Densities, Viscosities, and Refractive Indices of Binary Mixtures of Benzene with Isomeric Butanols at 30°C

A. Ali,[1,2] **A. K. Nain,**[1] **B. Lal,**[1] **and D. Chand**[1]

The densities, ρ, viscosities, η, and refractive indices, n, of binary mixtures of benzene with 1-butanol, 2-methyl-1-propanol, 2-butanol, and 2-methyl-2-propanol, including those of the pure liquids, were measured over the complete composition range at 30°C. The dependence of ρ, η, and n on composition was checked by using an empirical relation. The experimental data were used to calculate excess molar volumes, V^{E}, deviations in viscosity, $\Delta\eta$, excess free energies of activation of viscous flow, $G^{*\mathrm{E}}$, deviations in refractive index, Δn, apparent molar volumes, $V_{\phi,1}$ and $V_{\phi,2}$, and partial molar volumes, $\overline{V}_1^{\circ}$ and $\overline{V}_2^{\circ}$, of benzene in alcohols and alcohols in benzene, respectively, at infinite dilution. The variations of these parameters with composition and the effect of branching in alcohols were discussed from the point of view of intermolecular interactions in these mixtures.

KEY WORDS: 1-butanol; 2-methyl-1-propanol; 2-butanol; 2-methyl-2-propanol; benzene; density; binary mixtures; viscosity; refractive index; molecular interactions.

1. INTRODUCTION

The volumetric and viscometric properties of mixed solvent systems and their dependence on composition find applications in many important chemical, industrial, and biological processes. The study of functions such as excess molar volume, deviation in viscosity, excess free energy of activation of viscous flow, deviation in refractive index, etc. of binary liquid mixtures are useful in understanding the nature and strength of molecular

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[1]Department of Chemistry, Jamia Millia Islamia (Central University), Jamia Nagar, New Delhi-110025, India.
[2]To whom correspondence should be addressed. E-mail: anwar.ch@jmi.ernet.in

interactions between the component molecules [1–3]. Benzene is a nonpolar and unassociated liquid, whereas alcohols are polar and self-associated through hydrogen bonding in the pure state [4]. The benzene + alcohol binary mixtures are interesting because of the possibility of weak hydrogen bonding involving π-electrons of the benzene ring and the proton of the hydroxyl group of alcohols. Weak hydrogen bonding of aromatic rings with proton donors (like alcohols) appears to play an important role in the structure of certain biomolecules [5,6]. To the best of our knowledge, there has been no study on benzene + isomeric butanol binary mixtures from the point of view of their volumetric, viscometric and refractive index behavior at 30°C except for the volumetric study of these mixtures by Brown and Smith [7] and Bhardwaj et al. [8] at 25 and 30°C. These considerations led us to undertake the present study.

In continuation of our previous studies [9–12], in this paper we report the densities, ρ, viscosities, η, and refractive indices, n, of pure benzene, 1-butanol, 2-methyl-1-propanol, 2-butanol, 2-methyl-2-propanol, and those of their thirty six binary mixtures, with benzene as a common component, at 30°C, covering the entire composition range. The dependence of ρ, η, and n on the composition of the mixtures has been checked by using an empirical relation. The experimental values of ρ, η, and n were used to calculate excess molar volumes, V^E, deviations in viscosity, $\Delta\eta$, excess free energies of activation of viscous flow, G^{*E}, deviations in refractive index, Δn, apparent molar volumes, $V_{\phi,1}$ and $V_{\phi,2}$, and partial molar volumes, $\overline{V}_1^\circ$ and $\overline{V}_2^\circ$, of benzene in alcohols and alcohols in benzene, respectively, at infinite dilution.

2. EXPERIMENTAL

Benzene was the same as used in our earlier study [9]. 1-Butanol, 2-methyl-1-propanol, 2-butanol, and 2-methyl-2-propanol (all s.d. fine chemicals, India, analytical grade reagents) were purified by using the methods described in the literature [13]. Before use, all the liquids were stored over 0.4 nm molecular sieves to reduce the water content, if any, and were degassed. The mixtures of benzene with alcohols were prepared by mass and were kept in special air-tight stopper glass bottles to avoid evaporation. The weighings were done on an Afcoset ER-120A electronic balance with a precision of ± 0.1 mg. The probable error in the mole fraction was estimated to be less than ± 0.0001.

The densities of the pure liquids and their binary mixtures were measured by using a single-capillary pycnometer (made of Borosil glass) having a bulb capacity of 8×10^{-6}m^3 . The capillary, with graduated

marks, had a uniform bore and could be closed by a tight- fitting glass cap. The marks on the capillary were calibrated by using triple distilled water.

The viscosities of the pure liquids and their binary mixtures were measured by using a Ubbelohde-type suspended level viscometer calibrated with triple-distilled water. The viscometer containing the test liquid was allowed to stand for about 30 min in a thermostatic water bath so that the thermal fluctuations in the viscometer were minimized.

The refractive indices of pure liquids and their binary mixture were measured by using a thermostated Abbe refractometer. The values of refractive index were obtained for sodium D light.

The temperature of the test liquids during the measurements was maintained at $30 \pm 0.02°C$ in an electronically controlled thermostatic water bath. The reliability of the experimental measurements of ρ, η, and n were ascertained by comparing the experimental results for the pure liquids with corresponding literature values [14–22] at 30°C. The comparisons are given in Table I. The estimated total uncertainties in the determination of ρ, η, and n were $\pm 0.017 \, \text{kg·m}^{-3}$, ± 0.0135 mPa·s, and ± 0.0001, respectively.

3. RESULTS AND DISCUSSION

The experimental values of density, viscosity, and refractive index of binary mixtures of benzene with 1-butanol, 2-methyl-1-propanol, 2-butanol, and 2-methyl-2-propanol, over the complete composition range expressed by mole fraction, x_1 of benzene, at 30°C are presented in Table II. The values of ρ, η, and n of all four binary systems were

Table I. Comparisons of Measured Densities, ρ, Viscosities, η, and Refractive Indices, n, of Pure Components with Corresponding Literature Values at 30°C

Component	$\rho(\text{kg·m}^{-3})$		$\eta(\text{mPa·s})$		n	
	measured	lit.	measured	lit.	measured	lit.
Benzene	868.2	868.31 [14]	0.5632	0.5620 [15]	1.4942	1.4940 [16]
1-Butanol	801.9	801.91 [17]	2.2611	2.2853 [18]	1.3946	1.3947 [19]
		801.94 [18]				
2-Methyl-1-propanol	794.2	794.37 [20]	2.8760	2.8720 [20]	1.3914	–
2-Butanol	798.8	798.88 [21]	2.5532	2.4920 [20]	1.3903	–
2-Methyl-2-propanol	776.1	775.75 [21]	3.3435	3.3180 [20]	1.3899	–
		776.16 [22]				

least-squares fitted to the polynomial equation,

$$a = \sum_{i=0}^{4} a_i x_1^i \qquad (1)$$

where a is ρ or η or n. The coefficients, a_i along with the standard deviations, $\sigma(a)$ and average errors, s between calculated and experimental values, are listed in Table III. It is found that Eq. (1) represents ρ values with an average error of ± 0.030, ± 0.022, ± 0.029 and $\pm 0.040\,\mathrm{kg \cdot m^{-3}}$; η values with an average error of $\pm 1.8 \times 10^{-3}$, $\pm 1.6 \times 10^{-3}$, $\pm 3.8 \times 10^{-3}$ and $\pm 3.7 \times 10^{-3}$ mPa·s; and n values with an average error of ± 0.00002, ± 0.00006, ± 0.00002, and ± 0.00008; for benzene + 1-butanol, + 2-methyl-1-propanol, +2-butanol, and +2-methyl-2-propanol binary mixtures, respectively.

The values of V^E, $\Delta\eta$, G^{*E}, and Δn have been calculated by using the following relations:

$$V^E = x_1 M_1 (1/\rho - 1/\rho_1) + x_2 M_2 (1/\rho - 1/\rho_2) \qquad (2)$$

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

$$G^{*E} = RT[\ln(\eta V) - x_1 \ln(\eta_1 V_1) - x_2 \ln(\eta_2 V_2)] \qquad (4)$$

$$\Delta n = n - (x_1 n_1 + x_2 n_2) \qquad (5)$$

where x and M are the mole fraction and molar mass, respectively; subscripts 1 and 2 stand for pure components, benzene and alcohol, respectively; R is the universal gas constant; T is the absolute temperature; and V is the molar volume calculated by using the relation,

$$V = (x_1 M_1 + x_2 M_2)/\rho \qquad (6)$$

The values of V^E, $\Delta\eta$, G^{*E}, and Δn were fitted to a Redlich–Kister [23] type polynomial equation,

$$Y^E = x_1 x_2 \sum_{i=1}^{5} A_i (1 - 2x_1)^{i-1} \qquad (7)$$

where Y^E is V^E or $\Delta\eta$ or G^{*E} or Δn. The values of coefficients, A_i, and the corresponding standard deviations, $\sigma(Y^E)$, calculated by using the relation,

$$\sigma(Y^E) = \left[\sum (Y_{\mathrm{expt}}^E - Y_{\mathrm{cal}}^E)^2 / (h - k) \right]^{1/2} \qquad (8)$$

where h is the number of experimental data points and k is the number of coefficients considered ($k = 5$ in the present calculation) are listed in

Table II. Experimental Values of Densities, ρ, Viscosities, η, and Refractive Indices, n as a Function of Mole Fraction, x_1 of Benzene for the Binary Mixtures at 30°C

x_1	$\rho(\text{kg·m}^{-3})$	$\eta(\text{mPa·s})$	n
Benzene + 1-Butanol			
0.1035	808.2	1.9001	1.4018
0.2024	814.3	1.6015	1.4090
0.3037	820.5	1.3440	1.4166
0.4000	826.4	1.1450	1.4245
0.5047	833.0	0.9759	1.4340
0.5931	838.7	0.8562	1.4427
0.7016	845.9	0.7412	1.4545
0.7660	850.5	0.6921	1.4634
0.8859	859.1	0.6110	1.4790
Benzene + 2-Methyl-1-propanol			
0.1012	800.9	2.3224	1.3998
0.2055	807.8	1.8805	1.4082
0.3024	814.3	1.5512	1.4160
0.4124	821.9	1.2603	1.4261
0.5110	828.8	1.0571	1.4357
0.6016	835.4	0.9052	1.4455
0.7060	843.3	0.7704	1.4573
0.8037	851.1	0.6783	1.4691
0.8987	859.0	0.6131	1.4812
Benzene + 2-Butanol			
0.1032	804.6	1.9502	1.3998
0.2077	810.6	1.5202	1.4092
0.3084	816.5	1.2037	1.4184
0.4086	822.7	0.9692	1.4279
0.5110	829.3	0.8012	1.4382
0.5989	835.2	0.6995	1.4473
0.7027	842.7	0.6211	1.4588
0.7981	850.3	0.5854	1.4699
0.8973	858.6	0.5709	1.4818
Benzene + 2-Methyl-2-propanol			
0.1001	782.4	2.4871	1.3992
0.2137	790.9	1.7873	1.4105
0.3033	798.0	1.3950	1.4198
0.4117	807.4	1.0802	1.4320
0.5129	816.5	0.8736	1.4440
0.6126	825.9	0.7351	1.4560
0.7187	836.5	0.6441	1.4684
0.8046	845.7	0.6103	1.4775
0.8998	856.2	0.5822	1.4865

Table III. Coefficients, a_i, of Eq.(1) along with Standard Deviations, $\sigma(a)$, and Average Error in Calculated Values of ρ, η, and n ($\pm$Units) for the Binary Mixtures

Benzene +	a_o	a_1	a_2	a_3	a_4	$\sigma(a)$	$\pm$units
$\rho(\mathrm{kg \cdot m^{-3}})$							
	ρ_o	$\rho_1 \times 10^2$	$\rho_2 \times 10^2$	$\rho_3 \times 10^2$	$\rho_4 \times 10^2$	(10^{-5})	
1-Butanol	0.80190	6.12083	−0.07511	0.05197	0.53083	4.429	0.030
2-Methyl-1-propanol	0.79421	6.55353	0.26063	0.06109	0.52151	3.183	0.022
2-Butanol	0.79881	5.55947	0.42655	0.43990	0.51337	4.215	0.029
2-Methyl-2-propanol	0.77606	5.93137	5.44763	−4.29467	2.13089	5.453	0.040
$\eta(\mathrm{mPa \cdot s})$							
	η_o	η_1	η_2	η_3	η_4	(10^{-3})	(10^{-3})
1-Butanol	2.26264	−3.82405	2.87931	−0.69281	−0.06306	2.423	1.8
2-Methyl-1-propanol	2.87447	−6.05424	6.69057	−4.15623	1.21020	2.214	1.6
2-Butanol	2.54825	−6.47149	8.22906	−5.10519	1.36596	5.752	3.8
2-Methyl-2-propanol	3.34278	−9.87139	14.13887	−9.76776	2.72322	5.086	3.7
n							
	n_o	n_1	n_2	n_3	n_4	(10^{-4})	(10^{-5})
1-Butanol	1.39448	0.07643	−0.04485	0.12206	−0.05378	2.903	2
2-Methyl-1-propanol	1.39147	0.08328	−0.02821	0.08946	−0.04181	0.978	6
2-Butanol	1.39030	0.09415	−0.02762	0.06722	−0.02985	0.372	2
2-Methyl-2-propanol	1.38993	0.08939	0.02300	0.04189	−0.05009	1.093	8

Table IV. $Y_{\mathrm{cal}}^{\mathrm{E}}$ has been obtained from Eq. (7) by using the best-fit values of coefficient A_i. The variations of smoothed values of V^{E}, $\Delta\eta$, $G^{*\mathrm{E}}$, and Δn, with mole fraction, x_1, of benzene are shown graphically in Figs. 1–4.

The curves in Fig. 1 show that the values of the excess molar volume, V^{E}, are positive, for all four binary systems (benzene + 1-butanol/2-methyl-1-propanol/2-butanol/2-methyl-2-propanol) under study, over the entire composition range. The general shape of our experimental V^{E} against x_1 curves are similar to those reported by Brown and Smith [7] at 25°C and Bhardwaj et al. [8] at 35°C for benzene + isomeric butanol mixtures. The variation of V^{E} with composition has been qualitatively interpreted as follows. As stated above, the alcohols are self-associated through hydrogen bonding in the pure state [4]. Mixing of benzene with alcohols (isomeric butanols) would induce dissociation of the hydrogen-bonded structures of these alcohols leading to expansion in volume, and thus a positive contribution to V^{E} values. On the other hand, there is the possibility of formation of weak $\pi \cdots \mathrm{H}$ bonding between π-electrons of the benzene ring and the hydrogen atom of the hydroxyl group of alcohols leading to a contraction in volume, and hence resulting in a negative contribution to V^{E} values. Larson et al. [24] have also suggested the presence

Table IV. Coefficients, A_i of Eq. (7) along with Standard Deviations, $\sigma(Y^E)$ for the Binary Mixtures

Properties	A_1	A_2	A_3	A_4	A_5	$\sigma(Y^E)$
Benzene + 1-Butanol						
V^E (10^{-6} m$^3\cdot$mol^{-1})	0.8280	−0.5755	0.1529	−0.0176	0.0851	0.0025
$\Delta\eta$ (mPa·s)	−1.7199	−0.4361	−0.1588	0.1278	0.2600	0.0007
G^{*E} (kJ·mol^{-1})	−1.3726	0.6023	−0.3109	0.1241	0.5494	0.0021
Δn (10^{-2})	−4.3074	1.3276	−0.6393	−1.8734	3.0837	0.0110
Benzene + 2-Methyl-1-propanol						
V^E (10^{-6}m$^3\cdot$mol^{-1})	1.1039	−0.5004	0.0704	−0.1916	0.0914	0.0020
$\Delta\eta$ (mPa·s)	−2.5692	−0.8236	−0.3875	−0.1737	0.1140	0.0006
G^{*E} (kJ·mol^{-1})	−1.6486	0.6917	−0.4964	−0.2606	0.5229	0.0015
Δn (10^{-2})	−3.2393	0.2034	0.5105	0.3350	0.9636	0.0097
Benzene + 2-Butanol						
V^E (10^{-6}m$^3\cdot$mol^{-1})	1.9374	−0.7114	0.3614	−0.0199	−0.3826	0.0012
$\Delta\eta$ (mPa·s)	−2.9669	−1.0772	−0.3056	−0.4519	−0.1460	0.0006
G^{*E} (kJ·mol^{-1})	−3.8212	1.2580	0.4315	−1.2430	0.5095	0.0021
Δn (10^{-2})	−2.0691	0.4181	0.4365	−0.1157	0.5619	0.0024
Benzene + 2-Methyl-2-propanol						
V^E (10^{-6} m$^3\cdot$mol^{-1})	2.4853	0.1680	0.7384	0.3116	0.0610	0.0022
$\Delta\eta$ (mPa·s)	−4.2261	−2.1108	−1.0771	−0.1550	0.6549	0.0024
G^{*E} (kJ·mol^{-1})	−4.2228	0.8694	0.0279	−0.5852	1.5177	0.0046
Δn (10^{-2})	0.1605	−3.0974	1.5461	0.7260	−0.6368	0.0022

of weak $\pi\cdots$H bonding between π-electrons of benzene and the hydroxyl group of *tert*-butyl alcohol.

Another equally important contribution leading to the positive V^E values arises from the molecular sizes of benzene (molar volume $= 8.9968 \times 10^{-5}$m$^3\cdot$mol^{-1}) and 1-butanol/2-methyl-1-propanol/2-butanol/2-methyl-2-propanol (molar volumes $= 9.2430 \times 10^{-5}/9.3327 \times 10^{-5}/9.2789 \times 10^{-5}/9.5503 \times 10^{-5}m^3\cdot$mol^{-1}) that are not too different, suggesting that the component molecules do not pack well into each other's structure resulting in expansion in volume, and hence, positive V^E values. Assarson and Eirich [25] suggested that the liquids of similar molecular sizes usually mix with positive excess volumes. It is clear from Fig. 1 that for the binary mixtures under study, V^E values become increasingly positive in the order: 1-butanol < 2-methyl-1-propanol < 2-butanol < 2-methyl-2-propanol, i.e., V^E values become more positive as the branching in the alcohol molecule increases. The presence of three methyl groups at $3°$ carbon atom in 2-methyl-2-propanol, one methyl and one ethyl groups at $2°$ carbon atom in 2-butanol, one isobutyl group at $1°$ carbon atom in 2-methyl-1-propanol, and one propyl group at $1°$ atom in 1-butanol create steric hindrance
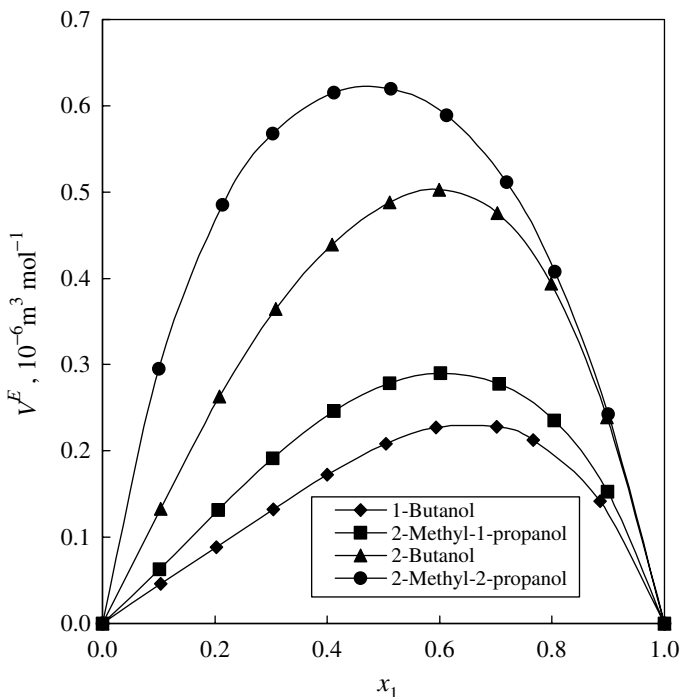
Fig. 1. Variation of excess molar volume, V^E, with mole fraction, x_1, of benzene for the binary mixtures at 30°C.

near the hydroxyl group in the order: 2-methyl-2-propanol > 2-butanol > 2-methyl-1-propanol > 1-butanol. As a result, the observed positive deviations in V^E (Fig. 1) suggest that the closer approach of unlike molecules in the mixtures is sterically hindered (due to the branching in alcohol molecules); therefore, the strength of interaction ($\pi \cdots H$ bonding) between benzene and alcohol molecules should follow the order: 1-butanol > 2-methyl-1-propanol > 2-butanol > 2-methyl-2-propanol. This is in accordance with the view proposed by Fort and Moore [26] that V^E values become increasingly positive as the strength of interaction between the component molecules of the mixture decreases.

Figures 2 and 3 show that $\Delta\eta$ and G^{*E} values are negative for all four binary systems (benzene + 1-butanol/2-methyl-1-propanol/2-butanol/2-methyl-2-propanol) over the complete composition range. The negative $\Delta\eta$ and G^{*E} values indicate the presence of weak interactions between the unlike molecules in the mixture [27,28]. As expected, the values of $\Delta\eta$ and G^{*E} become more negative as the branching in the alcohol molecule
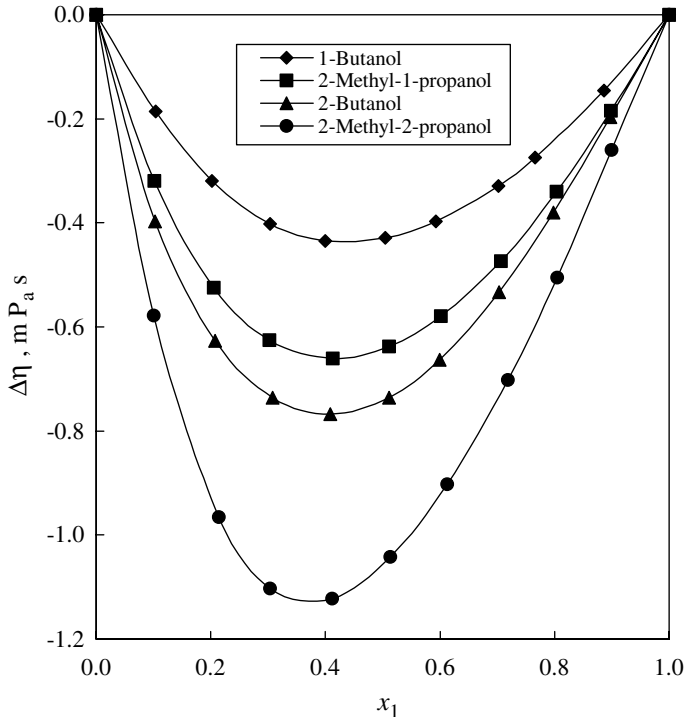
Fig. 2. Variation of deviations in viscosity, $\Delta\eta$, with mole fraction, x_1, of benzene for the binary mixtures at 30°C.

increases from 1-butanol to 2-methyl-2-propanol. This suggests that the strength of interaction in these mixtures is in the order: 1-butanol > 2-methyl-1-propanol > 2-butanol > 2-methyl-2-propanol.

The curves in Fig. 4 show that for all the systems under study the deviations in refractive index, Δn, are negative and tend to become less negative on going from 1-butanol to 2-methyl-2-propanol over the complete composition range. In general, the magnitude of Δn increases with decreases in the strength of interaction between the component molecules in the mixture [29], as in the present case. During their recent study Brocos et al. [30] have suggested that the deviation Δn from ideality is physically more interpretable when defined on a volume fraction basis instead of mole fraction basis. However, in the present work and in the work reported by others [31,32], the mole fraction basis has been used to calculate Δn values. Thus, the functions V^E, $\Delta\eta$, G^{*E}, and Δn truly
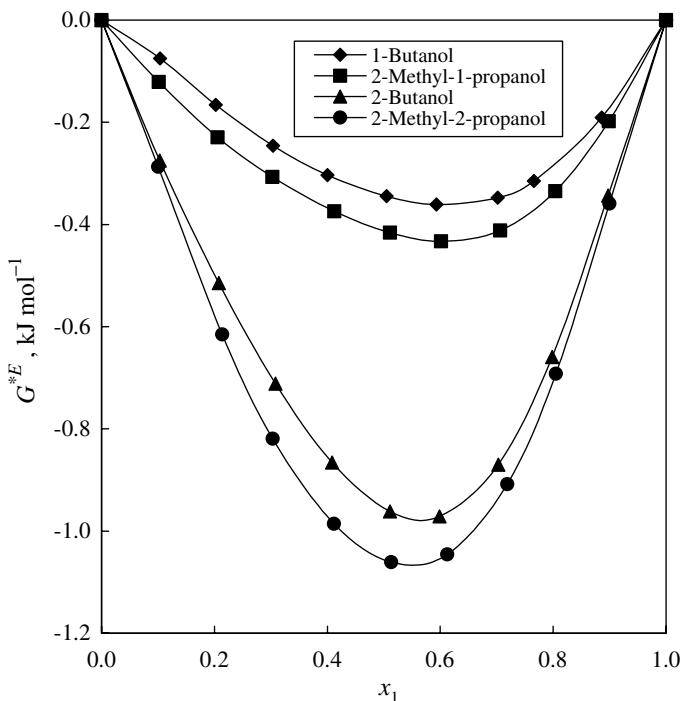
Fig. 3. Variation of excess free energies of activation of viscous flow, G^{*E}, with mole fraction, x_1, of benzene for the binary mixtures at 30°C.

complement each other in describing the behaviors of the present binary liquid mixtures.

The apparent molar volumes, $V_{\phi,1}$ and $V_{\phi,2}$, of benzene and alcohol, respectively, in these mixtures have been calculated by using the following relations [33]:

$$V_{\phi,1} = V_1^* + V^E/x_1 \tag{9}$$

$$V_{\phi,2} = V_2^* + V^E/x_2 \tag{10}$$

where V_1^* and V_2^* are the molar volumes of pure benzene and alcohol, respectively. The partial molar volumes, $\bar{V}_1^\circ$ and $\bar{V}_2^\circ$, of benzene in alcohol and alcohol in benzene, respectively, at infinite dilution were obtained by using the method described elsewhere [25, 33]. The deviations in V_ϕ at infinite dilution, ΔV_1 and ΔV_2, for benzene and alcohol, respectively, were
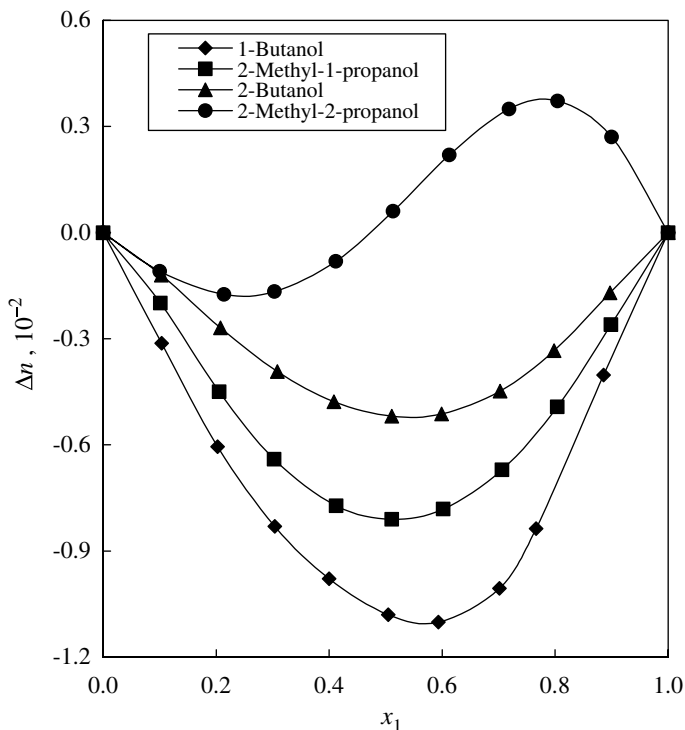
Fig. 4. Variation of deviations in refractive index, Δn, with mole fraction, x_1, of benzene for the binary mixtures at 30°C.

calculated by using the relations [33],

$$\Delta V_1 = \bar{V}_1^{\circ} - V_1^{*} \tag{11}$$

$$\Delta V_2 = \bar{V}_2^{\circ} - V_2^{*} \tag{12}$$

The values of $\bar{V}_1^{\circ}$, V_1^{*}, ΔV_1, $\bar{V}_2^{\circ}$, V_2^{*}, and ΔV_2 are listed in Table V. A perusal of Table V indicates that the partial molar volumes, $\bar{V}_1^{\circ}$ and $\bar{V}_2^{\circ}$, of benzene and alcohols at infinite dilution are larger than their corresponding molar volumes, i.e., there is an expansion in volume of the components, benzene and alcohols, on mixing. It is clear from Table V that the deviations, ΔV_1 and ΔV_2, are positive for all four binary systems and follow the order: 1-butanol < 2-methyl-1-propanol < 2-butanol < 2-methyl-2-propanol, which is also the order of branching in alcohol molecules. This again suggests that the strength of interaction between benzene and

Table V.   Values of $\bar{V}_1^\circ$, V_1^*, ΔV_1, $\bar{V}_2^\circ$, V_2^*, and ΔV_2 for the Binary Mixtures

Benzene +	$\bar{V}_1^\circ$	V_1^*	ΔV_1	$\bar{V}_2^\circ$	V_2^*	ΔV_2
	$(10^{-5}\ \mathrm{m^3 \cdot mol^{-1}})$			$(10^{-5}\mathrm{m^3 \cdot mol^{-1}})$		
1-Butanol	9.0472	8.9968	0.0504	9.3431	9.2430	0.1001
2-Methyl-1-propanol	9.0702	8.9968	0.0734	9.4583	9.3327	0.1256
2-Butanol	9.1381	8.9968	0.1413	9.4838	9.2632	0.2048
2-Methyl-2-propanol	9.2465	8.9968	0.2497	9.7730	9.5503	0.2267

alcohol molecules follow the order: 1-butanol > 2-methyl-1-propanol > 2-butanol > 2-methyl-2-propanol.

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