

THERMO-ACOUSTICAL AND EXCESS THERMODYNAMIC STUDIES OF TERNARY LIQUID MIXTURES OF SUBSTITUTED BENZENES IN AQUEOUS MIXED SOLVENT SYSTEMS AT 303.15, 308.15 AND 313.15 K

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Received: April 15, 2011; Accepted: May 06, 2011

Abstract: The observed experimental parameters such as density (ρ), viscosity (η) and ultrasonic velocity (U) of ternary liquid mixtures of substituted benzenes such as chlorobenzene, bromobenzene, nitrobenzene, ethylbenzene and toluene added in the fixed binary solvent mixtures of (ratio 4:2) acetophenone and 1-heptanol at 303.15, 308.15 and 313.15K were determined. The observed experimental data have been utilized to evaluate some of the excess thermo acoustical parameters such as adiabatic compressibility (β^E), intermolecular free length (L^E), free volume (V_f^E), internal pressure (π^E), Gibb's energy (ΔG^E), viscosity (η^E) molar volume (V_m^E) and Grunberg's Nissan interaction parameter (d). It is eventually observed that weak molecular association between acetophenone and 1-heptanol is through hydrogen bonding and substituted benzenes through weak dipole-dipole interactions. The existence of formation of donor – acceptor complexes in the present study has been identified and promptly discussed. Further, the impact due to the elevation of temperature on these liquid systems has also been interpreted.

Keywords: adiabatic compressibility, dipole-dipole interaction, hydrogen bonding, internal pressure, intermolecular free length

1. INTRODUCTION

In recent years, the measurement of ultrasonic velocity has been adequately employed in understanding the nature of molecular interactions in pure liquids and liquid mixtures. The ultrasonic velocity measurements are highly sensitive to molecular interactions and can be used to provide qualitative information about the physical nature and strength of molecular interaction in the liquid mixtures [1-3]. Ultrasonic velocity of a liquid is fundamentally related to the binding forces between the atoms or the molecules and has been adequately employed in understanding the nature of molecular interaction in pure liquids [4-6]. The variation of ultrasonic velocity and related parameters throw much light upon the structural changes associated with the liquid mixtures having weakly interacting components [7-8] as well as strongly interacting components.

The study of molecular association in organic ternary mixtures having alcohol as one of the components is of particular interest, since alcohols are strongly self-associated liquid having a three dimensional network of hydrogen bond and can be associated with any other group having some degree of polar attractions. Accurate knowledge of thermodynamic mixing properties such as adiabatic compressibility, intermolecular free length, free volume, internal pressure and molar volume and their excess values for mixtures of protic, non-protic and associated liquids has a great importance in theoretical

and applied areas of research. The deviations from ideality and specific or non-specific interactions have been revealed. Alcohols [1-3] and aromatic ketones such as acetophenone [9-10] exist as associated structures in liquid state. Thus, upon mixing of ketones with alcohols provide interesting properties due to specific interactions arising from charge-transfer, dipole-dipole, donor-acceptor and hydrogen bond formation forces may be observed.

The thermodynamic properties of systems containing acetophenone are helpful to better understanding the molecular interaction and to design and simulate the different process of separation [11]. Acetophenone is typical aromatic ketone has been used in perfumery and fragrances. 1-heptanol is one of the primary alkanols with a seven carbon chain and it is a clear colourless liquid, slightly soluble in water, but miscible with ether and ethanol. It is commonly used in cardiac electrophysiology experiments. Nitrobenzene have higher dipole moment and dielectric constant values than those of chlorobenzene and bromobenzene. Nitrobenzene is supposed to be a relatively complex molecule and its non-ideality in all probability may be due to the polarity arisen out of nitro-group is concerned, it rotates freely along the C-N axis where it likely to give more flexibility to the interactions arising due to the two highly polar N→O bonds. Chlorine atom in chlorobenzene is an electron-withdrawing atom, tends to

attract to π -electrons of the benzene ring, thereby, decreases the electron density of ring. As a result, the benzene ring in chlorobenzene becomes relatively poor electron-donor towards the electron-seeking proton of any groups. Chlorobenzene has low dielectric constant $\epsilon' = 5.649$ and dipole moment $\mu = 1.69\text{D}$. Its boiling point and melting points are 132°C and -45°C respectively. Chlorobenzene is neither acidic nor basic colourless liquid with a pleasant smell. Chlorobenzene is insoluble in water and soluble in alcohol, benzene and ether. Chlorobenzene is more reactive because the chlorine atom is bonded with SP^3 hybridized carbon atom and consequently can be removed easily. So, the rate of reaction with chlorobenzene is faster. Bromobenzene is less reactive because of its double bond character between carbon and bromine atom. The slow reactions in case of bromobenzene may also be attributed to its being heavier. Thus, the molecular interaction is likely to be more affected resulting in a greater degree of variation with respect to the ultrasonic related parameters.

Ethyl benzene, which is one aromatic hydrocarbon, is important in the petrochemical industry as an intermediate in the production of styrene, which in turn is used for making polystyrene, a common plastic material. Toluene, which is also one of the aromatic hydro carbons – also known as mono-substituted benzene, i.e. one in which a single hydrogen atom from the benzene molecule has been replaced by a univalent group. It is widely used as a solvent and also used as an inhalant drug for its intoxicating properties. Toluene is an important organic solvent, but is also capable of dissolving a number of notable inorganic chemicals such as sulphur.

Owing to these physical characteristics of substituted benzenes, ketones and primary alcohols which motivated the authors to carry out the present study of molecular interactions of ternary liquids mixtures of substituted benzenes with binary solvent mixtures of acetophenone and 1-heptanol (in the ratio of 4:2) at different temperatures. Though, a number of workers have studied the behaviours of substituted benzenes with alcohols [12-13], almost no systematic study was carried out the combined effect of ketones with alcohols as binary solvents in which substituted benzenes were added as solutes on varying temperatures.

The present ternary liquid systems taken up by the authors at different temperatures, say at 303.15, 308.15 and 313.15 K are

System I	acetophenone	+	1-heptanol	+	chlorobenzene
System II	acetophenone	+	1-heptanol	+	bromobenzene
System III	acetophenone	+	1-heptanol	+	nitrobenzene
System IV	acetophenone	+	1-heptanol	+	ethylbenzene, and
System V	acetophenone	+	1-heptanol	+	toluene

2. MATERIALS AND METHODS

In the present work, we have used chemicals, which are analytical reagent (AR) and spectroscopic reagent (SR) grades of minimum assay of 99.9% obtained from E-Merk, Germany and Sd fine chemicals, India. The purities of the above chemicals were checked by the density measurements at 303.15, 308.15 and 313.15K and were compared with available literature values [14-25]. The ternary liquid mixtures of different known composition were prepared by mole fraction basis. The density, viscosity and ultrasonic velocity were measured as a function of the ternary liquid mixture of substituted benzenes which were added to the binary mixtures of acetophenone and 1-heptanol at 303.15, 308.15 and 313.15K. For this purpose, binaries with fixed ratio $X_1/X_2 = 4:2$ respectively were prepared by volume. The density was determined using a specific gravity bottle by relative measurement method. The weight of the sample was measured using a electronic digital balance with an accuracy of ± 0.1 mg (Model: Shimadzu, Japan Make, AX-200). An Ostwald's viscometer (10ml) was used for the viscosity measurement. Efflux time was determined by using a digital chronometer to within ± 0.001 s. An Ultrasonic Interferometer having a frequency of 2 MHz (Mittal Enterprises, New Delhi. Model: F-81) has been used for velocity measurement. The overall accuracy in the velocity measurement is $\pm 2\text{ms}^{-1}$. An electronically digital operated constant temperature water bath (RAAGA Industries, Chennai) 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 $\pm 0.1\text{K}$.

3. THEORY AND CALCULATION

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

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

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

where K is a temperature dependent constant. Its values are 631×10^{-6} , 636×10^{-6} and 642×10^{-6} respectively at 303.15, 308.15 and 313.15K.

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

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.

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

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.

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

Where K is the Boltzmann's constant ($1.23 \times 10^{-23} \text{ JK}^{-1}$), T the absolute temperature, 'h' the Planck's constant and

τ is the relaxation time ($\tau = \frac{4}{3} \eta \beta$).

Excess values of the above parameters can be determined using

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

where $A_{id} = \sum A_i X_i$, A_i is any acoustical

parameters and X_i the molefraction of the liquid component. Grunberg[26] and Nissan formulated the following relation between the viscosity of a binary liquid mixture and pure components.

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

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

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

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

4. RESULTS AND DISCUSSION

The Tables 1 and 2 provide the experimentally determined values of density (ρ), viscosity (η) and ultrasonic velocity (U) of pure liquids and for the ternary systems at 303.15, 308.15, and 313.15 K. The excess values of adiabatic compressibility (β^E), free length (L^E), free volume (V^E), and internal pressure (π^E) are tabulated in Table 3. The excess viscosity (η^E), excess Gibbs Energy (ΔG^E), excess molar volume (V_m^E) and Grunberg and Nissan's interaction parameter (d) are listed in Table 4.

It is seen that in all the five ternary liquid systems, the density (ρ) and the ultrasonic velocity (U) increases with increasing concentrations of substituted benzenes, such as chlorobenzene, bromobenzene, nitrobenzene, ethyl benzene and toluene, whereas the viscosity (η) shows the decreasing trend. These are tabulated in Table 2. The variation of ultrasonic velocity in a mixture depends

upon the increase (or) decrease of intermolecular free length after mixing the components. On the basis of a model, for sound propagation proposed by Eyring and Kincaid [27], ultrasonic velocity should decrease, if the intermolecular free length increase and vice-versa. This is in fact observed in the present investigation for all the five liquid systems.

The thermodynamic excess properties are found to be more sensitive towards intermolecular interaction between the component molecules of liquid mixtures. The sign and magnitude of deviation of excess properties depend on strength of interaction between unlike molecules. That is in order to understand the nature of molecular interactions between the components of the liquid mixtures, it is of interest to discuss the same in term of excess parameter rather than actual values. Non-ideal liquid mixtures show considerable deviation from linearity in their physical behaviour with respect to concentration and these have been interpreted as arising from the presence of strong or weak interactions [28].

The perusal of Table 3 exhibits the values of excess adiabatic compressibility (β^E) for all the five liquid systems. The negative values of β^E is associated with a structure-forming tendency, while positive values are an indication of structure-breaking tendency due to hetero-molecular interaction between the component molecules of the mixtures. The positive values of excess adiabatic compressibility which indicates the loosely packed molecules in the mixtures resulting due to shape and size. From Table 3, it is evident that the excess adiabatic compressibility are all almost negative in all the five ternary liquid system, except in systems I and II, where a slight positive deviations are seen in lower mole fraction range [See Fig.1(a,b)] and becomes negative on going to the higher mole fraction range. The negative values of the β^E decrease with increasing concentrations of substituted benzenes in all the five liquid systems as well as increase of temperature. This may be ascribed as that in acetophenone, the carbonyl group is highly polar and hence has a high percentage of ionic character [29] and there exists is a negative charge on the carbonyl oxygen atom of acetophenone. Hence, the decreasing trend of negative values will make one would expect a weak interactions with substituted benzenes such as chlorobenzene, bromobenzene, nitrobenzene, ethyl benzene and toluene due to weak dipole-dipole interactive forces. Also, in the case of substituted benzenes, the bromine, chlorine and nitrogen all having a tendency to act as an electron acceptors. The decrease in adiabatic compressibility may further be attributed to the internal interactions between π -electrons of C = O bond and π -electrons of the benzene ring [30]. A plausible explanation may be that the oxygen atom of carbonyl group of acetophenone being strongly electronegative would act as a good electron acceptor towards the π -electrons of the aromatic ring, forming donor-acceptor complexes [31]. It is observed that decreasing negative deviations in β^E increasing with molar concentrations seems to be an indication of weak

molecular interaction between unlike molecules increases. Such decreasing negative values of β^E may also be due to the dominance of dispersive interaction forces resulting from the breaking up of the hydrogen bonds of substituted benzenes makes the compressibility to decrease. The magnitude of the β^E values are of the order: Systems III > I > II > IV > V.

The perusal of Table 3 shows the values of excess free length (L_f^E) for all the five ternary liquid systems. It was noticed that the L_f^E values are negative in all the five liquid systems and found to be decreased with increasing molar concentration of substituted benzenes and elevation of temperature, except in systems I, II, and III, where a slight positive deviations are seen in lower mole fraction range, and however such positive deviation are found to be decreasing and becomes negative on going to the higher mole fraction range [See Fig.2(a,b)]. According to Kannappan *et al.* [32], the negative values of L_f^E indicate 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[33] indicated that the positive excess values of free length should be attributed to the dispersive forces and negative excess values of should be due to charge transfer and hydrogen bond formation. In the present investigation, one can notice that the decreasing trend of negative excess values of intermolecular free length (L_f^E) with the increasing concentration of substituted benzenes as well as increase of temperature may be interpreted as aromatic ketones such as acetophenone which is being a highly polar molecule ($\mu = 2.96$) and it may enhance the polarities of the substituted benzenes. The π - electron density in derivatives of benzene ring depends upon that is attached to it. The heteromolecular interaction between component molecules necessarily depends upon the net electron density in the ring. Further, these interactions seemed to depend on relative orientation of the two groups in the ring. As the separation between the two groups increases, the intermolecular interaction is expected to decrease. Hence, a decrease in excess free length (L_f) is noticed. The magnitude of the L_f^E values are in the order: Systems III > I > II > IV > V.

The perusal of Table 3 provides a qualitative picture of excess free volume (V_f^E) values for all the five ternary liquid systems. The present parameter indicates the extent of deviation from ideal with the mole fraction of the mixtures. The excess values for all the five ternary liquid systems found to be negative. These values found to be decreased on increasing the molar concentration of substituted benzenes as well as elevation of temperature. The results can be explained in terms of molecular interaction, structural effect and interstitial accommodation along with the changes in free volume. The sign of the V_f^E depends on the relative strength between the contractive forces and expansive forces. The factors responsible for volume contraction are (i) specific interactions between the component molecules and (ii) weak physical forces, such as dipole-dipole or

dipole-induced dipole interactions or Vanderwaal's forces. The factors that cause expansion in volume are dispersive forces, steric hindrance of component molecules, unfavourable geometric fitting and electrostatic repulsion. The negative values of excess free volume in all the systems assert that the combined effect of the factors responsible for volume contraction and vice-versa [34]. Adgaonkar *et al.* [35] showed positive values of V_f^E indicating the existence of weak molecular interactions in the liquid mixtures and the negative values of excess free volume suggesting the specific interactions among unlike molecules.

In our binary mixture solvent, 1-heptanol, a primary alcohol also a polar one which is in association with acetophenone forms hydrogen bonding. Grace selvarani *et al.* [36] had made it clear that the molecular association is due to the hydroxyl group of 1-alkanol and acetophenone. According to them, aromatic ketones such as acetophenone are more associated with alcohols than the aliphatic one. Since alkanols are liquids which are associated through hydrogen bonding and in the pure state, they exhibit an equilibrium between multimer and monomer species. Further, when the substituted benzenes such as Nitrobenzene is mixed with 1-heptanol, the NO_2 group can interact with OH group[37,38]. The aromatic derivatives set up on interaction between the π -electrons cloud and the hydroxyl group. Though, this interaction is of minor intensity compared with hydrogen bonding, but they may lead to formation of intermolecular complexes[39].

In the study of liquid mixtures, the variation of internal pressure may give some suitable information regarding the nature and strength of the forces existing between the molecules. In fact, the internal pressure is a broader concept and it is a measure of the totality of forces of the dispersion, ionic and dipolar interaction that contribute to be overall cohesion of the liquid systems. The present study shows that the excess internal pressure values (π_f^E) are negative in all the five liquid systems. Such negative values of π_f^E are increasing on increasing the molar concentration of substituted benzenes indicate that only dipolar forces are operating between the unlike molecules. Further, the observed increasing behavior of excess internal pressure values reveals that the weakening of cohesive forces resulting perhaps due to making up of the structure of the solvent.

One should observe from the Table 4, the variation of excess Gibbs energy ΔG^E for the five ternary liquid systems. The values of ΔG^E are all negative in all the systems concerned and increase with molar concentration of substituted benzenes [See Fig.3(a,b)]. According to Read *et al.* [40], the positive values of excess Gibbs energy values may be attributed to specific interactions like hydrogen bonding and charge transfer, while negative ΔG^E values may be ascribed to the dominance of dispersion forces [41]. In the present investigation, our close observation suggest that the increasing negative values of ΔG^E shows that the strength of interaction gets weakened with increasing of substituted benzenes. Further, the negative values of

ΔG^E indicate the easier flow of ternary mixture compared with the behaviour of pure components [42].

According to Fort *et al.* [33] the variation of excess viscosity (η^E) gives the strength of molecular interaction between the molecules. For systems, where dispersion, induction, and dipolar forces which are operated by 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. In the present study, from Table 4- the excess viscosity are negative in all the five ternary liquid systems and increase with the further addition of substituted benzenes as well as elevation of temperature. Hence, our present close observation of excess viscosity reveals that presence of weak dipolar forces are existing in the liquid mixtures. The increasing negative values of η^E further suggests that the dominance of dispersive interaction resulting from the breaking up of the hydrogen bonds of substituted benzenes make the mixture to flow more easily. Further, the strength of interaction get weakened in view of rise in temperature due to the presence of thermal dispersion forces in the liquid mixtures. The strength of the molecular interactions among the liquid systems by making use of the magnitude of the η^E values are of the order: Systems III > I > II > IV > V.

The perusal of Table 4 exhibits the excess molar volume (V_m^E) are all negative over the entire range of concentration range for the mixtures of acetophenone and 1-heptanol and substituted benzenes. According to V_m^E data for ternary mixtures of acetophenone and 1-heptanol with substituted benzenes depend upon the balance between two opposing contributions [43, 44].

- (a) A positive term from the rupture of alkanol-alkanol hydrogen bonds and physical dipole-dipole interactions between alkanol monomers and multimers, and
- (b) A negative term from the formation of the OH π -electron hydrogen bonded complexes, changes of free volume and interstitial accommodation.

The experimental data in the present investigation suggest that factor (b) which is responsible for the negative excess volume is dominant over the entire composition range in the mixtures of acetophenone and 1-heptanol with substituted benzenes. On the other hand, both the factors, (a) and (b) compete with each other to varying degrees in the mixtures which contain benzene and toluene as non- common components. According to Awwad *et al.* [45] measured excess molar volume obtained in the present study can be qualitatively explained as arising to due to differences in the sizes of the component molecules and in the interactions between them. Differences are mainly due to negative contributions arising from changes of free volume in the real mixtures, comprising alkanol monomers and multimers and acetophenone, benzene and substituted benzene molecules. The experimental data in the present study suggest that the factors which account for the existence of dipole-dipole interactions present in the

liquid mixtures. The decreasing trend of negative values of V_m^E for the further addition of substituted benzenes in all the five liquid systems clearly predicting this.

The interaction parameter d in the Grunberg and Nissam equation is a measure of strength of interaction between the mixing components. d -values were said to indicate various type of interactions [46]. Large and positive d -values indicate strong specific interaction, small positive values indicate weak specific interaction and large negative values indicate no specific interaction. It is evident from Table 4. The d -values are negative (except in system – III) and decrease with increasing of molefraction X_3 , and the same varies non-linearly with the rise of temperature. Further, the decreasing behavior of d -values exhibits weaker interaction between the unlike molecules, and also their non-linear behaviour towards the rise of temperature which may suggest the weakening of intermolecular interaction due to thermal dispersion forces.

5. CONCLUSION

Ultrasonic method is a powerful probe for characterising the physico-chemical properties and existence of molecular interactions in the liquid mixtures. The evaluated ultrasonic excess values of ternary mixtures of substituted benzenes in the binary solvent mixture of acetophenone and 1-heptanol (in the fixed ratio of 4:2) at 303.15, 308.15 and 313.15K which may eventually concluded that there exist a molecular interaction between the acetophenone and 1-heptanol is through hydrogen bonding and with substituted benzenes through dipole-dipole interactions. The strength of the molecular interactions gets weakened on further addition of substituted benzenes as well as rise in temperature. Weak dipolar and cohesive forces are existing in the present systems of liquid mixtures. Our present study also predicts the formation of donor-acceptor complexes in the component mixtures. Further, the elevation of temperature in the liquid mixtures lead to the weakening of molecular interactions due to weak intermolecular forces and thermal dispersion forces. From the magnitude of some of our excess parameters [β^E , L^E and η^E] which confirms the present molecular interaction is of the order: Systems III > I > II > IV > V.

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Fig. 1 (a) - Excess adiabatic compressibility Vs mole fraction

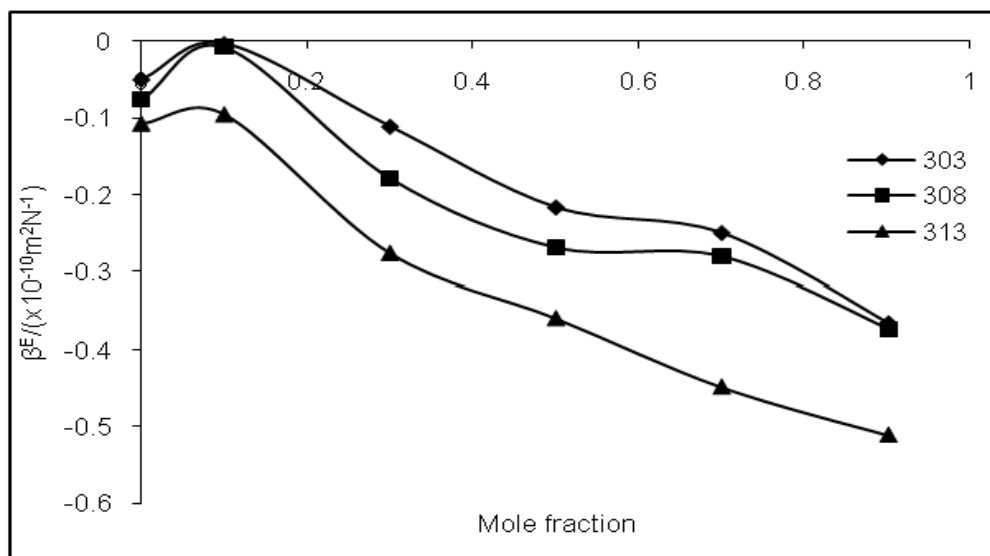
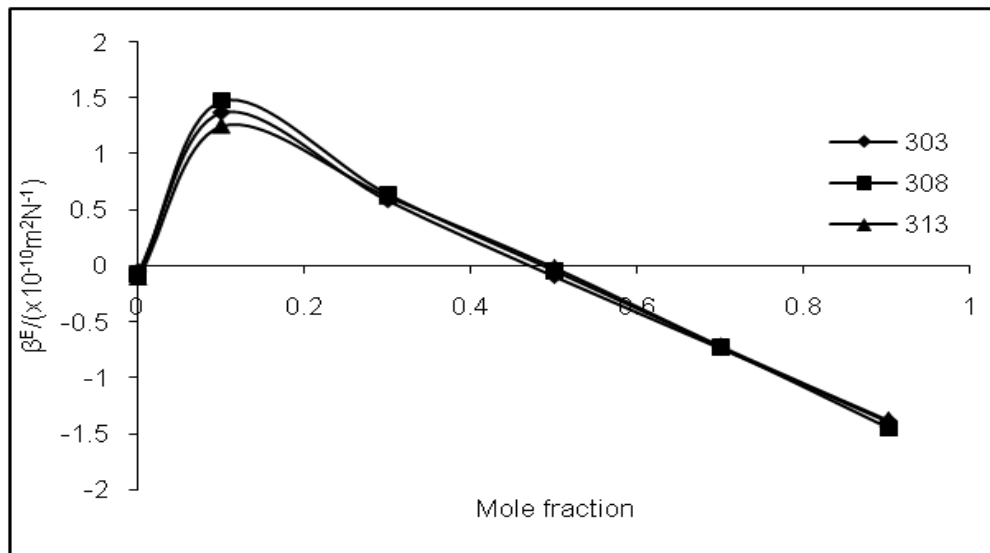
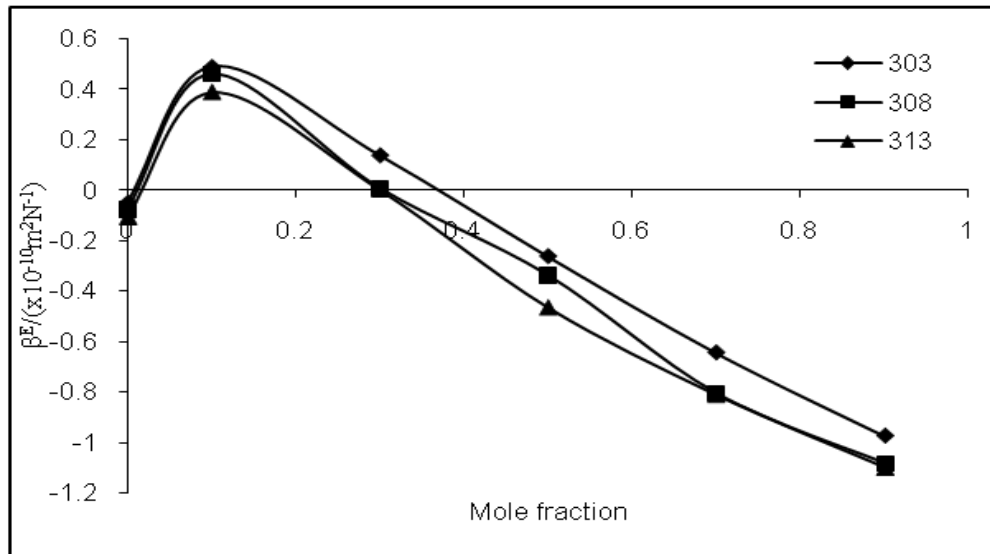


Fig. 1 (b) - Excess adiabatic compressibility Vs mole fraction

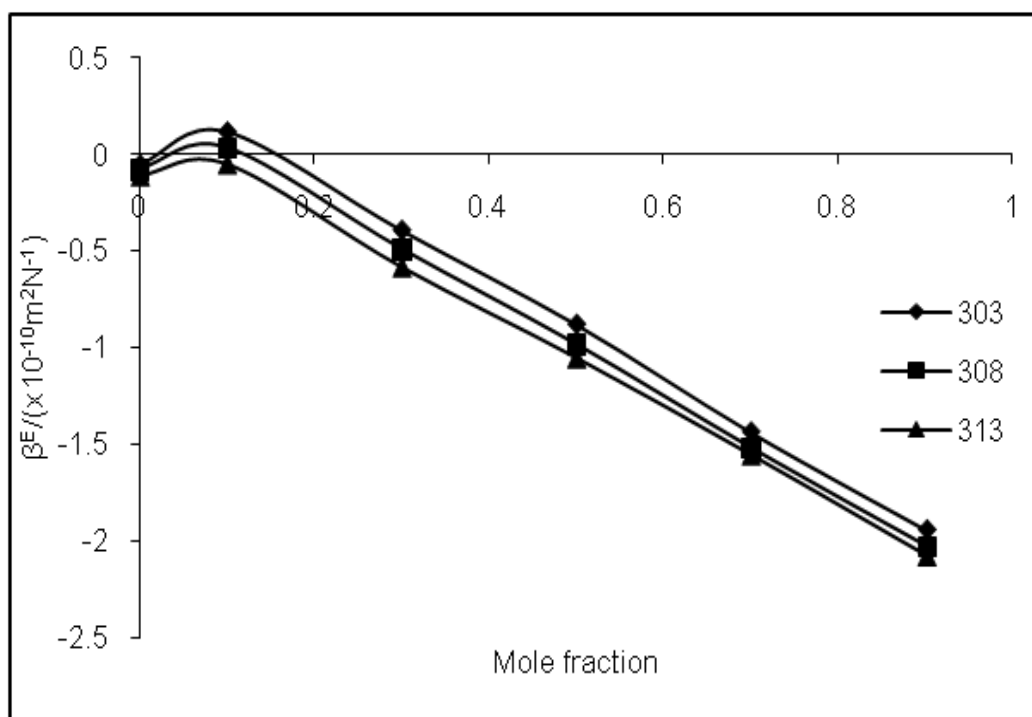
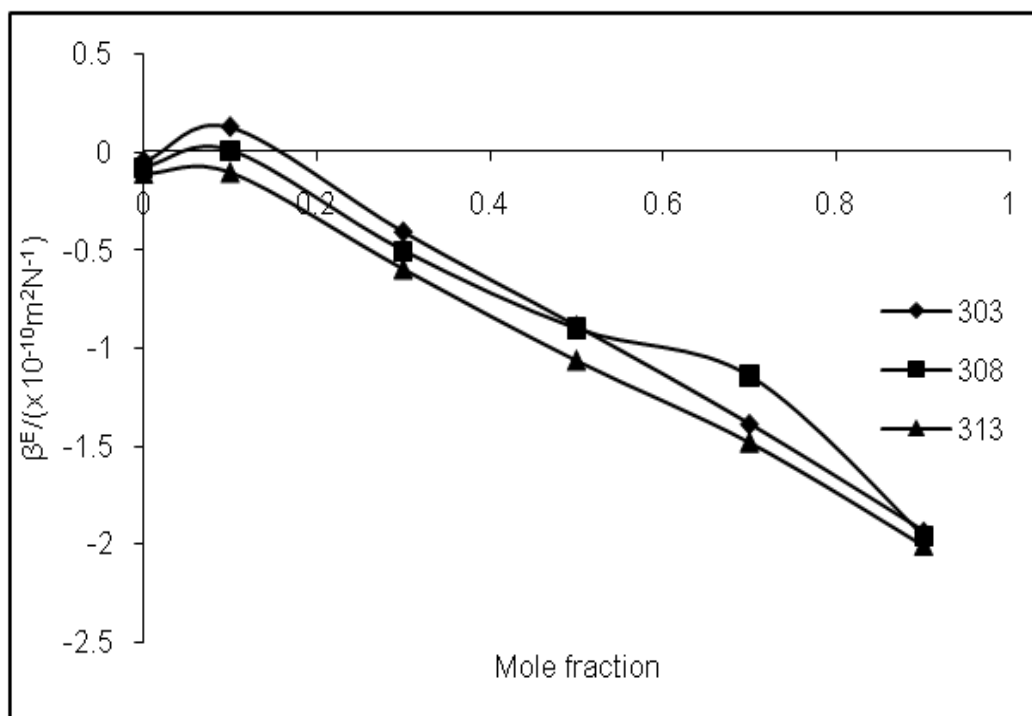


Fig. 2(a) - Excess free length Vs mole fraction

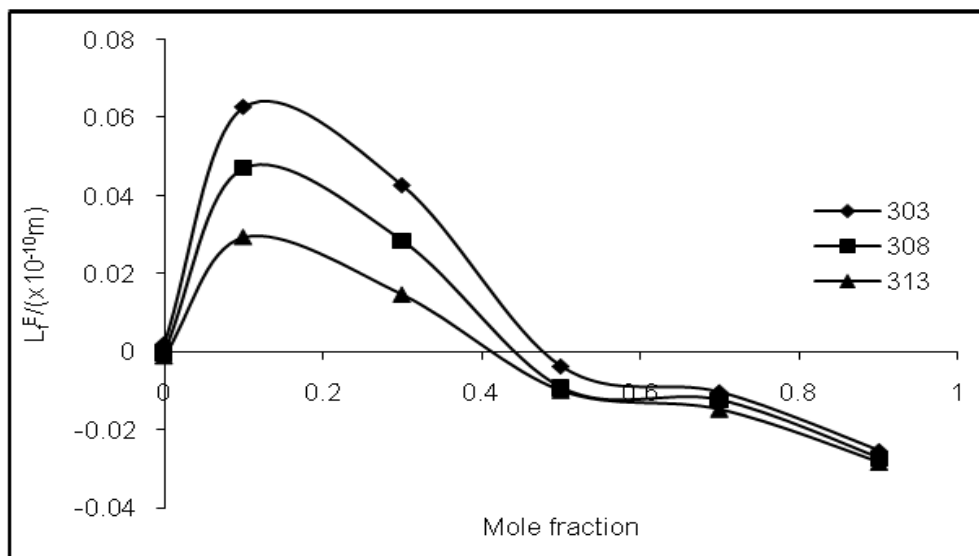
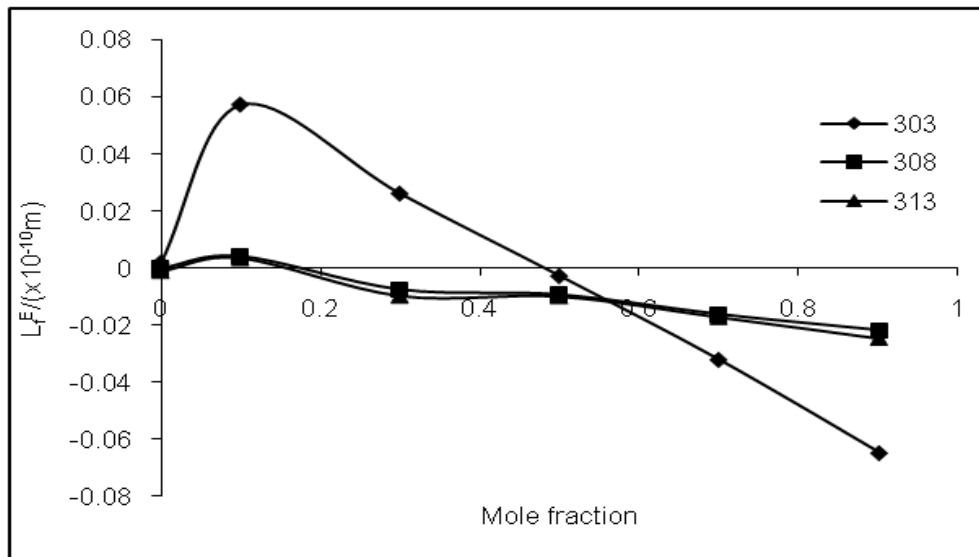
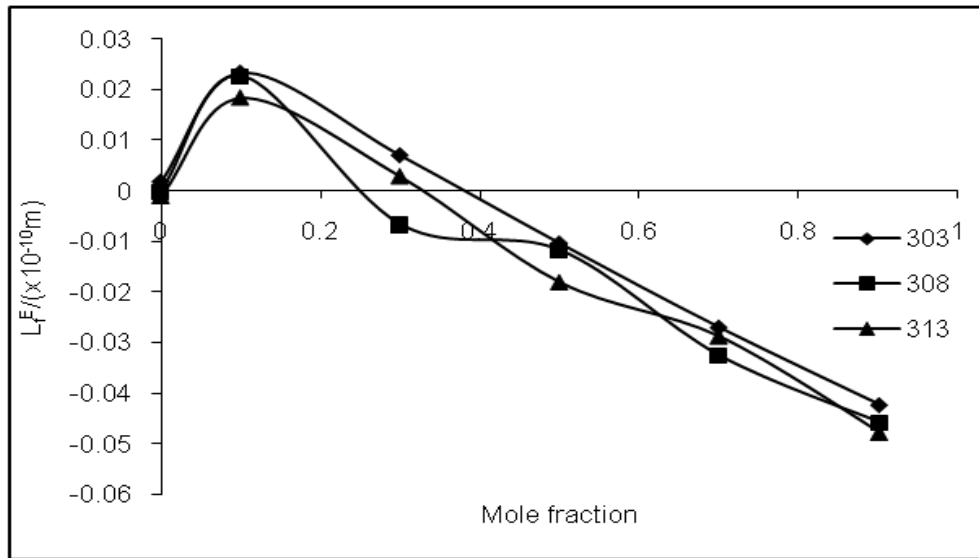


Fig. 2(b) - Excess free length Vs mole fraction

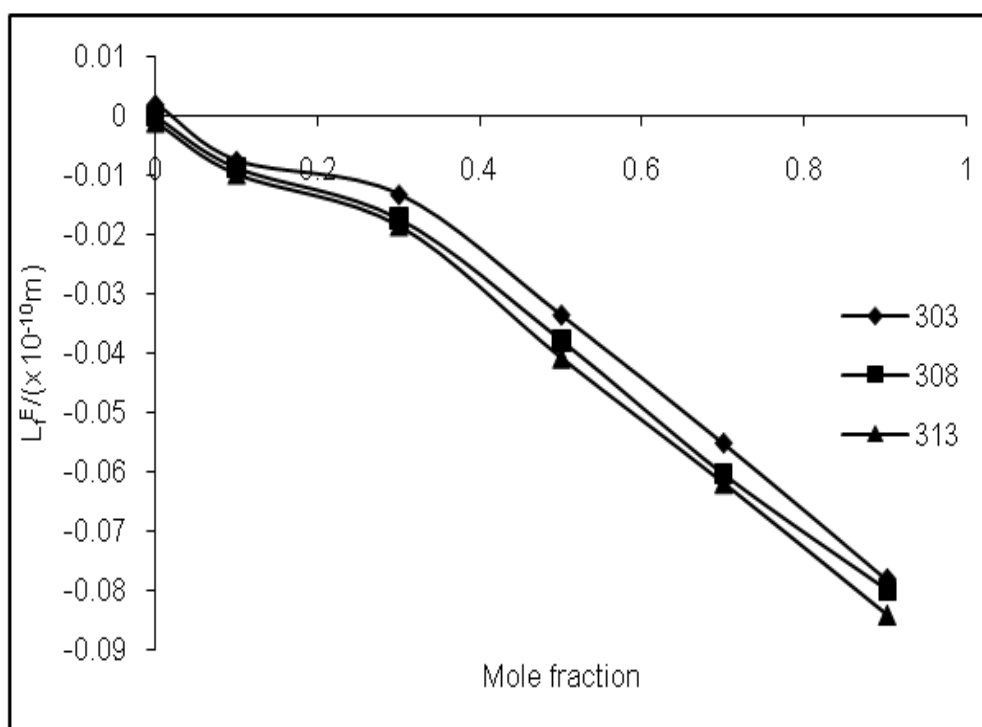
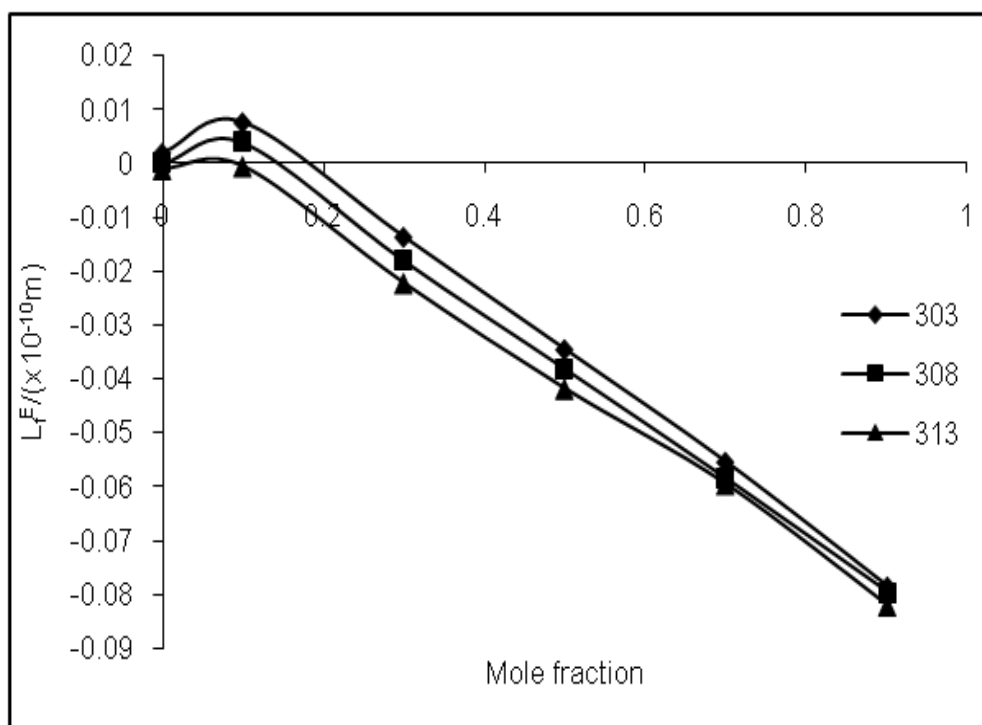


Fig. 3(a) - Excess Gibb's free energy Vs mole fraction

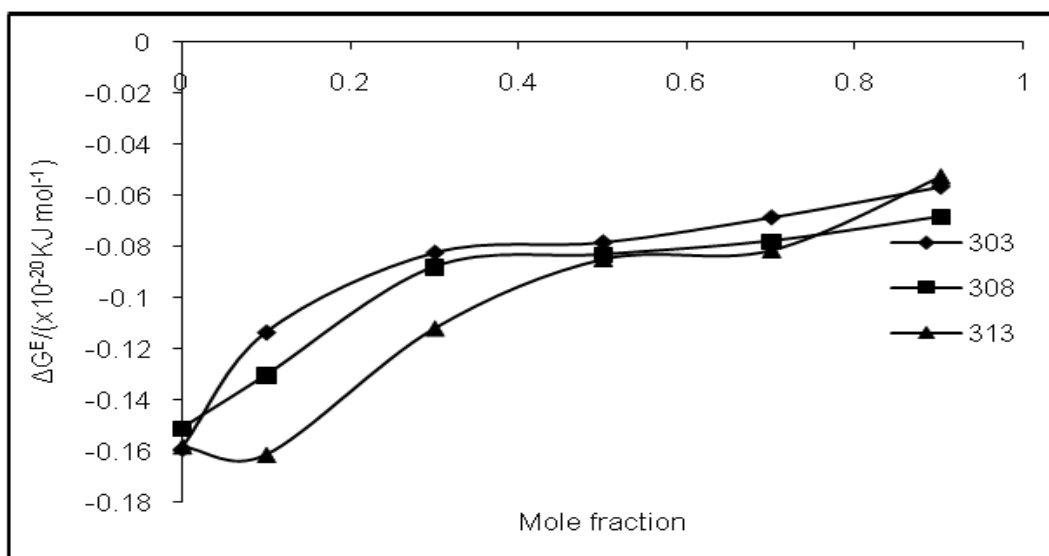
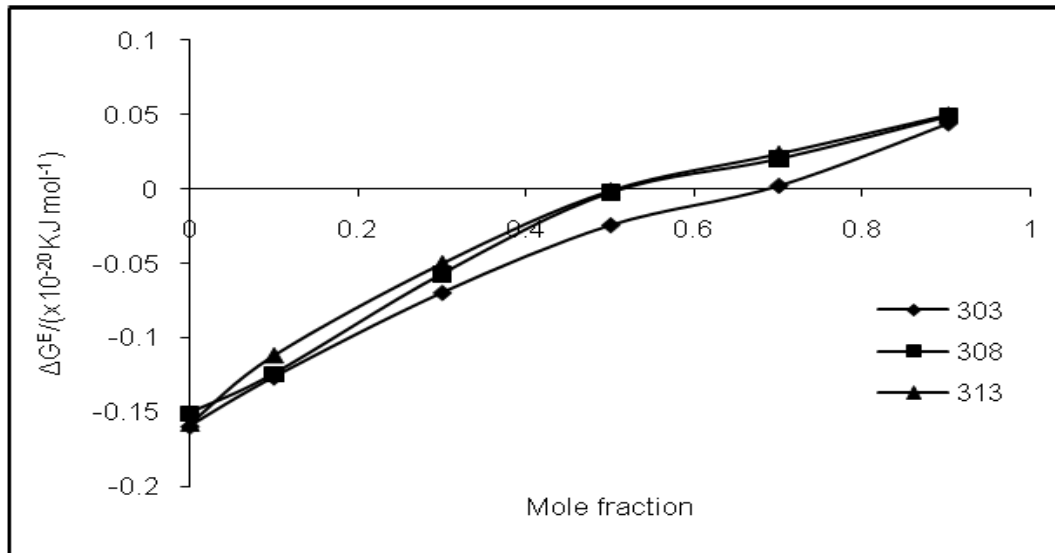
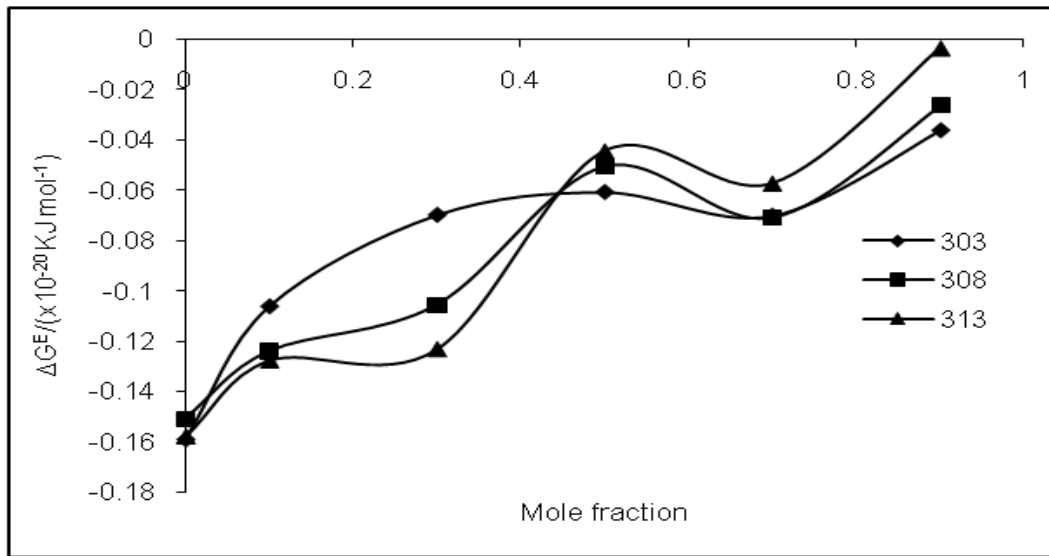


Fig. 3(b) - Excess Gibb's free energy Vs mole fraction

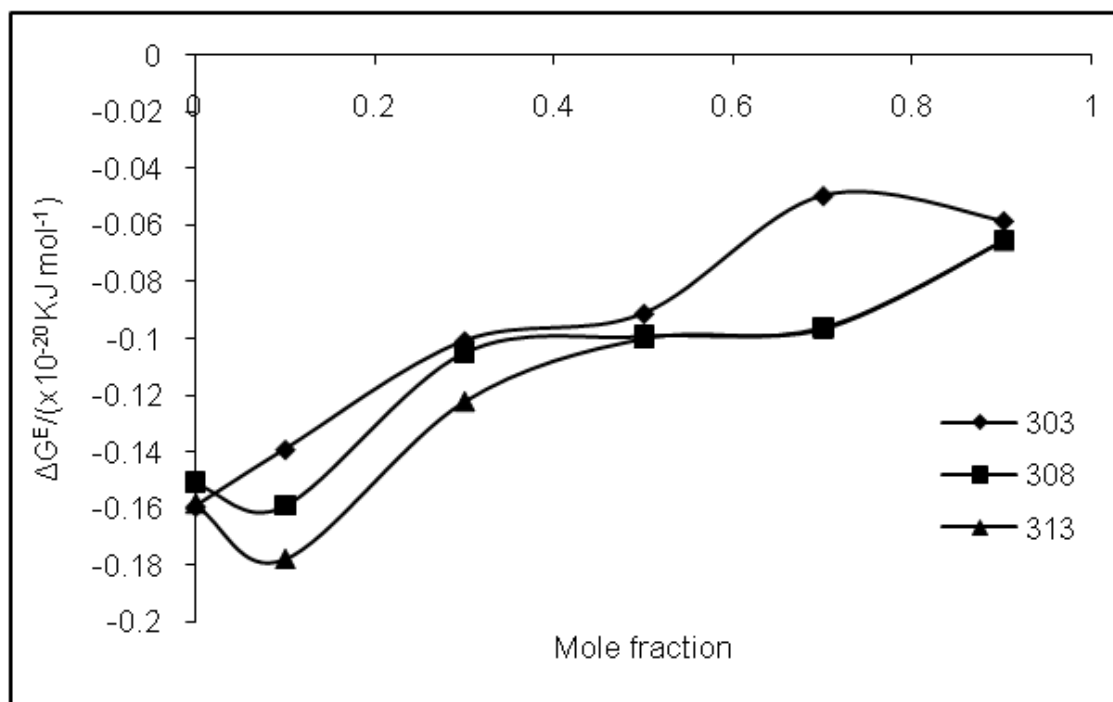
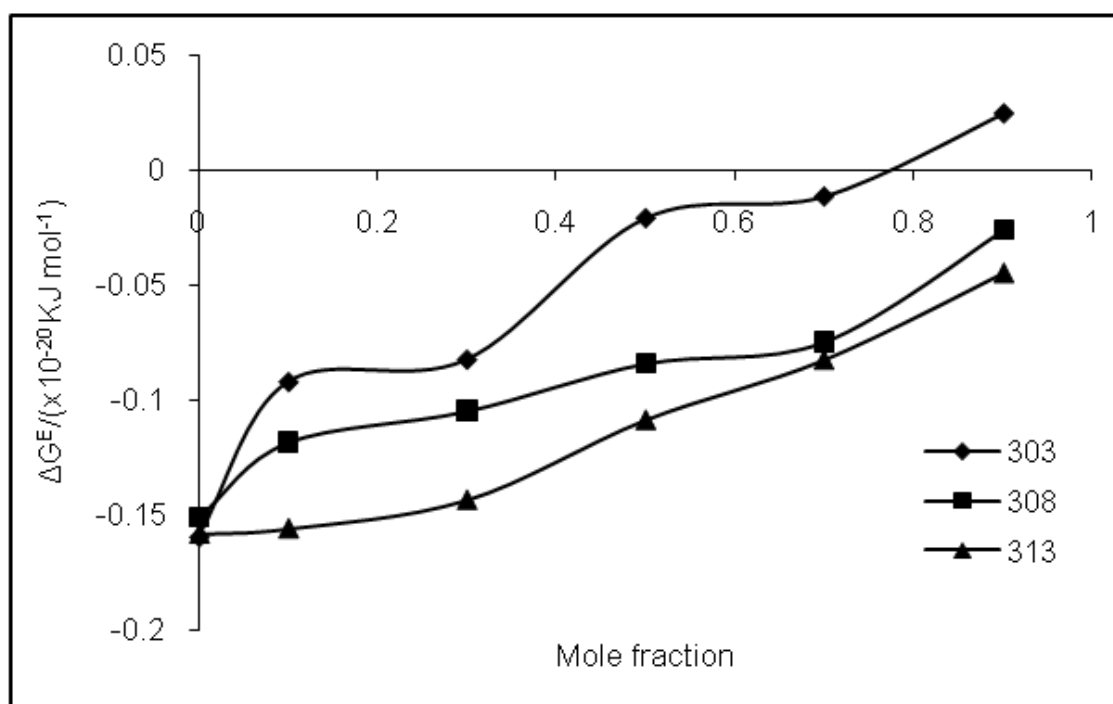


Table 1 - Comparison of experimental densities (ρ), viscosities (η) and velocities (U) of pure liquids

Organic Liquids	Temperature (K)	Density ρ /(kg/m ⁻³)			Viscosity η /($\times 10^{-3}$ Nsm ⁻²)			Velocity U /(m/s)		
		P.W*	Ref1	Ref2	P.W*	Ref1	Ref2	P.W*	Ref1	Ref2
acetophenone	303.15	1019.7	1019.4 ^[14]	1019.9 ^[21]	1.5323	1.5400 ^[21]		1453.6	1454.4 ^[19]	
	308.15	1015.2	1014.9 ^[15]	1014.8 ^[18]	1.4118	1.4110 ^[15]	1.410 ^[21]	1444.4	1445.0 ^[20]	
	313.15	1010.5	1011.2 ^[21]		1.3119	1.3100 ^[21]		1423.7	–	
1-heptanol	303.15	815.7	815.5 ^[17]		4.9781	5.0100 ^[17]		1324.9	1327.2 ^[17]	
	308.15	812.0	811.9 ^[15]		4.4684	–		1290.8	1292.0 ^[16]	
	313.15	808.7	808.4 ^[17]		3.6519	3.6542 ^[17]		1282.0	1282.4 ^[17]	
chlorobenzene	303.15	1095.1	1095.4 ^[14]	1095.4 ^[22]	0.7292	0.7258 ^[24]		1239.6	...	
	308.15	1090.0	1089.7 ^[22]	1089.3 ^[24]	0.6779	0.6770 ^[24]		1224.7	1224.0 ^[22]	
	313.15	1084.0	1084.6 ^[22]	1083.9 ^[24]	0.6281	0.6240 ^[24]		1213.4	...	
bromobenzene	303.15	1481.9	1481.4 ^[14]	1481.5 ^[24]	1.0138	1.0146 ^[24]		1131.6	...	
	308.15	1474.5	1475.3 ^[22]	1475.8 ^[24]	0.9638	0.9540 ^[24]		1122.4	1122.0 ^[22]	
	313.15	1467.7	1467.3 ^[22]	1467.3 ^[24]	0.8711	0.8780 ^[24]		1110.4	...	
nitrobenzene	303.15	1193.9	1193.4 ^[14]	1193.4 ^[22]	1.6089	1.6060 ^[24]		1432.8	...	
	308.15	1187.6	1188.2 ^[22]		1.5408	1.5430 ^[22]		1422.9	1423.0 ^[22]	
	313.15	1183.0	1183.2 ^[22]		1.4270	...		1409.0	...	
Ethylbenzene	303.15	855.1	856.5 ^[23]		0.6094	0.6006 ^[23]		1280.4	...	
	308.15	853.2	853.8 ^[16]		0.5729	...		1276.2	1276.0 ^[16]	
	313.15	849.5	848.9 ^[23]		0.5378	0.5355 ^[23]		1272.6	...	
toluene	303.15	856.8	857.5 ^[22]	857.6 ^[14]	0.5264	0.5229 ^[25]		1270.8	...	
	308.15	852.9	852.8 ^[16]	852.8 ^[22]	0.4987	0.4980 ^[22]		1263.2	1262.0 ^[16]	1262.0 ^[22]
	313.15	847.2	848.3 ^[22]		0.4513	...		1259.4	...	

P.W*:Presentwork

Table 2 - Density (ρ), viscosity (η) and ultrasonic velocity (U) at 303.15, 308.15 and 313.15K for

Mole Fraction X_3	Density $\rho/(\text{kg/m}^{-3})$			Viscosity $\eta/(\times 10^{-3} \text{ Nsm}^{-2})$			Ultrasonic Velocity $U/(\text{m/s})$		
	303.15K	308.15K	313.15K	303.15K	308.15K	313.15K	303.15K	308.15K	313.15K
System I: Acetophenone (X_1) + 1-heptanol (X_2) + chlorobenzene (X_3) [$X_1/X_2=4:2$]									
0.0000	938.57	935.36	933.43	1.5370	1.4549	1.2916	1409.7	1391.0	1384.8
0.0999	951.01	947.15	945.38	1.5201	1.3805	1.2113	1329.8	1314.5	1307.6
0.3000	976.40	975.14	972.64	1.3705	1.1853	1.0551	1339.5	1324.8	1317.4
0.5000	1009.16	1005.68	1001.90	1.1532	1.1365	1.0184	1353.2	1341.8	1327.0
0.7000	1039.85	1035.55	1033.55	0.9263	0.8702	0.8310	1368.4	1358.8	1338.7
0.9013	1066.65	1064.61	1063.38	0.8253	0.7899	0.7267	1381.2	1372.8	1350.6
System II: Acetophenone (X_1) + 1-heptanol (X_2) + bromobenzene (X_3) [$X_1/X_2=4:2$]									
0.0000	938.57	935.36	933.43	1.5370	1.4549	1.2916	1409.7	1391.0	1384.8
0.0999	978.85	976.75	974.60	1.4973	1.4058	1.2935	1229.3	1214.8	1206.0
0.3000	1076.00	1074.05	1072.66	1.4619	1.4031	1.2891	1249.6	1227.2	1215.2
0.5000	1180.03	1178.38	1176.42	1.4441	1.3800	1.2762	1271.2	1243.2	1229.6
0.6999	1290.28	1285.99	1283.55	1.3815	1.2899	1.1624	1303.2	1283.9	1274.2
0.9012	1381.40	1378.91	1375.83	1.3415	1.2387	1.1492	1364.0	1353.6	1338.0
System III: Acetophenone (X_1) + 1-heptanol (X_2) + nitrobenzene (X_3) [$X_1/X_2=4:2$]									
0.0000	938.57	935.36	933.43	1.5370	1.4549	1.2916	1409.7	1391.0	1384.8
0.0999	954.24	947.09	945.61	2.3143	2.1476	1.9902	1411.9	1401.9	1397.8
0.2999	1018.04	1015.16	1012.24	2.2372	2.1047	1.9658	1415.1	1405.3	1401.5
0.5000	1097.01	1094.73	1091.17	2.1815	2.0008	1.9219	1419.6	1408.0	1404.7
0.6999	1171.39	1166.40	1164.19	2.0972	1.9891	1.8478	1421.4	1411.1	1407.9
0.9012	1276.97	1274.17	1271.91	1.9899	1.9358	1.8129	1424.9	1413.7	1410.0
System IV: Acetophenone (X_1) + 1-heptanol (X_2) + ethylbenzene (X_3) [$X_1/X_2=4:2$]									
0.0000	938.57	935.36	933.43	1.5370	1.4549	1.2916	1409.7	1391.0	1384.8
0.1000	928.90	924.12	921.84	1.4893	1.3494	1.1947	1372.3	1369.2	1366.4
0.2999	949.60	946.54	943.21	1.3281	1.2601	1.1284	1378.5	1376.1	1373.8
0.5000	973.08	970.08	968.14	1.1149	1.0984	0.9612	1381.4	1379.2	1376.5
0.6999	999.81	995.24	993.24	0.9298	0.8793	0.8109	1384.6	1382.4	1379.7
0.9013	1031.45	1028.40	1025.58	0.8153	0.7849	0.7298	1389.4	1384.1	1381.5
System V: Acetophenone (X_1) + 1-heptanol (X_2) + toluene (X_3) [$X_1/X_2=4:2$]									
0.0000	938.59	935.36	933.43	1.5370	1.4549	1.2916	1409.7	1391.0	1384.8
0.0999	925.94	915.36	911.93	1.3748	1.2349	1.0709	1373.6	1370.7	1365.0
0.3000	943.38	940.48	936.74	1.2136	1.1546	0.9925	1378.8	1373.1	1370.2
0.5000	961.86	958.62	954.52	1.0004	0.9823	0.8453	1381.1	1377.9	1373.8
0.7000	987.98	985.28	981.88	0.8253	0.7632	0.6957	1384.6	1382.8	1375.4
0.9012	1016.27	1012.69	1008.93	0.7108	0.6688	0.6106	1387.6	1385.0	1380.9

Table 3 - Excess values of adiabatic compressibility (β^E), free length (L^E) free volume (V^E) and internal pressure (π^E) at 303.15, 308.15 and 313.15K for

Mole Fraction X_3	Adiabatic compressibility $\beta^E (\times 10^{-10} \text{m}^2 \text{N}^{-1})$			Free length $L^E (\times 10^{-10} \text{m})$			Free volume $V^E (\times 10^{-7} \text{m}^3 \text{mol}^{-1})$			Internal pressure $\pi^E (\times 10^6 \text{Nm}^{-2})$		
	303.15K	308.15K	313.15K	303.15K	308.15K	313.15K	303.15K	308.15K	313.15K	303.15K	308.15K	313.15K
System I: Acetophenone (X_1) + 1-heptanol (X_2) + chlorobenzene (X_3) [$X_1/X_2=4:2$]												
0.0000	-0.0500	-0.0765	-0.1075	0.0019	-0.0001	-0.0011	0.03032	0.3673	0.4027	-102.75	-84.47	-39.18
0.0999	0.4881	0.4597	0.3872	0.0234	0.0226	0.0183	0.0134	0.1201	0.0756	-75.43	-72.52	-48.96
0.3000	0.1376	0.0084	0.0016	0.0071	-0.0067	0.0028	0.0986	0.0029	-0.1574	-55.09	-51.16	-41.87
0.5000	-0.2630	-0.3366	-0.4648	-0.0103	-0.0118	-0.0181	-0.1324	-0.1548	-0.2005	-41.66	-37.44	-29.11
0.7000	-0.6451	-0.8039	-0.8111	-0.0269	-0.0326	-0.0288	-0.1613	-0.1988	-0.2861	-33.48	-31.15	-16.57
0.9011	-0.9757	-1.0825	-1.0998	-0.0422	-0.0458	-0.0478	-0.1998	-0.2643	-0.2995	12.98	14.62	17.97
System II: Acetophenone (X_1) + 1-heptanol (X_2) + bromobenzene (X_3) [$X_1/X_2=4:2$]												
0.0000	-0.0500	-0.0765	-0.1075	0.0019	-0.0001	-0.0011	0.3032	0.3673	0.4027	-102.75	-84.47	-39.18
0.0999	1.3688	1.4765	1.2486	0.0574	0.0041	0.0034	-0.0364	-0.0404	-0.0494	-87.25	-71.42	-27.62
0.2999	0.5832	0.6427	0.6181	0.0262	-0.0076	-0.0097	-0.1988	-0.2352	-0.2775	-59.45	-49.67	-16.28
0.5000	-0.0943	-0.0415	-0.0154	-0.0027	-0.0094	-0.0099	-0.2917	-0.3836	-0.5601	-29.26	-26.77	-07.51
0.6999	-0.7372	-0.7251	-0.7181	-0.0321	-0.0163	-0.0171	-0.4481	-0.5140	-0.6208	-06.12	-02.47	06.64
0.9012	-1.3928	-1.4474	-1.3797	-0.0648	-0.0219	-0.0245	-0.5622	-0.5981	-0.6711	10.33	12.34	14.25
System III: Acetophenone (X_1) + 1-heptanol (X_2) + nitrobenzene (X_3) [$X_1/X_2=4:2$]												
0.0000	-0.0500	-0.0765	-0.1075	0.0019	-0.0001	-0.0011	0.3032	0.3673	0.4027	-102.75	-84.47	-39.18
0.0999	-0.0037	-0.0086	-0.0956	0.0628	0.0471	0.0294	-0.3230	-0.3444	-0.3671	-107.11	-81.62	-74.80
0.2999	-0.1111	-0.1793	-0.2759	0.0427	0.0285	0.0147	-0.3752	-0.3878	-0.3919	-80.69	-56.51	-53.35
0.5000	-0.2160	-0.2690	-0.3591	-0.0038	-0.0091	-0.0099	-0.3913	-0.3998	-0.4011	-67.44	-36.55	-35.00
0.6999	-0.2492	-0.2804	-0.4483	-0.0104	-0.0123	-0.0147	-0.4055	-0.4107	-0.4171	-46.98	-20.04	-10.81
0.9012	-0.3645	-0.3731	-0.5109	-0.0255	-0.0272	-0.0283	-0.4147	-0.4198	-0.4207	-27.05	-14.34	-8.03
System IV: Acetophenone (X_1) + 1-heptanol (X_2) + ethylbenzene (X_3) [$X_1/X_2=4:2$]												
0.0000	-0.0500	-0.0765	-0.1075	0.0019	-0.0001	-0.0011	0.3032	0.3673	0.4027	-102.75	-84.47	-39.18
0.0999	0.1238	0.0065	-0.1027	0.0076	0.0038	-0.0006	-0.0142	-0.1058	-0.2140	-82.62	-66.74	-27.35
0.2999	-0.4121	-0.5045	-0.5964	-0.0137	-0.0182	-0.0223	-0.3408	-0.4104	-0.4291	-44.30	-33.29	-11.97
0.5000	-0.8918	-0.9012	-1.0638	-0.0346	-0.0383	-0.0419	-0.5106	-0.5323	-0.5455	-11.57	-02.56	-1.08
0.6999	-1.3926	-1.1434	-1.4818	-0.0555	-0.0586	-0.0596	-0.5607	-0.6369	-0.6397	09.45	13.67	26.92
0.9013	-1.9412	-1.9628	-2.0106	-0.0787	-0.0798	-0.0822	-0.6836	-0.8582	-0.8899	48.22	52.63	54.73
System V: Acetophenone (X_1) + 1-heptanol (X_2) + toluene(X_3) [$X_1/X_2=4:2$]												
0.0000	-0.0500	-0.0765	-0.1075	0.0019	-0.0001	-0.0011	0.3032	0.3673	0.4027	-102.75	-84.47	-39.18
0.0999	0.1210	0.0330	-0.0454	-0.0076	-0.0087	-0.0098	-0.1726	-0.2609	-0.2913	-90.44	-79.39	-41.22
0.2999	-0.3900	-0.4895	-0.5798	-0.0133	-0.0173	-0.0185	-0.2176	-0.2715	-0.2960	-54.09	-42.10	-29.18
0.5000	-0.8780	-0.9826	-1.0507	-0.0336	-0.0378	-0.0408	-0.3812	-0.4324	-0.4853	-21.76	-10.57	-06.93
0.6999	-1.4370	-1.5167	-1.5539	-0.0552	-0.0603	-0.0619	-0.4623	-0.5710	-0.5919	-10.69	13.56	34.69
0.9013	-1.9457	-2.0272	-2.0782	-0.0781	-0.0798	-0.0841	-0.6359	-0.6374	-0.6493	44.29	46.47	55.36

Table 4- Excess values of Gibb's free energy (ΔG^E), viscosity (η^E), molar volume (V_m^E) and interaction parameter (d) at 303.15, 308.15 and 313.15K

Mole Fraction	Excess Gibb's free energy ΔG^E ($\times 10^{-20}$ KJmol $^{-1}$)			Excess Viscosity η^E ($\times 10^{-12}$ s)			Excess Molar volume V_m^E ($\times 10^{-3}$ m 3 /mol)			Interaction parameter (d)		
	X_3	303.15K	308.15K	313.15K	303.15K	308.15K	313.15K	303.15K	308.15K	313.15K	303.15K	308.15K
System I: Acetophenone (X_1) + 1-heptanol (X_2) +chlorobenzene (X_3) [$X_1/X_2=4:2$]												
0.0000	-0.1592	-0.1509	-0.1581	-1.1412	-0.9733	-0.7982	-0.0003	-0.0007	-0.0012	-1.81	-1.70	-1.60
0.0999	-0.1063	-0.1241	-0.1277	-0.9649	-0.8744	-0.7335	-0.0017	-0.0037	-0.0042	-15.96	-12.40	-11.90
0.3000	-0.0700	-0.1057	-0.1233	-0.7179	-0.7177	-0.5955	-0.0021	-0.0012	-0.0047	-7.01	-6.80	-4.30
0.5000	-0.0609	-0.0507	-0.0645	-0.5480	-0.4153	-0.3387	-0.0054	-0.0059	-0.0068	-3.09	-1.50	-1.10
0.7000	-0.0703	-0.0711	-0.0773	-0.3869	-0.3290	-0.2349	-0.0079	-0.0081	-0.0084	-3.30	-9.70	-11.82
0.9011	-0.0364	-0.0266	-0.0396	-0.0960	-0.0624	-0.0455	-0.0099	-0.0101	-0.0106	-65.90	-60.57	-56.98
System II: Acetophenone (X_1) + 1-heptanol (X_2) + bromobenzene (X_3) [$X_1/X_2=4:2$]												
0.0000	-0.1592	-0.1509	-0.1581	-1.1412	-0.9733	-0.7982	-0.0003	-0.0007	-0.0012	-1.81	-1.70	-1.60
0.0999	-0.1264	-0.1245	-0.1121	-1.0159	-0.8491	-0.6754	-0.0009	-0.0013	-0.0017	-18.60	-17.30	-15.50
0.2999	-0.0697	-0.0572	-0.0502	-0.6308	-0.5850	-0.4345	-0.0014	-0.0019	-0.0025	-6.10	-4.90	-3.70
0.5000	-0.0244	-0.0023	-0.0012	-0.4025	-0.3138	-0.2024	-0.0027	-0.0031	-0.0033	-1.80	-0.85	0.20
0.6999	0.0020	0.0204	0.0236	-0.1309	-0.1125	-0.0736	-0.0041	-0.0042	-0.0047	-4.40	-4.60	-4.50
0.9012	0.0438	0.0490	0.0497	-0.0635	-0.0494	-0.0217	-0.0067	-0.0074	-0.0081	-74.91	-89.99	-104.21
System III: Acetophenone (X_1) + 1-heptanol (X_2) + nitrobenzene (X_3) [$X_1/X_2=4:2$]												
0.0000	-0.1592	-0.1509	-0.1581	-1.1412	-0.9733	-0.7982	-0.0003	-0.0007	-0.0012	-1.81	-1.70	-1.60
0.0999	-0.1136	-0.1302	-0.1614	-0.2594	-0.1940	-0.0952	-0.0011	-0.0018	-0.0023	3.00	3.60	5.60
0.3000	-0.0823	-0.0879	-0.1120	-0.1218	-0.0588	-0.0413	-0.0018	-0.0031	-0.0037	8.10	3.20	4.20
0.4999	-0.0783	-0.0831	-0.0849	-0.0369	-0.0154	-0.0094	-0.0044	-0.0048	-0.0050	4.80	4.10	6.10
0.7000	-0.0685	-0.0778	-0.0814	0.1668	0.1814	0.2214	-0.0061	-0.0063	-0.0085	11.21	11.80	12.90
0.9012	-0.0565	-0.0683	-0.0526	0.2758	0.3076	0.3207	-0.0094	-0.0100	-0.0104	91.83	102.26	109.96
System IV: Acetophenone (X_1) + 1-heptanol (X_2) + ethylbenzene (X_3) [$X_1/X_2=4:2$]												
0.0000	-0.1592	-0.1509	-0.1581	-1.1412	-0.9733	-0.7982	-0.0003	-0.0007	-0.0012	-1.81	-1.70	-1.60
0.0999	-0.0920	-0.1183	-0.1558	-0.9844	-0.8955	-0.7418	0.0007	0.0006	0.0005	-13.80	-16.70	-17.80
0.2999	-0.0823	-0.1047	-0.1434	-0.7311	-0.6129	-0.4971	-0.0043	-0.0045	-0.0046	-4.30	-3.40	-3.70
0.5000	-0.0210	-0.0840	-0.1087	-0.5299	-0.4030	-0.3534	-0.0087	-0.0095	-0.0099	-1.90	0.80	-1.30
0.6999	-0.0116	-0.0747	-0.826	-0.3009	-0.2508	-0.1929	-0.0138	-0.0145	-0.0150	-2.00	-3.05	-2.90
0.9013	0.0244	-0.0261	-0.0449	-0.1164	-0.0663	-0.0389	-0.0195	-0.0202	-0.0207	-82.89	-96.51	-94.30
System V: Acetophenone (X_1) + 1-heptanol (X_2) + toluene(X_3) [$X_1/X_2=4:2$]												
0.0000	-0.1592	-0.1509	-0.1581	-1.1412	-0.9733	-0.7982	-0.0003	-0.0007	-0.0012	-1.81	-1.70	-1.60
0.0999	-0.1391	-0.1590	-0.1777	-1.0906	-1.0053	-0.8569	0.0001	0.0019	0.0018	-26.80	-20.90	-12.38
0.2999	-0.1008	-0.1049	-0.1221	-0.8207	-0.6962	-0.6070	-0.0037	-0.0038	-0.0039	-5.70	-4.80	-3.50
0.5000	-0.0911	-0.0990	-0.0997	-0.6026	-0.4823	-0.4260	-0.0082	-0.0083	-0.0086	-3.20	-1.20	-0.70
0.6999	-0.0495	-0.0959	-0.0964	-0.3473	-0.3149	-0.2476	-0.0131	-0.0134	-0.0137	-0.80	-0.92	-3.40
0.9013	-0.0584	-0.0653	-0.0653	-0.0278	-0.0201	-0.0033	-0.0179	-0.0181	-0.0184	-80.25	-81.44	-83.83