

List of Publications

1. Thermodynamics and Transport Behavior of Non Aqueous Binary Mixtures of Benzene with Carbontetrachloride and Chloroform at Different Temperatures. *J. Indian Chem. Soc.*, 81, 330-334 (2004).
2. Physico-Chemical Studies on the Solute-Solvent Interactions and Ultrasonic Speed of Resorcinol in 2-Methoxyethanol and Tetrahydrofuran at Different Temperatures. *J. Tech. Res. Chem.*, 11(2), 7-17 (2005).
3. Study of Densities, Viscosity Deviations, and Isentropic Compressibilities of Ternary Liquid Mixtures of Water and Ethane-1, 2-diol with Some Monoalcohols at Various Temperatures. *J. of Physics and Chemistry of Liquids*, 1-13 (2006).
4. Study of Densities, Viscosities and Ultrasonic Speeds of Binary Mixtures Containing 1, 2-Dimethoxyethane and Alkan-1-ol at 298.15 K. Accepted for Publication in *J. Solution Chemistry*, Acceptance letter enclosed.
5. Study of Some Metal Halides in Glycerol + Water Mixtures. Accepted for Publication in *International J. of Thermophysics*. Acceptance letter enclosed.

Thermodynamic and transport behavior of non-aqueous binary mixtures of benzene with carbontetrachloride and chloroform at different temperatures

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Density and viscosity of pure benzene and its binary mixtures with carbontetrachloride and chloroform have been measured as a function of composition over the entire range at 298, 308 and 318 K. The excess volume, excess viscosity, excess free energy of activation of viscous flow and interaction parameter of Grunberg and Nissan have been calculated from the experimental data as a function of composition. All the excess functions are found to be either negative or positive over the entire range of composition depending on the molecular interactions and the nature of liquid mixtures. These properties are discussed in terms of nature of the molecular interactions between the component molecules.

In continuation of our earlier study^{1,2} on binary systems, we now report here the density and viscosity data of binary mixtures formed by benzene, carbontetrachloride and chloroform at various temperatures. There has been a recent upsurge of interest³⁻⁵ in the thermodynamic properties of binary liquid mixtures. These have been extensively used to obtain information on the intermolecular interactions and stereochemical effects in these systems⁵. The various thermodynamic properties such as excess molar volume (V^E), excess viscosity (η^E) etc. obtained from experimental observations have been rationalised. The main aim of the study is to correlate the data with the nature and type of interactions between the mixing components. In this paper the nature of various types of interactions in these binary systems are discussed.

Results and discussion

The excess functions V^E , η^E , G^{*E} and d were calculated from the experimentally determined ρ and η using equations⁹ (1-4).

$$V^E = V - (X_1 V_1 + X_2 V_2) \quad (1)$$

$$\eta^E = \eta - (X_1 \eta_1 + X_2 \eta_2) \quad (2)$$

$$G^{*E} = RT (\ln \eta V - X_1 \ln \eta_1 V_1 - X_2 \ln \eta_2 V_2) \quad (3)$$

$$\ln \eta = X_1 \ln \eta_1 + X_2 \ln \eta_2 + X_1 X_2 d \quad (4)$$

where X , V and η represent mole-fraction, molar volume and viscosity respectively, and subscripts 1, 2 represent the pure components. The values of these functions and d are recorded in Table 1 along with the values of ρ , η and mole fraction. The molar value (V) of pure liquid/mixture

Table 1. Density (ρ), viscosity (η), excess viscosity (η^E), volume (V), excess volume (V^E), excess free energy of activation of viscous flow (G^{*E}), interaction parameter (d) and mole-fraction of benzene (X_1) with carbontetrachloride and chloroform at 298, 308 and 318 K

X_1	ρ g cm ⁻³	η Cp	$\times 10^3 \eta^E$ Cp	V cm ³ mol ⁻¹	$\times 10^2 V^E$ cm ³ mol ⁻¹	$\times 10^{-7} G^{*E}$ ergs mol ⁻¹	d
Benzene + carbontetrachloride at 298 K							
0.00000	1.58522	0.91284	0.000	103.34212	0.000	0.00000	0.00000
0.18899	1.47057	0.88139	25.045	100.38398	-34.129	94.29401	0.26041
0.27013	1.41938	0.86261	30.521	99.10473	-49.704	117.82761	0.25647
0.34397	1.37121	0.84496	34.941	97.97036	-60.898	136.97908	0.26228
0.47336	1.28305	0.80843	37.092	96.05881	-72.894	151.95467	0.26604
0.58302	1.20449	0.77512	36.565	94.52053	-74.881	154.57804	0.27868
0.63181	1.16821	0.76013	36.161	93.87636	-71.741	154.59647	0.29049
0.67714	1.13395	0.74553	35.112	93.28686	-67.925	151.64939	0.30264

Note

Table-1 (contd.)

0.71937	1.10169	0.73086	33.061	92.73280	-64.857	144.21095	0.31229
0.75880	1.07101	0.71756	31.547	92.23338	-60.202	138.13864	0.32958
0.83033	1.01433	0.69173	27.101	91.34345	-50.151	119.08712	0.36937
0.89349	0.96332	0.66704	21.292	90.56053	-40.989	92.30070	0.42818
0.92239	0.93950	0.65666	19.551	90.22011	-35.015	82.81818	0.51028
0.94969	0.91652	0.64245	13.502	89.92904	-26.321	57.17201	0.53333
1.00000	0.87278	0.61391	0.000	89.49563	0.000	0.00000	0.00000
at 308 K							
0.00000	1.57396	0.78877	0.000	104.08143	0.000	0.00000	0.00000
0.18899	1.45826	0.75179	10.123	101.23160	-20.091	59.75659	0.15504
0.27013	1.40632	0.73548	14.044	100.02518	-27.006	81.85908	0.16559
0.34397	1.35777	0.72016	17.126	98.94018	-32.011	99.19281	0.17568
0.47336	1.26961	0.69188	21.098	97.07574	-37.100	121.66077	0.19546
0.58302	1.19190	0.66515	21.704	95.51904	-39.068	126.64292	0.20969
0.63181	1.15613	0.65237	21.088	94.85476	-37.111	124.61643	0.21540
0.67714	1.12262	0.63913	19.145	94.22843	-36.209	116.02392	0.21416
0.71937	1.09072	0.62697	17.509	93.66561	-33.301	108.20761	0.21622
0.75880	1.06055	0.61585	16.219	93.14384	-30.212	101.03189	0.22255
0.83033	1.00479	0.59394	12.140	92.21036	-23.302	78.53985	0.22483
0.89349	0.95465	0.57527	9.216	91.38292	-17.520	58.49386	0.24929
0.92239	0.93123	0.56608	7.221	91.02143	-13.162	45.91814	0.25978
0.94969	0.90897	0.55730	5.251	90.67660	-9.381	32.83340	0.27908
1.00000	0.86726	0.53951	0.000	90.06526	0.000	0.00000	0.00000
at 318 K							
0.00000	1.55478	0.67879	0.000	105.36539	0.000	0.00000	0.00000
0.18899	1.43995	0.64829	7.301	102.51830	-15.066	53.16862	0.13052
0.27013	1.38848	0.63458	9.823	101.31017	-20.112	71.42070	0.13673
0.34397	1.34047	0.62170	11.711	100.21725	-24.052	85.08949	0.14283
0.47336	1.25355	0.59903	14.922	98.31969	-29.200	105.54218	0.16148
0.58302	1.17693	0.57748	15.311	96.93410	-31.301	109.09294	0.17236
0.63181	1.14176	0.56648	14.066	96.05098	-30.002	103.31145	0.17067
0.67714	1.10857	0.55655	13.209	95.42202	-28.223	98.46345	0.17310
0.71937	1.07721	0.54690	12.006	94.84031	-26.142	91.27643	0.17384
0.75880	1.04756	0.53731	10.304	94.29805	-24.111	80.90671	0.17931
0.83033	0.99266	0.52092	8.221	93.33758	-18.102	65.47513	0.17861
0.89349	0.94326	0.50531	5.242	92.48625	-13.121	43.40566	0.17642
0.92239	0.92030	0.49921	4.926	92.10301	-10.212	38.10922	0.20580
0.94969	0.89801	0.49204	3.221	91.74340	-7.222	25.21784	0.20504
1.00000	0.85743	0.47876	0.000	91.09782	0.000	0.00000	0.00000
Benzene + chloroform at 298 K							
0.00000	1.47393	0.54268	0.000	80.99435	0.000	0.00000	0.00000
0.14517	1.37566	0.54309	-9.93046	82.42514	19.666	-35.00007	-0.13819
0.21242	1.33167	0.54573	-12.085	83.06369	26.350	-41.04441	-0.12311
0.27646	1.29010	0.54783	-14.542	83.69168	34.707	-48.27207	-0.12323
0.39577	1.21502	0.55323	-17.645	84.81081	45.191	-57.01902	-0.12713

Table-1 (contd.)

0.50468	1.14903	0.55904	-19.593	85.76952	48.474	-63.49582	-0.13017
0.55565	1.11897	0.56204	-20.216	86.19365	47.556	-66.19287	-0.13558
0.60449	1.09040	0.56598	-19.754	86.60387	47.058	-64.12259	-0.13599
0.65132	1.06339	0.57025	-18.827	86.98563	45.423	-60.49306	-0.13550
0.69628	1.03783	0.57512	-17.156	87.34018	42.656	-54.20199	-0.13152
0.78100	0.99060	0.58438	-13.935	87.97603	34.218	-43.49345	-0.13031
0.85942	0.94773	0.59431	-9.584	88.53954	23.902	-29.47952	-0.12506
0.89649	0.92827	0.60021	-6.326	88.74784	13.218	-19.47952	-0.10564
0.93223	0.90900	0.60443	-4.654	89.00548	8.598	-14.69747	-0.11404
1.00000	0.87278	0.61391	0.000	89.49563	0.000	0.00000	0.00000
at 318 K							
0.00000	1.45991	0.48812	0.00000	81.77216	0.000	0.00000	0.00000
0.14517	1.36404	0.49175	-3.83029	83.12732	15.125	-12.05804	-0.05739
0.21242	1.32082	0.49388	-5.15374	83.74621	21.243	-15.86915	-0.05698
0.27646	1.28021	0.49580	-6.52721	84.33843	27.356	-20.14491	-0.06030
0.39577	1.20681	0.50010	-8.35961	85.38522	33.090	-26.53942	-0.06427
0.50468	1.14181	0.50495	-9.10254	86.31221	35.469	-29.03089	-0.06649
0.55565	1.11210	0.50767	-9.00564	86.72594	34.572	-28.70186	-0.06622
0.60449	1.08396	0.51083	-8.35240	87.11829	33.303	-25.85351	-0.06288
0.65132	1.05741	0.51360	-7.98992	87.47791	30.429	-25.04433	0.06303
0.69628	1.03208	0.51680	7.04568	87.82682	28.034	21.29448	0.05908
0.78100	0.98514	0.52247	-5.78559	88.46250	21.343	-17.85518	-0.05947
0.85942	0.94241	0.52821	-4.07559	89.03923	13.981	-12.73463	-0.05873
0.89649	0.92254	0.53113	-3.06210	89.29913	9.229	-9.82571	-0.05704
0.93223	0.90331	0.53355	-2.47300	89.56693	6.369	-8.51867	-0.06845
1.00000	0.86726	0.53951	0.00000	90.06526	0.000	0.00000	0.00000
at 318 K							
0.00000	1.44510	0.44869	0.00000	82.64394	0.000	0.00000	0.00000
0.14517	1.35045	0.45096	-2.10021	83.96374	9.255	-7.04657	-0.04004
0.21242	1.30799	0.45198	-3.09654	84.56712	12.741	-10.99165	-0.03869
0.27646	1.26810	0.45319	-3.81023	85.14341	16.231	-13.44011	-0.03976
0.39577	1.19539	0.45577	-4.82001	86.20360	21.387	-16.89676	-0.04189
0.50468	1.13083	0.45873	-5.13224	87.15030	23.986	-17.62623	-0.04243
0.55565	1.10122	0.46020	-5.20051	87.58315	24.181	-17.93445	-0.04340
0.60449	1.07319	0.46177	-5.10005	87.99255	23.829	-17.55326	-0.04382
0.65132	1.04665	0.46347	-4.80221	88.37682	22.670	-16.40578	-0.04333
0.69628	1.02139	0.46517	-4.45623	88.74631	21.610	-14.98282	-0.04301
0.78100	0.97442	0.46854	-3.63297	89.43625	18.983	-11.76751	-0.04310
0.85942	0.93182	0.47202	-2.51106	90.05024	14.087	-7.75642	-0.04187
0.89649	0.91193	0.47383	-1.82231	90.38824	11.548	-5.09829	-0.03919
0.93223	0.89298	0.47552	-1.20011	90.60279	7.789	-3.28636	-0.03790
1.00000	0.85743	0.47876	0.00000	91.09782	0.000	0.00000	0.00000

is calculated using the following equation.

$$V = M/\rho \quad (5)$$

where M is the molecular weight and for mixture is given by $X_1M_1 + X_2M_2$.

From Table 1, it is found that the value of V^E for

Table 2. Physical properties of benzene, carbontetrachloride and chloroform

T/K	$\rho / \text{g cm}^{-3}$		η/Cp	
	This Work	Lit.	This work	Lit.
Benzene				
298	0.87278	0.87278 ^a	0.61391	0.61391 ^a
308	0.86726	0.86726 ^a	0.53951	0.53950 ^a
318	0.85743	0.85742 ^a	0.47876	0.47876 ^a
Carbontetrachloride				
298	1.58522	1.58522 ^a	0.91284	0.91284 ^a
308	1.57396	1.57396 ^a	0.78877	0.78877 ^a
318	1.55478	1.55479 ^a	0.67879	0.67879 ^a
Chloroform				
298	1.47393	1.47393	0.54268	0.54628 ^a
308	1.45991	1.45990	0.48812	0.48812 ^a
318	1.44451	1.44452	0.44869	0.44868 ^a

^aRefs. = 1, 9.

benzene and carbontetrachloride mixture is negative where it is positive for benzene and chloroform mixture at various temperatures over the entire composition. The negative value of V^E indicates that the main contribution to V^E is the decrease in volume due to hydrogen bond formation between unlike molecules⁶. There may be another source of negative contribution to V^E from the difference in size and shape of component molecules in the mixture. The molar volume of benzene is much smaller than that of carbontetrachloride shown in Table 1. Because of appreciable difference in the molar volumes of the components, benzene will fit into the structures of the carbontetrachloride molecule thereby reducing the volume of the mixture⁷. Muller⁸ made a similar report from the V^E studies of binary liquid mixtures.

The observed positive value of V^E for benzene and chloroform mixture over the entire composition range shown in Table 1 indicate the mutual dissociation of the component molecules. Because of the small difference in the molar volumes of the components, benzene will not fit into the structure of chloroform thereby increasing the volume of the mixture.

A correlation between the sign of η^E and V^E has been observed for a number of binary solvent systems^{9,10}, i.e. if η^E is positive then V^E is negative and *vice versa*. In the present observation this is found to hold good which is evident from Table 1.

The value of G^{*E} for the mixture of benzene + carbontetrachloride is positive whereas it is negative for the mixture of benzene + chloroform (Table 1). This indi-

cates that inter molecular complex is formed between benzene and carbontetrachloride through H-bonding and this is not favourable in the mixture of benzene and chloroform. Subha *et al.*^{6,11} made a similar observation from their G^{*E} studies for the mixtures of propionic acid and alcohols.

The positive value of Grunberg and Nissan parameter (d) gives an indication of specific hydrogen bonding interaction¹² between unlike molecules. This parameter d is found to be positive in the mixture of benzene and carbontetrachloride and negative in case of benzene and chloroform mixture (Table 1). This indicates that there is formation of inter-molecular complexes between benzene and carbontetrachloride through H-bonding in their mixture whereas such complex formation is not favourable in the mixture of benzene and chloroform. These conclusions are in excellent agreement with that drawn from G^{*E} values as reported^{6,11} earlier. A similar result was reported¹³ by the workers in the case of thermodynamic studies of formamide with various glycols at 308 K.

Experimental

Extrapure A.R. grade AN, benzene, chloroform and carbontetrachloride procured from Sisco Research Laboratories, Mumbai, were purified further as described earlier¹⁴.

The densities (ρ) were measured with an Ostwald-Sprengel type pycnometer having a bulb volume of 25 cm³ and an internal diameter of the capillary of about 0.1 cm. The pycnometer was calibrated at (298, 308 and 318) K with doubly-distilled water and benzene. The pycnometer with the test solution was equilibrated in a water bath maintained at ± 0.01 K of the desired temperature by means of a mercury glass thermoregulator and the temperature was determined by a calibrated thermometer and Muller bridge. The pycnometer was then removed from the thermostatic bath, properly dried and weighed. The evaporation losses remained insignificant during the time of actual measurements. An average of three measurements were taken into account. The density values are reproducible to $\pm 3 \times 10^{-5}$ g cm⁻³. Details have been described earlier^{1,15}.

The viscosities were measured by means of suspended level Ubbelohde¹⁶ viscometer at the desired temperature (accuracy ± 0.01 K). The precision of the viscosity measurements was 0.05%. Details have been discussed earlier¹.

The physical properties such as density and viscosity of pure benzene, carbontetrachloride and chloroform are

reported in Table 2. These results are in excellent agreement with the literature values¹⁷.

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Physico-Chemical Studies on the Solute-Solvent Interactions and Ultrasonic Speed of Resorcinol in 2-Methoxy Ethanol and Tetrahydrofuran at Different Temperatures

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ABSTRACT

The densities, viscosities and ultrasonic speeds of resorcinol in pure 2-methoxy ethanol and pure tetrahydrofuran have been investigated in 303.15K, 313.15K and 323.15K. Apparent molar volume (V_{ϕ}), viscosity parameters of these solutions are obtained from these data supplemented with their densities and viscosities respectively. The limiting apparent molar volumes (V_{ϕ}^0) and experimental slopes (S_{ϕ}^) derived from the Masson equation have been interpreted in terms of solute-solvent and solute-solute interactions respectively. The viscosity data have been interpreted in terms of solute-solvent and solute-solute interactions, respectively. The structure making/breaking capacity of this solute investigated here has been discussed. The compressibility data also indicate the electrostriction of the solvent molecules around the solute particles.*

Introduction

Studies on viscosities, densities and ultrasonic speeds of solutions assist in characterizing the structure and thermodynamic properties of solutions. Various types of interactions exist between the solutes in solutions and of these solute-solute and solute-solvent interactions are of current interest in all branches of chemistry. These interactions help in better understanding the nature of solute and solvent, that is, whether the solute modifies or distorts the structure of the solvent.

Tetrahydrofuran (THF), and 2-methoxy ethanol (ME) are very important

solvents widely used in various industries. These are industrial solvents and figure prominently in the high-energy battery technology and have also found wide applications in the organic synthesis as manifested from the physico-chemical studies in these media¹⁻¹⁰. In this present work, an attempt has been made to provide an unequivocal interpretation of solute-solvent and solute-solute interactions prevailing in the studied solutions. Several workers have reported volumetric, viscometric and ultrasonic studies of this compound in non-aqueous solutions¹¹⁻¹⁵ but such studies in pure THF and 2-methoxy ethanol are still scanty.

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Experiments

Method

Densities, ρ at 303.15, 313.15 and 323.15 K were measured with Sprengel type Pycnometer having a bulb volume of 25 cm³ and an internal diameter of the capillary of ~0.1 cm. It was calibrated at 303.15, 313.15 and 323.15 K with double-distilled water and benzene. The pycnometer with the test solution was equilibrated in a water-bath maintained at the desired temperature (± 0.01 K) by means of a mercury-in-glass thermo-regulator, and the absolute temperature was determined by a calibrated platinum resistance thermometer and Muller bridge. The pycnometer was then removed from the thermometer and Muller bridge. The pycnometer was then removed from the thermostatic bath, properly dried and weighed. The evaporation losses remained insignificant during time of actual measurements. An average of triplicate measurements was taken into account. The density values were reproducible to $\pm 3 \times 10^{-5}$ g cm⁻³. Details have been described earlier¹⁶. The viscosity was measured by means of a suspended level Ubbelohde¹⁷ viscometer at the desired temperature with a thermostat bath controlled to ± 0.01 K.

Sound velocities were determined with an accuracy of 0.3% using a single

crystal variable path ultrasonic interferometer (Mittal Enterprises, New Delhi, India) working at 4 MHz which was calibrated with water, methanol and benzene at each temperature, described in detail elsewhere^{18,19}. The solutions studied here were made by mass, the conversion of molality into molarity was done²⁰.

Source and purity of samples

Tetrahydrofuran (Merck, India) was kept several days over KOH, refluxed for 24 hours and distilled over LiAlH₄ described earlier¹. 2-Methoxy ethanol was allowed to stand overnight with CaSO₄ and distilled from sodium. Before fractional distillation the solvent was treated with 2, 4-dinitrophenhydrazine to remove aliphatic ketones. Resorcinol (A.R.) was purified by the reported procedure¹¹ and the compound was dried and stored in a vacuum desiccator.

Results

The experimental values of concentration(c), densities (ρ), viscosities (η) and derived parameters at 303.15, 313.15 and 323.15 K are recorded in Table - 1.

The apparent molar volume (V_ϕ) were determined from the solution densities using the following equation,

$$V_\phi = \frac{M}{\rho_0} - \frac{1000(\rho - \rho_0)}{c\rho_0} \quad (1)$$

Table -1

Molar concentration, densities, viscosities, apparent molar volumes (v_{ϕ}), limiting apparent molar volume (v_{ϕ}^0), experimental slope (S_{ϕ}^*) and values of A and B of resorcinol in various solvents at different temperatures

Resorcinol in ME							
$c/\text{mol dm}^{-3}$	$\rho/\text{g cm}^{-3}$	η/cp	$v_{\phi}/\text{cm}^3 \text{mol}^{-1}$	$v_{\phi}^0/\text{cm}^3 \text{mol}^{-1}$	$S_{\phi}^*/\text{cm}^3 \text{dm}^{1/2} \text{mol}^{-3/2}$	$A/\text{dm}^{3/2} \text{mol}^{-1/2}$	$B/\text{dm}^3 \text{mol}^{-1}$
303.15K							
0.008029	0.95834	1.40147	34.21621				
0.05621	0.96104	1.43588	53.28075				
0.10438	0.96271	1.46825	65.04447				
0.15256	0.96348	1.50134	75.54179	21.7866	135.4508	0.02071	0.46342
0.20074	0.96382	1.53721	83.23679				
0.24891	0.96401	1.57063	88.58110				
313.15Ks							
0.007954	0.94935	1.21587	44.36590				
0.05566	0.95166	1.23826	62.06549				
0.10333	0.95303	1.26285	72.99709				
0.15105	0.95395	1.28926	80.17937	34.1912	118.3329	-0.0273	0.48472
0.19873	0.95418	1.31665	87.56597				
0.24644	0.95444	1.34376	91.96859				
323.15K							
0.007893	0.94208	1.04675	55.04547				
0.05522	0.94401	1.06603	70.97192				
0.10247	0.94509	1.08673	80.97367				
0.14972	0.94556	1.10875	88.98941	46.1865	106.6030	-0.0344	0.4940
0.19699	0.94584	1.13195	94.18618				
0.24482	0.94624	1.15483	96.85475				
Resorcinol in THF							
303.15K							
0.0080634	0.8768	0.44945	5.36021				
0.05644	0.88083	0.45897	26.99526				
0.10482	0.88381	0.46680	40.09853	-7.46164	145.60272	0.079406	0.21908
0.15320	0.88611	0.47375	49.99311				
0.20159	0.88787	0.48075	58.19976				
0.24997	0.88934	0.48838	64.55114				

313.15K

0.007974	0.86708	0.41034	9.84666				
0.05580	0.87090	0.41594	31.32406				
0.10363	0.87381	0.42164	43.11721				
0.15415	0.87599	0.42677	53.02089	-2.14684	140.23313	0.0152	0.25078
0.19927	0.87764	0.43228	61.24157				
0.24715	0.87930	0.43740	66.24838				

323.15K

0.007906	0.86129	0.38944	14.45223				
0.05542	0.86494	0.39331	35.06637				
0.10291	0.86768	0.39780	46.99245				
0.15039	0.86981	0.40258	56.09555	2.16829	139.40834	-0.01988	0.28058
0.19782	0.87128	0.40736	64.69020				
0.24526	0.87258	0.41245	70.76840				

where M is the molecular weight of the solute, c is the molarity of the solution and the other symbols have their usual significance.

squares treatment of the plot of V_ϕ^* vs $c^{1/2}$ using the Masson equation²¹.

The limiting apparent molar volumes (V_ϕ^0) were calculated using the least

$$V_\phi = V_\phi^0 + S_\phi^* c^{1/2} \tag{2}$$

Table – 2

Molal concentration (m), sound velocity (u), adiabatic compressibility (β), apparent molal adiabatic compressibility (ϕ_k), limiting apparent molal adiabatic compressibility (ϕ_k^0) and experimental slope (S_k^*) of resorcinol in various solvents at different temperatures

Resorcinol in ME

m/mol kg ⁻¹	u/ms ⁻¹	$\beta \times 10^{10}/\text{pa}^{-1}$	$\phi_k \times 10^7/\text{m}^3 \text{mol}^{-1} \text{pa}^{-1}$	$\phi_k^0 \times 10^7/\text{m}^3 \text{mol}^{-1} \text{pa}^{-1}$	$S_k^* \times 10^7/\text{m}^3 \text{mol}^{-3/2} \text{pa}^{-1} \text{kg}^{-1}$
303.15K					
0.008386	1302.42222	6.15144	-3.53056		
0.05889	1315.10976	0.01637	-2.60571		
0.10973	1323.69716	5.92826	-2.02055	-4.0552	6.01661
0.16115	1329.89488	5.86844	-1.58099		
0.21316	1335.37726	5.81830	-1.29114		
0.26576	1338.06352	5.79382	-1.00556		

313.15K					
0.008386	1268.73567	6.54382	-4.17088		
0.05887	1282.8593	6.38552	-3.07110		
0.10973	1292.53715	6.28069	-2.4066	-4.8330	7.2967
0.16115	1299.04835	6.21189	-1.90114		
0.21316	1301.84487	6.18374	-1.4104		
0.26576	1304.96485	6.15253	-1.12253		
323.15K					
0.008386	1240.67703	6.89596	-4.91245		
0.58870	1256.37339	6.71098	-3.61256		
0.10973	1267.15845	6.58969	-2.83223	-5.7239	8.7415
0.16115	1274.50031	6.51075	-2.23143		
0.21316	1277.51300	6.47816	-1.67623		
0.26576	1279.76619	6.45264	-1.30988		
Resorcinol in THF					
303.15K					
0.0092057	1252.90681	7.26544	-4.55220		
0.06453	1263.84373	7.10757	-3.25120		
0.12017	1270.90584	7.00510	-2.5363	-5.2802	7.9261
0.17625	1273.69507	6.95635	-1.8841		
0.23287	1274.80837	6.93044	-1.4088		
0.29005	1276.09679	6.90502	-1.10556		
313.15K					
0.0092057	1193.59694	8.09516	-5.32146		
0.06453	1205.16882	7.90562	-3.90844		
0.12017	1211.94033	7.79149	-2.9865	-6.1470	8.9247
0.17625	1216.28610	7.71666	-2.3412		
0.23287	1218.31345	7.67654	-1.80601		
0.29005	1219.65271	7.64523	-1.4413		
323.15K					
0.0092057	1148.06843	8.80897	-7.32857		
0.06453	1162.03754	8.56198	-5.20866		
0.12017	1169.80470	8.42197	-3.91044	-8.5072	13.02574
0.17625	1173.01666	8.35540	-2.90088		
0.23287	1174.41862	8.3514	-2.17714		
0.29005	1175.04828	8.33394	-1.67397		

Where V_{ϕ}^0 is the partial molar volume at infinite dilution and S_v^* the experimental slope.

The values of V_{ϕ}^0 and S_v^* along with temperature of resorcinol in solvents follows the polynomial-

$$V_{\phi}^0 = a_0 + a_1 T + a_2 T^2 \quad (3)$$

Over the temperature range under investigation, where T is the temperature in Kelvin.

Values of coefficients of the above equation for resorcinol in pure THF and 2-methoxy ethanol are recorded in Table - 3.

Table - 3

Values of coefficient for resorcinol in THF and ME

Solvent	$a_0/\text{cm}^3 \text{ mol}^{-1}$	$a_1/\text{cm}^3 \text{ mol}^{-1} \text{ K}^{-1}$	$A_2/\text{cm}^3 \text{ mol}^{-1} \text{ K}^{-2}$
THF	-643.094	3.61205	-4.9985×10^{-3}
ME	-548.5366	2.05172	-2.0465×10^{-3}

From the values of coefficients the following equations are obtained

Resorcinol in THF

$$V_{\phi}^0 = -643.0947 / \text{cm}^3 \text{ mol}^{-1} + 2.50172T / \text{cm}^3 \text{ mol}^{-1} \text{ K}^{-1} - 4.9985 \times 10^{-3} / \text{cm}^3 \text{ mol}^{-1} \text{ K}^{-2} \quad (4)$$

Resorcinol in ME

$$V_{\phi}^0 = -548.53663 / \text{cm}^3 \text{ mol}^{-1} + 2.50172T / \text{cm}^3 \text{ mol}^{-1} \text{ K}^{-1} - 2.0465 \times 10^{-3} / \text{cm}^3 \text{ mol}^{-1} \text{ K}^{-2} \quad (5)$$

The apparent molar expansibilities (ϕ_E^0) can be obtained by the following equation.

$$\phi_E^0 = \left(\frac{\delta V_{\phi}^0}{\delta T} \right)_p = a_1 + 2a_2 T \quad (6)$$

The values of ϕ_E^0 of the studied compound at 303.13, 313.15 and 323.15K are determined and reported in table - 4.

Table - 4

Limiting apparent molar expansibilities (ϕ_E^0) for resorcinol in THF and ME at different temperatures

Solvents	303.15K	313.15K	323.15K	$(\delta^2 V_{\phi}^0 / dT^2)_p$
THF	0.57262	0.49035	0.40809	Negative
ME	1.26093	1.21999	1.17907	Negative

Helper²² developed a technique of examining the magnitudes of $(\delta^2 V_\phi^0 / \delta T^2)_p$ for solute in terms of long range structure-making and breaking capacity of the solutes in mixed solvent systems using the general thermodynamic expression.

$$(\delta c_p / \delta P) = -(\delta^2 V_\phi^0 / \delta T^2)_p \quad (7)$$

The viscosity of resorcinol in pure THF and in pure-2-methoxy ethanol have been analyzed using the Jones-Dole²³ equation:

$$\frac{\eta}{\eta_0} = 1 + Ac^{1/2} + Bc \quad (8)$$

$$\left(\frac{\eta}{\eta_0} - 1 \right) / c^{1/2} = A + Bc^{1/2}$$

$$\text{where } \eta = \left(Kt - \frac{L}{t} \right) \rho$$

where, η_0 and η are the viscosities of solvent and solution respectively. K and L are the constants for a particular viscometer. The values of A and B are estimated by computerized least square method and recorded in table 1.

Adiabatic compressibility (β) was calculated from the following relation.

$$\beta = \frac{1}{u^2 \rho} \quad (9)$$

where ρ is the solution density and u is the sound velocity in the solution. The apparent molal adiabatic compressibility (ϕ_k) of the solution was determined from the relation.

$$\phi_k = M\beta / \rho_0 + 1000(\beta\rho_0 - \beta_0\rho) / m\rho\rho_0 \quad (10)$$

The limiting apparent molal adiabatic compressibility (ϕ_k^0) was obtained by extrapolating the plots of (ϕ_k) versus the square root of molal concentration of the solute to zero concentration by the computerized least square method.

$$\varphi_k = \phi_k^0 + S_k^* m^{1/2} \quad (11)$$

where S_k^* is the experimental slope.

The values of u , β , ϕ_k , ϕ_k^0 and S_k^* are recorded in Table 2.

Discussion

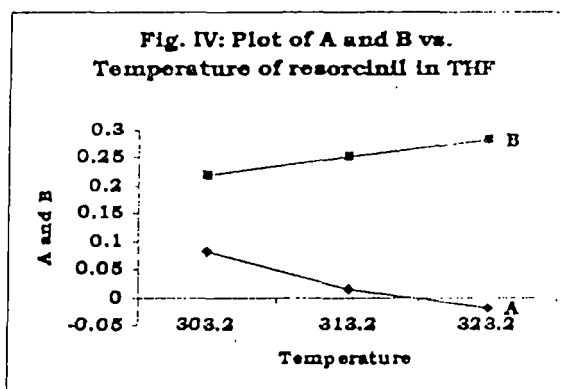
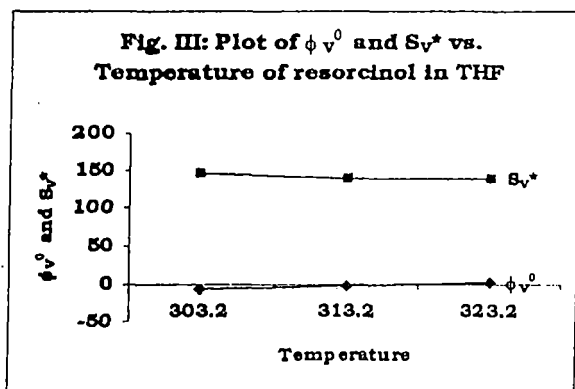
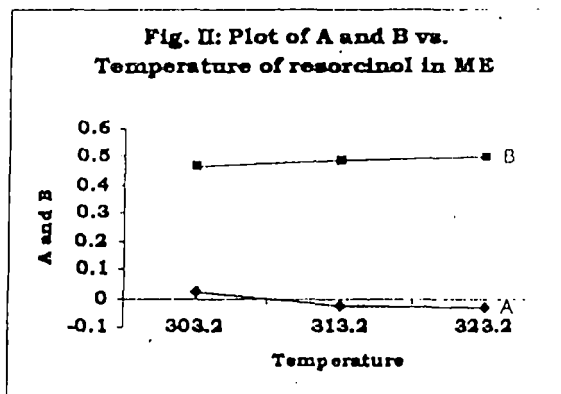
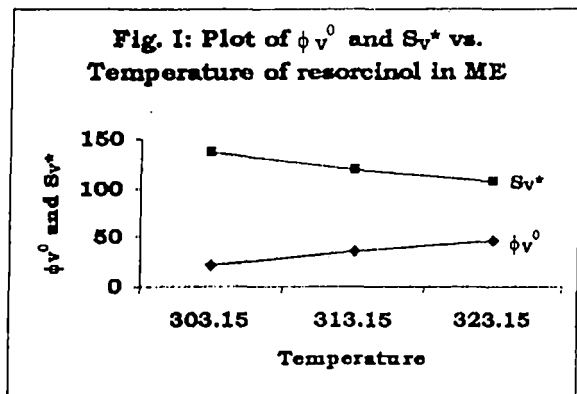
We have determined the ρ and η and calculated the V_ϕ , V_ϕ^0 , S_v^* , ϕ_k , ϕ_k^0 , B and A at 303.15, 313.15 and 323.15K using appropriate equations and graphical diagrams.

As the investigated systems are characterized by hydrogen bond, the solute-solvent and solute-solute interactions can be interpreted in terms of structural changes which arise due to hydrogen bond interactions present between various components of the solvent and solute-solute interactions can be interpreted in terms of structural changes which arise due to hydrogen bond interactions present between various components of the solvent and solution systems.

To examine the solute-solvent interactions, the V_ϕ^0 can be used. Table-I and Figs. I and III in case of ME reveals that the V_ϕ^0 values are positive and increase with rise in temperature. This indicates the presence

of strong solute-solvent interaction and these interactions are strengthened with rise in temperature. Whereas in case of THF the V_{ϕ}^0 values are small at various temperatures and the values of V_{ϕ}^0 increase with increase of temperature. This indicates the presence

of weak solute-solvent interaction and such interaction increases with rise of temperature. Similar results were obtained for some 1:1 electrolyte in aqueous DMF²⁴ and aqueous THF¹.



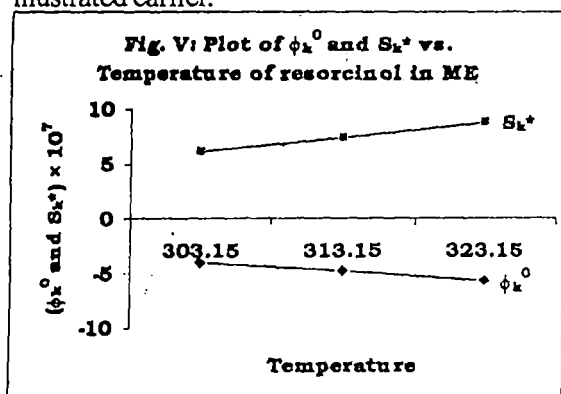
It is also evident (Table-I and Figs. I and III) that S_v^* are positive in both systems at different temperatures. Since S_v^* is a measure of solute-solute interactions, the results indicate the presence of strong solute-solute interaction. As expected, the S_v^* values decrease with increasing temperature in these solvents for the studied solute, which is

attributed to more violent thermal agitation at higher temperature resulting in diminishing the force of solute-solute interactions²⁵.

It is found from Table 4 that the value of ϕ_E^0 of solute decreases with rise in temperature in studied solvents, which can be ascribed to the absence of caging or packing effect.²⁶

In our present investigations, it is evident from Table 4 that the $(\delta^2 V_{\phi}^0 / \delta T^2)_{\rho}$ values are negative for resorcinol in studied solvents, suggesting thereby that resorcinol acts as a structure-breaker in such solvents.

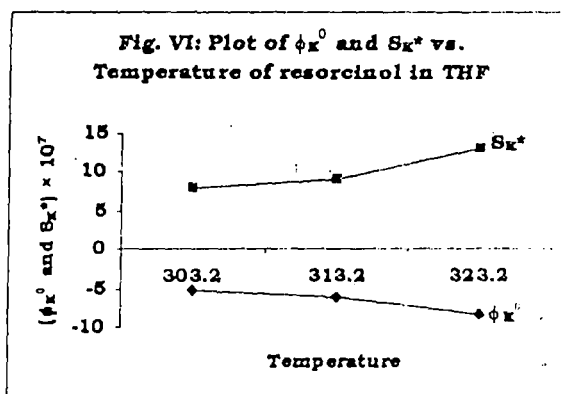
It is observed (Table 1 and Figs. II and IV) that the values of B of resorcinol in the studied solvent systems are positive and these values increase with increasing temperature. This indicates that this solute acts structure-breaker in such solvents. These conclusions are excellent agreement with that drawn from magnitude of $(\delta^2 V_{\phi}^0 / \delta T^2)$ illustrated earlier.



A perusal of Table 2 and Figs. V and VI show that the values of ϕ_k^0 are negative and become more negative on increasing the temperature. Negative ϕ_k^0 values of resorcinol can be interpreted in terms of the loss of compressibility of solvents due to electrostrictive forces in the vicinity of the solute particles. On raising the temperature of the system, the solute particles lose some solvent molecules from their first coordination sphere in a process, which is expected to increase the compressibility. But at higher

It has been reported by a number of workers that dB/dT is a better criterion^{22,28} determining the structure making/breaking nature of any solute rather than simply the value of B. It is found from Table 1 that the values of B increase with rise in temperature (positive dB/dT) suggesting structure-breaking tendency.

A similar result was reported by some workers²⁹ in studied solvents in case of viscosity of some salts in propionic acid + ethanol mixtures.



temperature, breakdown of the non-covalent bonding between the solvent molecules also takes place more effectively resulting in a loss of compressibility. Thus it may be concluded that for the solute solution under study, the latter effect is growing faster and overriding the former as far as the present temperature range is concerned. From Table 2 (figs. V and VI), it is evident that S_k^* have positive values indicating the existence of strong solute-solute interactions in the studied solvent system which resembles the agreement

drawn from S_v^* discussed earlier. A similar results were reported by work³⁰ in the case of ultrasonic studies of some alkali metal halides and nitrates in THF + Water mixture.

List of Symbols

- ρ : Density of solution
- ρ_0 : Density of solvent
- η : Viscosity of solution
- η_0 : Viscosity of solvent
- c : Molar concentration of solution
- m : Molal concentration of solution
- u : Sound velocity of solution
- u_0 : Sound velocity of solvent
- β : Adiabatic compressibility of solution
- β_0 : Adiabatic compressibility of solvent
- ϕ_K : Apparent molal adiabatic compressibility
- ϕ_K^0 : Limiting apparent molal adiabatic compressibility
- S_K^* : Experimental slope
- V_ϕ : Apparent molar volume
- V_ϕ^0 : Limiting apparent molar volume
- S_V^* : Experimental slope
- M : Molecular weight of solute
- ϕ_E : Apparent molar expansibility
- ϕ_E^0 : Limiting apparent molar expansibility
- ME : 2-Methoxy ethanol
- THF : Tetrahydrofuran

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Study of densities, viscosity deviations, and isentropic compressibilities of ternary liquid mixtures of water and ethane-1,2-diol with some monoalcohols at various temperatures

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Excess molar volume (V^E), viscosity deviation ($\Delta\eta$), and excess Gibbs energy of activation ($\Delta G^{\ddagger E}$) of viscous flow have been investigated from the density (ρ) and viscosity (η) measurements of ternary liquid mixture of water + ethane-1,2-diol + methanol, water + ethane 1,2-diol + ethanol, and water + ethane-1,2-diol + 1-propanol over the entire range of composition at 298.15, 308.15 and 318.15 K. The speeds of sound are also observed for these mixtures and thus, the isentropic compressibility (K_S) and excess isentropic compressibility (K_S^E) calculated at 298.15 K. The results are discussed in terms of specific interactions and nature of liquid mixtures. The system studied here exhibits a very strong cross association through hydrogen bonding.

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Keywords: Densities; Viscosities; Excess molar volume; Viscosity deviation; Excess Gibbs energy of activation of viscous flow; Ethane-1,2-diol; Monoalcohol; Sound speed; Isentropic compressibility; Excess isentropic compressibility; Specific interactions

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1. Introduction

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A knowledge of the thermodynamic behavior of liquid mixtures has been the main aim during recent years. The investigated mixtures were chosen in order to obtain information about the molecular interactions between their components [1–4]. This is the case for the systems studied in this research, which contain water, ethane-1,2-diol, and some monoalcohols.

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Rheology is a branch of science [5] that studies material deformation and flow, and is increasingly applied to analyze the viscous behavior of many pharmaceutical products [6] and to establish their stability and even their bio-availability, since it has been firmly established that viscosity influences the drug absorption rate in the body.

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The liquids were selected on the basis of their industrial use [7]. Ethane-1,2-diol and monoalcohols are important liquids which find a variety of applications in pharmaceuticals, cosmetics etc. In our systematic investigation of the thermodynamics, acoustic, and transport properties of ternary mixtures, we have reported viscosities (η),

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49 densities (ρ), speeds of sound (u), isentropic compressibilities and excess molar volumes
50 for the ternary liquid mixtures of water + ethane-1,2 diol + methanol, water + ethane-
51 1,2-diol + ethanol, and water + ethane-1,2-diol + 1-propanol. Viscosity and density of
52 these ternary liquid mixtures are useful in understanding molecular interactions
53 between the components of the mixture which may be used to develop new theoretical
54 models and also for engineering applications [8]. In our previous investigation of the
55 properties, we have reported viscosities, densities and speeds of sound of various polar
56 mixtures [3,9,10].

57 The present work contributes to the study of various thermodynamics and transport
58 properties viz. excess molar volumes (V^E), viscosity deviations ($\Delta\eta$), excess Gibbs
59 energy of activation (ΔG^{*E}) of viscous flow, Grunberg and Nissan parameters (d) and
60 isentropic compressibility (K_S) of various ternary mixtures.

61 To the best of our knowledge, the properties of mixtures of this liquid have not been
62 reported earlier.

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2. Experimental

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2.1. Source and purity of samples

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2.2. Method

The speeds of sound (u) in pure liquids and in ternary mixtures were measured with multi-frequency ultrasonic interferometer supplied by Mittal Enterprise, New Delhi. In the present work, a steel cell fitted with a quartz crystal of 2 MHz [11] frequency was employed. Densities (ρ) were measured with an Ostwald-Sprengel type pycnometer having a bulb volume of 25 cm³ and an internal diameter of the capillary of about 1 mm. The pycnometer was calibrated at 298.15, 308.15, 318.15 K with triply distilled water and benzene. The pycnometer with the test solution was equilibrated in a thermostatic water bath maintained at ± 0.01 K of the desired temperature by means of a mercury in glass thermoregulator and the temperature was determined with a calibrated thermometer and a Muller bridge [12]. The viscosities were measured by means of a suspended Ubbelohde type viscometer [13] which was calibrated at the desired temperatures with water and methanol. The solutions were prepared by mixing known volumes of pure liquids in air-tight, narrow-mouth ground stoppered bottles taking due precautions to minimize the evaporation losses. The masses were determined by using a Mettler electronic analytical balance (AG285, Switzerland) accurate to 0.0002 g. The uncertainties in the liquid composition, density, viscosity, and speeds of sound measurements were estimated to be 1×10^{-4} , 2×10^{-4} g cm⁻³, 3×10^{-4} m Pa.s, and 0.2 m s^{-1} respectively.

Viscosity deviations and other properties of some aqueous ternary mixtures

Table 1. Comparison of experimental densities (ρ), viscosities (η), and ultrasonic speeds (u) of pure liquids with literature values.

Liquids	T(K)	$\rho \times 10^{-3} \text{ (kg m}^{-3}\text{)}$		$\eta \times 10^3 \text{ (kg m}^{-1}\text{s}^{-1}\text{)}$		$u \text{ (m s}^{-1}\text{)}$	
		Expt	Lit.	Expt	Lit.	Expt	Lit.
Water	298.15	0.99707	0.9971 [15]	0.8904	0.890 [15]	1498.2	1497.4 [21]
	308.15	0.99406	0.9940 [15]	0.7194	0.719 [15]		
	318.15	0.99025	0.9902 [15]	0.596	0.597 [15]		
Ethane-1,2-diol	298.15	1.10998	1.1100 [16,21]	16.47362	16.472 [16,21]	1660.7	1662.0 [21]
	308.15	1.10299	1.1029 [16,21]	10.47212	10.470 [16,21]		
	318.15	1.09764	1.0956 [16,21]	7.69443	7.694 [16,21]		
Methanol	298.15	0.78664	0.78656 [17,20]	0.54230	0.5422 [17,20]	1105.1	1103.0 [21]
	308.15	0.77728	0.7772 [17,20]	0.47424	0.4742 [17,20]		
	318.15	0.76775	0.7677 [17,20]	0.41739	0.4174 [17,20]		
Ethanol	298.15	0.78514	0.7851 [18,20]	1.08805	1.088 [18,20]	1144.9	—
	308.15	0.77658	0.7765 [18,20]	0.90421	0.904 [18,20]		
	318.15	0.76781	0.7677 [18,20]	0.76304	0.763 [18,20]		
Propanol	298.15	0.79958	0.79954 [19,20]	1.93968	1.9324 [19,20]	1207.2	1206.5 [19,20]
	308.15	0.79166	0.79162 [19,20]	1.56776	1.560 [19,20]		
	318.15	0.78456	—	1.14998	—		

3. Results and discussion

The comparison of the experimentally determined densities, viscosities at 298.15, 308.15, and 318.15 K, and sound velocities at 298.15 K of the pure components with the literature values [15–21] have been presented in table 1.

The experimentally determined density, viscosity, and calculated excess thermodynamic properties for ternary liquid mixtures are listed in table 2. Representative plots of η , $\Delta\eta$, V^E , ΔG^{*E} , and K_S^E against mole fraction of water (x_1) at 298.15 K are shown in figures 1–5, since for other temperature the curves are similar.

3.1. Viscosity deviations

In table 2, the measured η and calculated $\Delta\eta$ of the ternary mixtures are shown along with the mole fraction of water (x_1) and ethane-1,2-diol (x_2) at the three temperatures.

The viscosity deviations from linear dependence on mole fraction were calculated [22] by,

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

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

It is observed that $\Delta\eta$ values increases to attain a maximum and then decreases over the entire range of composition for each temperature (figure 2). This trend is observed for all the ternary mixtures examined here. The values become more and more positive as the temperature rises from 298.15 to 318.18 K. The positive value of $\Delta\eta$ support the existence of the specific interactions [23,18,24] between the unlike molecules.

For (1)+(2)+methanol, the maxima of $\Delta\eta$ is observed at $x_1=0.89$, for (1)+(2)+ethanol at $x_1=0.92$, and for (1)+(2)+1-propanol at $x_1=0.93$ for all

Table 2. Densities (ρ), viscosities (η), excess molar volumes (V^E), Gibbs energy of activation (ΔG^{*E}) of viscous flow, interactions parameters (d), and viscosity deviation ($\Delta\eta$) for water (1) + ethane-1,2-diol (2) + methanol, ethanol, and 1-propanol at 298.15, 308.15 and 318.15 K.

Mole fraction of water (x_1)	Mole fraction of ethane-1,2-diol (x_2)	$\rho \times 10^{-3}$ (kgm ⁻³)	$\eta \times 10^3$ (kgm ⁻¹ s ⁻¹)	$V^E \times 10^3$ (m ³ mol ⁻¹)	ΔG^{*E} (J mol ⁻¹)	d	$\Delta\eta \times 10^3$ (kgm ⁻¹ s ⁻¹)
Water (1) + ethane-1,2-diol (2) + methanol							
298.15 K							
0	0.29693	0.91470	1.99757	-0.46412	720.91933	-	-3.27522
0.20125	0.23344	0.92896	2.12681	-0.71701	1279.13480	17.69101	-2.20455
0.36081	0.18311	0.94147	2.31497	-0.83732	1776.10648	21.48512	-1.27011
0.49042	0.14222	0.95227	2.34765	-0.86844	2018.48838	28.75390	-0.63112
0.59779	0.10835	0.96148	2.18441	-0.84101	1991.38454	38.20262	-0.29214
0.68819	0.07983	0.96959	2.02624	-0.78402	1915.170859	55.28743	-0.02740
0.76536	0.05549	0.97666	1.78830	-0.70712	1684.88063	82.03357	0.09554
0.81399	0.03447	0.98221	1.58294	-0.60502	1440.21583	141.25689	0.20187
0.89010	0.01613	0.98664	1.34916	-0.49132	1084.85606	307.40161	0.24005
0.94125	0	0.98699	1.10039	-0.31205	614.79326	-	0.23044
1.00000	0	0.99707	0.89040	0	0	-	0
308.15 K							
0	0.29693	0.90956	1.63445	-0.66112	805.38420	-	-1.80846
0.20125	0.23344	0.92350	1.70923	-0.85766	1329.56768	19.91352	-1.14827
0.36081	0.18311	0.93652	1.84429	-0.96310	1813.45279	21.27794	-0.54912
0.49042	0.14222	0.94782	1.87559	-0.98087	2064.8945	28.51184	-0.14078
0.59779	0.10835	0.95720	1.74204	-0.93115	2027.76717	37.66037	0.03798
0.68819	0.07983	0.96560	1.62360	-0.85810	1957.06212	54.69905	0.18251
0.76536	0.05549	0.97289	1.43721	-0.76545	1723.15866	81.22125	0.22055
0.81399	0.03447	0.97915	1.27898	-0.66005	1479.61422	140.66394	0.25614
0.89010	0.01613	0.98286	1.11393	-0.52812	1167.45246	320.81800	0.26021
0.94125	0	0.98455	0.93606	-0.34503	752.71494	-	0.23106
1.00000	0	0.99406	0.71940	0	0	-	0
318.15 K							
0	0.29693	0.90411	1.35988	-0.83101	813.73667	-	-1.21828
0.20125	0.23344	0.91846	1.45387	-1.00971	1417.12126	18.67391	-0.69822
0.36081	0.18311	0.93217	1.53852	-1.11006	1867.14629	21.32097	-0.27581
0.49042	0.14222	0.94476	1.57187	-1.14125	2138.52520	28.76058	0.03195
0.59779	0.10835	0.95575	1.44264	-1.11025	2066.86750	37.38858	0.13001
0.68819	0.07983	0.96437	1.34133	-1.01401	1989.14140	54.12886	0.22010

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0.76536	0.05549	0.97049	1.20811	-0.86732	1798.47426	82.58307	0.25021
0.81399	0.03447	0.97689	1.08745	-0.74521	1578.72382	146.41136	0.27061
0.89010	0.01613	0.97978	0.96463	-0.56405	1308.5924	351.88657	0.27088
0.94125	0	0.98109	0.81807	-0.37325	906.93254	-	0.23256
1.00000	0	0.99026	0.59600	0	0	-	0
Water (1) + ethane 1,2-diol (2) + ethanol							
298.15 K							
0	0.37783	0.91083	3.56697	-0.41845	380.77010	-	-3.33421
0.24310	0.28199	0.92474	3.12739	-0.70440	1048.99560	10.38940	-2.25118
0.41901	0.21264	0.93725	3.00454	-0.83334	1632.95151	15.90263	-1.27228
0.55220	0.16013	0.94806	2.80252	-0.85564	1888.59731	24.43785	-0.64008
0.65654	0.11900	0.95738	2.41869	-0.81704	1844.05730	34.61152	-0.37048
0.74051	0.08590	0.96553	2.10754	-0.74732	1732.96578	52.18853	-0.15577
0.80953	0.05869	0.97251	1.79958	-0.65658	1508.47676	80.82641	-0.03146
0.86726	0.03593	0.97881	1.60019	-0.56232	1338.15418	152.96767	0.13075
0.91627	0.01661	0.98401	1.38094	-0.45611	1060.62018	369.5337	0.21843
0.95839	0	0.98560	1.10848	-0.29501	585.19882	-	0.20986
1.00000	0	0.99707	0.89040	0	0	-	0
308.15 K							
0	0.37783	0.90524	2.68819	-0.60101	394.65607	-	-1.83106
0.24310	0.28199	0.91908	2.40732	-0.83314	1094.98909	10.56769	-1.15001
0.41901	0.21264	0.93176	2.31123	-0.93250	1637.93848	15.64411	-0.55006
0.55220	0.16013	0.94334	2.18552	-0.95389	1936.65108	24.15345	-0.14875
0.65654	0.11900	0.95296	1.88388	-0.89701	1867.49864	33.78925	-0.03758
0.74051	0.08590	0.96138	1.66572	-0.81121	1773.99394	51.61752	0.07648
0.80953	0.05869	0.96891	1.43802	-0.71412	1555.92148	80.71840	0.12187
0.86726	0.03593	0.97543	1.28085	-0.60612	1373.02040	151.82249	0.19314
0.91627	0.01661	0.98029	1.11946	-0.47732	1110.40185	374.85924	0.22565
0.95839	0	0.98260	0.93895	-0.31504	722.16454	-	0.21186
1.00000	0	0.99406	0.71940	0	0	-	0
318.15 K							
0	0.37783	0.89962	2.12960	-0.77730	371.63570	-	-1.25233
0.24310	0.28199	0.91399	1.97635	-0.99220	1167.22148	11.05933	-0.70066
0.41901	0.21264	0.92775	1.89041	-1.10098	1700.11398	15.82461	-0.27653
0.55220	0.16013	0.94058	1.77488	-1.13257	1975.02648	24.00011	-0.00584
0.65654	0.11900	0.95086	1.53876	-1.06302	1908.31704	33.55912	0.06055

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(continued)

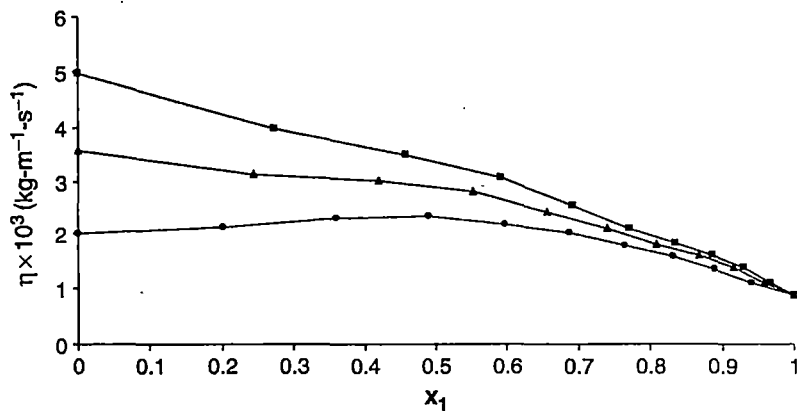
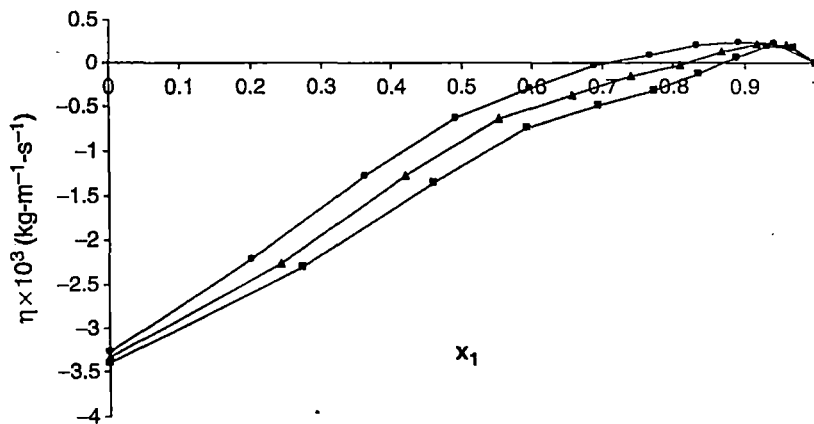
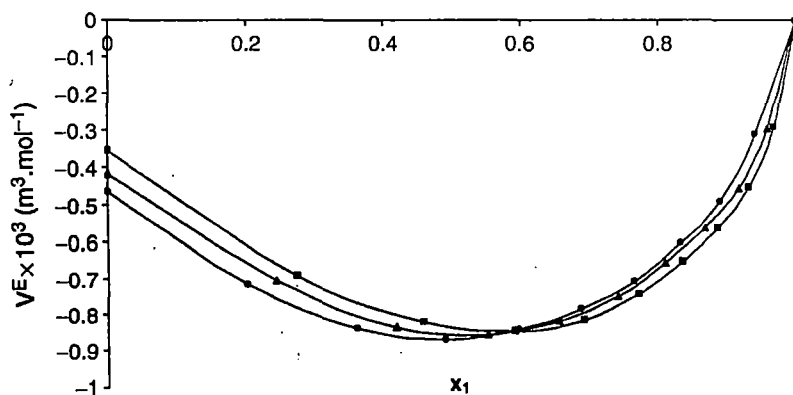
Table 2. Continued.

Mole fraction of water (x_1)	Mole fraction of ethane-1,2-diol (x_2)	$\rho \times 10^{-3}$ (kgm $^{-3}$)	$\eta \times 10^3$ (kgm $^{-1}$ s $^{-1}$)	$V^E \times 10^3$ (m 3 mol $^{-1}$)	ΔG^{*E} (Jmol $^{-1}$)	d	$\Delta\eta \times 10^3$ (kgm $^{-1}$ s $^{-1}$)
0.74051	0.08590	0.95945	1.35820	-0.95124	1799.70709	50.81977	0.12345
0.89010	0.01613	0.97978	0.96463	-0.56405	1308.5924	351.88657	0.27088
0.94125	0	0.98109	0.81807	-0.37325	906.93254	—	0.23256
1.00000	0	0.99026	0.59600	0	0	—	0
Water (1) + ethane-1,2-diol (2) + 1-propanol							
298.15 K							
0	0.44203	0.91960	4.97192	-0.35625	2.32850	—	-3.39220
0.27341	0.31716	0.93291	3.96902	-0.69250	941.93126	7.05350	-2.29336
0.45843	0.23265	0.94443	3.48948	-0.81935	1552.54048	13.55001	-1.35050
0.59195	0.17166	0.95460	3.07220	-0.84733	1847.28747	23.04513	-0.74125
0.69285	0.12558	0.96333	2.54730	-0.81229	1799.3841	34.38759	-0.49056
0.77179	0.08953	0.97073	2.11686	-0.74135	1632.04139	51.85844	-0.31424
0.83522	0.06055	0.97709	1.81609	-0.65433	1459.10388	86.32559	-0.12724
0.88731	0.03676	0.98262	1.60514	-0.56084	1300.44857	170.52862	0.06223
0.93085	0.01687	0.96878	1.39411	-0.45225	1057.36129	436.56056	0.18598
0.96780	0	0.98762	1.10989	-0.29355	573.64575	—	0.18570
1.00000	0	0.99707	0.89040	0	0	—	0
308.15 K							
0	0.44203	0.91402	3.67039	-0.52805	40.17761	—	-1.83336
0.27341	0.31716	0.92731	2.99794	-0.80909	993.19384	7.29452	-1.16198
0.45843	0.23265	0.93940	2.66833	-0.92484	1604.68756	13.56883	-0.58212
0.59195	0.17166	0.95027	2.37968	-0.94851	1904.69620	22.99942	-0.21442
0.69285	0.12558	0.95916	1.98952	-0.89065	1848.98043	34.14452	-0.10866
0.77179	0.08953	0.96701	1.67581	-0.80955	1688.28789	51.96625	-0.03442
0.83522	0.06055	0.97373	1.46391	-0.71122	1537.92542	88.63395	0.06556
0.88731	0.03676	0.97929	1.29579	-0.60058	1363.71786	173.73409	0.15346
0.93085	0.01687	0.98337	1.12109	-0.47466	1091.16953	435.73864	0.19283
0.96780	0	0.98469	0.94006	-0.31244	713.02579	—	0.19334
1.00000	0	0.99406	0.71940	0	0	—	0
318.15 K							
0	0.44203	0.90955	2.75457	-0.70445	90.0749	—	-1.28825
0.27341	0.31716	0.92308	2.31963	-0.95303	1072.66058	7.84557	-0.75452
0.45843	0.23265	0.93593	2.07145	-1.07006	1652.91411	13.58418	-0.34714

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0.59195	0.17166	0.94812	1.85600	-1.11458	1929.55597	22.54220	-0.08947
0.69285	0.12558	0.95793	1.57359	-1.05621	1875.13960	33.56327	-0.01442
0.77179	0.08953	0.96592	1.35940	-0.95068	1751.11126	52.65067	0.05104
0.83522	0.06055	0.97122	1.21703	-0.81212	1642.74673	93.08035	0.13348
0.88731	0.03676	0.97722	1.10035	-0.67539	1503.70099	189.19690	0.20135
0.93085	0.01687	0.98076	0.96084	-0.52344	1234.59604	487.27481	0.21614
0.96780	0	0.98193	0.81900	-0.34666	876.46701	-	0.20516
1.00000	0	0.99025	0.59600	0	0	-	0

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157158 Figure 1. Viscosity, η , for water + ethane-1,2-diol + alkanol at 298.15 K. Experimental points:
159 Alkanol: CH_3OH (●), $\text{C}_2\text{H}_5\text{OH}$ (▲), and $\text{C}_3\text{H}_7\text{OH}$ (■).160
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174175 Figure 2. Viscosity deviation, $\Delta\eta$, in water + ethane-1,2-diol + alkanol at 298.15 K. Experimental points:
176 Alkanol: CH_3OH (●), $\text{C}_2\text{H}_5\text{OH}$ (▲), and $\text{C}_3\text{H}_7\text{OH}$ (■).177
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190191 Figure 3. Excess molar volume, V^E , of water + ethane-1,2-diol + alkanol at 298.15 K. Experimental points:
192 Alkanol: CH_3OH (●), $\text{C}_2\text{H}_5\text{OH}$ (▲), and $\text{C}_3\text{H}_7\text{OH}$ (■).

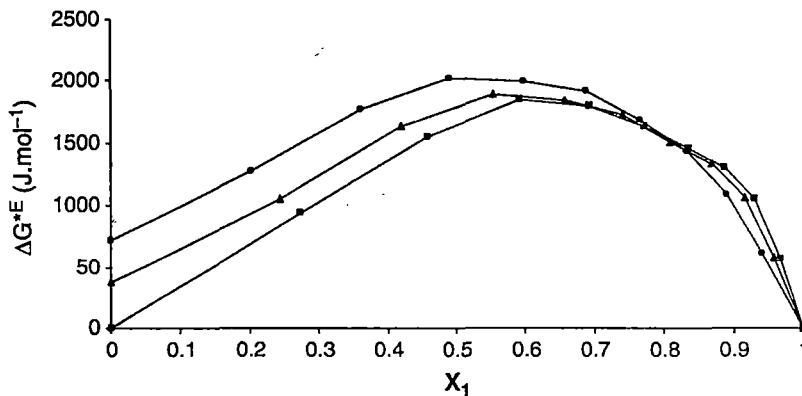


Figure 4. Excess Gibbs energy, ΔG^{*E} , of activation of viscous flow for water + ethane-1,2-diol + alkanol at 298.15 K. Experimental points: Alkanol: CH_3OH (●), $\text{C}_2\text{H}_5\text{OH}$ (▲), and $\text{C}_3\text{H}_7\text{OH}$ (■).

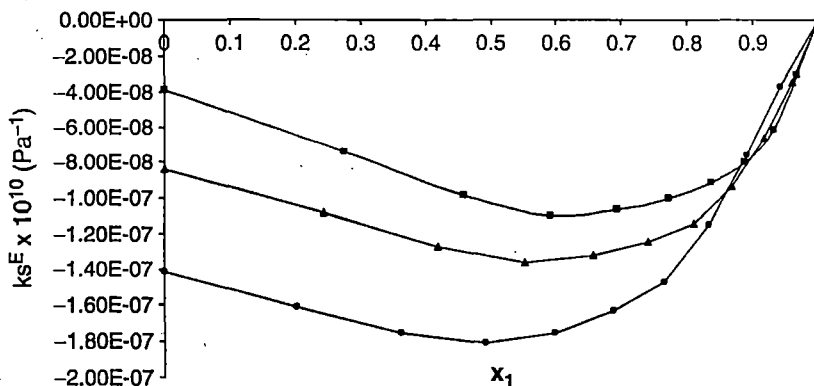


Figure 5. Excess isentropic compressibility, K_s^E for water + ethane-1,2-diol + alkanol at 298.15 K. Experimental points: Alkanol: CH_3OH (●), $\text{C}_2\text{H}_5\text{OH}$ (▲), and $\text{C}_3\text{H}_7\text{OH}$ (■).

the temperatures. From the value of $\Delta\eta$ as shown in table 2, the positive values of $\Delta\eta$ follows the trend:

$$(1) + (2) + \text{methanol} > \text{ethanol} > 1\text{-propanol}.$$

Here, dispersion and dipolar interactions are operating between water, ethane-1,2-diol, and $\text{CH}_3\text{OH}/\text{C}_2\text{H}_5\text{OH}/\text{C}_3\text{H}_7\text{OH}$ molecules resulting in negative $\Delta\eta$ but with the increase in temperature and mole fraction of water, the hydrogen bonding interactions come into play leading to the formation of complex species between unlike molecules thereby resulting in positive $\Delta\eta$ [25–27].

3.2. Excess molar volume

The excess molar volumes, V^E , are calculated from density data according to the following equation [28]:

$$V^E = \sum_{i=1}^n x_i M_i (1/\rho - 1/\rho_i) \quad (2)$$

241 where, M_i , ρ_i and ρ are the molar mass, density of the i th component and density
242 of the mixture respectively.

243 It can be seen from table 2 and figure 3 that V^E is negative for all the water (1)+
244 ethane-1,2-diol (2) + monoalcohol mixtures at all temperatures and over the entire
245 range of composition. The negative values of V^E increases with rising temperature from
246 298.15 to 318.15 K. The magnitude of the negative values of V^E decreases with
247 increasing chain length of the monoalcohol in water (1) + ethane-1,2-diol (2) + mono-
248 alcohol mixture series. The values of V^E at first decreases to minima and then increases
249 over the entire range of compositions for all ternary mixtures.

250 For (1)+(2) + methanol mixture, the minima is observed at $x_1=0.49$, for
251 (1)+(2) + ethanol, the minima is found at $x_1=0.55$, and for (1)+(2) + 1-propanol,
252 the minima observed at $x_1=0.59$.

253 From the values of V^E as shown in table 2, the observed trend is as follows:

254 (1) + (2) + methanol < ethanol < 1-propanol.
255

256 The negative V^E indicates the presence of strong molecular interactions between the
257 components of the mixture. Volume changes for a mixed system result from changes
258 in the free volume of the liquids, since the bond lengths and bond distances in the
259 molecules themselves do not change. The optimum packing condition is directly related
260 to differences in molecular sizes and intermolecular attractions, in particular when
261 hydrogen bonding occurs between unlike molecules creating association complexes,
262 as well as being effected by the breaking of interactions between like molecules [24].

263 To account for temperature effect on V^E , the following considerations are important.
264 It is known that pure monoalcohol can form either ring or chain-like complexes, and
265 while the fraction of ring complexes increases with the number of CH_2 groups in the
266 monoalcohol, the degree of association decreases [23,30]. The associated rings break
267 down to chains with increasing temperature, which is then followed by a total break
268 down of the associated species. The degree of association of water and ethane-1,2-diol
269 also decreases with increasing temperature. The formation of mixed complexes is an
270 exothermic process and hence, the equilibrium constant decreases with increasing
271 temperature. Thus, the number of the mixed associated species is influenced by two
272 opposing effects with rise of temperature: (i) The number of species which are able
273 to form mixed complexes increases with temperature, while the self-associated mono-
274 alcohol rings, water and also ethane-1,2-diol break down. (ii) The equilibrium constant
275 of the association resulting mixed complexes decreases with increasing temperature.
276 The first effect seems to be dominant in presently investigated mixtures, i.e., the number
277 of hydrogen bonds increases with temperature so the contraction of mixture increases.
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280 3.3. Excess Gibbs energy of activation of viscous flow

281 On the basis of the theory of absolute reaction rates [22], the excess Gibbs energy,
282 ΔG^{*E} , of viscous flow was calculated from
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$$284 \Delta G^{*E} = RT \ln(\eta M / \rho) - RT \sum_{i=1}^n x_i \ln(\eta_i M_i / \rho_i), \quad (3)$$

286
287 where n stands for the number of components of the mixture and M_i are the molar mass
288 of the mixture and of the pure components i .

289 The values of ΔG^{*E} are positive for all the ternary mixtures and decreases from
290 methanol to propanol (figure 4 and table 2), and attain a maximum value and then
291 decreases over the entire range of composition. With the increase in temperature, the
292 values of ΔG^{*E} show an increasing trend. The values of ΔG^{*E} decreases with the chain
293 length of the monoalcohol.

294 The positive values of ΔG^{*E} over the entire range of mole fraction indicate the
295 presence of strong interactions [23,] between the mixing components accompanied
296 by the complex formation.

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299 3.4. Grunberg and Nissan interactions parameter

300 Grunberg and Nissan [31] suggested a logarithmic relation between the viscosity of
301 liquid mixture and that of its pure components,

$$303 \ln \eta = \sum_{i=1}^n x_i \ln \eta_i + d \prod_{i=1}^n x_i \quad (4)$$

305 where d is a constant, regarded as a measure of the strength of molecular interactions
306 between the mixing components.

307 The positive value of Grunberg and Nissan parameter (d) gives an indication of
308 specific hydrogen bonding interaction between unlike molecules [32,33]. This parameter
309 (d) has been calculated (vide eq. 4) for the ternary liquid mixtures under discussion as a
310 function of the composition of the mixtures. It is seen from table 2 that the values
311 of d are positive over the entire range of composition for all the ternary liquid mixtures
312 at all temperatures. The observed value of d indicates the presence of strong molecular
313 interactions between the mixing components [32, 33].

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317 3.5. Isentropic compressibility

318 Table 3 contains the sound velocity (u), isentropic compressibility (K_S) and excess
319 isentropic compressibility (K_S^E) data for the mixtures at 298.15 K.

320 Isentropic compressibility (K_S) and excess isentropic compressibility (K_S^E) are
321 obtained using the following equation [29]:

$$323 K_S = 1/u^2 \cdot \rho \quad (5)$$

$$325 K_S^E = K_S - \sum_{i=1}^n x_i K_{S,i} \quad (6)$$

327 where, $K_{S,i}$ gives the isentropic compressibility for the i th component of the mixture.

328 As can be seen from table 3 and figure 5 that K_S^E is negative and such magnitude of
329 negative values decreases with increasing number of carbon atoms of monoalcohol over
330 the whole composition range for all the mixtures. However, the values of K_S^E decreases
331 to reach a minima at $x_1 = 0.49, 0.55$ and 0.59 for (1) + (2) + methanol, + ethanol, and
332 +1-propanol, respectively and thereafter increases for every mixture. The trend follows
333 the sequence:

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$$(1) + (2) + \text{methanol} < \text{ethanol} < 1\text{-propanol.}$$

Table 3. Speeds of sound (u), isentropic compressibility (K_S), and excess isentropic compressibility (K_S^E) of ternary liquid mixtures of various compositions (mole fraction) at 298.15 K.

x_1	u (ms ⁻¹)	$K_S \times 10^{10}$ (Pa ⁻¹)	$K_S^E \times 10^{10}$ (Pa ⁻¹)	x_1	u (ms ⁻¹)	$K_S \times 10^{10}$ (Pa ⁻¹)	$K_S^E \times 10^{10}$ (Pa ⁻¹)
Water (1) + ethane-1,2-diol (2) + methanol				Water (1) + ethane-1,2-diol(2) + ethanol			
0	1261.26	46.87243	-1.42	0	1305.72	6.43967	-0.84
0.20125	1347.07	5.93228	-1.61	0.24310	1396.89	5.54184	-1.08
0.36081	1428.55	4.20481	-1.75	0.41901	1479.25	4.87599	-1.27
0.49042	1499.42	4.67085	-1.81	0.55220	1544.33	4.42264	-1.36
0.59779	1550.40	4.32688	-1.76	0.65654	1580.53	4.1813	-1.32
0.68819	1581.90	4.12151	-1.63	0.74051	1602.50	4.03309	-1.24
0.76536	1600.12	3.9990	-1.47	0.80953	1613.36	3.95044	-1.14
0.81399	1580.98	4.07328	-1.15	0.86726	1598.63	3.99765	-0.94
0.89010	1544.20	4.25043	-0.755	0.91627	1566.91	4.13914	-0.66
0.94125	1509.34	4.44746	-0.37	0.95839	1528.71	4.34161	-0.35
1.00000	1498.20	4.46822	0	1.00000	1498.20	4.46822	0
Water (1) + ethane-1,2-diol (2) + 1-propanol							
0	1363.82	5.84636	-0.39				
0.27341	1435.34	5.20294	-0.74				
0.45843	1538.32	4.47445	-0.99				
0.59195	1591.98	4.13337	-1.10				
0.69285	1610.91	4.00023	-1.06				
0.77179	1618.78	3.93120	-1.00				
0.83522	1617.83	3.91023	-0.91				
0.88731	1608.20	3.93490	-0.80				
0.93085	1582.44	4.04692	-0.62				
0.96780	1535.64	4.29368	-0.31				
1.00000	1498.20	4.46822	0				

These results can be explained in terms of molecular interactions [20, 29] between unlike molecules. It appears from the sign and magnitude of K_S^E that specific interactions exist between mixing components [34]. The exaltation of polarization supported the specific interactions between components in the mixtures [34, 35]. This graded behavior is consistent with the present viscosity results.

5. Conclusions

The investigated mixtures were chosen in order to obtain information about the molecular interactions between their components. In this work, the mixed systems have been studied in terms of excess molar volumes, viscosity deviations, excess Gibbs energy of activation of viscous flow, Grunberg and Nissan parameter, and excess isentropic compressibility. The measured data and calculated values of all systems are in good accordance, and are theoretically and statistically satisfying.

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