Excess Volume, Viscosity, Heat of Mixing and IR Studies of some Binary and Ternary Liquid Mixtures involving H-Bond

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Abstract - Experimental densities and viscosities of the ternary liquid mixtures of isobutanol(1) + 1,2-propanediol (2) + cyclohexanone (3) and their constituent binaries such as, isobutanol (1) + 1,2,-propanediol (2), isobutanol (1) + cyclohexanone (3) and 1,2-propanediol (2) + cyclohexanone (3) were measured at (308.15 K and 318.15 K) over the whole composition range. From the measured data, excess volume (V^E), deviation in viscosity ($\Delta\eta$), interaction parameter 'd' and excess Gibbs free energy of activation of viscous flow (ΔG^{*E}) were calculated. The calculated parameters were fitted to Redlich-Kister type polynomial equation. Furthermore, heat of mixing (ΔH) and *IR* spectra were studied at equimolar concentration. The obtained results have been explained on the basis of the intermolecular interactions present in the binary and ternary liquid mixtures.

Keywords - Excess volume. Deviation in viscosity. Excess Gibbs free energy of activation of viscous flow. Heat of mixing and IR-Study

I. INTRODUCTION

Measurements of density and viscosity are of great interest in the development of Physico-chemical properties of liquid mixtures. The experimental data of liquid mixtures provide useful information about the insight into the nature of intermolecular interaction between the like and unlike molecules. Knowledge of the excess properties of binary and ternary liquid mixtures are essential in many chemical engineering processes such as separation process, design calculation, heat transfer, mass transfer, fluid flow and so forth [1-7].

Isobutanol is self-associated liquid through hydrogen bonding and it is used as solvent for coating, flavouring agent, precursors, additives and chemical extractant in production of organic compounds. 1,2-propanediol is also self-associated through intermolecular and intramolecular hydrogen bonding. It is used as medical lubricant, moisturizer in medicines, tobacco products and cosmetics. Cylohexanone exhibit dipole-dipole interaction in pure state and its derivatives are used for the synthesis of pharmaceuticals, dyes, herbicides, pesticides, plasticizers and rubber technology. Keeping in view of the importance, the liquids, measurement of density and viscosity of the given binary and ternary liquid mixtures were reported over the entire range of composition at (308.15 and 318.15)K. A complete survey of literatures has shown that no experimental physico-chemical studies for the isobutanol + 1,2-propanediol + cyclohexanone ternary liquid mixtures were reported earlier.

II. EXPERIMENTAL

2.1 Materials

The chemicals used in the present study are of analytical grade (Merck) and further purified by standard methods [8, 9]. Before use, the chemicals were dried by suitable drying agents to remove the water content, if any. The binary and ternary mixtures were prepared by knowing mass and were kept in special air tight glass stoppered conical type bottles to avoid evaporation. The weighing measurements were performed on a Shimasdzu Auy 220 Japan electronic digital balance with precision of ± 0.0001 g.

2.2 Density and Viscosity Measurements

Densities of pure liquids and their liquid mixtures at (308.15 and 318.15)K were measured by using special type specific gravity bottle of 10 ml capacity with an accuracy ± 0.001g. Each reported value is taken as the average of at least three measurements. The viscosity of pure liquids and their liquid mixtures were measured by using in Ostwald's viscometer of 10 ml capacity. The viscometer was calibrated with fresh conductivity water. The viscosities were measured at temperature (308.15 and 318.15) K. The time given to attain thermal equilibrium for the content of viscometer was 15 min. The time of flow was measured with an accurate stop watch which is capable of measuring time to within 0.01 s. Three sets of reading for the flow time were taken and the average values were taken for pure liquids and liquid mixtures.

2.3 Heat of Mixing

Heat of mixing of binary and ternary liquid mixtures were measured with calorimeter (Dewar Flask) at equimolar concentration.

2.4 IR study

IR spectra for pure liquids and at equimolar concentration of binary and ternary liquid mixtures were recorded by Perkin Elmer FT-IR.

III. RESULTS AND DISCUSSION

Comparison of experimental density (ρ) and viscosity (η) values with literature values for pure isobutanol(1), 1,2-propanediol(2) and cyclohexanone (3) are presented in Table – 1.

Table 1. Comparison of measured density (ρ) and viscosity (η) values with literature values of pure liquids at (308.15 K and 318.15) K.

Name of the Components	T(K)	ρ(g. cm^{-3})		η(m Pa.s)	
		Expt	Lit	Expt	Lit
Isobutanol	308.15	0.7967	0.7980 [10]	1.9282	2.4990 [11]
	318.15	0.7875	0.7851 [12]	1.3609	1.8610[13]
1,2-Propanediol	308.15	1.0256	1.0251 [14]	21.3250	25.2200 [15]
	318.15	1.0199	1.0175 [16]	12.7391	12.7800 [17]
Cyclohexanone	308.15	0.9290	0.9306 [18]	1.3140	1.6562 [19]
	318.15	0.9237	0.9225 [20]	1.0640	1.3700 [21]

3.1 Calculation of various parameters

Excess volume (V^E) [22]

$$V_{bin}^{E} = \left(\frac{x_1M_1 + x_2M_2}{\rho_{mix}}\right) - \left(\frac{x_1M_1}{\rho_1} + \frac{x_2M_2}{\rho_2}\right)$$

$$V_{tern}^{E} = \left(\frac{x_1M_1 + x_2M_2 + x_3M_3}{\rho_{mix}}\right) - \left(\frac{x_1M_1}{\rho_1} + \frac{x_2M_2}{\rho_2} + \frac{x_3M_3}{\rho_3}\right) \quad (1)$$

where x_1, x_2 and x_3 are mole fraction of component 1,2 and 3, ρ_1, ρ_2, ρ_3 and ρ_{mix} are densities of pure liquids and mixtures respectively, M_1, M_2 and M_3 are molecular weight of components 1,2 and 3 respectively.

Molar volume *(V) [23]*

$$V = \sum_{i=1}^{n}\left(\frac{x_iM_i}{\rho}\right) \quad (2)$$

V refers to molar volume that can be calculated from the density of mixtures, Here ρ is density of mixtures.

Viscosity *(η) [24]*

$$\eta = \left(At - \frac{B}{t}\right)\rho \quad (3)$$

A and B are characteristic constants of viscometer calculated using standard liquids like water and nitrobenzene, t is time of flow.

Deviation in viscosity *($\Delta\eta$)* [25]

$$\Delta\eta_{mix} = \eta_{mix} - (x_1\eta_1 + x_2\eta_2 + \cdots) \quad (4)$$

Viscosity difference ($\Delta ln\eta$) [26]

$$\Delta ln\eta_{mix} = 1n\eta_{mix} - (x_1\,1n\eta_1 + x_2\,1n\eta_2 + \cdots) \quad (5)$$

Interaction parameter *('d')* [27]

$$\ln\eta_{mix} = x_1\ln\eta_1 + x_2\ln\eta_2 + x_1x_2d + \cdots \quad (6)$$

The equation (6) can be rearranged in the form of

$$d = [\ln\eta_{mix} - (x_1\ln\eta_1 + x_2\ln\eta_2 + \cdots)]\,/(x_1x_2\cdots) \quad (7)$$

where '*d*' is Grunberg Nissan interaction parameter
Excess Gibbs free energy of activation of viscous flow (ΔG^{*E}) [28]

$$\Delta G^{*E} = RT\,[\ln(\eta_{mix}V) - (x_1 \ln(\eta_1 V_1) + x_2 \ln(\eta_2 V_2) + \cdots)] \quad (8)$$

where η_1, η_2, η_3 and η_{mix} are the viscosities of pure liquids and liquid mixtures, x_1, x_2 and x_3 are molefraction of component 1,2 and 3 respectively. R is gas constant, T is temperature, V_1, V_2, V_3 and V are molar volumes of pure components and mixtures respectively.
Heat of mixing (ΔH) [29]

$$q = m\, s\, \Delta T + w\, \Delta T \quad (9)$$

where, m is mass of liquid solution, s is specific heat of solution, w is water equivalent calorimeter, q is the heat loss or gain, ΔT is the change in temperature on mixing of pure components.

The calculated values of V^E, $\Delta\eta$ and ΔG^{*E} were fitted to Redlich-Kister type polynomial equation

$$A^E_{bin} = x_1 x_2 [(a + b(x_1 - x_2) + c\,(x_1 - x_2)^2]$$
$$A^E_{tern} = x_1 x_2 x_3 [(a + b x_1 (x_2 - x_3) + c x_1^{\,2}\,(x_2 - x_3)^2] \quad (10)$$

The method of least squares was used to derive the adjustable parameters a, b and c. The standard deviation [30] σ values were calculated using the relation.

$$\sigma = \left[\left(A^E_{\exp} - A^E_{cal} \right)^2 / (n - m) \right]^{1/2} \quad (11)$$

where '*n*' is number of experimental data, 'm' is number of adjustable parameters, A is the parameter studied.

Measurements of densities and viscosities for the binary and ternary liquid mixtures under study over the entire mole fraction range at (308.15 and 318.15) K are given in Table 2 – 5. From these measured values of density and viscosity, the values of excess volume (V^E), molar volume (V), deviation in viscosity ($\Delta\eta$), interaction parameter '*d*' and excess Gibbs free energy of activation of viscous flow (ΔG^{*E}) were calculated. The excess volumes were plotted against mole fractions at (308.15 and 318.15) K over the entire composition range. The adjustable parameters a, b and c evaluated by the method of least squares, along with standard deviation (σ) are presented in Table 6 and 7.

The variation in these excess parameters values reflect the interactions between the mixing components depending upon the composition, molecular size, shape of the components and temperature [31]. The effect which influences the values of excess properties may be the result of physical, chemical and structural contributions such as (1) physical contribution arises from dispersion forces or weaker interaction causing the positive values of V^E and negative ΔG^{*E}, (2) The chemical effect such as breaking of self associated compounds present in pure liquids which results in the positive value of V^E and negative ΔG^{*E}. On the other hand, charge transfer process, formation of hydrogen bond and other complex forming interactions results in the negative values of V^E and positive ΔG^{*E}. (3) The structural contribution arising from the geometrical fitting of one molecule into other molecule because of difference in their molar volumes[32]. From the Table 2-5, it is observed that there is non-linear variation of densities and viscosities with mole fractions of the both binaries and ternary liquid mixture at (308.15 and 318.15)K temperatures. This confirms the presence of molecular interaction among the molecules[33].

3.2 Excess volume of Mixing

The excess volume (V^E) data for the binary isobutanol(1) + 1,2-propanediol (2), isobutanol (1) + cyclohexanone (3), 1,2-propanediol(2) + cyclohexanone(3) and the ternary mixture isobutanol(1) + 1,2-propanediol(2) + cyclohexanone (3) are negative over the entire composition range at (308.15 and 318.15)K which are recorded in Tables 2-5. The variation of excess volumes for the three binary liquid mixtures with respect to mole fractions are graphically represented in Figures 1 and 2 and the curves are unsymmetrical.

The excess volumes for the ternary liquid mixtures are plotted against mole fractions x_1 x_2 and x_3. These are graphically displayed as 3D contour triangle and 3D contour surface triangle diagrams in Figures 3-6. Negative excess volume values for all the studied three binary and ternary liquid mixtures indicate the presence of specific interactions [34,35], like hydrogen bonding formation and dipole-dipole interaction between unlike molecules.

Table 2. Densities, viscosities and excess properties of binary liquid mixtures of isobutanol (1)+ 1,2-propanediol (2), isobutanol(1)+cyclohexanone(3) and 1,2-propanediol(2)+cyclohexanone (3) at 308.15 K..

x_1	ρ (g.cm^{-3})	V^E (cm^3.mol^{-1})	η (mPa.s)	$\Delta\eta$ (mPa.s)	$\Delta ln\eta$ (mPa.s)	d	V (cm^3.mol^{-1})	ΔG^{*E} (kJ. mol^{-1})
Isobutanol (1) + 1,2-propanediol (2)								
0.0000	1.0256		21.3250				74.2005	
0.1015	1.0177	-0.0484	17.5857	-1.8068	0.0459	-28.0089	74.8349	0.0619
0.2030	0.9841	-0.0725	14.8059	-2.5822	0.1230	-13.2021	76.6739	0.2817
0.2900	0.9573	-0.1236	11.9949	-3.7056	0.1216	-8.9916	77.9300	0.2692
0.3905	0.9279	-0.1772	8.9272	-4.8239	0.0677	-6.7240	78.9231	0.1055
0.4949	0.8991	-0.2366	7.0380	-4.6879	0.0808	-5.5314	79.4239	0.0949
0.6010	0.8703	-0.1826	5.5910	-4.0768	0.1057	-5.0157	79.4231	0.0971
0.6800	0.8500	-0.1499	4.7430	-3.3924	0.1310	-4.9950	79.0374	0.1039
0.7804	0.8252	-0.0849	3.8592	-2.3287	0.1661	-5.5604	78.0792	0.1044
0.8916	0.7996	-0.0563	3.1044	-0.9265	0.2157	-8.3562	76.3843	0.1109
1.0000	0.7967		1.9282				93.0086	
Isobutanol (1) + cyclohexanone (3)								
0.0000	0.9290		1.3140				105.4898	
0.1042	0.9161	0.0675	1.3614	-0.0166	-0.0045	-3.0451	104.2590	-0.0079
0.2001	0.9057	-0.0690	1.4020	-0.0349	-0.0119	-1.8476	102.9277	-0.0289
0.2997	0.8935	-0.0848	1.4381	-0.0600	-0.0247	-1.4854	101.6711	-0.0611
0.3966	0.8815	-0.1186	1.4726	-0.0849	-0.0381	-1.3896	100.4302	-0.0956
0.5002	0.8685	-0.1734	1.5038	-0.1175	-0.0569	-1.4517	99.0849	-0.1449
0.5969	0.8549	-0.0937	1.5835	-0.0971	-0.0424	-1.6267	97.9600	-0.1057
0.6900	0.8418	-0.0508	1.6601	-0.0777	-0.0308	-2.0069	96.8434	-0.0754
0.7907	0.8275	-0.0326	1.7304	-0.0692	-0.0279	-2.9340	95.6075	-0.0685
0.8900	0.8130	-0.0052	1.8202	-0.0404	-0.0154	-5.6768	94.3983	-0.0370
1.0000	0.7967		1.9282				93.0086	
1,2-Propanediol (2) + cyclohexanone (3)								
0.0000	0.9290		1.3140				105.4898	
0.1015	0.9369	-0.0851	2.7793	-0.5658	0.4663	-5.0737	102.2288	1.2056
0.2030	0.9452	-0.1612	4.5808	-0.7954	0.6831	-3.6626	98.97688	1.7698
0.2900	0.9531	-0.2576	6.1230	-0.9942	0.7308	-3.4393	96.1583	1.8964
0.3905	0.9625	-0.3347	8.0197	-1.1086	0.7206	-3.6377	92.9366	1.8735
0.4949	0.9728	-0.4091	10.1218	-1.0956	0.6624	-4.2951	89.5956	1.7248
0.6010	0.9824	-0.3290	12.4745	-0.8661	0.5757	-5.5996	86.3559	1.5040
0.6800	0.9900	-0.2617	14.2855	-0.6360	0.4911	-7.3045	83.9514	1.2861
0.7804	0.9999	-0.1505	16.4953	-0.4352	0.3552	-11.4807	80.9211	0.9341
0.8916	1.0123	-0.0744	18.9065	-0.2493	0.1817	-25.5944	77.5179	0.4799
1.0000	1.0256		21.3250				74.2005	

Table 3. Densities, viscosities and excess properties of binary liquid mixtures of isobutanol (1)+1, 2-propanediol (2), isobutanol (1)+cyclohexanone (3) and 1,2-propanediol (2)+cyclohexanone (3) at 318.15 K.

x_1	ρ (g.cm^{-3})	V^E (cm^3.mol^{-1})	η (mPa.s)	$\Delta\eta$ (mPa.s)	$\Delta ln\eta$ (mPa.s)	d	V (cm^3.mol^{-1})	ΔG^{*E} (kJ. mol^{-1})
			Isobutanol (1) + 1,2-propanediol (2)					
0.0000	1.0199		12.7391				74.6151	
0.1015	0.9918	-0.0658	11.0136	-0.5922	0.0771	-23.0139	76.6534	01937
0.2030	0.9648	-0.1118	9.3447	-1.0846	0.1442	-10.6872	78.4578	0.3896
0.2900	0.9428	-0.1657	7.6008	-1.8386	0.1322	-7.1805	80.0987	0.3592
0.3905	0.9185	-0.2206	5.7198	-2.5761	0.0726	-5.2781	82.0015	0.2022
0.4949	0.8944	-0.2802	4.4075	-2.7005	0.0455	-4.2686	83.9756	0.1293
0.6010	0.8701	-0.2388	3.5069	-2.3939	0.0542	-3.7516	86.0839	0.1528
0.6800	0.8522	-0.1618	2.9597	-2.0422	0.0613	-3.6200	87.6998	0.1722
0.7804	0.8310	-0.1153	2.4166	-1.4430	0.0831	-3.7816	89.7021	0.2280
0.8916	0.8086	-0.0773	1.8857	-0.7086	0.0837	-5.0624	91.9063	0.2257
1.0000	0.7875		1.3609				94.0952	
			Isobutanol (1) + cyclohexanone (3)					
0.0000	0.9237		1.0640				106.0950	
0.1042	0.9114	-0.0503	1.0902	-0.0047	-0.0013	-0.8530	104.7944	-0.0030
0.2001	0.8997	-0.0842	1.1155	-0.0079	-0.0020	-0.5859	103.6097	-0.0043
0.2997	0.8873	-0.1239	1.1415	-0.0115	-0.0034	-0.5146	102.3748	-0.0083
0.3966	0.8750	-0.1688	1.1612	-0.0205	-0.0102	-0.5176	101.1672	-0.0268
0.5002	0.8618	-0.2491	1.1784	-0.0341	-0.0210	-0.5764	99.8436	-0.0572
0.5969	0.8480	-0.1893	1.2205	-0.0207	-0.0096	-0.6691	98.7430	-0.0259
0.6900	0.8346	-0.1528	1.2541	-0.0148	-0.0055	-0.8575	97.6624	-0.0144
0.7907	0.8197	-0.1053	1.2895	-0.0093	-0.0024	-1.2965	96.5015	-0.0059
0.8900	0.8045	-0.0405	1.3218	-0.0064	-0.0021	-2.5920	95.3748	-0.0046
1.0000	0.7875		1.3609				94.0952	
			1,2-Propanediol (2) + cyclohexanone (3)					
0.0000	0.9237		1.0640				106.0950	
0.1015	0.9318	-0.1081	2.0362	-0.2128	0.3971	-2.7322	102.7917	1.0611
0.2030	0.9403	-0.2099	3.0674	-0.3667	0.5548	-2.3776	99.4948	1.4866
0.2900	0.9481	-0.2984	4.0184	-0.4313	0.6089	-2.4071	96.6675	1.6344
0.3905	0.9574	-0.3704	4.9804	-0.6427	0.5740	-2.7284	93.4317	1.5457
0.4949	0.9678	-0.4525	6.0926	-0.7494	0.5164	-3.3562	90.0631	1.3933
0.6010	0.9772	-0.3610	7.5468	-0.5339	0.4670	-4.4597	86.8147	1.2643
0.6800	0.9847	-0.2853	8.6715	-0.3316	0.4098	-5.8833	84.4034	1.1121
0.7804	0.9947	-0.1905	9.9173	-0.2579	0.2948	-9.3730	81.3377	0.8035
0.8916	1.0070	-0.0977	11.3180	-0.1556	0.1508	-21.1181	77.9298	0.4130
1.0000	1.0199		12.7391				74.6151	

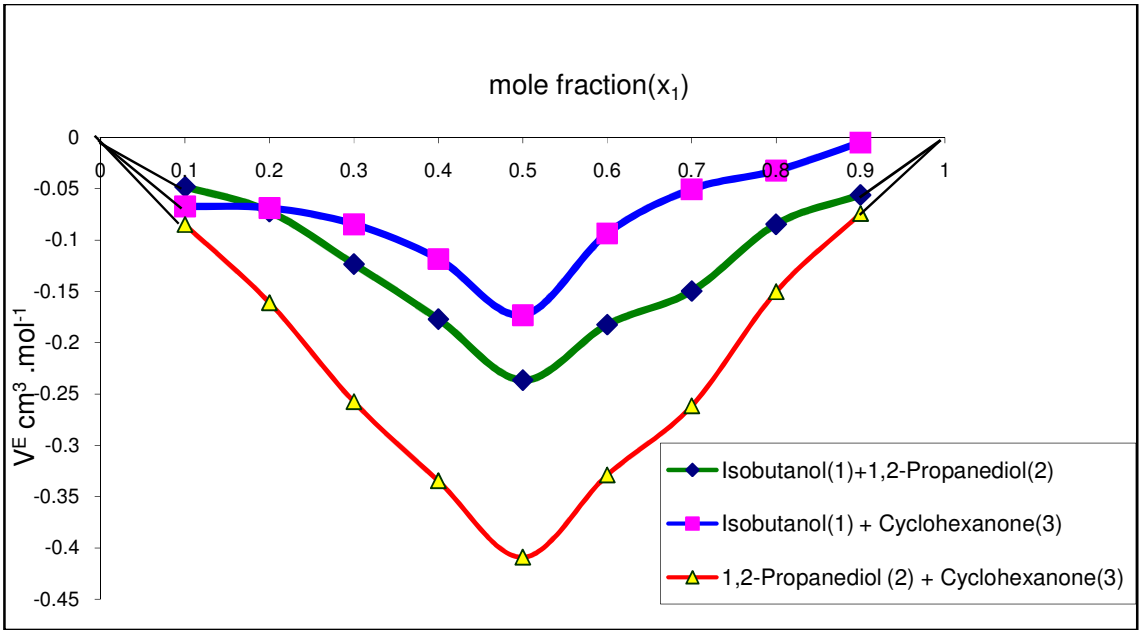


Fig.1 V^E versus mole fraction(x_1)plot for binary liquid mixutres of isobutanol(1) + 1,2-propanediol(2), 1,2-propanediol(2)+cyclohexanone(3) and isobutanol(1)+cyclohexanone (3) at 308.15K

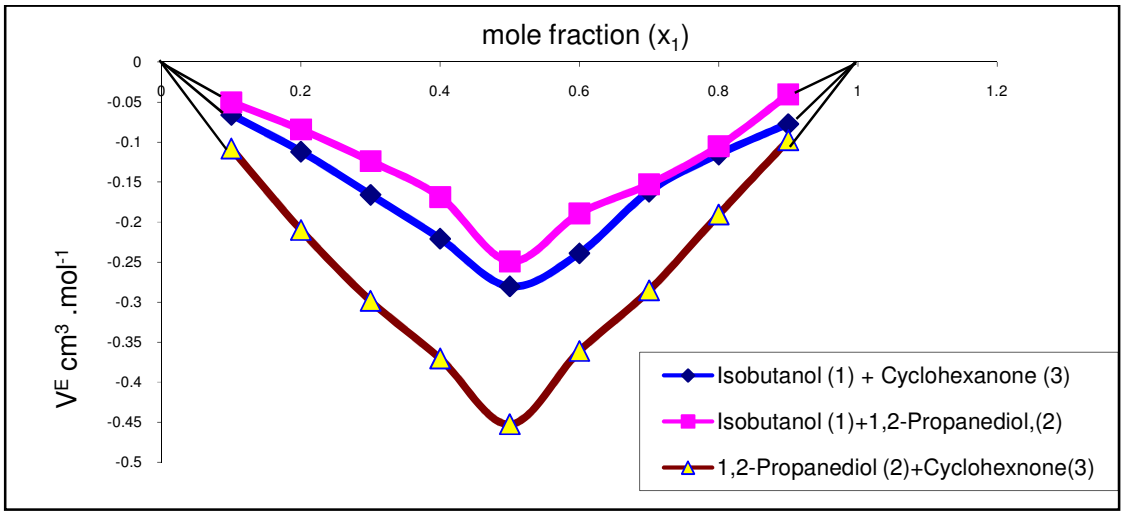


Fig. 2 V^E versus mole fraction (x_1) plot for binary liquid mixutres of isobutanol(1) + 1,2-propanediol (2), 1,2-propanediol(2) + cyclohexanone(3) and isobutanol (1)+cyclohexanone(3) at 318.15K

Table 4. Densities, viscosities and excess properties of ternary liquid mixtures of isobutanol (1)+1, 2-propanediol (2) + cyclohexanone (3) at 308.15 K.

x_1	x_2	ρ (g.cm^{-3})	V^E (cm^3.mol^{-1})	η (mPa.s)	$\Delta\eta$ (mPa.s)	$\Delta ln\eta$ (mPa.s)	d	V (cm^3.mol^{-1})	ΔG^{*E} (kJ. mol^{-1})
0.1138	0.2077	0.9303	-0.0419	2.0554	-3.4867	-0.1755	-55.0232	97.6874	-0.4404
0.2032	0.2786	0.9245	-0.0866	2.4214	-4.5924	-0.2431	-37.5464	94.1505	-0.5973
0.3034	0.4031	0.9211	-0.1542	3.7856	-5.7799	-0.1813	-40.9500	88.8317	-0.4317
0.3923	0.4919	0.9165	-0.2590	5.4238	-5.9748	-0.1036	-78.4712	84.9657	-0.2522
0.4507	0.1537	0.8840	-0.1419	2.5714	-2.1068	0.0678	-30.3426	95.8534	0.1059
0.5738	0.3031	0.8751	-0.1473	3.0481	-4.6771	-0.2219	-63.9518	88.1708	-0.5100
0.6980	0.1953	0.8505	-0.0912	3.4954	-2.1546	0.1666	-73.7557	90.5137	0.4429
0.7902	0.1018	0.8323	-0.0677	2.3729	-1.4634	0.0043	-98.1937	92.3645	0.0186
0.8710	0.0674	0.8187	-0.0367	2.4759	-0.7220	0.1116	-218.5702	92.4842	0.2894
0.6982	0.1007	0.8461	-0.0254	2.6561	-1.1150	0.1527	-54.5544	94.6551	0.3101
0.4944	0.1958	0.8798	-0.1098	2.9134	-2.6223	0.0610	-32.5635	93.0734	0.1726
0.2938	0.2963	0.9145	-0.1647	3.4984	-3.9250	0.0409	-32.7140	92.3670	0.1286
0.0974	0.3872	0.9471	-0.0915	5.2848	-3.8338	0.2761	-70.1457	91.7935	0.7624
0.3983	0.4562	0.9127	-0.2454	6.5812	-4.1085	0.1866	-61.6704	86.1585	0.4813
0.1940	0.5947	0.9527	-0.1154	7.9383	-5.3865	0.0688	-82.8216	83.6287	0.2610
0.1060	0.7162	0.9766	-0.0876	8.6847	-7.0322	-0.1493	-164.8121	82.1450	-0.3972
0.1049	0.8012	0.9858	-0.0536	13.9501	-3.4635	0.0889	-313.9783	79.2483	0.2295
0.0570	0.8839	1.0021	-0.0291	17.3161	-1.7250	0.0925	-874.7629	77.4411	0.2209
0.1906	0.7069	0.9641	-0.1266	10.0309	-5.5459	-0.0105	-165.4066	80.8667	-0.0092
0.3071	0.4960	0.9289	-0.2326	5.3403	-6.0947	-0.0993	-55.9688	86.4544	-0.2799
0.3858	0.3118	0.9026	-0.1405	3.1955	-4.5885	-0.1269	-34.8464	90.2623	-0.2605
0.4907	0.1025	0.8751	-0.1719	1.8818	-1.7781	-0.1133	-36.2670	95.4493	-0.2349
0.3977	0.1525	0.8911	-0.1913	1.8634	-2.7472	-0.2283	-30.5278	95.6169	-0.5769
0.1944	0.2068	0.9208	-0.1285	1.8159	-3.7558	-0.3274	-37.7844	96.4649	-08187
0.1924	0.1018	0.9132	-0.0784	1.6364	-1.8237	-0.1361	-45.4381	99.0870	-0.2718
0.1081	0.1042	0.9235	-0.0339	1.4864	-1.9734	-0.2074	-68.0282	100.3825	-0.4771
0.0568	0.0510	0.9259	-0.0155	1.2017	-1.1650	-0.2527	-169.0708	102.9482	-0.6205

ρ_1 = 0.7967 g.cm^{-3} η_1 = 1.9282 mPa.s V_1 = 93.0086 cm^3. mol^{-1}
ρ_2 = 1.0256 g.cm^{-3} η_2 = 21.3250 mPa.s V_2 = 74.2005 cm^3. mol^{-1}
ρ_3 = 0.9290 g.cm^{-3} η_3 = 1.3140 mPa.s V_3 = 105.4898 cm^3. mol^{-1}

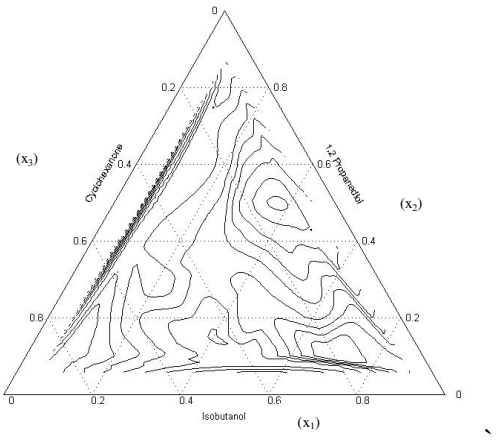


Fig. 3 V^E versus mole fractions x_1, x_2 and x_3 plots of ternary liquid mixtures of isobutanol (1) + 1, 2-propanediol (2) + cyclohexanone (3) at 308.15 K

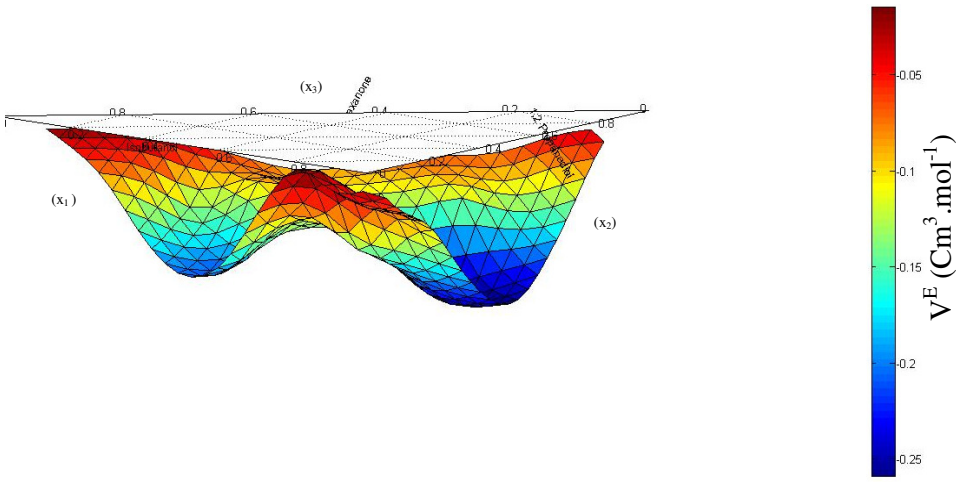


Fig. 4 V^E versus mole fractions x_1, x_2 and x_3 plots of ternary liquid mixtures of isobutanol (1) + 1, 2-propanediol (2) + cyclohexanone (3) at 308.15 K

Table 5. Densities, viscosities and excess properties of ternary liquid mixtures of isobutanol (1) + 1, 2-propanediol (2) + cyclohexanone (3) at 318.15 K.

x_1	x_2	ρ (g.cm^{-3})	V^E (cm^3.mol^{-1})	η (mPa.s)	$\Delta\eta$ (mPa.s)	$\Delta ln\eta$ (mPa.s)	d	V (cm^3.mol^{-1})	ΔG^{*E} (kJ. mol^{-1})
0.1138	0.2077	0.9246	-0.0611	1.8836	-1.6407	0.0274	-37.0569	98.2896	0.0813
0.2032	0.2786	0.9186	-0.1319	2.2759	-2.1011	0.0187	-26.5742	94.7553	0.0742
0.3034	0.4031	0.9144	-0.1772	3.5586	-2.3007	0.1320	-30.5255	89.4826	0.3826
0.3923	0.4919	0.9093	-0.2867	4.9795	-1.9442	0.2255	-60.0351	85.6384	0.6100
0.4507	0.1537	0.8770	-0.1752	2.6750	-0.3267	0.4289	-18.8260	96.6185	1.0633
0.5738	0.3031	0.8671	-0.1546	3.1933	-1.5745	0.2056	-45.3551	88.9843	0.604.7
0.6980	0.1953	0.8425	-0.1354	3.4542	-0.0965	0.5210	-48.4471	91.3732	1.3937
0.7902	0.1018	0.8239	-0.0922	2.3373	-0.1497	0.3398	-57.8209	93.3062	1.9059
0.8710	0.0674	0.8099	-0.0436	1.8887	-0.2209	0.1922	-121.8707	93.4891	0.5118
0.6982	0.1007	0.8380	-0.0387	2.3773	-0.0803	0.3815	-31.7770	95.5700	0.9248
0.4944	0.1958	0.8724	-0.1263	2.9686	-0.5281	0.4183	-21.2537	93.8629	1.1228
0.2938	0.2963	0.9080	-0.1881	3.4703	-1.1403	0.3743	-23.1357	93.0549	1.0123
0.0974	0.3872	0.9417	-0.1419	4.9635	-0.6473	0.5550	-52.5426	92.3199	1.5236
0.3983	0.4562	0.9054	-0.2617	6.3159	-0.1941	0.5503	-46.5548	86.8532	1.4591
0.1940	0.5947	0.9466	-0.1578	7.5670	-0.4906	0.4380	-65.1889	84.1676	1.2453
0.1060	0.7162	0.9704	-0.0853	9.0712	-0.3908	0.3386	-132.6583	82.6698	0.8808
0.1049	0.8012	0.9797	-0.0643	10.1513	-0.2998	0.2406	-255.9202	79.7418	0.6380
0.0570	0.8839	0.9962	-0.0367	11.1295	-0.2745	0.1389	-719.8483	77.8997	0.3507
0.1906	0.7069	0.9576	-0.1400	9.0163	-0.3574	0.3351	-132.7666	81.4156	0.9047
0.3071	0.4960	0.9221	-0.2567	5.4906	-1.4610	0.3337	-42.7785	87.0920	0.8558
0.3858	0.3118	0.8956	-0.1636	3.2357	-1.5779	0.2435	-24.8343	90.9678	0.7103
0.4907	0.1025	0.8679	-0.1989	1.7032	-0.6977	0.0956	-21.0945	96.2412	0.3091
0.3977	0.1525	0.8842	-0.2131	1.8013	-1.1618	0.0500	-19.1310	96.3631	0.1397
0.1944	0.2068	0.9148	-0.1553	1.8636	-1.6725	-0.0008	-25.2692	97.0976	0.0178
0.1924	0.1018	0.9073	-0.1083	1.5745	-0.7277	0.0923	-25.9717	99.7313	0.3226
0.1081	0.1042	0.9179	-0.0563	1.4511	-0.8568	0.0253	-38.9628	100.9949	0.1222
0.0568	0.0510	0.9205	-0.0332	1.2365	-0.4376	0.0098	-78.3231	103.5522	0.0531

ρ_1 = 0.7875 g.cm^{-3} η_1 = 1.3609 mPa.s V_1 = 94.0952 cm^3. mol^{-1}
ρ_2 = 1.0199 g.cm^{-3} η_2 = 13.7391 mPa.s V_2 = 74.6151 cm^3. mol^{-1}
ρ_3 = 0.9237 g.cm^{-3} η_3 = 1.0640 mPa.s V_3 = 106.0950 cm^3. mol^{-1}

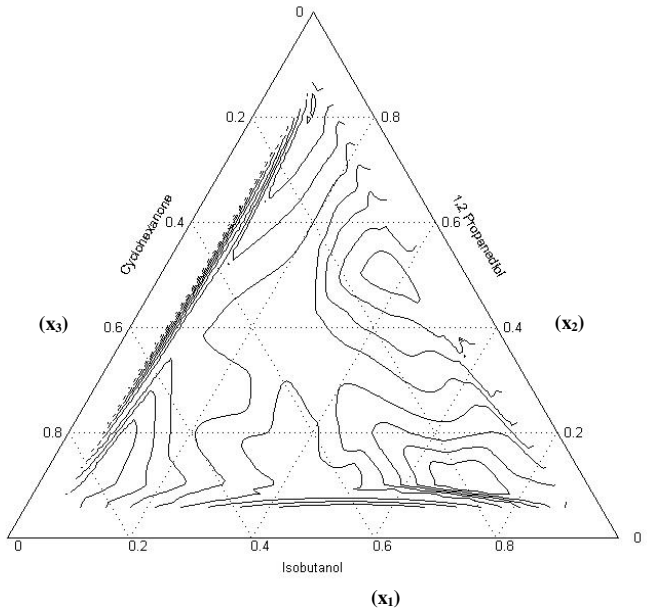


Fig.5 V^E versus mole fractions x_1, x_2 and x_3 plots of ternary liquid mixtures of isobutanol (1) + 1, 2-propanediol (2) + cyclohexanone (3) at 318.15K.

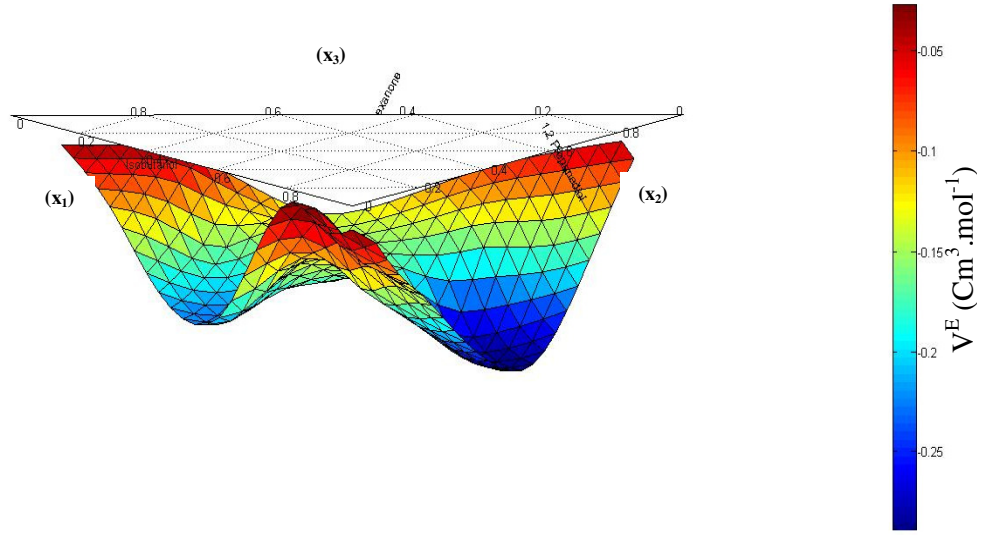


Fig. 6 V^E versus mole fractions x_1, x_2 and x_3 plots of ternary liquid mixtures of isobutanol (1) + 1, 2-propanediol (2)+ cyclohexanone (3) at 318.15K

Table 6. Values of a, b and c and corresponding standard deviation for the binary liquid mixtures of isobutanol (1) + 1, 2 – propanediol (2), 1,2-propanediol (2) + cyclohexanone (3) and isobutanol (1) + cyclohexanone (3) at (308.15 and 318.15)K.

Temperature	Parameters	a	b	c	σ
	Binary liquid mixtures of isobutanol (1) + 1,2-propanediol (2)				
308.15K	V^E ($cm^3.mol^{-1}$)	-0.8131	0.0340	0.7637	0.0107
	$\Delta\eta$ (mPa.s)	-18.5912	4.2647	9.2561	0.1324
	ΔG^{*E} (kJ. mol^{-1})	0.4514	-0.5676	1.5854	0.0221
318.15K	V^E ($cm^3.mol^{-1}$)	-0.9878	0.0230	0.7268	0.0014
	$\Delta\eta$ (mPa.s)	-10.5859	-0.3895	7.5597	0.1092
	ΔG^{*E} (kJ. mol^{-1})	0.6284	0.6163	3.2457	0.0475
	Binary liquid mixtures of 1,2-propanediol (2) + cyclohexanone (3)				
308.15K	V^E (cm^3. mol^{-1})	-1.4839	0.1358	1.3782	0.0197
	$\Delta\eta$ (mPa.s)	-4.0999	2.2807	0.5427	0.0645
	ΔG^{*E} (kJ. mol^{-1})	6.8825	-4.4890	3.5493	0.0514
318.15K	V^E (cm^3. mol^{-1})	-1.6261	1.8799	1.1432	0.0172
	$\Delta\eta$ (mPa.s)	-2.5170	0.7377	1.8252	0.0252
	ΔG^{*E} (kJ . mol^{-1})	5.6931	-3.7644	3.6806	0.0540
	Binary liquid mixtures of isobutanol (1) + cyclohexanone (3)				
308.15K	V^E($cm^3.mol^{-1}$)	-0.5596	0.0649	1.1650	0.0171
	$\Delta\eta$(mPa.s)	-0.4040	-0.1369	0.2517	0.0035
	ΔG^{*E} (kJ.mol^{-1})	-0.4660	-0.1449	0.5017	0.0069
318.15K	V^E($cm^3.mol^{-1}$)	-0.8317	-0.0718	0.7349	0.0102
	$\Delta\eta$(mPa.s)	-0.0995	-0.0083	0.1214	0.0016
	ΔG^{*E} (kJ.mol^{-1})	-0.1421	-0.0106	0.2912	0.0039

Table 7. The values of a, b and c and corresponding standard deviation for the ternary liquid mixtures of isobutanol (1)+1,2-propanediol (2)+ cyclohexanone (3) at (308.15 and 318.15)K.

Temp	Parameters	a	b	c	σ
	Ternary liquid mixtures of isobutanol (1) + 1,2-propanediol (2) + cyclohexanone (3)				
308.15K	V^E ($cm^3.mol^{-1}$)	-3.8164	-11.2768	-270.2620	0.0002
	$\Delta\eta$ (mPa.s)	-157.2890	-575.5710	-486.4600	0.0735
	ΔG^{*E} (kJ.mol^{-1})	-3.2862	44.3965	-275.758	0.0007
318.15K	V^E ($cm^3.mol^{-1}$)	-4.7879	-10.2279	-275.5260	0.0008
	$\Delta\eta$ (mPa.s)	-44.6171	-32.4977	209.2180	0.0082
	ΔG^{*E} (kJ.mol^{-1})	30.5649	98.3892	-75.9508	0.0182

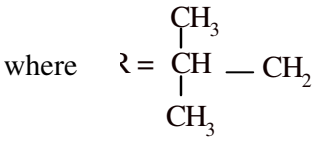
Isobutanol is self associated through intermolecular hydrogen bonding and the diploemoment value is 1.79D. 1,2-Propanediol has both inter and intra molecular hydrogen bonding and its dipolemoment value is 2.27D. Cyclohexanone is a class of organic compound that contains carbonyl (C=O) group. The greater electronegativity of an oxygen atom and high dipolemoment value make cyclohexanone polar. The dipolemoment value of cyclohexanone is 3.06D. Further the presence of oxygen atom with its non-bonding electron pairs makes cyclohexanone molecule to involve in intermolecular H-bond with molecule like alcohols. Cyclohexanone exhibits dipole – dipole interaction in pure state and does not compete with high degree of association between alcohol molecules.

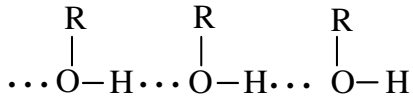
The strength of interaction between the participating molecules depends upon the dipolement, polarisability and geometry of the interacting molecules. Due to polar nature of isobutanol, 1,2-propanediol and cyclohexanone, the lesser attractive interactions like hydrogen bonding formation among the molecules and dipole-dipole interaction exist in these mixtures. When the components are mixed, the changes that occur in association with the unlike molecules rupture the hydrogen bond in pure isobutanol and 1,2-propanediol, dipole-dipole interaction and the formation of OH OH between isobutanol, 1,2-propanediol molecules; and perhaps OH O=C between H atom of isobutanol and 1,2-propanediol molecules with oxygen atom of cyclohexanone molecule.

Upon mixing of isobutanol with 1,2-propanediol, it would release the dipoles of isobutanol due to breaking up of H-bonds as a result the free dipole released from the isobutanol molecules is associated with 1,2-propanedial molecules forming H-bonds. Hence molecular interaction exists between isobutanol and 1,2-propanediol molecule through hydrogen bonding.

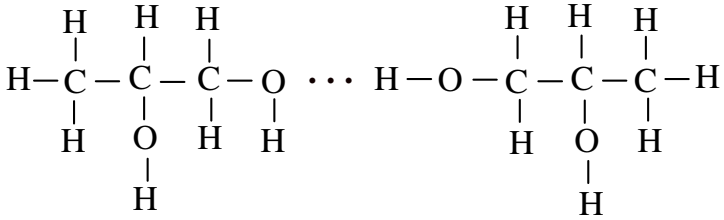
Further addition of cyclohexanone with the mixtures causes dissociation of hydrogen bonded structure of isobutanol as well as decrease in interaction between isobutanol and 1, 2-propanediol molecules and the subsequent formation new type of hydrogen bonding between proton acceptor oxygen atom of C=O group of cyclohexanone and proton of OH group of isobutanol.

Isobutanol is associated through hydrogen bonding

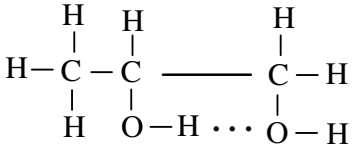




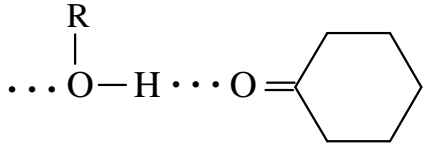
The 1,2-propanediol is associated through intermolecular hydrogen bonding.



The intramolecular hydrogen bonding in 1, 2-propanediol molecule is as follows:



Intermolecular hydrogen bonding can be formed between isobutanol and 1, 2 propanediol. The formation of hydrogen bond between the proton of OH group of isobutanol and the oxygen atom of (C=O) ketone of cyclohexanone is given below.



The formation of Hydrogen bonding between unlike molecules will lead to reduction in volume during the mixing process.

The negative V^E values become more negative with increasing temperature from (308.15 to 318.15) K of the mixture for all the three binaries and ternary system under study. These are represented in Tables 2-5 and Figures 1- 6. This may arise due to increase in the rate of association of unlike molecules with the stimulation of those molecules by thermal energy resulting the enhancement of the dipolar association between the molecules. Hence the interaction increases with rising temperature [35]. This will imply that the constituent binaries and ternary liquid mixtures are more compact than that of corresponding pure liquids at 318.15K.

While observing otherwise, the contraction in volumes of the mixtures is due to negative contribution of V^E which has come from geometrical fitting of one component molecule into each other structure because of the considerable difference in their molar volumes. The molar volumes of isobutanol (1), 1,2-propanediol (2) and cyclohexanone (3) at 318.15k are 94.0952 $cm^3. mol^{-1}$, 74.6151 $cm^3. mol^{-1}$ and 106.0950 $cm^3. mol^{-1}$ respectively. The above differences in their molar volumes of pure liquids may favour fitting of one molecule in to other molecules.

3.3 Viscosities

Viscosity data of binary and ternary liquid mixtures have been employed to yield information regarding the nature of interaction in the present systems, all the three binary and one ternary liquid mixtures are deviated negatively from mixture law, that is, the measured viscosity values of the mixtures are less than the values expected from the mixture law. The values of $\Delta\eta$, $\Delta ln\eta$ and 'd' as function of composition of the given constituents binary and ternary liquid mixtures were calculated using equation (4), (5) and (7) which are listed in Tables 2-5.

From the Tables, it can be seen that the values of $\Delta\eta$, 'd' are negative and $\Delta ln\eta$ are positive except isobutanol(1) + cyclohexanone (3) system at all concentration range for their constituent liquid mixtures. In the case ternary liquid mixtures, the $\Delta\eta$, 'd' values are negative and $\Delta ln\eta$ is positive except at 308.15K. The negative values of $\Delta\eta$ also indicate complex formation [36], between the components of the liquid mixture. These negative values of $\Delta\eta$ change with composition of all the constituent binaries and ternary mixtures confirm the presence of molecular interaction between the mixing components. The deviation in viscosity ($\Delta\eta$) values can be used to detect the molecular interaction. As temperature is raised from (308.15 to 318.15)K, the values of $\Delta\eta$ and 'd' become less negative. The breaking of self associated isobutanol, and 1,2- propanediol, by the formation of hydrogen bond and dipole – dipole interaction between the unlike molecules, makes the mixtures less viscous and flows more easily when compared with the behaviours of pure liquids.

Excess Gibbs free energy of activation of viscous flow (ΔG^{*E}) values are calculated and recorded in Tables 2 – 5 over the entire composition range. The ΔG^{*E} values are found to be positive and negative can be observed at (308.15 and 318.15)K for all the three binary liquid mixtures. The positive values of ΔG^{*E} may be due to size effect and as reliable measure to detect the presence of molecular interaction between the molecules [37]. In the case of ternary liquid mixtures, the values of ΔG^{*E} are negative for some mole fraction range at 308.15K. When temperature is increased from (308.15 to 318.15)K, the negative value of ΔG^{*E} changes to positive and positive value becomes more positive. The increase in the values of ΔG^{*E} with increasing temperature indicates a weak specific interaction (like H-bond) between the component molecules.

3.4 Heat of Mixing

Heat of mixing of binary and ternary liquid mixtures namely, isobutanol (1) + 1,2-propanediol(2), isobutanol(1) + cyclohexanone (3), 1,2-propanediol (2) + cyclohexanone (3) and isobutanol (1) + 1,2-propanediol (2) + cyclohexanone (3) are endothermic. The heat of mixing (ΔH) of these liquids mixtures may be influenced by factors such as (1) rupture of hydrogen bonds and physical interaction between unlike or like molecules (endothermic) and (2) the formation of hydrogen bonded complexes (exothermic)[38]. The heat of mixing of above studied binary and ternary liquid mixtures were measured with Dewar Flask at equimolar concentration at room temperature. The calculated heat of mixing values for all the constituents binary and ternary liquid mixtures are presented in Table 8.

Table 8. Heat of mixing values for binary and ternary liquid mixtures

S. No.	Name of the mixtures	ΔH (J. mol^{-1})
1.	Isobutanol (1) + 1,2-Propanediol (2)	216.32
2.	1,2-Propanediol (2) + Cyclohexanone (3)	1066.50
3.	Isobutanol (1) + Cyclohexanone (3)	871.26
4.	Isobutanol (1) + 1,2-Propanediol (2) + Cyclohexanone (3)	1055.07

From the Table 8, it is seen that there is a remarkable change of heat of mixing value and all the values are less positive for these liquid mixtures. This observation suggests that there is attractive interaction among unlike molecules(That is the order of joules only). This is also supporting evidence of V^E.

3.5 FT-IR spectral studies

FT-IR spectra of pure liquids, binary and ternary liquid mixtures (Fig. 7-13) of isobutanol + 1,2-propanediol + cyclohexanone at equimolar concentration were recorded and the characteristic peaks are tabulated in Table 9.

Table 9. FT-IR spectral data for pure liquids, binary and ternary liquid mixtures of isobutanol(1) + 1,2-propanediol(2) + cyclohexanone(3) at equimolar concentration

S. No.	Name of the pure liquids	Before mixing $\overline{\upsilon}$ -OH (cm^{-1})	Name of the binary and ternary liquid mixtures	After mixing $\overline{\upsilon}$ -OH (cm^{-1})
1.	Isobutanol	3314.46	Isobutanol(1) + 1,2-Propanediol (2)	3369.00
2.	1,2-Propanediol	3339.85	Isobutanol (1) + Cyclohexanone (3)	3411.18
3.	Cyclohexanone	1706.85 (C=O)	1,2-Propanediol (2) + Cyclohexanone (3)	3328.20
			Isobutanol (1) + 1,2-Propanediol (2) + Cyclohexanone (3)	3392.88

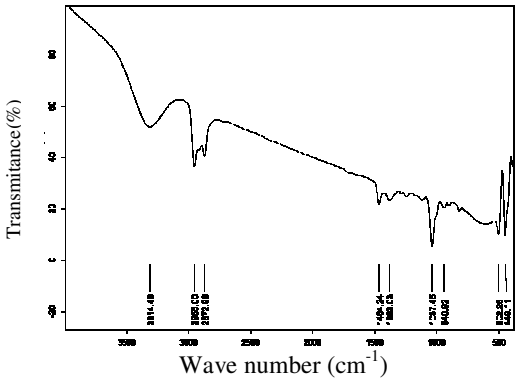


Fig. 7 FT-IR spectra for pure isobutanol

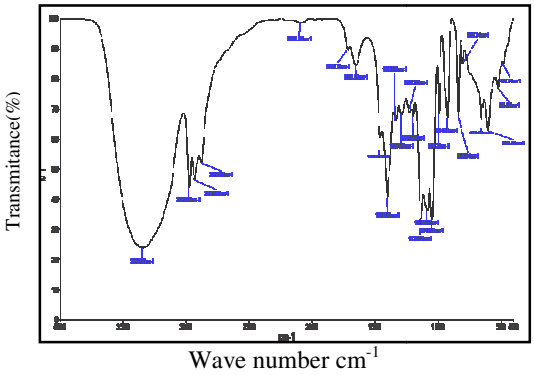


Fig. 8 FT-IR spectra for pure 1,2-propanediol molecule

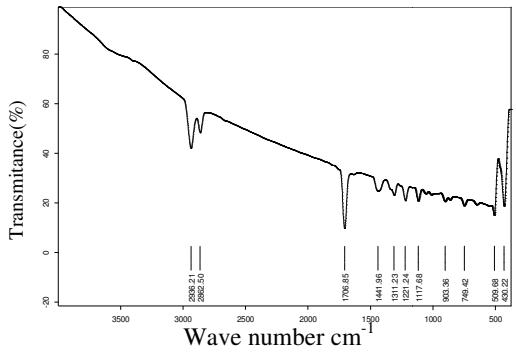


Fig. 9 FT-IR spectra for pure cyclohexanone molecule

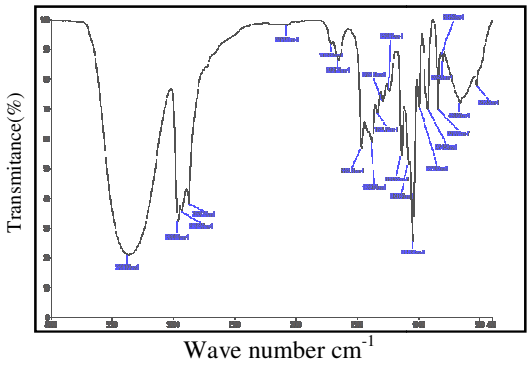


Fig. 10 FT-IR spectra for binary liquid mixtures of isobutanol (1) + 1,2-propanediol (2)

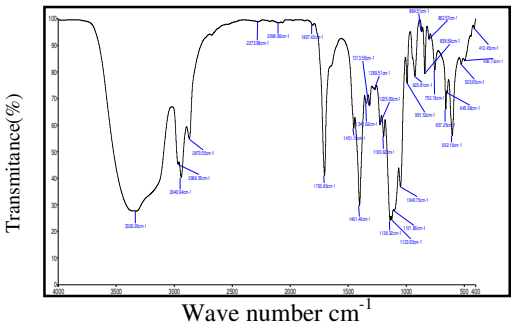


Fig. 11 FT-IR spectra for binary liquid mixtures of 1,2-propanediol (1) + cyclohexanone (2)

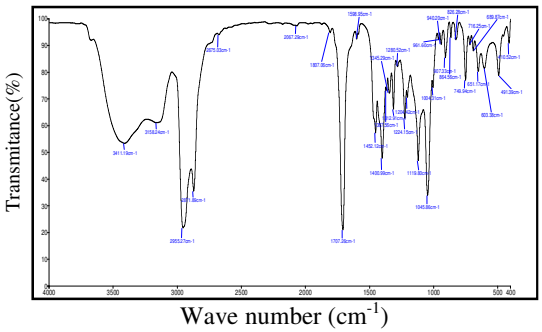


Fig. 12 FT-IR spectra for binary liquid mixtures of isobutanol (1) + cyclohexanone (2)

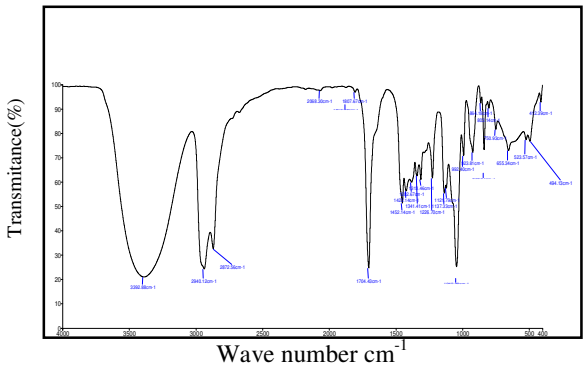


Fig. 13 FT-IR spectra for ternary liquid mixtures of isobutanol (1) + 1,2-propanediol (2) + cyclohexanone (3)

Hydrogen bonding changes both stretching and bending vibrations of functional group. The sharp band around 3650 cm^{-1} is due to free –OH group of alcohol molecules and the weak band observed at 3350 cm^{-1} is due to hydrogen bonded –OH. The intermolecular hydrogen bonding involves association of two or more molecules of same compound or different compounds. The band that results from intermolecular hydrogen bonding appears at lower energies [39].

When isobutanol was mixed with 1,2-propanediol and cyclohexanone, the absorption bands are shifted to longer wave number when compared to those of pure liquids. Hence there is significant shift in the wavenumber caused by intermolecular hydrogen bonding between the unlike molecules after mixing. These are clearly observed in Figs. 7-13. From the Tables and Figures, hydrogen bonded alcohols i.e. (pure liquids) for isobutanol shows strong broad peak at 3314.46 cm^{-1} 1,2-propanediol shows a broad moderate peak at 3339.85 cm^{-1} and cyclohexanone has a strong peak at 1706.84 cm^{-1} for C=O group. At equimolar concentration, the–OH peaks of components were shifted to 3369.0 cm^{-1}, 3411.19 cm^{-1}, 3328.20 cm^{-1} and 3392.88 cm^{-1} longer wave number. This shows the formation of weaker intermolecular hydrogen bonding between the unlike molecules in the present investigation. This supports the conclusion reached from V^E, viscosity and ΔH data.

4. CONCLUSIONS

The densities and viscosities for binary and ternary liquid mixtures of isopropanol + 1,2-propanediol + cyclohexanone are determined experimentally, at (308.15 and 318.15)K over whole composition range. The value of V^E, $\Delta\eta$ and ΔG^{*E} are calculated from experimental results at both temperatures. The excess deviation functions are fitted to Redlich-Kister type polynomial equation and corresponding standard deviations (σ) are calculated. A close observation of negative values of V^E, $\Delta\eta$ and positive values of ΔG^{*E} suggest the presence of weaker intermolecular hydrogen bonding and dipole-dipole interaction between the unlike molecules. The less positive values of (ΔH) and FT-IR spectral data reveals the formation of weaker intermolecular hydrogen bonding between the mixing components.

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