Densities and Viscosities of Binary Mixtures of Toluene with Methanol, Ethanol, Propan-1-ol, Butan-1-ol, Pentan-1-ol, and 2-Methylpropan-2-ol at (303.15, 308.15, 313.15) K

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Densities and viscosities of the binary mixtures of toluene with methanol, ethanol, propan-1-ol, butan-1-ol, pentan-1-ol, and 2-methylpropan-2-ol have been measured at 303.15, 308.15, and 313.15 K. From these data, excess molar volumes (V^E) and deviations in viscosity ($\Delta\eta$) have been calculated. These results are fitted to Redlich−Kister type polynomial equations to estimate the binary coefficients and standard errors.

Introduction

Thermodynamic and transport properties of binary liquid mixtures containing protic, aprotic, and associated liquids have been studied in earlier literature.[1−5] The calculated excess quantities from such data have been interpreted in terms of the differences in the size of molecules and the strength of specific and nonspecific interactions taking place between components of the mixtures. In this study, the interaction of alkanols with toluene has been investigated. Alkanols exist in associated form whereas toluene has a nonassociated structure in the liquid state. When alkanols are mixed with toluene, mixing properties with varying intermolecular interactions may be generated. To investigate this effect, in the present investigation, the density (ρ) and the viscosity (η) of binary mixtures of methanol, ethanol, propan-1-ol, butan-1-ol, pentan-1-ol, or 2-methylpropan-2-ol with toluene have been studied over the entire range of composition at (303.15, 308.15, and 313.15) K.

Before this study, very few reports[6] were available in the literature on the density of the mixtures, but no reports on the viscosity were available.

Experimental Section

Toluene (Qualigens Fine Chemicals, purity > 99%) was double distilled, and the middle fraction was used.[7] Ethanol (Maharashtra Government Distillary) was refluxed over calcium oxide for 5 h and distilled at atmospheric pressure. This procedure was repeated two or three times.[8] Methanol (Quilignens Fine Chem, purity 99%), propan-1-ol (E. Merck, purity 99.5%), butan-1-ol (s. d. fine chem., purity 99%), and 2-methylpropan-2-ol (s. d. fine chem., purity 99.2%) were used after single distillation. The purity of the solvents after purification was ascertained by GLC and also by comparing their densities and viscosities with the corresponding literature values at 303.15 K (Table 1). Binary mixtures were prepared by mass in airtight, stoppered glass bottles with a precision of ±0.01 mg on a Metler balance (Switzerland, model AE-240). Care was taken to avoid evaporation and solvent contamination during mixing. The estimated error in mole fraction was $<1 \times 10^{-4}$.

The densities of degassed pure liquids and binary mixtures were measured using the bicapillary pycnometer having a bulb volume of 15 cm^3 and a capillary bore with an internal diameter of 1 mm, in a transparent glass-walled water bath having a thermal stability of ±0.01 K.[9,10] The pycnometer was calibrated using the conductivity water having a conductivity $< 1 \times 10^{-6}\ \Omega^{-1}\ cm^{-1}$. Density values were reproducible within $\pm 5 \times 10^{-5}$ g·cm^{-3}.

Dynamic viscosities were measured using an Ubbelohde suspended-level viscometer, calibrated with conductivity water.[11] An electronic digital stopwatch with a readability of ±0.01 s was used for flow time measurements. At least three repetitions of each data point obtained were reproducible to ±0.05 s, and the results were averaged. Since all flow times were > 200 s and the capillary radius (0.5 mm) was far less than its length (50−60 mm), the kinetic energy and the end corrections were negligible. The dynamic viscosity (η) of the liquids was calculated using

$$\eta/\eta_0 = (\rho t)/(\rho_0 t_0) \quad (1)$$

where ρ, ρ_0 and t, t_0 refer to the density and the flow time of the liquids and water, respectively. The values of the dynamic viscosities (η) thus obtained are reproducible to ±0.003 mPa·s. The other experimental details of the measurements of density and viscosity remain the same as those described previously.[12]

Results and Discussion

Experimental values of densities (ρ) and viscosities (η) of mixtures at (303.15, 308.15, and 313.15) K are listed as a function of mole fraction of toluene in Table 2. The ρ values have been used to calculate the excess molar volumes (V^E) using the following equation:

$$V^E/(cm^3 mol^{-1}) = (x_1M_1 + x_2M_2)/\rho_{12} - (x_1M_1/\rho_1) - (x_2M_2/\rho_2) \quad (2)$$

where ρ_{12} is the density of the mixture and x_1, M_1, ρ_1 and x_2, M_2, ρ_2 are the mole fraction, the molecular weight, and the density of pure components 1 and 2, respectively.

The viscosity deviations ($\Delta\eta$) were calculated using

$$\Delta\eta/(mPa\cdot s) = \eta_{12} - x_1\eta_1 - x_2\eta_2 \quad (3)$$

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Table 1. Comparison of Experimental Densities (ρ) and Viscosities (η) of Pure Liquids with Literature Values along with Deviations (δ) at 303.15 K

	ρ/(g·cm^{-3})			η/(mPa·s)		
liquid	expt	lit.	δ (%)	expt	lit.	δ (%)
toluene	0.8566	0.8569(2)	0.03	0.526	0.527(7)	0.19
methanol	0.7816	0.7816(16), 0.7817(8)	0.00	0.514	0.5146(8), 0.515(12)	0.12
ethanol	0.7806	0.7806(8), 0.7805(16)	0.01	0.965	0.976(19)	1.45
propan-1-ol	0.7954	0.7956(8), 0.7955(12)	0.01	1.705	1.705(16), 1.725(8)	0.00
butan-1-ol	0.8024	0.8022(8), 0.8020(12)	0.25	2.246	2.262(15)	0.71
pentan-1-ol	0.8068	0.8072(8), 0.8068(18)	0.00	2.973	2.909(8), 2.961(18)	0.40
2-methylpropan-2-ol	0.7750	0.7755(8), 0.7755(12)	0.06	3.359	3.378(8), 3.318(17)	0.56

Table 2. Density (ρ), Viscosity (η), and Excess Molar Volume (V^E) for Toluene (1) + Alkanols (2) at (303.15, 308.15, and 313.15) K

	ρ (×10^{-3})	V^E	η		ρ (×10^{-3})	V^E	η		ρ (×10^{-3})	V^E	η
x_1	(kg·m^{-3})	(cm^3·mol^{-1})	(mP·s)	x_1	(kg·m^{-3})	(cm^3·mol^{-1})	(mPa·s)	x_1	(kg·m^{-3})	(cm^3·mol^{-1})	(mPa·s)
					Toluene (1) + Methanol (2)						
					303.15 K						
0.0000	0.7816		0.514	0.3723	0.8285	−0.096	0.540	0.8880	0.8535	−0.033	0.516
0.0912	0.7978	−0.034	0.531	0.4963	0.8368	−0.097	0.529	0.9511	0.8553	−0.016	0.520
0.2105	0.8136	−0.073	0.539	0.6944	0.8466	−0.078	0.515	1.0000	0.8566		0.526
0.2773	0.8204	−0.085	0.542	0.8307	0.8516	−0.047	0.513				
					308.15 K						
0.0000	0.7769		0.480	0.3723	0.8236	−0.090	0.502	0.8880	0.8486	−0.027	0.483
0.0912	0.7930	−0.031	0.495	0.4963	0.8319	−0.088	0.494	0.9511	0.8505	−0.011	0.488
0.2105	0.8087	−0.065	0.502	0.6944	0.8418	−0.071	0.482	1.0000	0.8518		0.496
0.2773	0.8155	−0.077	0.504	0.8307	0.8468	−0.043	0.481				
					Toluene (1) + Ethanol (2)						
					303.15 K						
0.0000	0.7806		0.965	0.5285	0.8320	−0.035	0.596	0.8565	0.8500	0.030	0.519
0.0692	0.7900	−0.031	0.893	0.5858	0.8353	0.003	0.575	0.9213	0.8531	0.019	0.524
0.1849	0.8036	−0.062	0.803	0.6901	0.8414	0.027	0.546	1.0000	0.8566		0.526
0.3191	0.8136	−0.068	0.699	0.7906	0.8467	0.033	0.527				
					308.15 K						
0.0000	0.7762		0.881	0.5285	0.8272	−0.027	0.552	0.8565	0.8452	0.037	0.486
0.0692	0.7855	−0.025	0.814	0.5858	0.8306	0.014	0.533	0.9213	0.8483	0.023	0.493
0.1849	0.7989	−0.053	0.736	0.6901	0.8366	0.034	0.507	1.0000	0.8518		0.496
0.3191	0.8117	−0.062	0.642	0.7906	0.8419	0.040	0.491				
					Toluene (1) + Propan-1-ol (2)						
					303.15 K						
0.0000	0.7954		1.705	0.3778	0.8240	−0.017	0.866	0.8737	0.8506	0.048	0.541
0.1141	0.8052	−0.034	1.376	0.4778	0.8299	0.012	0.761	0.9517	0.8543	0.023	0.527
0.1926	0.8114	−0.045	1.182	0.6242	0.8380	0.045	0.651	1.0000	0.8566		0.526
0.2912	0.8184	−0.039	0.982	0.7421	0.8441	0.056	0.587				
					308.15 K						
0.0000	0.7913		1.526	0.3778	0.8195	−0.012	0.795	0.8737	0.8458	0.053	0.514
0.1141	0.8010	−0.032	1.251	0.4778	0.8254	0.016	0.703	0.9517	0.8495	0.028	0.499
0.1926	0.8071	−0.042	1.078	0.6242	0.8334	0.050	0.608	1.0000	0.8518		0.496
0.2912	0.8140	−0.036	0.899	0.7421	0.8394	0.062	0.553				
					313.15 K						
0.0000	0.7871		1.372	0.3778	0.8150	−0.005	0.736	0.8737	0.8411	0.057	0.491
0.1141	0.7967	−0.030	1.150	0.4778	0.8208	0.024	0.655	0.9517	0.8447	0.033	0.476
0.1926	0.8027	−0.038	0.990	0.6242	0.8288	0.057	0.570	1.0000	0.8470		0.471
0.2912	0.8095	−0.030	0.830	0.7421	0.8347	0.067	0.521				
					Toluene (1) + Butan-1-ol (2)						
					303.15 K						
0.0000	0.8024		2.246	0.3877	0.8248	0.073	0.999	0.8121	0.8470	0.073	0.571
0.0859	0.8078	−0.004	1.860	0.5025	0.8310	0.090	0.833	0.9470	0.8540	0.023	0.526
0.2006	0.8144	0.034	1.452	0.6013	0.8362	0.094	0.726	1.0000	0.8566		0.526
0.2972	0.8198	0.056	1.175	0.7617	0.8445	0.084	0.590				
					308.15 K						
0.0000	0.7985		1.982	0.3877	0.8204	0.084	0.911	0.8121	0.8423	0.080	0.534
0.0859	0.8038	−0.001	1.656	0.5025	0.8264	0.109	0.770	0.9470	0.8492	0.027	0.498
0.2006	0.8102	0.041	1.311	0.6013	0.8316	0.109	0.675	1.0000	0.8518		0.496
0.2972	0.8155	0.068	1.065	0.7617	0.8398	0.095	0.557				
					313.15 K						
0.0000	0.7945		1.764	0.3877	0.8159	0.106	0.840	0.8121	0.8376	0.097	0.507
0.0859	0.7996	0.003	1.489	0.5025	0.8219	0.129	0.713	0.9470	0.8444	0.035	0.478
0.2006	0.8059	0.053	1.193	0.6013	0.8269	0.130	0.629	1.0000	0.8470		0.471
0.2972	0.8112	0.080	0.979	0.7617	0.8351	0.109	0.524				

Table 2 (Continued)

x_1	ρ (×10^{-3}) (kg·m^{-3})	V^E (cm^3·mol^{-1})	η (mP·s)	x_1	ρ (×10^{-3}) (kg·m^{-3})	V^E (cm^3·mol^{-1})	η (mPa·s)	x_1	ρ (×10^{-3}) (kg·m^{-3})	V^E (cm^3·mol^{-1})	η (mPa·s)
					Toluene (1) + Pentan-1-ol (2)						
					303.15 K						
0.0000	0.8068		2.973	0.5202	0.8317	0.106	0.925	0.8629	0.8492	0.058	0.564
0.0745	0.8103	0.026	2.508	0.6090	0.8361	0.113	0.787	0.9443	0.8536	0.032	0.536
0.2114	0.8167	0.065	1.810	0.7058	0.8410	0.101	0.678	1.0000	0.8566		0.526
0.4291	0.8272	0.099	1.101	0.8063	0.8462	0.079	0.592				
					308.15 K						
0.0000	0.8030		2.592	0.5202	0.8273	0.122	0.846	0.8629	0.8445	0.073	0.530
0.0745	0.8064	0.031	2.202	0.6090	0.8316	0.132	0.726	0.9443	0.8488	0.039	0.507
0.2114	0.8126	0.077	1.611	0.7058	0.8364	0.118	0.630	1.0000	0.8518		0.496
0.4291	0.8229	0.114	1.001	0.8063	0.8416	0.093	0.554				
					313.15 K						
0.0000	0.7992		2.262	0.5202	0.8229	0.140	0.778	0.8629	0.8397	0.093	0.500
0.0745	0.8025	0.036	1.940	0.6090	0.8271	0.150	0.670	0.9443	0.8440	0.047	0.484
0.2114	0.8086	0.088	1.439	0.7058	0.8318	0.140	0.587	1.0000	0.8470		0.471
0.4291	0.8186	0.128	0.911	0.8063	0.8368	0.116	0.520				
					Toluene (1) + 2-Methylpropan-2-ol (2)						
					303.15 K						
0.0000	0.7750		3.359	0.3886	0.8047	0.541	0.854	0.8135	0.8405	0.290	0.549
0.1003	0.7820	0.257	1.981	0.4942	0.8135	0.535	0.727	0.9107	0.8484	0.213	0.521
0.1723	0.7872	0.415	1.455	0.6059	0.8228	0.496	0.644	1.0000	0.8566		0.526
0.3109	0.7984	0.505	0.986	0.7118	0.8317	0.419	0.588				
					308.15 K						
0.0000	0.7698		2.623	0.3886	0.7996	0.550	0.771	0.8135	0.8355	0.305	0.518
0.1003	0.7768	0.268	1.622	0.4942	0.8084	0.541	0.666	0.9107	0.8435	0.223	0.490
0.1723	0.7820	0.420	1.239	0.6059	0.8178	0.500	0.594	1.0000	0.8518		0.496
0.3109	0.7933	0.513	0.878	0.7118	0.8266	0.436	0.547				
					313.15 K						
0.0000	0.7645		2.014	0.3886	0.7945	0.554	0.696	0.8135	0.8305	0.322	0.486
0.1003	0.7716	0.264	1.328	0.4942	0.8034	0.549	0.611	0.9107	0.8386	0.234	0.460
0.1723	0.7767	0.431	1.059	0.6059	0.8127	0.515	0.548	1.0000	0.8470		0.471
0.3109	0.7882	0.520	0.781	0.7118	0.8216	0.454	0.510				

Table 3. Parameters and Standard Deviations (σ) of Eqs 3 and 4 for Toluene + Alkanols

system		temp/K	a_o	a_1	a_2	a_3	σ
toluene + methanol	V^E/(cm^3·mol^{-1})	303.15	−0.394	0.100	0.026	−0.080	0.002
		308.15	−0.368	0.058	0.071	0.020	0.002
	$\Delta\eta$/(mPa·s)	303.15	0.037	−0.186	0.012	0.017	0.002
		308.15	0.025	−0.153	−0.006	−0.040	0.001
toluene + ethanol	V^E/(cm^3·mol^{-1})	303.15	−0.116	0.607	0.024	−0.238	0.008
		308.15	−0.087	0.621	0.083	−0.286	0.008
	$\Delta\eta$/(mPa·s)	303.15	−0.546	−0.036	−0.044	0.141	0.005
		308.15	−0.502	−0.062	−0.048	0.195	0.005
toluene + propan-1-ol	V^E/(cm^3·mol^{-1})	303.15	0.054	0.549	−0.012	−0.059	0.003
		308.15	0.068	0.537	0.035	0.018	0.004
		313.15	0.093	0.501	0.059	0.130	0.005
	$\Delta\eta$/(mPa·s)	303.15	−1.525	0.692	−0.076	−0.366	0.006
		308.15	−1.331	0.619	0.065	−0.425	0.006
		313.15	−1.172	0.537	0.238	−0.502	0.008
toluene + butan-1-ol	V^E/(cm^3·mol^{-1})	303.15	0.384	0.184	−0.232	0.163	0.007
		308.15	0.443	0.181	−0.243	0.199	0.006
		313.15	0.521	0.175	−0.218	0.289	0.005
	$\Delta\eta$/(mPa·s)	303.15	−2.227	0.922	−0.284	−0.270	0.008
		308.15	−1.897	0.756	−0.195	−0.241	0.008
		313.15	−1.640	0.559	−0.056	−0.094	0.008
toluene + pentan-1-ol	V^E/(cm^3·mol^{-1})	303.15	0.427	0.077	0.063	0.053	0.004
		308.15	0.493	0.094	0.113	0.076	0.005
		313.15	0.561	0.177	0.186	0.0299	0.003
	$\Delta\eta$/(mPa·s)	303.15	−3.180	1.158	−0.132	−0.233	0.005
		308.15	−2.688	0.908	−0.061	−0.158	0.005
		313.15	−2.276	0.671	0.051	−0.080	0.007
toluene + 2-methylpropan-2-ol	V^E/(cm^3·mol^{-1})	303.15	2.093	−0.830	0.915	0.878	0.027
		308.15	2.118	−0.773	1.054	0.805	0.025
		313.15	2.166	−0.739	1.084	0.895	0.026
	$\Delta\eta$/(mPa·s)	303.15	−4.784	3.9127	−4.421	2.608	0.017
		308.15	−3.516	2.765	−3.150	1.840	0.014
		313.15	−2.490	1.850	−2.073	1.060	0.011

where η_{12} is the viscosity of the mixture and x_1, x_2 and η_1, η_2 are the mole fraction and the viscosity of pure components 1 and 2, respectively. The estimated uncertainties are ±0.002 cm^3·mol^{-1} for V^E and ±0.005 mPa·s for $\Delta\eta$.

The results of V^E and $\Delta\eta$ are fitted to the Redlich and Kister equation:[13]

$$Y = x_1x_2\sum a_i(x_1 - x_2)^i \qquad (4)$$

where Y refers to V^E/(cm^3·mol^{-1}) or $\Delta\eta$/(mPa·s) and x_1 and x_2 are the mole fractions of toluene and alkanol, respectively. The coefficients a_i were obtained by fitting eq 4 to experimental results using a least-squares regression

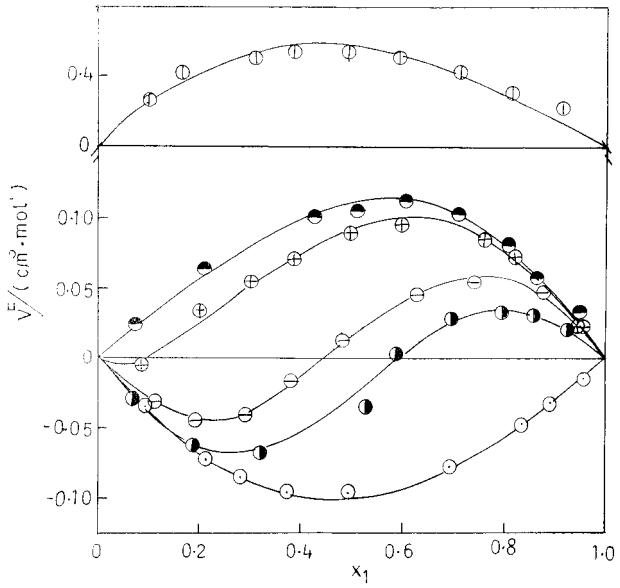

Figure 1. Excess molar volumes (V^E) at 303.15 K for (x_1) toluene + (1 − x_1) alkanols: (⊙) methanol; (◑) ethanol; (⊖) propan-1-ol; (⊕) butan-1-ol; (◓) pentan-1-ol; (Φ) 2-methylpropan-2-ol.

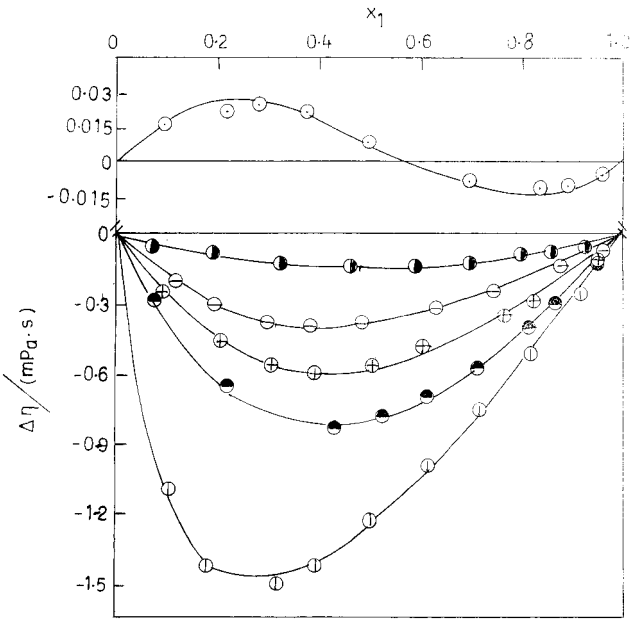

Figure 2. $\Delta\eta$ values at 303.15 K for (x_1) toluene + (1 − x_1) alkanols: (⊙) methanol; (◑) ethanol; (⊖) propan-1-ol; (⊕) butan-1-ol; (◓) pentan-1-ol; (Φ) 2-methylpropan-2-ol.

method. In each case, the optimum number of coefficients is ascertained from an examination of the variation in the standard deviation (σ). The calculated values of a_i along with the tabulated standard deviations (σ) are given in Table 3.

The σ was calculated using

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

where n is the number of data points and m is the number of coefficients.

The variations of V^E and $\Delta\eta$ with mole fraction of toluene (x_1) for the six systems studied at 303.15 K are presented in Figures 1 and 2, respectively. The V^E values are negative for the mixtures of toluene with methanol. For the mixtures of ethanol, propan-1-ol, or butan-1-ol, V^E values are negative at lower mole fractions of toluene and positive at higher mole fractions of toluene. The V^E values are positive over the entire composition range of binary mixtures containing pentan-1-ol or 2-methylpropan-2-ol. For equimolar mixtures ($x_1 \cong x_2$), V^E varies as per the following order: methanol < ethanol < propane-1-ol < butan-1-ol < pentan-1-ol < 2-methylpropan-2-ol. Also at equimolar composition, the V^E values for toluene + propan-1-ol, + butan-1-ol, or + pentan-1-ol (0.014, 0.096, and 0.107 cm³/mol, respectively) agree with the reported values (0.035, 0.096, and 0.090 cm³/mol) of Swami et al.[6]

Table 4. Excess Molar Volumes (V^E) and Deviations in Viscosity ($\Delta\eta$) at Equimolar Concentrations at (303.15, 308.15, and 313.15) K

system	temp/K	V^E/(cm³·mol⁻¹)	$\Delta\eta$/(mPa·s)
toluene + methanol	303.15	−0.098	0.009
	308.15	−0.092	0.006
toluene + ethanol	303.15	−0.029	−0.136
	308.15	−0.022	−0.125
toluene + propan-1-ol	303.15	0.014	−0.381
	308.15	0.017	−0.333
	313.15	0.023	−0.293
toluene + butan-1-ol	303.15	0.096	−0.557
	308.15	0.112	−0.447
	313.15	0.130	−0.410
toluene + propan-1-ol	303.15	0.107	−0.795
	308.15	0.123	−0.672
	313.15	0.140	−0.569
toluene + 2-methylpropan-2-ol	303.15	0.523	−1.196
	308.15	0.529	−0.879
	313.15	0.541	−0.623

It has been reported[14] that the V^E values of binary mixtures result from the chemical, physical, and structural characteristics of liquids. Physical effects contribute to positive V^E; chemical and structural effects contribute to negative V^E. In the present systems, the negative V^E values are attributed to changes in free volume in the mixture of electron donor−acceptor type interactions between alkanols and toluene (toluene being a π-electron donor). The positive V^E arises due to breaking of H-bonds in the self-associated alcohol and steric hindrance due to the bulky methyl group in 2-methylpropan-2-ol. The sigmoidal shape of V^E observed in the toluene + ethanol, + propan-1-ol, and + butan-1-ol systems is attributed to varying interactions between relatively large negative and positive contributions; their magnitudes are sensitive to the length of the component molecules.

The V^E values in the present study are found to increase with temperature, but this is not displayed in Figure 1.

Figure 2 shows that $\Delta\eta$ is negative in all systems except for toluene + methanol and becomes more negative with increasing length and branching of alkanols. The temperature dependence of V^E and $\Delta\eta$ at equimolar composition of all the systems studied is included in Table 4.

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