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Densities, Viscosities, Sound Speeds, Refractive Indices, and Excess Properties of Binary Mixtures of Isoamyl Alcohol with Some Alkoxyethanols

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Abstract Densities and viscosities were measured for binary mixtures of isoamyl alcohol with 2-methoxyethanol, 2-ethoxyethanol, and 2-butoxyethanol over the entire range of composition at 303.15 K, 313.15 K, and 323.15 K and ultrasonic speeds and refractive indices at 303.15 K under atmospheric pressure. From the experimental values of density, viscosity, ultrasonic speed, and refractive index, the values of excess molar volume (V^{E}), viscosity deviations ($\Delta\eta$), deviations in isentropic compressibility (ΔK_S), and excess molar refraction (ΔR) have been calculated. The excess or deviation properties were found to be either negative or positive, depending on the molecular interactions and the nature of liquid mixtures.

Keywords Excess molar volume · Refractive index · Ultrasonic speed · Viscosity deviation

1 Introduction

Grouping of solvents into classes is often based on the nature of the intermolecular forces because the manner whereby solvent molecules are associated with each other brings about a marked effect on the resulting properties. The determination of density, viscosity, speed of sound, and refractive index is a valuable tool to develop new theoretical models and learn about the liquid state [1] because of the close

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connection between liquid structure and macroscopic properties. Ultrasonic properties and refractive index find extensive applications resulting from their ability of characterizing the physicochemical behavior of liquid systems. On the other hand, excess thermodynamic functions and deviations of non-thermodynamic properties of binary liquid mixtures are fundamental for understanding the interactions between molecules in these types of binary mixtures.

There has been a recent upsurge of interest [2,3] in the study of thermodynamic properties of binary liquid mixtures which has been used extensively to obtain information on intermolecular interactions and stereochemical effects in these solvents. The present work reports the density (ρ) and viscosity (η) for binary mixtures of isoamyl alcohol (I.A.A) with 2-methoxyethanol (2-M.E), 2-ethoxyethanol (2-E.E), and 2-butoxyethanol (2-B.E) over the entire range of composition at 303.15 K, 313.15 K, and 323.15 K. Also, the ultrasonic speed (u) and refractive index (n_{D}) have been reported for the binary mixtures at 303.15 K.

The amyl alcohols are used for the composition of perfumes and the synthesis of fruit essences. They are also used as solvents for surfaces and lacquer baths, inks for print, and dyes for wool as well as in the chemical production of photographic and pharmaceutical substances. Furthermore, they are an intermediate in the production of amyl acetate and other amyl esters. In some of these uses, a knowledge of their physical properties is very important. It is well known that alkoxyethanols have wide use as monomers in the production of polymers and emulsion formulations. They are also of considerable interest for studying the heteroproximity effects of the etheric oxygen on the –OH bond and, hence their influence on the associated nature of the species in these molecules. This work provides a test of various empirical equations to correlate viscosity, density, acoustic, and refractive index data of binary mixtures in terms of pure component properties.

2 Experimental

2.1 Chemicals

2-methoxyethanol, 2-ethoxyethanol, and 2-butoxyethanol (S.D. Fine Chemicals, AR, India) were purified as described in the literature [4]. Isoamyl alcohol was procured from Merck, India and was used as purchased. The pure chemicals were stored over activated 4 Å molecular sieves to reduce water content before use. The chemicals after purification were 99.8 % pure, and their purity was ascertained by GLC and also by comparing experimental values of density, viscosity, and refractive index with those reported in the literature when available, as presented in Table 1.

2.2 Measurements

Densities (ρ) were measured with an Ostwald–Sprengel type pycnometer having a bulb volume of about 25 cm^3 and an internal diameter of the capillary of about 0.1 cm. The measurements were carried out in a thermostat bath controlled to ±0.01 K. The viscosity (η) was measured by means of a suspended Ubbelohde type viscometer,

Table 1 Density (ρ), viscosity (η), sound speed (u), and refractive index (n_D) of the pure component liquids at different temperatures

T (K)	$\rho \times 10^{-3}(\mathrm{kg \cdot m^{-3}})$		$\eta(\mathrm{mPa \cdot s})$		$\mathrm{u(m \cdot s^{-1})}$		n_D	
	This work	Literature	This work	Literature	This work	Literature	This work	Literature
Isoamyl alcohol								
303.15	0.8031	0.8032 [19]	3.2622	3.1111[19]	1197.4	1197.0 [19]	1.4035	–
313.15	0.7964	–	2.4409	–	–	–	–	–
323.15	0.7898	–	1.9396	–	–	–	–	–
2-Methoxyethanol								
303.15	0.9568	0.9558 [20]	1.5496	1.476 [20]	1324.3	1359.2 [21]	1.3983	–
313.15	0.9463	0.9462 [20]	1.2883	1.189 [20]	–	–	–	–
323.15	0.9378	–	1.0824	–	–	–	–	–
2-Ethoxyethanol								
303.15	0.9195	0.9212 [20]	1.6226	1.643 [20]	1301.5	1319.9 [21]	1.4065	–
313.15	0.9120	0.9123 [20]	1.3554	1.293 [20]	–	–	–	–
323.15	0.9038	–	1.1432	–	–	–	–	–
2-Butoxyethanol								
303.15	0.8920	0.8923 [20]	2.4864	2.408 [20]	1288.4	1322.0 [21]	1.4150	–
313.15	0.8842	0.8839 [20]	1.9788	1.869 [20]	–	–	–	–
323.15	0.8775	–	1.6525	–	–	–	–	–

calibrated at 298.15 K with triply distilled water and purified methanol using density and viscosity values from the literature. The flow times were accurate to ±0.1 s, and the uncertainty in the viscosity measurements was $2 \times 10^{-4}\,\mathrm{mPa \cdot s}$. The mixtures were prepared by mixing known volume of pure liquids in airtight-stopper bottles, and each solution thus prepared was distributed into three recipients to perform all the measurements in triplicate, with the aim of determining possible dispersion of the results obtained. Adequate precautions were taken to minimize evaporation losses during the actual measurements. The reproducibility in mole fractions was within ±0.0002. The mass measurements were carried out on a Mettler AG-285 electronic balance with a precision of ±0.01 mg. The uncertainty of density measurements was less than $0.0002\,\mathrm{g \cdot cm^{-3}}$.

Ultrasonic speeds of sound (u) were determined by a multifrequency ultrasonic interferometer (Mittal Enterprise, New Delhi, M-81) working at 1 MHz, calibrated with triply distilled and purified water, methanol, and benzene at 303.15 K. The uncertainty of the ultrasonic speed measurements was $0.8\,\mathrm{m \cdot s^{-1}}$. The details of the methods and techniques have been described in earlier articles [5–8]. The refractive index was measured with the help of an Abbe refractometer (USA), which works with the wavelength corresponding to the D line of sodium. The uncertainty of refractive index measurements was 0.0002 units. The refractometer was calibrated using twice distilled and deionized water, and calibration was checked after every few measurements. All experimental measurements were done under atmospheric pressure.

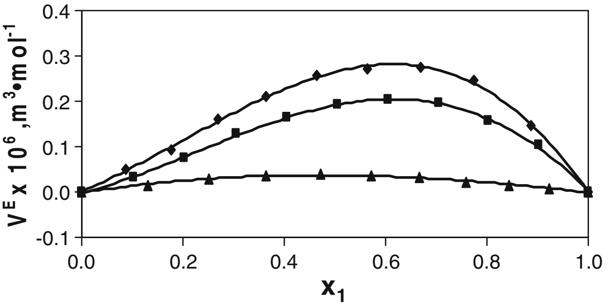

Fig. 1 Plots of excess molar volumes $V^{\mathrm{E}} \times 10^6 (\mathrm{m}^3 \cdot \mathrm{mol}^{-1})$ against mole fraction (x_1) of isoamyl alcohol with 2-methoxyethanol (◆), 2-ethoxyethanol (■), and 2-butoxyethanol (▲) at 303.15 K

3 Results and Discussion

3.1 Excess Molar Volumes

The experimental densities (ρ), viscosities (η), excess molar volumes (V^{E}), and viscosity deviations ($\Delta\eta$) for the binary mixtures studied at 303.15 K, 313.15 K, and 323.15 K are reported in Table 2.

The excess molar volumes, V^{E}, for the mixtures were calculated using the following equation [9]:

$$V^{\mathrm{E}} = \sum_{i=1}^{2} x_i M_i \left(1/\rho - 1/\rho_i\right) \tag{1}$$

where ρ is the density of the mixture and M_i, x_i, and ρ_i are the molar mass, mole fraction, and density of the ith component in the mixture, respectively.

Figure 1 illustrates that the excess molar volumes, V^{E}, for the binary systems of I.A.A with 2-M.E, 2-E.E, and 2-B.E. are positive over the entire range of composition at 303.15 K and follows the order 2-M.E > 2-E.E > 2-B.E. Similar trends were found for higher temperatures.

The sign of the excess volume (V^{E}) of a system depends on the relative magnitude of expansion/contraction on mixing of two liquids. If the factors causing expansion dominate the factors causing contraction, then V^{E} becomes positive. On the other hand, if the contractive factors dominate the expansive factors, then V^{E} becomes negative.

Mixing of I.A.A with alkoxyethanols induces a decrease in the molecular order in the latter, resulting in an expansion in volume and hence positive V^{E} values. The values of excess molar volumes V^{E} were found to decrease with an increase in the carbon chain length of alkoxyethanols. A similar result was found in the study of binary mixtures of chloroform with propan-1-ol and butan-1-ol [10]. The V^{E} values were found to increase with an increase in temperature over the complete composition range. A similar dependence of V^{E} values on temperature was reported elsewhere [11,12].

Table 2 Experimental values of density, ρ, viscosity, η, excess molar volume, V^{E}, and deviations in viscosity, $\Delta\eta$, for the binary mixtures under investigation at 303.15 K, 313.15 K, and 323.15 K

Mole fraction of I.A.A (x_1)	$\rho \times 10^{-3}$ ($\mathrm{kg \cdot m^{-3}}$)	η(mPa · s)	$V^{\mathrm{E}} \times 10^6$ ($\mathrm{m^3 \cdot mol^{-1}}$)	$\Delta\eta$(mPa · s)
I.A.A (1) + 2-M.E (2) 303.15 K				
0.0000	0.9568	1.5496	0.0000	0.0000
0.0875	0.9383	1.5784	0.0510	−0.1211
0.1775	0.9205	1.6106	0.0930	−0.2430
0.2701	0.9032	1.6521	0.1600	−0.3600
0.3653	0.8867	1.7492	0.2100	−0.4260
0.4633	0.8708	1.8200	0.2580	−0.5230
0.5643	0.8558	1.9339	0.2720	−0.5820
0.6683	0.8415	2.1190	0.2740	−0.5750
0.7754	0.8278	2.3539	0.2460	−0.5237
0.8860	0.8151	2.7148	0.1460	−0.3520
1.0000	0.8031	3.2622	0.0000	0.0000
313.15 K				
0.0000	0.9463	1.2883	0.0000	0.0000
0.0875	0.9282	1.3120	0.0530	−0.0772
0.1775	0.9108	1.3412	0.1070	−0.1517
0.2701	0.8939	1.3763	0.1790	−0.2232
0.3653	0.8776	1.4323	0.2560	−0.2771
0.4633	0.8622	1.4823	0.2940	−0.3400
0.5643	0.8476	1.5537	0.3100	−0.3850
0.6683	0.8336	1.6675	0.3060	−0.3910
0.7754	0.8205	1.8341	0.2550	−0.3480
0.8860	0.8081	2.0595	0.1520	−0.2500
1.0000	0.7964	2.4409	0.0000	0.0000
323.15 K				
0.0000	0.9378	1.0824	0.0000	0.0000
0.0875	0.9198	1.1114	0.0640	−0.0460
0.1775	0.9028	1.1526	0.1110	−0.0820
0.2701	0.8859	1.1689	0.1950	−0.1450
0.3653	0.8700	1.1921	0.2550	−0.2035
0.4633	0.8547	1.2236	0.3060	−0.2560
0.5643	0.8401	1.2801	0.3360	−0.2860
0.6683	0.8263	1.3552	0.3390	−0.3000
0.7754	0.8132	1.4811	0.2930	−0.2660
0.8860	0.8011	1.6708	0.1770	−0.1710
1.0000	0.7898	1.9396	0.0000	0.0000
I.A.A (1) + 2-E.E (2) 303.15 K				
0.0000	0.9195	1.6226	0.0000	0.0000
0.1020	0.9061	1.6885	0.0313	−0.1013

Table 2 continued

Mole fraction of I.A.A (x_1)	$\rho \times 10^{-3}$ $(\mathrm{kg \cdot m^{-3}})$	$\eta(\mathrm{mPa \cdot s})$	$V^E \times 10^6$ $(\mathrm{m^3 \cdot mol^{-1}})$	$\Delta\eta(\mathrm{mPa \cdot s})$
0.2036	0.8929	1.7610	0.0750	−0.1953
0.3047	0.8801	1.8121	0.1288	−0.3101
0.4053	0.8677	1.8916	0.1644	−0.3956
0.5055	0.8558	1.9937	0.1920	−0.4578
0.6053	0.8443	2.1244	0.2020	−0.4906
0.7046	0.8333	2.3012	0.1973	−0.4767
0.8035	0.8228	2.4957	0.1560	−0.4443
0.9020	0.8126	2.8067	0.1040	−0.2947
1.0000	0.8031	3.2622	0.0000	0.0000
313.15 K				
0.0000	0.9120	1.3554	0.0000	0.0000
0.1020	0.8986	1.3851	0.0385	−0.0810
0.2036	0.8856	1.4245	0.0796	−0.1518
0.3047	0.8729	1.4688	0.1320	−0.2173
0.4053	0.8605	1.5295	0.1740	−0.2659
0.5055	0.8487	1.6041	0.1976	−0.3001
0.6053	0.8373	1.6804	0.2087	−0.3320
0.7046	0.8264	1.8119	0.2008	−0.3084
0.8035	0.8159	1.9451	0.1698	−0.2825
0.9020	0.8058	2.1431	0.1156	−0.1760
1.0000	0.7964	2.4409	0.0000	0.0000
323.15 K				
0.0000	0.9038	1.1432	0.0000	0.0000
0.1020	0.8905	1.1695	0.0400	−0.0550
0.2036	0.8777	1.1963	0.0830	−0.1090
0.3047	0.8651	1.2221	0.1370	−0.1637
0.4053	0.8530	1.2627	0.1790	−0.2033
0.5055	0.8413	1.3098	0.2020	−0.2360
0.6053	0.8300	1.3771	0.2180	−0.2482
0.7046	0.8192	1.4598	0.2090	−0.2445
0.8035	0.8088	1.5657	0.1850	−0.2174
0.9020	0.7990	1.7275	0.1230	−0.1340
1.0000	0.7898	1.9396	0.0000	0.0000
I.A.A (1) + 2-B.E (2) 303.15 K				
0.0000	0.8920	2.4864	0.0000	0.0000
0.1297	0.8822	2.4826	0.0142	−0.1043
0.2510	0.8725	2.4959	0.0282	−0.1852
0.3649	0.8631	2.5135	0.0370	−0.2560
0.4720	0.8539	2.5572	0.0400	−0.2953
0.5728	0.8450	2.5937	0.0364	−0.3370

Table 2 continued

Mole fraction of I.A.A (x_1)	$\rho \times 10^{-3}$ $(\mathrm{kg \cdot m^{-3}})$	$\eta(\mathrm{mPa \cdot s})$	$V^{\mathrm{E}} \times 10^6$ $(\mathrm{m^3 \cdot mol^{-1}})$	$\Delta\eta(\mathrm{mPa \cdot s})$
0.6679	0.8386	2.6532	0.0313	−0.3514
0.7578	0.8277	2.7480	0.0231	−0.3262
0.8428	0.8193	2.8774	0.0158	−0.2629
0.9235	0.8111	3.0576	0.0080	−0.1452
1.0000	0.8031	3.2622	0.0000	0.0000
313.15 K				
0.0000	0.8842	1.9788	0.0000	0.0000
0.1297	0.8745	1.9694	0.0165	−0.0693
0.2510	0.8654	1.9674	0.0335	−0.1274
0.3649	0.8561	1.9873	0.0422	−0.1601
0.4720	0.8472	1.9969	0.0460	−0.2002
0.5728	0.8379	2.0284	0.0449	−0.2151
0.6679	0.8295	2.0671	0.0368	−0.2204
0.7578	0.8210	2.1207	0.0294	−0.2083
0.8428	0.8125	2.2001	0.0195	−0.1682
0.9235	0.8046	2.3034	0.0092	−0.1021
1.0000	0.7964	2.4409	0.0000	0.0000
323.15 K				
0.0000	0.8775	1.6525	0.0000	0.0000
0.1297	0.8677	1.6233	0.0190	−0.0664
0.2510	0.8582	1.6286	0.0356	−0.0960
0.3649	0.8488	1.6363	0.0495	−0.1209
0.4720	0.8398	1.6498	0.0540	−0.1382
0.5728	0.8310	1.6729	0.0514	−0.1440
0.6679	0.8223	1.7022	0.0473	−0.1420
0.7578	0.8139	1.7371	0.0383	−0.1329
0.8428	0.8057	1.7985	0.0280	−0.0960
0.9235	0.7976	1.8507	0.0190	−0.0669
1.0000	0.7898	1.9396	0.0000	0.0000

I.A.A Isoamyl alcohol, *2-M.E* 2-methoxyethanol, *2-E.E* 2-ethoxyethanol, *2-B.E* 2-butoxyethanol

3.2 Viscosity Deviations

The viscosity deviations ($\Delta\eta$) from a linear dependence on mole fraction were calculated [13] by

$$\Delta\eta = \eta - \sum_{i=1}^{2} x_i \eta_i \tag{2}$$

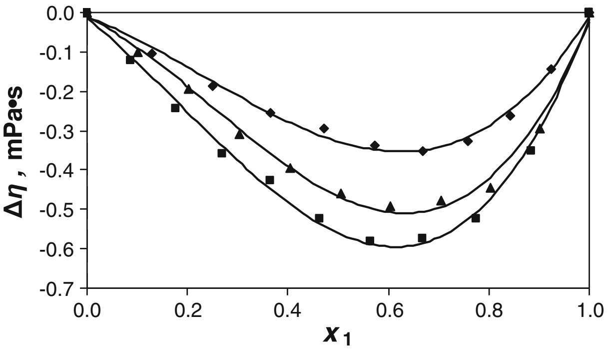

Fig. 2 Plots of viscosity deviations, $\Delta\eta(\mathrm{mPa}\cdot\mathrm{s})$ against mole fraction (x_1) of isoamyl alcohol with 2-methoxyethanol (■), 2-ethoxyethanol (▲), and 2-butoxyethanol (♦) at 303.15 K

where η is the viscosity of the mixture and x_i and η_i are the mole fraction and viscosity of pure component i, respectively.

Deviations in viscosity ($\Delta\eta$) for the mixture of I.A.A with alkoxyethanols are negative as depicted in Fig. 2, and the magnitude of the negative deviation increases with the increasing chain length of alkoxyalkanols. The trend in negative deviation of $\Delta\eta$ is 2-M.E > 2-E.E > 2-B.E at 303.15 K and also at higher temperatures. The $\Delta\eta$ values show a systematic increase with increase in temperature for the binary mixtures. Similar results have been reported earlier [11]. Also, the deviations in $\Delta\eta$ values are found to be opposite to the sign of excess molar volumes V^E for all three binary mixtures, which is in agreement with the view proposed by Brocos et al. [14,15]. A correlation between the sign of $\Delta\eta$ and V^E (Table 2) has been observed for a number of binary solvent systems [16,17], i.e., $\Delta\eta$ is positive when V^E is negative and vice-versa.

3.3 Deviations in Isentropic Compressibility

The isentropic compressibility, K_S, and deviation in isentropic compressibility, ΔK_S, were calculated using the following relations:

$$K_S = (u^2\rho)^{-1} \tag{3}$$

$$\Delta K_S = K_S - \sum_{i=1}^{2} x_i K_{Si} \tag{4}$$

where u and K_S are the speed of sound and isentropic compressibility of the mixture, respectively, and K_{Si} is the isentropic compressibility of the ith component in the mixture. The experimental speed of sound, isentropic compressibility, and deviation in isentropic compressibility are listed in Table 3 and are graphically represented in Fig. 3 as a function of mole fraction of I.A.A. From Fig. 3, it is evident that the ΔK_S values are positive and the magnitude of the positive values decreases with increasing chain length of the alcohols. The order of ΔK_S values is: 2-M.E > 2-E.E > 2-B.E.

Table 3 Experimental values of ultrasonic speed, u, isentropic compressibility, K_S, deviation in isentropic compressibility, ΔK_S, refractive index, n_D, and excess molar refraction, ΔR, for the binary mixtures at 303.15 K

Mole fraction of I.A.A (x_1)	$u(\mathrm{m \cdot s^{-1}})$	$K_S \times 10^{12}$ ($\mathrm{Pa^{-1}}$)	$\Delta K_S \times 10^{12}$ (Pa^{-1})	n_D	$\Delta R \times 10^{-6}$ ($\mathrm{m^3 \cdot mol^{-1}}$)
I.A.A (1) + 2-M.E (2) 303.15 K					
0.0000	1324.3	595.9	0.0	1.3983	0.0000
0.0875	1312.1	619.1	21.1	1.3988	0.0076
0.1775	1299.2	643.6	39.2	1.3993	0.0137
0.2701	1287.4	668.0	52.6	1.3999	0.0314
0.3653	1274.8	694.0	62.2	1.4004	0.0412
0.4633	1261.2	721.9	67.6	1.4009	0.0513
0.5643	1248.8	749.2	66.8	1.4014	0.0540
0.6683	1235.7	778.3	60.5	1.4020	0.0600
0.7754	1223.3	807.3	47.5	1.4025	0.0545
0.8860	1210.5	837.2	27.5	1.4030	0.0322
1.0000	1197.4	868.5	0.0	1.4035	0.0000
I.A.A (1) + 2-E.E (2) 303.15 K					
0.0000	1301.5	1301.5	0.0	1.4065	0.0000
0.1020	1292.4	1292.4	16.8	1.4062	0.0175
0.2036	1282.4	1282.4	31.0	1.4059	0.0290
0.3047	1272.7	1272.7	41.3	1.4055	0.0392
0.4053	1262.9	1262.9	47.9	1.4052	0.0432
0.5055	1251.2	1251.2	51.6	1.4049	0.0396
0.6053	1240.6	1240.6	50.3	1.4046	0.0320
0.7046	1229.3	1229.3	44.9	1.4043	0.0223
0.8035	1219.2	1219.2	34.5	1.4040	0.0148
0.9020	1208.7	1208.7	19.6	1.4037	0.0065
1.0000	1197.4	1197.4	0.0	1.4035	0.0000
I.A.A (1) + 2-B.E (2) 303.15 K					
0.0000	1288.4	675.3	0.0	1.4150	0.0000
0.1297	1279.8	692.1	14.6	1.4138	0.0136
0.2510	1270.3	710.2	26.2	1.4127	0.0194
0.3649	1261.8	727.7	33.3	1.4115	0.0223
0.4720	1251.8	747.3	38.0	1.4104	0.0221
0.5728	1242.3	766.8	39.1	1.4093	0.0208
0.6679	1233.5	785.9	36.7	1.4080	0.0176
0.7578	1224.2	806.1	31.7	1.4069	0.0140
0.8428	1215.3	826.4	23.7	1.4058	0.0116
0.9235	1206.4	847.1	13.1	1.4047	0.0047
1.0000	1197.4	868.4	0.0	1.4035	0.0000

I.A.A Isoamyl alcohol, *2-M.E* 2-methoxyethanol, *2-E.E* 2-ethoxyethanol, *2-B.E* 2-butoxyethanol

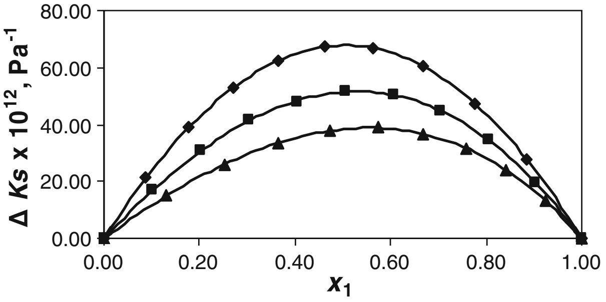

Fig. 3 Plots of deviation in isentropic compressibility $\Delta K_S \times 10^{12}(\mathrm{Pa}^{-1})$ against mole fraction (x_1) of isoamyl alcohol with 2-methoxyethanol (◆), 2-ethoxyethanol (■), and 2-butoxyethanol (▲) at 303.15 K

3.4 Excess Molar Refraction

The molar refraction, $[R]$, can be evaluated from the Lorentz–Lorenz relation [18] and gives more information than n_D about mixture phenomena because it takes into account the electronic perturbation of a molecular orbital during the liquid mixture process and $[R]$ is also directly related to the dispersion forces,

$$[R] = \left(n_\mathrm{D}^2 - 1/n_\mathrm{D}^2 + 2\right)(M/\rho) \tag{5}$$

where $[R]$, n_D^2, and M are, respectively, the molar refraction, the refractive index, and the molar mass of the mixture. Deviations for the molar refraction were calculated from the following relation:

$$M = x_1 M_1 + (1 - x_1) M_2 \tag{6}$$

$$\Delta R = [R] - (x_1[R]_1 + x_2[R]_2) \tag{7}$$

The value of ΔR is positive (Table 3) for all systems indicating that the dispersion forces are higher in the mixture than in the pure liquids. The deviations in refractive

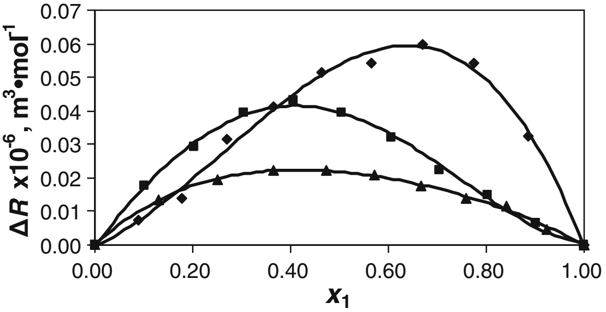

Fig. 4 Plots of molar refraction $\Delta R \times 10^{-6}(\mathrm{m}^3 \cdot \mathrm{mol}^{-1})$ against mole fraction (x_1) of isoamyl alcohol with 2-methoxyethanol (◆), 2-ethoxyethanol (■), and 2-butoxyethanol (▲) at 303.15 K

indices are shown in Fig. 4. The ΔR values for the binary mixtures under study follows the order 2-M.E > 2-E.E > 2-B.E with a maximum at around $x_1 \sim 0.3$ to 0.4.

4 Conclusion

After a thorough study of the behavior of alkoxyethanols and isoamyl alcohol, we get a clear idea about the molecular interactions between the components and it was found that the interactions between the solvent molecules increase with increasing chain length of alkoxyethanols.

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