at a number of mole fractions at $T = (308.15 \text{ and } 318.15)$ K and atmospheric pressure. The excess volumes and the viscosity deviations from the mole fraction average were calculated from the experimental density and viscosity data. The experimental data were correlated with Redlich−Kister equation. Variations in the calculated excess and deviation properties for the liquid mixtures were studied in terms of intermolecular interactions.

Introduction

The density and viscosity of liquid mixtures are required in most engineering calculations and different analytical applications where fluid flow or mixing is an important factor.[1−4] Knowledge about the density and the viscosity of multicomponent mixtures is also important for understanding and rationalizing the behavior of mixed liquids.[5−9] The studies of excess and deviation properties are of considerable interest in understanding the intermolecular interactions in the multicomponent liquid mixtures.[10−15] Therefore, the evaluation and prediction of these properties of solvent mixtures as functions of temperature and composition are of theoretical and practical importance.[16]

We are interested in the accumulation of the binary physical property data of organic liquid mixtures[10,12−15] to explore the possible interactions in the mixed liquid systems. Here we report the values of densities and viscosities for some binary systems of hexan-1-ol with isomeric xylenes (*o*-xylene, *m*-xylene, and *p*-xylene) at (308.15 and 318.15) K and atmospheric pressure, working with the pure species and with number of mixtures covering the entire miscibility range. All liquids are widely used as solvents in scientific studies and industrial applications. The aliphatic polar liquid and monohydric alcohol, hexan-1-ol, herein has been considered as the solute, and the other three liquids, *o*-xylene, *m*-xylene, and *p*-xylene, are considered as solvents. Thus, there is a possibility of favorable interaction with the hydroxyl group of hexan-1-ol with the delocalized π-electrons of aromatic rings in the xylenes,[17] and the experimental results will allow us to study the interactions.

The excess molar volume and viscosity deviation have been calculated from the experimental density and viscosity measurements. The calculated results were fitted to the Redlich−Kister type polynomial equation,[18] to derive the binary coefficients and estimate the standard deviations between experimental and calculated results. Some previous observations for the binary systems were used to interpret the experimental data and the derived quantities.

In the literature, there are reports on the volumetric and viscometric behavior of xylenes with different alkanols[19−25] other than hexan-1-ol. Excess molar volumes of hexan-1-ol + xylenes at 298.15 K was reported by Rodriguez-Nuñez et al.[26] Das et al.[13] and Habibullah et al.[15] reported the excess and deviation properties of the hexan-1-ol + *m*-xylene and + *p*-xylene systems, respectively, at $T = (303.15, 313.15, \text{ and } 323.15)$ K. However, no experimental data are available at the comparable conditions for hexan-1-ol with *o*-xylene, *m*-xylene, or *p*-xylene at $T = (308.15 \text{ and } 318.15)$ K.

Experimental Section

Materials. Hexan-1-ol and *o*-, *m*-, and *p*-xylenes from Merck-Schuchard, Germany (0.98 mass fraction purity), were used without further treatment. The density and viscosity values of pure solvents were compared with the literature data and are shown in Table 1.

Apparatus and Procedure. The liquid mixtures were prepared by mass on an analytical balance (B 204-S, Mettler Toledo, Switzerland) with an uncertainty of ± 0.0001 g which was operated in a drybox. The average uncertainty in the mole fraction of the mixtures was estimated to be less than $\pm 1.0 \cdot 10^{-4}$. Caution was taken to prevent evaporation of the samples after preparation.

The density of the pure components and their mixtures was measured with a 10 mL bicapillary pycnometer. Before each series of measurements the instrument was calibrated at atmospheric pressure with double-distilled water. The uncertainty in density measurements was $0.0004 \text{ g} \cdot \text{cm}^{-3}$.

The viscosity was measured using an A-type Ostwald viscometer, calibrated with double-distilled water. An electronic digital stopwatch with a readability of ± 0.01 s was used for flow-time measurement. The overall uncertainty of the viscosity measurements was $\pm 0.005 \text{ mPa} \cdot \text{s}$.

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* Author(s) for correspondence. E-mail: mhu786@gmail.com (M.H.); kamalctgu@hotmail.com (K.N.D.); i.m.m.rahman@gmail.com (I.M.M.R.). Tel./Fax: +81-76-234-4792.
[†] University of Chittagong.
[‡] Kanazawa University.
[§] Prime Polymer Co., Ltd.

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Table 1. Comparison of Experimental Density (ρ) and Viscosity (η) of Pure Liquids with Literature Values at T = (308.15 and 318.15) K

		ρ/g·cm^{-3}			η/mPa·s		
liquid	T/K	exp.	lit.	ref	exp.	lit.	ref
hexan-1-ol	308.15	0.8085	0.80854	33	3.318	3.30	34
			0.80836	35			
	318.15	0.8012	0.80123	33	2.518	2.5416	36
			0.80105	35			
o-xylene	308.15	0.8667	0.86662	37	0.666	0.665	37
			0.86705	38			
	318.15	0.8584	0.85812	37	0.591	0.589	37
			0.85853	38			
m-xylene	308.15	0.8513	0.85119	37	0.521	0.524	37
			0.85145	38		0.5201	39
			0.85157	40			
			0.85140	41			
	318.15	0.8427	0.84249	37	0.470	0.471	37
			0.84288	38			
			0.8424	39			
p-xylene	308.15	0.8482	0.84796	37	0.541	0.541	37
			0.84812	38		0.540	40
			0.8479	42		0.539	42
	318.15	0.8395	0.83920	37	0.486	0.484	37
			0.83942	38		0.4882	39

Table 2. Densities (ρ), Viscosities (η), Excess Molar Volumes, (V_m^E), and Viscosity Deviations ($\Delta\eta$) for the Binary Mixtures of Hexan-1-ol (1) with *o*-, *m*-, and *p*-Xylenes (2) at T = (308.15 and 318.15) K

x_1	ρ g·cm^{-3}	V_m^E cm^3·mol^{-1}	η mPa·s	$\Delta\eta$ mPa·s	x_1	ρ g·cm^{-3}	V_m^E cm^3·mol^{-1}	η mPa·s	$\Delta\eta$ mPa·s
Hexan-1-ol (1) + *o*-Xylene (2)									
T = 308.15 K									
0.0999	0.8601	0.0886	0.709	−0.073	0.6000	0.8307	0.0970	1.475	−0.270
0.1999	0.8539	0.1260	0.789	−0.129	0.7000	0.8251	0.0730	1.799	−0.250
0.2999	0.8479	0.1403	0.899	−0.178	0.8000	0.8195	0.0540	2.217	−0.189
0.4001	0.8421	0.1292	1.043	−0.222	0.8999	0.8140	0.0254	2.738	−0.087
0.4997	0.8364	0.1136	1.230	−0.255					
T = 318.15 K									
0.0999	0.8517	0.1203	0.621	−0.062	0.6000	0.8227	0.1471	1.199	−0.211
0.1999	0.8456	0.1593	0.683	−0.107	0.7000	0.8172	0.1228	1.431	−0.199
0.2999	0.8397	0.1747	0.768	−0.145	0.8000	0.8118	0.0878	1.730	−0.154
0.4001	0.8339	0.1790	0.879	−0.177	0.8999	0.8064	0.0583	2.104	−0.074
0.4997	0.8283	0.1636	1.020	−0.200					
Hexan-1-ol (1) + *m*-Xylene (2)									
T = 308.15 K									
0.1002	0.8464	0.0826	0.562	−0.065	0.5999	0.8247	0.1201	1.298	−0.284
0.2002	0.8417	0.1397	0.638	−0.117	0.6999	0.8206	0.0959	1.629	−0.275
0.3007	0.8372	0.1660	0.742	−0.168	0.7999	0.8166	0.0577	2.072	−0.219
0.4001	0.8330	0.1564	0.875	−0.218	0.8999	0.8126	0.0207	2.643	−0.114
0.5000	0.8288	0.1451	1.057	−0.259					
T = 318.15 K									
0.1002	0.8376	0.1349	0.502	−0.054	0.5999	0.8166	0.1683	1.066	−0.221
0.2002	0.8330	0.1987	0.561	−0.097	0.6999	0.8127	0.1326	1.306	−0.215
0.3007	0.8287	0.2158	0.643	−0.135	0.7999	0.8088	0.0981	1.623	−0.176
0.4001	0.8246	0.2110	0.747	−0.173	0.8999	0.8050	0.0489	2.031	−0.098
0.5000	0.8206	0.1890	0.885	−0.202					
Hexan-1-ol (1) + *p*-Xylene (2)									
T = 308.15 K									
0.1001	0.8436	0.0878	0.578	−0.071	0.5999	0.8237	0.0905	1.279	−0.327
0.2001	0.8394	0.1187	0.647	−0.131	0.6999	0.8199	0.0667	1.602	−0.324
0.2999	0.8354	0.1222	0.742	−0.190	0.8000	0.8161	0.0431	2.042	−0.266
0.4001	0.8314	0.1246	0.870	−0.248	0.9000	0.8123	0.0211	2.618	−0.149
0.5009	0.8275	0.1092	1.046	−0.296					
T = 318.15 K									
0.1001	0.8349	0.1117	0.513	−0.060	0.5999	0.8156	0.1320	1.051	−0.252
0.2001	0.8308	0.1501	0.567	−0.108	0.6999	0.8119	0.1138	1.287	−0.250
0.2999	0.8269	0.1604	0.643	−0.153	0.8000	0.8083	0.0801	1.604	−0.207
0.4001	0.8231	0.1540	0.743	−0.196	0.9000	0.8047	0.0477	2.017	−0.118
0.5009	0.8193	0.1450	0.877	−0.231					

All measurements were carried out in a transparent glass-walled thermostatic water bath. The Thermo Haake DC10 controller (Thermo Fisher Scientific, MA, USA) was used to control the bath temperature, and a minimum−maximum thermometer (Brannan Thermometers, Cumberland, UK) was used for temperature monitoring. The uncertainty in the temperature during the measurements was ± 0.05 K.

Triplicate measurements were performed for all mixture compositions and pure solvents, and the average of these values was considered in all calculations. Microsoft Office Excel 2007

Table 3. Coefficients, A_i, of the Redlich−Kister Equation (eq 3), Expressing V_m^E and $\Delta\eta$, and the Standard Deviation, σ, for the Binary Mixtures of Hexan-1-ol (1) with *o*-, *m*-, and *p*-Xylenes (2) at T = (308.15 and 318.15) K

	T = 308.15 K					T = 318.15 K				
function	A_0	A_1	A_2	A_3	σ	A_0	A_1	A_2	A_3	σ
					Hexan-1-ol (1) + *o*-Xylene (2)					
$V_m^E/cm^3\cdot mol^{-1}$	0.4592	0.3566	0.2836	0.1098	0.0019	0.6508	0.2905	0.4280	0.2191	0.0048
$\Delta\eta/mPa\cdot s$	−1.0343	0.5415	0.1585	−0.6712	0.0033	−0.8117	0.4014	0.0259	−0.4786	0.0030
					Hexan-1-ol (1) + *m*-Xylene (2)					
$V_m^E/cm^3\cdot mol^{-1}$	0.5882	0.4023	0.0511	0.0526	0.0044	0.7666	0.4491	0.4181	0.2253	0.0019
$\Delta\eta/mPa\cdot s$	−1.0473	0.7338	0.0318	−0.5971	0.0026	−0.8164	0.5311	−0.0764	−0.3474	0.0017
					Hexan-1-ol (1) + *p*-Xylene (2)					
$V_m^E/cm^3\cdot mol^{-1}$	0.4311	0.3085	0.2240	0.2356	0.0030	0.5782	0.2248	0.4405	0.3573	0.0021
$\Delta\eta/mPa\cdot s$	−1.1974	0.8693	−0.0920	−0.4864	0.0028	−0.9299	0.6284	−0.1296	−0.3310	0.0020

(The Microsoft Corporation, USA) software was used for tabulation and data processing. LAB Fit[27] and a LSM[28] curve-fitting program were used for least-squares regression analyses. Sma4 for Windows[29] was used for graphical representation of the data.

Results and Discussion

The binary compositions and the experimental densities (ρ) at all the investigated temperatures are summarized in Table 2. The excess molar volumes of mixing (V_m^E) were calculated using the following relation:

$$V_m^E = [(x_1M_1 + x_2M_2)/\rho - \{(x_1M_1)/\rho_1 + (x_2M_2)/\rho_2\}] \tag{1}$$

where x_1, M_1, and ρ_1 represent the mole fraction, molar mass, and density of component 1 (hexan-1-ol), respectively, and x_2, M_2, and ρ_2 are the corresponding quantities of component 2 (*o*-xylene, *m*-xylene, or *p*-xylene).

Table 2 summarizes the binary composition of the solvent mixtures and the experimental viscosities (η). The viscosity deviations ($\Delta\eta$) at T = (308.15 and 318.15) K were calculated from the following relation:

$$\Delta\eta = \eta - \exp(x_1 \ln \eta_1 + x_2 \ln \eta_2) \tag{2}$$

where x_1 and x_2 are the mole fractions of the first and second components having viscosities η_1 and η_2, respectively.

The composition dependence of V_m^E or $\Delta\eta$ can be represented by a Redlich−Kister type smoothing equation:[18]

$$Y = x_1x_2 \sum_{i=0}^{n} A_i(1 - 2x_1)^i \tag{3}$$

where Y refers to V_m^E or $\Delta\eta$, and x_1 and x_2 are the mole fractions of hexan-1-ol and *o*-xylene, *m*-xylene, or *p*-xylene. The variables A_i are the equation coefficients, which can be obtained by fitting the equation to the experimental values with a least-squares regression method. The values of the coefficients with standard deviation at each studied temperature are summarized in Table 3. The following relation was used to calculate the standard deviation values:

$$\sigma(Y) = [\sum (Y_{expt} - Y_{calc})^2/(n - p)]^{(1/2)} \tag{4}$$

where n is the number of experimental points, p is the number of coefficients of eq 3, and Y_{expt} and Y_{calc} are the experimental and calculated values of the properties.

Figure 1 shows the trend of V_m^E as a function of the binary composition at T = (308.15 and 318.15) K. As it appears, V_m^E is always positive under all experimental conditions. A broad maximum at about $x_1 \approx 0.3000$, which becomes more positive as temperature increases, with a sharp increase followed by gradual decrease, is observed from the curves. Factors contributing to the expansion or contraction of volume define the sign

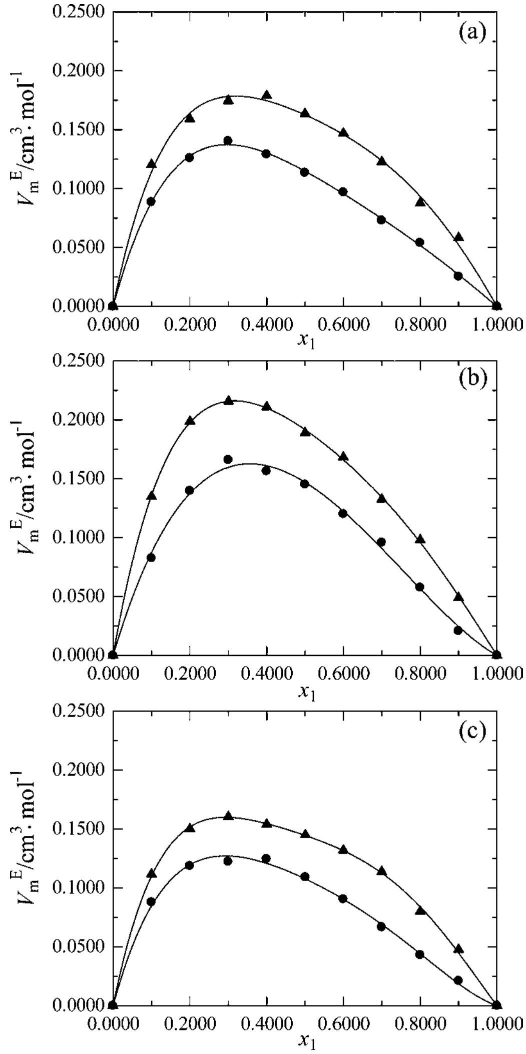

Figure 1. Excess molar volumes for the binary mixtures at 308.15 K (●) and 318.15 K (▲): (a) hexan-1-ol (1) + *o*-xylene (2); (b) hexan-1-ol (1) + *m*-xylene (2); (c) hexan-1-ol (1) + *p*-xylene (2).

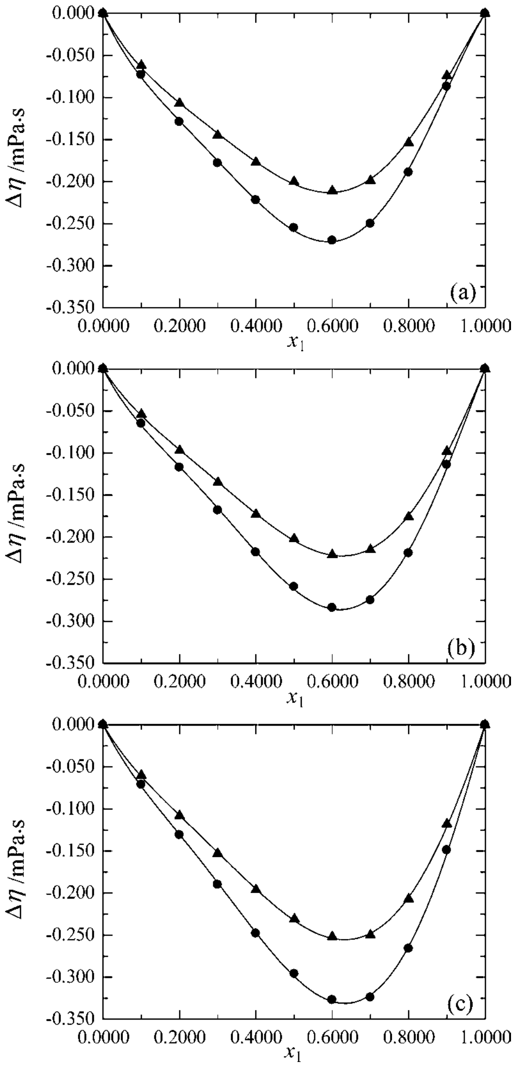

Figure 2. Viscosity deviations for the binary mixtures at 308.15 K (●) and 318.15 K (▲): (a) hexan-1-ol (1) + *o*-xylene (2); (b) hexan-1-ol (1) + *m*-xylene (2); (c) hexan-1-ol (1) + *p*-xylene (2).

Table 4. Boiling Point (t_b), Relative Permittivity (ε), and Dipole Moment (μ) of Isomeric Xylenes[43]

	t_b/°C	ε	μ/D
o-xylene	144.5	2.562	0.64
m-xylene	139.1	2.359	≈ 0
p-xylene	138.4	2.274	0

of V_m^E. The existence of a weak interaction between the components is apparent from the positive values of V_m^E. Hexan-1-ol has an associated structure because of hydrogen bonding and is mixed with the nonassociating solvents, *o*-xylene, *m*-xylene, or *p*-xylene. In the hexan-1-ol low concentration region, a high content of the apolar components in the mixed solution systems may cause disintegration in the associated structure of hexan-1-ol causing the sharp changes in V_m^E. On the contrary, in the hexan-1-ol-rich region, associated structures of the alcohol remain unchanged, which may facilitate the accommodation of the xylenes into the network of alcohol causing a gradual decrease in the V_m^E. The comparative order of V_m^E is as follows: hexan-1-ol + *m*-xylene > hexan-1-ol + *o*-xylene > hexan-1-ol + *p*-xylene. The patterns indicate the variation in the interactions between the delocalized π-electrons of the xylene ring and the −OH group of hexan-1-ol beside the disruption of H-bonding. The variation in the interaction may be due to the effect of the varying position of the two methyl groups on the aromatic ring. For the binary systems of different alkanols with xylenes, a similar explanation for the volumetric properties was suggested by Yadav et al.,[19] Letcher et al.,[20] Rodriguez-Nuñez et al.,[21,26] Sreenvasulu and Naidu,[30] Singh et al.,[31] Saleh et al.,[32] and so forth.

A negative trend in the $\Delta\eta$ values throughout the whole range of mole fraction are observed for the binary mixtures of hexan-1-ol with the xylenes at T = (308.15 and 318.15) K (Figure 2) with a minimum at around 0.6000 mole fraction of alcohol. The negative $\Delta\eta$ values represent repulsion-type interactions between the polar and the apolar components of the binary mixtures. When mixed with the xylenes, in the low concentration region, the associated structure of hexan-1-ol undergoes disruption to form less flow-resistant monomeric, oligomeric, or small-scaled multimeric species resulting in the negative viscosity deviations. The interstitial accommodation of the xylenes into the structured network of hexan-1-ol may occur in the high concentration region. Such an effect may create compact and well-shaped flowing species with favorable geometric fitting having reduced frictional resistance with a subsequent decrease in $\Delta\eta$. The boiling point, dielectric constant, and dipole moment of the xylenes are in the following order: *o*-xylene > *m*-xylene > *p*-xylene (Table 4), while the depth of the negative viscosity deviations follows the reverse order. A dipole−dipole type interaction, which is purely physical in nature, is possible between hexan-1-ol and isomeric xylenes. *o*-Xylene, the most polar among the xylenes, interacts with hexan-1-ol more strongly than the less polar *m*-xylene or the nonpolar *p*-xylene. Therefore, it may be predicted from the comparative profile that the greater the interaction the lesser is the depth of the minima of the negative viscosity deviations. Saleh et al.[25] proposed the same argument to explain the viscometric behavior of the systems containing different alkanols other than hexan-1-ol and *m*-xylene.

Conclusion

Densities and viscosities of the binary systems of hexan-1-ol with *o*-xylene, *m*-xylene, and *p*-xylene were measured at several temperatures and for the whole composition range. The excess molar volumes (V_m^E) and viscosity deviations ($\Delta\eta$) were computed and fitted to the Redlich−Kister type equation. A systematic change with increasing temperature is observed for V_m^E and $\Delta\eta$. V_m^E are positive for all of the mixtures over the entire composition range and become more positive with increasing temperature. The $\Delta\eta$ are negative and become less negative with increasing temperature.

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