



ULTRASONIC STUDIES ON SOME TERNARY ORGANIC LIQUID MIXTURES AT 303, 308 AND 313K

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ABSTRACT

Density (ρ), viscosity (η) and Ultrasonic velocity (U) measurement have been carried out for three ternary organic liquid mixture of 1-alkanols in the binary mixtures of tetrahydrofuran (THF) and 1-chlorobutane (3:1) ratio at 303, 308, 313K. From the experimental data, the acoustical parameters such as adiabatic compressibility (β), free length (L_f), free volume (V_f), internal pressure (π_i), and Gibb's free energy (ΔG) have been calculated. The excess values of the above parameters and Grunberg-Nissan interaction parameter (d) have also been evaluated and discussed in the light of molecular interaction in the mixtures.

Keywords: ultrasonic velocity, adiabatic compressibilities, free volume, Gibb's free energy.

INTRODUCTION

The ultrasonic velocity measurements find wide applications in characterising the physico-chemical behaviour of liquid mixtures¹⁻³ and in the study of molecular interactions. Ultrasonic velocity of a liquid is related to the binding forces between the atoms or the molecules. Ultrasonic velocities have been adequately employed in understanding the nature of molecular interaction in pure liquids⁴, binary and ternary mixtures⁵⁻⁷. The method of studying the molecular interaction from the knowledge of variation of thermodynamic parameters and their excess values with composition gives an insight into the molecular process⁸⁻¹⁰. The number of studies on thermodynamic properties of multi-component liquid mixtures has increased in recent years due to industrial applications and the theoretical interest in studying the nature of molecular interaction and packing phenomena in ternary mixtures.

The investigations regarding the molecular association in organic ternary mixtures having 1-alkanols group as one of the components is of particular interest, since alkanols group is highly polar and can associate with any other group having some degree of polar attractions. Tetrahydrofuran also known as THF, is a heterocyclic organic compound with the formula [(CH₂)₄O]. It is a colourless low-viscosity liquid with a smell similar to diethyl ether. It is one of the most polar ethers. THF is the fully hydrogenated analogue of the aromatic compound furan. THF is an aprotic solvent with a dielectric constant of 7.6. It is a moderately polar, aprotic solvent that dissolves a wide range of non-polar and polar compounds. It is often used in polymer science.

Owing to these considerations, an attempt has been made to elucidate the molecular interactions in the mixtures of 1-propanol, 1-butanol and 1-pentanol with THF in 1-chlorobutane at 303, 308 and 313K. Further, the excess values of some of the acoustical and interaction parameters have been calculated from the measurements of ultrasonic velocity, density and viscosity of the mixtures. The excess parameters are used to explain intermolecular interactions in these ternary mixtures.

EXPERIMENTAL

All the chemicals used in this present research work are spectroscopic reagent (SR) grade and analytical reagent (AR) grades of minimum assay of 99.9% obtained from E-Merck, Germany and Sd Fine Chemicals, India, which are used as such without further purification. The purities of the above chemicals were checked by density determination at 303, 308 and 313K \pm 0.1K which showed an accuracy of \pm

1×10^{-4} g on a electronic digital balance (Model: SHIMADZU AX 200). 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 313K for 1-propanol, 1-butanol and 1-pentanol were added to binary mixtures of THF and 1-chlorobutane. For this purpose, binaries with fixed mole ratios $X_1/X_2 \cong 3:1$ were prepared by mass. The density was determined using a specific gravity bottle by relative measurement method with an accuracy of ± 0.01 $\text{kg}\cdot\text{m}^{-3}$. An Ostwald's Viscometer (10ml capacity) was used for the viscosity measurement and efflux time was determined using a digital Chronometer to within ± 0.01 s. An ultrasonic interferometer having the frequency 3MHz (MITTAL ENTERPRISES, NEW DELHI, MODEL F-81) with an overall accuracy of $\pm 0.1\%$ 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.

Theory and Calculations

Various physical and thermodynamical parameters are calculated from the measured data such as

$$\text{Adiabatic Compressibility } \beta = \frac{1}{U^2 \rho} \quad (1)$$

$$\text{Intermolecular free length } L_f = K\sqrt{\beta} \quad (2)$$

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

$$\text{Free volume } V_f = \left(\frac{M_{eff} U}{K\eta} \right)^{3/2} \quad (3)$$

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

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

where b is the cubic packing which is assumed to be 2 for all liquids and solutions, η is the viscosity, R is a gas constant and T absolute temperature.

The Gibb's free energy can be estimated from the following relation.

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

where K is the Boltzmaan's constant ($1.23 \times 10^{-23} \text{ JK}^{-1}$), T the absolute temperature, 'h' the Planck's constant (6.626×10^{-26} Js) and τ is the relaxation time ($\tau = \frac{4}{3} \eta \beta$).

Excess values of the above parameters can be determined using

$$A^E = A_{exp} - A_{id} \quad (6)$$

where $A_{id} = \sum A_i X_i$, A_i is any acoustical parameters and X_i the molefraction of the liquid component.

Grunberg and Nissan formulated the following relation between the viscosity of a binary liquid mixture and pure components.

$$\ln \eta_{mix} = X_1 \ln \eta_1 + X_2 \ln \eta_2 + X_1 X_2 d \quad (7)$$

On applying to a ternary liquid mixture, this equation takes up the form

$$\ln \eta_{mix} = X_1 \ln \eta_1 + X_2 \ln \eta_2 + X_3 \ln \eta_3 + X_1 X_2 X_3 d \quad (8)$$

where d is an interaction parameter regarded as a measure of the strength of molecular interactions between the mixing components.

RESULTS AND DISCUSSION

The experimental values of density (ρ), viscosity (η) and ultrasonic velocity (U), for pure liquids and three ternary liquid systems at 303, 308 and 313K are presented in Tables 1-2. The excess values of viscosity, adiabatic compressibility, free length, free volume, internal pressure, Gibb's free energy and Grunberg interaction parameter 'd' have been calculated and presented in Tables 3-4. The variation of excess acoustical parameters with mole fraction of 1-alkanols (X_3) in THF and 1-chlorobutane mixtures at 303, 308 and 313K are plotted in Figs.1-6, 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 liquid mixtures. The sign and extent of deviation of excess parameters depend on the strength of interaction between unlike molecules¹¹. The measurement of viscosity in ternary liquid mixture gives some reliable information in the study of intermolecular interaction. According to Fort *et.al.*,¹² the excess viscosity gives the strength of the molecular interaction between the interacting molecules. For system where dispersion, induction and dipolar forces are operating the values of excess viscosity are found to be negative, whereas the existence of specific interactions leading to the formation of complexes in liquid mixtures tends to make excess viscosity positive. 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 concentration of 1-alkanols (X_3) but it increases non-linearly with rising of temperature. The observed behaviour of η^E in all systems suggests the less strengthening of interaction between the components of mixture.

The Fig.2. shows that the excess value of adiabatic compressibilities are positive and it decreases non-linearly with increasing the concentration of X_3 as well as rising of temperature in all systems studied. Fort *et.al.*,¹² found that the increasing negative value of excess compressibilities indicates greater interaction between the components of the mixtures. Positive values in excess properties correspond mainly to the existence of dispersive forces. The negative value of β^E is associated with a structure forming tendency while a positive values is taken to indicate a structure breaking tendency due to heteromolecular interaction between the component molecules of the mixtures. The negative β^E values for ternary mixtures indicate, the formation of H bonds. The positive excess adiabatic compressibility which indicates loosely packed molecules in the mixtures results due to shape and size. In the present investigation, the positive contribution of β^E in all systems suggests the existence of dispersive forces, and the decreasing behaviour of β^E values with increasing of 1-alkanols as well as rise in temperature shows the weakening of interaction exists in the mixtures.

Fig.3 shows that the variations of excess free length are positive and decreases non-linearly with increasing the contents of 1-alkanols as well as rising temperature in all systems studied. According to Ramamoorthy *et.al.*¹³ negative values of excess intermolecular free length L_f^E indicates that sound waves cover longer distances due to decrease in intermolecular free length ascribing the dominant nature of hydrogen bond interaction between unlike molecules. Fort *et.al.*,¹² indicated that the positive values of excess free length should be attributed to the dispersive forces and negative excess values should be due to charge transfer, dipole-induced dipole and dipole-dipole interactions. In the present study the positive contribution of L_f^E in all systems shows the existence of dispersive forces between the component of the mixtures. Spencer *et.al.*⁶ have also reported a similar observation on the basis of excess values of free length.

Fig.4 gives a qualitative picture of the excess free volume for the ternary liquid mixtures. The values of V_f^E are found to be positive and increase with increasing the molefraction of X_3 upto 0.5 and beyond it decreases for every mixtures. Further, these values are increases with rising of temperature in all systems studied. For some ternary liquid mixtures, Fort *et.al.*¹² notice the negative excess free volume tends to decrease as the strength of the interaction between the unlike molecules increases although they do not parallel with the excess compressibilities. The values of V_f^E are the resultant of contributions from several opposing effects¹⁴. These may be divided arbitrarily into these types, namely, chemical, physical and structural contributions, physical contributions, which are non-specific interactions between the real species present in the mixture, contribute a positive term to V_f^E . The chemical or specific intermolecular interactions and structural (interstitial accommodation) contributes negative value V_f^E . In the present study, the positive contribution of V_f^E in all the systems show the existence of dispersive forces between the component molecules of the mixtures. Further, from the observed behaviour of V_f^E with rising of temperature suggests the strength of interaction get weakened in all systems studied. The magnitude of V_f^E values follows the sequence: 1-pentanol > 1-butanol > 1-propanol.

In the study of ternary liquid mixtures, the variation of the internal pressure may give some information regarding the nature and strength of the forces existing between the molecules. The excess internal pressure values (Fig.5) is found to be negative and it decreases with increasing the molefraction of (X_3) as well as temperature in all the three systems studied. The negative values of π_i^E indicates that only dispersion and dipolar forces are operating with complete absence of specific interaction¹⁵. In the present investigation, the observed behaviour of π_i^E shows the existence of dispersive forces in all systems studied, but the strength of interaction decreases with rising of temperature results the decreasing of π_i^E values.

Fig.6 shows the variation of excess Gibb's free energy is found to be negative in all systems studied and it decreases with increasing the molefraction of (X_3) as well as with elevation of temperature. According to Reed *et.al.*,¹⁶ the positive deviation in ΔG^E may be attributed to specific interactions like hydrogen bonding and charge transfer, whereas the negative deviations may be ascribed to dispersion forces with systems. In the present investigation, the negative excess Gibb's free energy (ΔG^E) obtained in all the systems shows the dominance of dispersion forces. Recently, Ali *et.al.*,¹⁷ attributes the increasing positive values of ΔG^E in few ternary liquid mixtures, to hydrogen bond formation between unlike molecules.

The interaction parameters 'd' in Grunberg and Nissan equation is a measure of the strength of interaction between the mixing components. Table 4 shows that the values of 'd' become negative and these values are decreases with increasing the content of 1-alkanols as well as rising of temperature in all the three system studied. 'd' values were said to indicate various types of interactions as follows¹⁸. Large and positive 'd' values indicate strong specific interaction; small positive value indicated weak specific interaction and large negative values indicated no specific interaction. The negative values of 'd' may be attributed to the dominance of dispersion forces. Kalara *et.al.*¹⁸ arising from the breaking of hydrogen bonds in the associated component of the mixtures. In the present investigation, the negative values of 'd' in the ternary systems have been attributed to the presence of no specific interactions between the unlike molecules. Further, the decreasing behaviour of 'd' values with rising of temperature shows the weakening of interaction between the component of mixtures.

CONCLUSION

Ultrasonic method is a powerful probe for characterising the physico-chemical properties and existence of molecular interaction in the mixture. In addition, the density, viscosity and the derived excess acoustical parameters provide evidence of confirmation. From the excess acoustical parameters, it is concluded that

the weak molecular interaction exist between the mixing components which may be due to the dominance of dispersion and dipolar forces. Further the strength of interaction tends to be weaker with rise in temperature due to weak intermolecular forces and thermal dispersion forces. From the magnitude of V_f^E the existence of molecular interaction in the mixture is in the order: 1-propanol > 1-butanol > 1-pentanol.

Table-1 : Values of density (ρ), viscosity (η) and ultrasonic velocity (U) of pure liquids at 303, 308 and 313K

Organic liquids	$\rho/(\text{kg m}^{-3})$			$\eta/(\times 10^{-3} \text{ Nsm}^{-2})$			U/(ms ⁻¹)		
	303K	308K	313K	303K	308K	313K	303K	308K	313K
Tetrahydrofuran (THF)	87	870.1	864.5	0.6455	0.6120	0.5600	1240.1	1222.1	1186.4
1-chlorobutane	87	875.6	862.7	0.5883	0.5218	0.4952	1084.5	1071.9	1041.8
1-propanol	79	790.6	786.7	1.7856	1.6320	1.4020	1190.2	1170.5	1154.4
1-butanol	80	796.3	792.6	2.0679	1.9230	1.8000	1233.2	1204.2	1191.5
1-pentanol	80	805.2	802.6	2.9580	2.9016	2.3923	1268.0	1225.1	1218.4

Table-2: Values of density (ρ) viscosity (η) and ultrasonic velocity (U) for

mole fraction X ₃	$\rho/(\text{kg m}^{-3})$			$\eta/(\times 10^{-3} \text{ Nsm}^{-2})$			U/(m.s ⁻¹)		
	303 K	308 K	313 K	303 K	308 K	313 K	303 K	308 K	313 K
System I: tetrahydrofuran (THF) (X ₁) + 1-chlorobutane (X ₂) + 1-propanol (X ₃)									
0	758.8	755.7	752.6	0.6174	0.5687	0.5613	1202.3	1192.4	1182.6
0.1	751.0	745.7	743.9	0.6477	0.5528	0.5204	1200.4	1188.3	1172.5
0.3	750.5	736.2	728.4	0.6700	0.5392	0.5033	1198.6	1176.6	1159.5
0.5	739.3	737.0	734.3	0.6914	0.5686	0.5249	1196.3	1167.6	1142.55
0.7	729.9	727.6	723.1	0.9001	0.6895	0.6174	1194.1	1177.35	1138.8
0.9	720.2	718.0	715.0	1.1005	0.9733	0.6806	1192.5	1166.55	1149.6
System II : tetrahydrofuran (THF) (X ₁) + 1-chlorobutane (X ₂) + 1-butanol (X ₃)									
0	758.8	755.7	752.6	0.6174	0.5687	0.5613	1202.3	1192.4	1182.6
0.1	755.9	755.6	748.8	0.6583	0.5669	0.5413	1204.2	1196.0	1186.4
0.3	746.0	710.6	704.7	0.7076	0.5649	0.5244	1210.3	1199.7	1194.2
0.5	740.9	738.4	735.9	0.7272	0.6733	0.6099	1218.2	1202.2	1196.3
0.7	732.8	729.3	725.1	1.0138	0.7638	0.6774	1224.6	1218.2	1202.2
0.9	711.9	707.9	700.2	1.6830	1.4861	1.1220	1236.2	1222.4	1218.4
System III : tetrahydrofuran (THF) (X ₁) + 1-chlorobutane (X ₂) + 1-pentanol (X ₃)									
0	758.8	755.7	752.6	0.6174	0.5687	0.5613	1202.3	1192.4	1182.6
0.1	758.3	753.6	752.7	0.6331	0.6075	0.5793	1203.0	1196.5	1187.6
0.3	746.4	742.4	738.4	0.7053	0.6668	0.6267	1220.9	1208.1	1194.1
0.5	742.1	740.4	736.6	0.7970	0.7664	0.7191	1234.2	1237.5	1198.8
0.7	731.5	727.8	724.9	1.1956	1.1236	1.0063	1245.8	1237.5	1210.6
0.9	717.2	715.0	711.5	1.7311	1.5831	1.4282	1259.0	1240.2	1228.6

Table-3: Excess values of viscosity (η^E), adiabatic compressibility (β^E), frelength (L_f^E), and free volume (V_f^E) for

mole fraction X ₃	$\eta^E/(\times 10^{-3} \text{ Nsm}^{-1})$			$\beta^E/(\times 10^{-10} \text{ m}^2 \text{ N}^{-1})$			$L_f^E/(\times 10^{-10} \text{ m})$			$V_f^E/(\times 10^{-8} \text{ m}^3 \text{ mol}^{-1})$		
	303 K	308 K	313 K	303 K	308 K	313 K	303 K	308 K	313 K	303 K	308 K	313 K
System I: tetrahydrofuran (THF) (X ₁) + 1-chlorobutane (X ₂) + 1-propanol (X ₃)												
0	-0.0238	-0.0208	-0.0175	1.1047	1.0506	0.6672	12.25	11.58	7.40	0.72	1.23	2.23
0.1	-0.0989	-0.1409	-0.1092	1.1420	1.1430	0.8741	12.57	12.47	9.49	0.32	3.20	3.37
0.3	-0.3075	-0.3630	-0.2980	1.0030	1.2627	1.1664	10.97	13.53	12.33	1.61	5.89	6.42

0.5	-0.5170	-0.5421	-0.4480	1.0067	1.2085	1.2462	10.85	12.80	12.98	3.14	6.29	6.84
0.7	-0.5392	-0.6297	-0.5271	0.9907	0.9757	1.3367	10.51	10.26	13.71	1.06	4.43	3.29
0.9	-0.5697	-0.5544	-0.6356	0.9732	1.1000	1.1148	10.14	11.30	11.35	1.60	2.12	6.14
System II : tetrahydrofuran (THF) (X ₁) + 1-chlorobutane (X ₂) + 1-butanol (X ₃)												
0	-0.0238	-0.0208	-0.0175	1.1047	1.0506	0.6672	12.25	11.58	7.40	0.72	1.23	2.23
0.1	-0.1166	-0.1559	-0.1281	1.0905	0.9555	0.6490	12.06	10.53	7.18	0.42	3.15	3.03
0.3	-0.3546	-0.4246	-0.3963	1.0780	1.4000	1.1008	11.83	15.01	11.76	1.77	6.79	8.13
0.5	-0.6224	-0.5829	-0.5620	0.9812	0.9121	0.6348	10.73	9.88	6.89	4.41	4.93	6.65
0.7	-0.6231	-0.7591	-0.7457	0.9453	0.7007	0.6712	10.26	7.57	7.20	1.39	5.68	7.54
0.9	-0.2412	-0.3035	-0.5524	0.9968	0.8339	0.7388	10.71	8.86	7.83	0.43	0.61	2.35
System III : tetrahydrofuran (THF) (X ₁) + 1-chlorobutane (X ₂) + 1-pentanol (X ₃)												
0	-0.0238	-0.0208	-0.0175	1.1047	1.0506	0.6672	12.25	11.58	7.40	0.72	1.23	2.23
0.1	-0.2308	-0.2132	-0.1494	1.1317	1.0108	0.6309	12.53	11.14	7.01	2.15	1.54	1.34
0.3	-0.6239	-0.6163	-0.4717	1.0709	0.9672	0.7985	11.87	10.61	8.74	3.72	3.70	3.87
0.5	-0.9976	-0.9791	-0.7490	0.9925	0.5539	0.8367	11.02	6.18	9.12	5.00	5.12	4.74
0.7	-1.0644	-1.0844	-0.8315	1.0177	0.7030	0.8924	11.26	7.70	9.68	1.52	1.80	2.17
0.9	-0.9942	-1.0872	-0.7793	0.1069	0.8202	0.8802	11.78	8.88	9.53	1.32	1.85	1.89

Table-4: Excess values of internal pressure (π_i^E), Gibb's free energy (ΔG^E) and Grunberg interaction parameter 'd' for

mole fraction X ₃	$(\pi_i^E) / (\times 10^8 \text{ Nm}^{-2})$			$(\Delta G^E) / (\times 10^{-22} \text{ KJ mol}^{-1})$			d		
	303 K	308 K	313 K	303 K	308 K	313 K	303 K	308 K	313 K
System I: tetrahydrofuran (THF) (X ₁) + 1-chlorobutane (X ₂) + 1-propanol (X ₃)									
0	-0.5167	-0.5525	-0.4408	4.33	4.29	3.51	-0.11	-0.02	-0.01
0.1	-0.8327	-1.0135	-0.9366	-2.29	-1.11	-1.44	-5.10	-10.79	-9.05
0.3	-1.5154	-1.8201	-1.6937	-4.88	-9.45	-9.01	-9.13	-14.26	-13.08
0.5	-2.2136	-2.4029	-2.1837	-11.58	-15.53	-14.51	-18.28	-23.21	-21.68
0.7	-2.2357	-2.6722	-2.4322	-9.72	-16.99	-15.36	-31.56	-47.02	-45.34
0.9	-2.2585	-2.2880	-2.7778	-10.20	-11.36	-19.85	-225.14	--245.81	-372.05
System II : tetrahydrofuran (THF) (X ₁) + 1-chlorobutane (X ₂) + 1-butanol (X ₃)									
0	-0.5167	-0.5525	-0.4408	4.33	4.29	3.51	-0.11	-0.02	-0.01
0.1	-0.7163	-0.8794	-0.8417	-3.66	-1.53	-1.78	-5.00	-10.22	-8.10
0.3	-1.2441	-1.6806	-1.6898	-4.12	-8.96	-10.49	-8.75	-14.35	-14.31
0.5	-1.8501	-1.8536	-1.9020	-12.43	-13.30	-15.80	-19.26	-19.50	-20.61
0.7	-1.7350	-2.2642	-2.3470	-9.12	-18.45	-20.90	-30.19	-48.08	-52.30
0.9	-1.0150	-1.2100	-1.8647	-1.06	-1.75	-10.48	-51.68	-82.52	-209.09
System III : tetrahydrofuran (THF) (X ₁) + 1-chlorobutane (X ₂) + 1-pentanol (X ₃)									
0	-0.5167	-0.5525	-0.4408	4.33	4.29	3.51	-0.11	-0.02	-0.01
0.1	-0.8472	-0.8354	-0.7324	-0.43	-0.22	-0.31	-9.93	-8.37	-5.51
0.3	-1.4559	-1.5060	-1.3277	-8.19	-9.02	-8.04	-12.77	-12.82	-10.94
0.5	-1.9967	-2.0762	-1.7545	-15.50	-17.62	-14.02	-22.99	-22.75	-19.65
0.7	-1.9191	-2.0886	-1.7900	-11.82	-14.64	-12.29	-37.44	-39.78	-35.65
0.9	-1.8390	-2.1072	-1.7386	-9.35	-13.30	-10.30	-225.90	-264.45	-217.81

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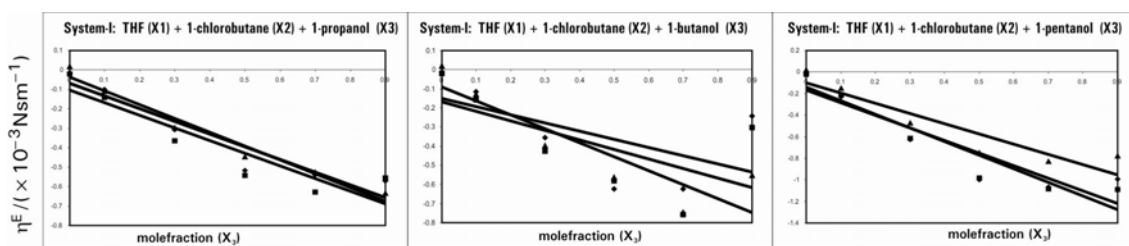


Fig. 1. The variation of excess viscosity with molefraction of 1-alkanols (X₃) in tetrahydrofuran and 1-chlorobutane mixture at 303, 308 and 313K

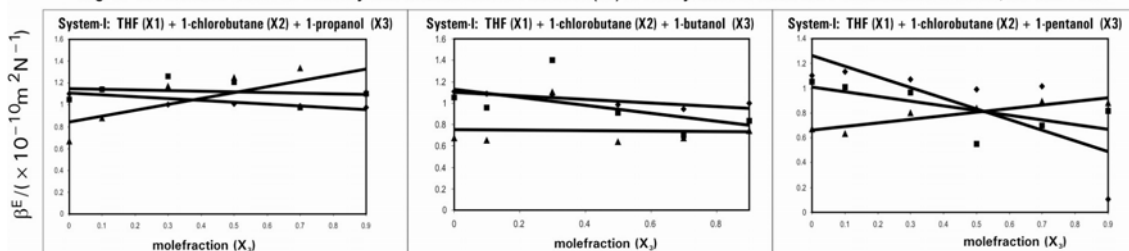


Fig. 2. The variation of excess adiabatic compressibility with molefraction of 1-alkanols (X₃) in tetrahydrofuran and 1-chlorobutane mixture at 303, 308 and 313K

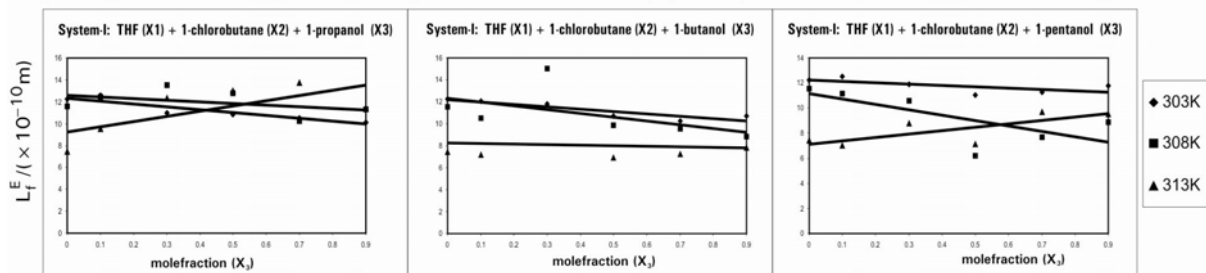


Fig. 3. The variation of excess frelength with molefraction of 1-alkanols (X₃) in tetrahydrofuran and 1-chlorobutane mixture at 303, 308 and 313K

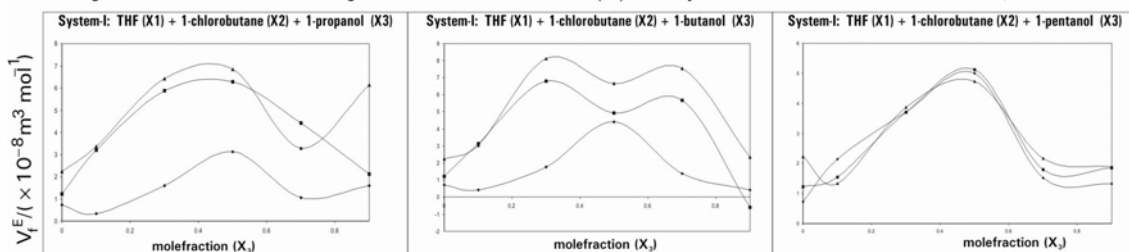


Fig. 4 The variation of excess free volume with molefraction of 1-alkanols (X₃) in tetrahydrofuran and 1-chlorobutane mixture at 303, 308 and 313K

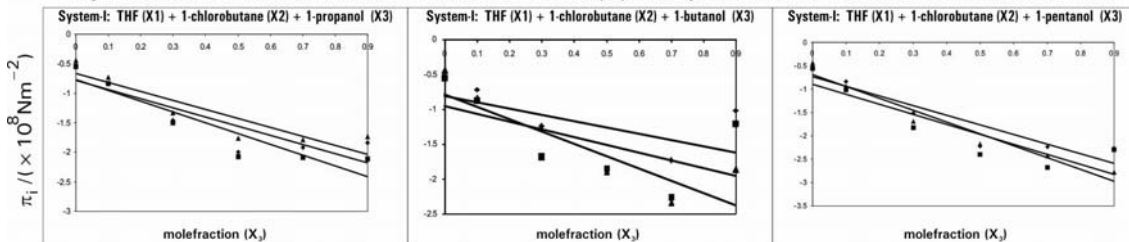


Fig. 5 The variation of internal pressure with molefraction of 1-alkanols (X₃) in tetrahydrofuran and 1-chlorobutane mixture at 303, 308 and 313K

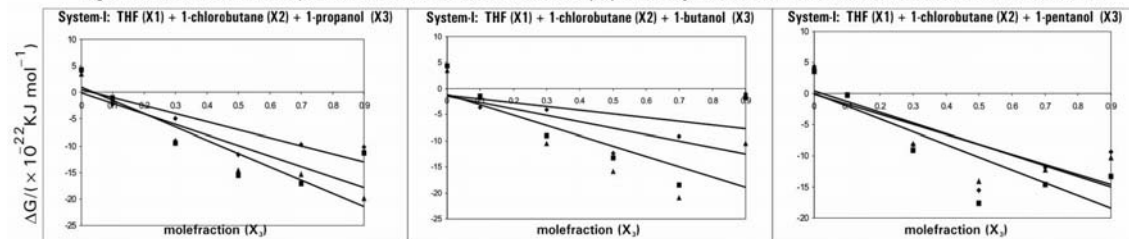


Fig. 6 The variation of Gibbs free energy with molefraction of 1-alkanols (X₃) in tetrahydrofuran and 1-chlorobutane mixture at 303, 308 and 313K