Acoustical properties of ternary mixtures of 1-alkanols in di-isopropyl ether and 2,2,2-trifluoroethanol mixed solvent

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Ultrasonic velocity (U), density (ρ) and viscosity (η) for the ternary liquid mixtures of diisopropyl ether+2,2,2-trifluoroethanol+1-propanol, diisopropyl ether+2,2,2-trifluoroethanol+1-butanol and diisopropyl ether+2,2,2-trifluoro-ethanol+1-pentanol have been measured as a function of the composition at 303, 308 and 313 K. The experimental data have been used to calculate some excess thermodynamical parameters such as viscosity (η^E), adiabatic compressibility (β^E), free length (L^E_f), free volume (V^E_f), internal pressure (π^E_i) and Gibb's free energy (ΔG^E). The results have been discussed and interpreted in terms of structural and specific interaction predominated by hydrogen bonding.

Keywords: Ultrasonic velocity, Adiabatic compressibility, Free length, Free volume, Gibb's free energy

1 Introduction

Ultrasonic studies have found wide applications owing to their ability to characterize the physico-chemical behaviour of solutions. The measurement of ultrasonic velocity in solutions can provide useful information regarding the degree of deviation from ideality, molecular association in solutions and important correlations with various acoustical parameters[1]. Ultrasonic velocity measurements are useful when dealing with the problems of structure and molecular interactions in liquids because of their accuracy[2]. The studies on volumetric, ultrasonic and viscometric properties of liquid mixtures and their dependence on composition and temperature are of importance in many fields of applied research and find applications in many important chemical, industrial and biological processes[3]. The excess properties have been claimed to be an aid in the characterization of the molecular interactions that are present in solutions and liquid mixtures. This is achieved through elevation of ideal quantities. In recent years, considerable efforts have been given for the evaluation of ideal and excess thermodynamic quantities of binary and ternary liquid mixtures[4].

The present work is part of our program to provide data for the characterization of the molecular interactions between solvents and commercially important alcohols, in particular the influence of the chemical structure of the solute in the system under consideration. The chemicals used in this work are chosen for their several applications in chemical industries. The 2,2,2-trifluoroethanol (commercially called fluorinol) is an effective solvent in bio-engineering[5,6] for catalytic polymerization reactions and for noble and non-polar gases. Branched (tertiary) ethers, such as diisopropyl ether are non-toxic and non-polluting chemicals. They are widely used octane enhancing additives in gasoline production processes, improving combustion and reducing emissions and contaminant agents of automobile catalysts, and are alternative oxygenated stabilizers in unleaded gasoline[7-9]. The knowledge of thermodynamic properties of these systems are valuable to improve their separation and recovery from product streams in order to reduce the costs of use as clean and efficient new energy fluids[10].

In the present study, the measurements on ultrasonic velocity, density and viscosity and their related excess thermodynamical and transport parameters for systems of diisopropyl ether+2,2,2-trifluoroethanol+1-propanol, diisopropyl ether+2,2,2-trifluoroethanol+1-butanol, diisopropyl ether+2,2,2-trifluoroethanol+1-pentanol at 303, 308 and 313 K have been undertaken. The dependence of excess parameters on composition has been used to explain the nature and extent of intermolecular interactions in these mixtures.

2 Experimental Details

All the chemical used are of analytical reagent (AR) and spectroscopic reagent (SR) without further purification. The purities of the above chemicals were

checked by density determination at 303, 308 and 313 ± 0.1 K, which showed an accuracy of $\pm1\times10^{-4}$ gcm^{-3} with the earlier values[10-18]. The ternary liquid mixtures of different known compositions were prepared in stopper measuring flasks. The density, viscosity and velocity were measured as a function of composition of the ternary liquid mixture at 303, 308 and 313 K for mixed solvent systems in which 1-propanol, 1-butanol and 1-pentanol were added to a binary mixtures of diisopropyl ether and 2,2,2-tri-fluroethanol. For this purpose binaries with fixed volume ratios $X_1/X_2 \cong 4{:}1$ were prepared. The density was determined using a specific gravity bottle by relative measurement method with an accuracy of ±0.01 kgm^{-3}. The weight of the sample was measured using electronic digital balance with an accuracy of ±0.1 mg (Model: SHIMADZU AX-200). An Ostwald's viscometer (10 ml) was used for the viscosity measurement. Efflux time was determined using a digital chronometer within ±0.01 s. An ultrasonic interferometer having the frequency of 3 MHz (Mittal Enterprises, New Delhi, Model: F-81) with an overall accuracy of $\pm0.1\%$ has been used for velocity measurement. An electronically digital operated constant temperature bath (RAAGA Industries) has been used to circulate water through the double walled measuring cell made up of steel containing the experimental solution at the desired temperature. The accuracy in the temperature measurement is ±0.1 K.

Various acoustical and thermodynamical parameters calculated from the measured data are as follows:

Adiabatic compressibility

$$\beta = \frac{1}{U^2\rho} \qquad \ldots(1)$$

Intermolecular free length

$$L_f = K\,\beta^{1/2} \qquad \ldots (2)$$

where K is temperature dependent constant. Its value is 631×10^{-6}, 636×10^{-6} and 642×10^{-6}, respectively at 303, 308 and 313 K.

Free volume

$$V_f = \left(\frac{M_{\text{eff}}\,U}{K\eta}\right)^{3/2} \qquad \ldots (3)$$

where M_{eff} is the effective molecular weight ($M_{\text{eff}}=\Sigma m_i x_i$, in which m_i and x_i are the molecular weight and the mole fraction of the individual constituents, respectively). K is a temperature independent constant which is equal to 4.28×10^9 for all liquids.

Internal pressure

$$\pi_i = bRT\left(\frac{K\eta}{U}\right)^{1/2}\left(\frac{\rho^{2/3}}{M_{\text{eff}}^{7/6}}\right) \qquad \ldots (4)$$

where b is the cubic packing which is assumed to be 2 for all liquids and solutions R. is the universal gas constant.

Gibb's free energy

$$\Delta G = KT\ln\left(\frac{KT\tau}{h}\right) \qquad \ldots (5)$$

where K is Boltzmann's constant (1.38×10^{-23} JK^{-1}), T is the absolute temperature, h the Planck's constant (6.626×10^{-34} Js) and τ is the relaxation time [$\tau=(4/3)\eta\beta$].

Excess values of the above parameters can be determined using:

$$A^E = A_{\text{exp}} - A_{\text{id}} \qquad \ldots (6)$$

where $A_{\text{id}} = \Sigma A_i X_i$, A_i, is any acoustical parameters and X_i the mole fraction of the liquid component.

3 Results and Discussion

The values of density (ρ), viscosity (η) and ultrasonic velocity (U) of pure liquids and liquid mixtures at 303, 308 and 313 K are given in Tables 1 and 2. The values of excess viscosity (η^E), excess adiabatic compressibility (β^E), excess free length (L^E_f), excess free volume (V^E_f), excess internal pressure (π^E_i) and excess Gibb's free energy (ΔG^E) have been calculated and presented in Tables 3 and 4. Further, the variation of excess viscosity values with mole fractions of 1-alkanols (X_3) at 303, 308 and 313 K is shown in Fig. 1 and the curves are drawn using least square fitting.

The thermodynamic excess properties are found to be more sensitive towards intermolecular interaction between the component molecules of the liquid mixtures. The sign and extent of deviation of excess properties depend on the strength of the interaction between unlike molecules[19]. The excess viscosity (Fig. 1) is negative through the whole range of concentration in all the three systems. From the analysis and close observation it is found that they decrease with increase in mole fraction of X_3 but it is

Table 1 — Comparison of experimental values of density (ρ), viscosity (η) and ultrasonic velocity (U) of pure liquids at 303, 308 and 313 K with literature values

Organic liquids	T/K	$\rho/(kg\ m^{-3})$		$\eta/(\times10^{-3}\ Nsm^{-2})$		$U/(ms^{-1})$	
		Exptl	Lit	Exptl	Lit	Exptl	Lit
Diisopropyl ether	298		718.7[10]				
	303	714.1		0.3690		958.9	
	308	691.3		0.3377		936.2	
	313	679.7		0.3019		927.0	
2,2,2-Trifluoroethanol	298		1382.11[10]				
	303	1364.4		1.5546		845.3	
	308	1358.1		1.3919		821.0	
	313	1352.8		1.1061		813.0	
1-Propanol	303	795.3	795.6[11]	1.7262	1.725[11]	1217.7	1180[18]
	308	791.3	791.2[11]	1.4919	1.526[15]	1182.7	
	313	787.8	787.4[12]	1.3624	1.366[16]	1180.5	
1-Butanol	303	799.2	801.8[11]	2.2753	2.268[11]	1225.8	1211[13]
	308	797.8	798.1[11]	1.9761	1.982[11]	1209.8	
	313	796.4	794.3[17]	1.7901	1.764[16]	1191.8	
1-Pentanol	303	806.9	807.2[14]	2.8955	2.909[14]	1254.9	
	308	798.8		2.5024		1240.2	
	313	795.9		2.2846		1228.5	

Table 2 — Values of density (ρ), viscosity (η) and ultrasonic velocity (U) at 303, 308 and 313 K

Mole fraction (X_3)	$\rho/(kg\ m^{-3})$			$\eta/(\times10^{-3}Nsm^{-2})$			$U/(ms^{-1})$		
	303 K	308 K	313 K	303 K	308 K	313 K	303 K	308 K	313K
System-I: Diisopropyl ether (X_1)+2,2,2-trifluoroethanol (X_2)+1-propanol (X_3) [(X_1)/(X_2)]=4:1									
0.0000	815.7	805.8	795.9	0.4642	0.4364	0.4020	922.3	918.8	893.7
0.0210	821.4	806.9	805.5	0.4662	0.4368	0.4062	924.3	920.0	894.7
0.0405	829.6	815.4	807.8	0.4685	0.4398	0.4089	931.2	921.4	898.2
0.0601	836.5	829.7	817.8	0.4696	0.4401	0.4091	936.4	925.2	904.5
0.0802	839.5	831.3	824.8	0.4700	0.4408	0.4096	941.5	931.0	913.5
0.0998	841.9	838.4	828.0	0.4702	0.4435	0.4102	946.9	943.5	923.5
System-II: Diisopropyl ether (X_1)+2,2,2-trifluoroethanol (X_2)+1-butanol (X_3) [(X_1)/(X_2)]=4:1									
0.0000	815.7	805.8	795.9	0.4642	0.4364	0.4020	922.3	918.8	893.7
0.0205	830.9	807.5	806.4	0.4679	0.4376	0.4082	931.0	922.0	895.3
0.0399	834.7	825.0	809.5	0.4709	0.4476	0.4112	933.5	923.5	912.6
0.0601	838.8	830.7	818.7	0.4738	0.4512	0.4174	940.8	926.5	916.3
0.0803	841.4	835.6	826.4	0.4759	0.4583	0.4255	943.0	935.5	921.7
0.1004	843.4	840.9	829.2	0.4855	0.4630	0.4274	956.0	948.5	926.2
System-III: Diisopropyl ether (X_1)+2,2,2-trifluoroethanol (X_2)+1-pentanol (X_3) [(X_1)/(X_2)]=4:1									
0.0000	815.7	805.8	795.9	0.4642	0.4364	0.4020	922.3	918.8	893.7
0.0205	834.5	809.6	808.1	0.4871	0.4416	0.4110	935.5	923.4	897.5
0.0409	836.6	826.0	811.3	0.4915	0.4610	0.4168	945.2	936.5	914.5
0.0603	839.4	831.1	820.0	0.4974	0.4693	0.4223	947.6	943.4	917.5
0.0800	842.7	836.3	829.8	0.4989	0.4778	0.4280	948.6	945.5	924.3
0.1000	844.0	841.6	830.8	0.5037	0.4920	0.4302	957.1	952.2	928.5

Table 3 — Excess values of viscosity (η^E), adiabatic compressibility (β^E) and free length (L_f^E) at 303, 308 and 313 K

Mole fraction (X_3)	$-\eta^E/(\times 10^{-3}$ N sm$^{-2})$			$-\beta^E/(\times 10^{-10}$ m^{2}N$^{-1})$			$-L_f^E/(\times 10^{-8}$ m$)$		
	303 K	308 K	313 K	303 K	308 K	313 K	303 K	308 K	313 K
System-I: Diisopropyl ether (X_1)+2,2,2-trifluoroethanol(X_2)+1-propanol (X_3) [(X_1)/(X_2)]=4:1									
0.0000	0.1429	0.1121	0.0607	0.1774	0.6875	0.2017	0.0213	0.0490	0.0087
0.0210	0.1623	0.1306	0.0705	0.1306	0.6185	0.2736	0.0186	0.0421	0.0180
0.0405	0.1825	0.1465	0.0898	0.1034	0.6884	0.3154	0.0001	0.0470	0.0150
0.0601	0.2037	0.1650	0.1076	0.2561	0.9266	0.5770	0.0121	0.0663	0.0302
0.0802	0.2257	0.1832	0.1251	0.3329	1.0161	0.8579	0.0178	0.0732	0.0581
0.0998	0.2479	0.1994	0.1425	0.4117	1.3539	1.0893	0.0238	0.1017	0.0690
System-II: Diisopropyl ether (X_1)+2,2,2-trifluoroethanol(X_2)+1-butanol (X_3) [(X_1)/(X_2)]=4:1									
0.0000	0.1429	0.1121	0.0607	0.1774	0.6875	0.2017	0.0213	0.0490	0.0087
0.0205	0.1716	0.1395	0.0811	0.2313	0.6836	0.3201	0.0118	0.0474	0.0164
0.0399	0.2020	0.1580	0.1046	0.2503	0.9024	0.8163	0.0124	0.0648	0.0545
0.0601	0.2325	0.1830	0.1249	0.4108	0.9548	0.9593	0.0252	0.0681	0.0671
0.0803	0.2638	0.2044	0.1434	0.3968	1.1675	1.1214	0.0231	0.0855	0.0799
0.1004	0.2875	0.2283	0.1681	0.6706	1.4875	1.1653	0.0461	0.1125	0.0828
System-III: Diisopropyl ether (X_1)+2,2,2-trifluoroethanol(X_2)+1-pentanol (X_3) [(X_1)/(X_2)]=4:1									
0.0000	0.1429	0.1121	0.0607	0.1774	0.6875	0.2017	0.0213	0.0490	0.0087
0.0205	0.1648	0.1460	0.0882	0.4147	1.7570	0.4180	0.0272	0.0534	0.0242
0.0409	0.2062	0.1657	0.1188	0.6007	1.2940	0.8901	0.0419	0.0975	0.0614
0.0603	0.2461	0.1965	0.1498	0.5854	1.4338	0.9894	0.0395	0.1086	0.0688
0.0800	0.2888	0.2270	0.1805	0.5380	1.4325	1.2183	0.0344	0.1076	0.0871
0.1000	0.3314	0.2519	0.2147	0.6638	1.5581	1.2103	0.0445	0.1178	0.0854

Table 4 — Excess values of free volume (V_f^E), internal pressure (π_i^E) and Gibb's free energy (ΔG^E) at 303, 308 and 313 K

Molefraction (X_3)	$-V_f^E/(\times 10^{-7}$ m^{3}mol$^{-1})$			$-\pi_i^E/(\times 10^8$ Nm$^{-2})$			$-\Delta G^E/(\times 10^{-21}$ KJ mol$^{-1})$		
	303 K	308 K	313K	303 K	308 K	313K	303 K	308 K	313K
System-I: Diisopropyl ether(X_1)+2,2,2-trifluroethanol(X_2)+1-propanol(X_3) [(X_1)/(X_2)]=4:1									
0.0000	1.5690	1.8940	2.6031	0.5477	0.4954	0.3050	0.1431	0.2554	0.1175
0.0210	1.5852	1.9048	2.6614	0.6118	0.5694	0.3435	0.2349	0.3263	0.0443
0.0405	1.5783	1.9414	2.6759	0.6762	0.6164	0.4094	0.3807	0.4124	0.0378
0.0601	1.5686	1.9325	2.6422	0.7440	0.6620	0.4701	0.5148	0.5766	0.2098
0.0802	1.5533	1.9145	2.6015	0.8232	0.7393	0.5371	0.6332	0.6936	0.3873
0.0998	1.5340	1.8860	2.5659	0.9037	0.8057	0.6127	0.7549	0.8706	0.5524
System-II: Diisopropyl ether(X_1)+2,2,2-trifluoroethanol(X_2)+1-butanol(X_3) [(X_1)/(X_2)]=4:1									
0.0000	1.5690	1.8984	2.6031	0.5477	0.4954	0.3050	0.1431	0.2554	0.1175
0.0205	1.5494	1.8458	2.6210	0.5858	0.5599	0.3534	0.3498	0.3593	0.0300
0.0399	1.5535	1.9806	2.5763	0.6434	0.5541	0.4082	0.4488	0.4455	0.2018
0.0601	1.5330	1.9919	2.6143	0.7077	0.5998	0.4386	0.5930	0.5457	0.3026
0.0803	1.5297	2.0095	2.6672	0.7696	0.6450	0.4678	0.6911	0.6638	0.3924
0.1004	1.5452	1.9797	2.6438	0.8258	0.5293	0.6814	0.8163	0.8420	0.5115
System-III: Diisopropyl ether (X_1)+ 2,2,2-trifluroethanol (X_2)+ 1-pentanol (X_3) [(X_1)/(X_2)]=4:1]									
0.0000	1.5690	1.8984	2.6031	0.5477	0.4954	0.3050	0.1431	0.2554	0.1175
0.0205	1.6997	1.9056	2.6647	0.5260	0.5391	0.2148	0.2553	0.3604	0.0339
0.0409	1.6628	2.0177	2.5975	0.5855	0.5200	0.3788	0.4143	0.4758	0.2030
0.0603	1.6745	2.0450	2.6127	0.6325	0.5396	0.4077	0.4993	0.5817	0.3180
0.0800	1.6530	2.0901	2.6044	0.6895	0.5798	0.4396	0.6116	0.6437	0.4726
0.1000	1.6254	2.1634	2.5673	0.7532	0.5941	0.5003	0.7521	0.6993	0.5923

System I: diisopropylether (X_1)+2,2,2-trifluoroethanol (X_2) +
1-propanol (X_3)

System II: diisopropylether (X_1)+2,2,2-trifluoroethanol (X_2) +
1-butanol (X_3)

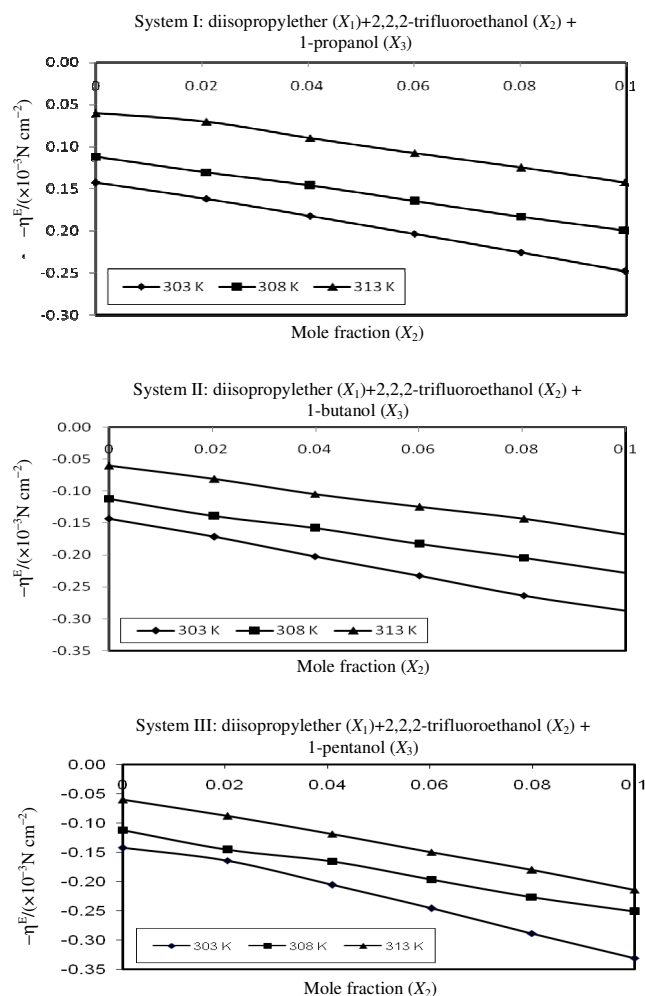System III: diisopropylether (X_1)+2,2,2-trifluoroethanol (X_2) +
1-pentanol (X_3)

Fig. 1 — Variation of excess viscosity (η^E) of diisopropyl ether and 2,2,2-trifluoroethanol mixed solvent system with mole fraction of 1-alkanols (X_3) at 303, 308 and 313 K.

found to be increases with the rising of temperature. This observed behaviour shows the existence of molecular interaction between the components of mixture for all the systems studied.

A deviation from Raoult's law on mixing of two non-electrolytes demonstrates a molecular interaction between unlike molecules, which is attributed to adhesive and cohesive forces[20]. Dispersion forces are responsible for positive deviations where the size of molecules are taken into considerations. Negative deviations of high magnitude lead to complex formation between the heteromolecules of the mixture through hydrogen bond formation[21]. To explain inter molecular interaction the sign and magnitude of β^E may be considered. A negative value of β^E indicates strong interaction between the components of the mixtures, while their positive values are indicative of weak interaction. When the strength of interaction

between unlike molecules increase, β^E becomes increasingly negative. A similar effect was observed in all the three systems studied at all temperatures (Table 3). In the present study the negative β^E values for ternary mixtures may be explained due to the formation of hydrogen bond between the fluorine of 2,2,2-trifluoroethanol and OH group of 1-alkanols in all the systems. When mixing ethers and alcohols together with fluoro-hydrocarbons, three types of specific hydrogen bonding are generated: O-H···O, H···O-H, O-H···F.

From the variation of excess free length (Table 3) it was observed that the L^E_f values are negative over the entire mole fraction range and even with the rising of temperature. But, however, these values are maintained with the same trend of excess adiabatic compressibility. According to Kannappan et al.[22], negative value of L^E_f indicates that sound waves cover long distances due to decrease in intermolecular free length describing the dominant nature of hydrogen bond interaction between unlike molecules. Fort and Moore[23] indicated that the positive excess values of free length should be attributed to the dispersive forces and negative excess values should be due to charge transfer and hydrogen bond formation. In the present study the negative contribution of L^E_f in all the three systems prevails the existence of greater interaction. Spencer et al.[24] have also reported a similar observation on the basis of excess values of free length.

Table 4 gives a qualitative picture of the excess free volume for the three ternary liquid systems. This indicates the extent of deviation from ideal with the mole fraction of the mixtures. The excess values for all the three systems are found to be negative. These values increase with increasing the mole fraction of X_3 but found to decrease with the rising of temperature. The results can be explained in terms of molecular interaction, structural effect and interstitial accommodation along with changes in free volume. The sign of V^E_f depends upon the relative strength between the contractive forces and expansive forces. The factors responsible for volume contraction are: (a) specific interaction between component molecules; (b) interstitial accommodation of molecules of one component into the vacant spaces of molecules of the other components. This occurs preferentially when the size difference between the components molecules is large, or when large gaps are available in the structural network of molecules and (c) weak physical forces, such as dipole-dipole or

dipole-induced dipole interactions or Vander waal's forces. The factors that cause expansion in volume are dispersion force, steric hindrance of component molecules, unfavourable geometric fitting and electrostatic repulsion.

The negative part of V^E_f curves of the system asserts that the combined effect of the factors responsible for volume contraction outweigh the combined effect of the factors causing volume expansion and vice-versa[25]. Adgaonkar *et al.*[26] showed positive value of V^E_f, indicating the existence of weak molecular interaction in the liquid mixtures. Fort *et al.*[23] noticed that negative excess free volume tends to decrease as the strength of the interaction between unlike molecules increases although they are not parallel with the excess compressibilities. However, in the present paper the observed behaviour of V^E_f shows that the strength of molecular interaction increases with increasing of 1-alkanols, but is lesser with increasing of temperature. The magnitude of V^E_f values follows the sequence: 1-pentanol < 1-butanol < 1-propanol.

In the study of liquid mixtures the variation of internal pressure may give some reliable information regarding the nature and strength of the forces existing between the molecules. In fact, the internal pressure is a broader concept and is a measure of the totality of forces of the dispersion, ionic and dipolar interaction that contribute to be overall cohesion of the liquid system. The excess internal pressure (Table 4) is negative but it is found to be decreasing with increasing the mole fraction of X_3 and increases with rising of temperature in all the three systems studied. The negative value of π^E_i indicate that only dipolar forces are operating between the unlike molecules. Further the observed behaviour of π^E_i values reveals that the strengthening of cohesive forces results perhaps due to making up of the structure of the solvent.

Table 4 shows the variation of ΔG^E for the three ternary liquid systems. The values of ΔG^E decrease with increasing the 1-alkanols content, but it is found to increase with rising of temperature. According to Reed *et al.*[27], the positive ΔG^E may be attributed to specific interactions like hydrogen bonding and charge transfer, while negative values may be ascribed to the dominance of dispersion forces[28]. In the present study the observed behaviour of ΔG^E shows the existence of molecular interactions between the components of unlike molecules

4 Conclusions

Ultrasonic method is a powerful tool for characterizing the physico-chemical properties and existence of molecular interaction in the mixture. The results of excess property reveals that, the strong molecular interaction exists in the mixtures which may be due to the dominance of hydrogen bonding and charge transfer between the mixing components. Alcohols, ethers and fluoro-hydrocarbons are highly polar molecules. Ether molecules do not associate among each other, while alcohols are strongly self-associated by means of hydrogen bonds. When mixing ethers and alcohols together with fluoro-hydrocarbons, three types of specific hydrogen bonding are generated: O-H⋯O, H⋯O-H, O-H⋯F. The strength of molecular interactions throughout formation/breaking of hydrogen bonding is strongly reflected by the magnitudes of η^E, β^E, L^E_f and V^E_f. From the magnitude of V^E_f, it can be concluded that the strength of interaction is in the order: 1-pentanol > 1-butanol > 1-propanol. But the strength of interaction tends to be weaker with rising of temperature due to the presence of weak intermolecular forces and thermal dispersion forces.

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