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Molecular Interactions in Certain Binary Liquid Systems Involving Toluene at Temperature 313K.

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Abstract: This paper presents experimental data for densities (ρ), viscosities (η) and ultrasonic speeds (U) of pure toluene (TOL), chlorobenzene (CB), bromobenzene (BB) and nitrobenzene (NB) and of their binary liquid mixtures at temperature 313 K. These parameters were used to determine the adiabatic compressibility (β), intermolecular free length (L_f), molar volume (V_m) and acoustic impedance (Z) and their excess values. The derived functions namely V_m^E , L_f^E , Z^E and β^E were used to have a better understanding of intermolecular interactions between the component molecules of the present liquid mixtures. The variations of these parameters with composition of mixture indicate the nature and extent of interaction between unlike molecules & suggest that the interactions occurring between toluene, chlorobenzene, bromobenzene and nitrobenzene molecules follow the sequence: Nitrobenzene > Chlorobenzene > Bromobenzene. The excess and deviation functions were fitted to Redlich-Kister type polynomial equation. The results provide information on the molecules in the pure liquids as well as in the binary mixtures.

Keywords: Molecular interactions, Acoustical properties, Molar volume, Binary Liquid mixtures.

I. Introduction:

Ultrasound waves are high frequency mechanical waves [14]. Ultrasonic wave propagation affects the physical properties of the medium and hence can furnish information about molecular interactions of the liquid and liquid mixtures. The measured ultrasonic parameters are being extensively useful to study intermolecular processes in liquid systems [21]. The sign and magnitude of the non-linear deviations from ideal values of velocities and adiabatic compressibility's of liquid mixtures with composition are attributed to the difference in molecular size and strength of interaction between unlike molecules.

Studies of thermodynamic properties of binary mixtures are of considerable interest in the fundamental understanding of the nature of interactions between the unlike molecules. In recent years, the theoretical and experimental investigations of excess and deviation functions are taken as interaction parameters to improve the results [1-4]. The thermodynamic properties of a binary mixture such as viscosity and density are important from practical and theoretical points of view to understand liquid theory. Accurate knowledge of thermodynamic properties of organic liquid mixtures has relevance in understanding the molecular interactions between the components of the mixture [14]. Binary liquid mixtures due to their unusual behavior have attracted considerable attention [19]. In chemical process industries materials are normally handled in fluid form and as a consequence, the physical, chemical, and transport properties of fluids assume importance. Thus data on some of the properties associated with the liquids and liquid mixtures like Density and viscosity find extensive application in solution theory and molecular dynamics[9][16][5]. The present work deals with the qualitative and quantitative study on the binary mixtures of toluene (TOL) with chlorobenzene (CB), bromobenzene (BB) and nitrobenzene (NB). Toluene is one of the aromatic hydro carbons also known as mono-substituted benzene. 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 [6]. The toluene molecules possess large quadrupole moments, causing an orientational ordering in this liquid. The orientational order is thought of as a partial alignment of neighbouring segments or possibly of whole molecules [7][23]. The binary mixtures containing aromatic hydrocarbons are interesting because they find applications in the studies of polymer phase diagrams and preferential interaction of polymers in mixed solvents [24] [25]. Therefore, the study of intermolecular interactions in toluene + substituted benzenes mixtures would be interesting owing to their industrial applications. Toluene, which is aprotic and polar in nature due to the presence of electron releasing methyl group. It is used as an octane booster in fuel, as a solvent for many organic compounds, paints, cleaning of polymer surface and electronic materials, as a raw material for toluene diisocyanate which is used for manufacturing the polyurethane, foams etc. [13]. The CH_3 group of toluene is an electron- donating group through induction enhances a π electron density of toluene ring [15] [26].

Nitrobenzene has 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. The nitrobenzene which is relatively complex molecule and its non-ideality in all probability may be due to polarity arisen out of C-N and N-O bonds. As far as the contribution of nitro group is concerned it rotates freely along the C-N axis which is likely to give more flexibility to the interactions arising due to the two highly polar N→O bonds [28] [29]. Chlorine atom in chlorobenzene is an electron-withdrawing atom, tends to attract to π electrons of the benzene ring, and 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 [6].

Therefore, a better understanding of the physicochemical properties of mixed solvent System I (Toluene & Nitrobenzene), System II (Toluene & Chlorobenzene) and System III (Toluene & Bromobenzene) are necessary for interpretation of data obtained from thermo chemical, electrochemical, biochemical and kinetic studies [9][18].

In view of their industrial importance, the present study reports the experimental values of densities (ρ), viscosities (η) and ultrasonic speeds (u) of pure TOL, NB, CB, BB and those of their binary mixtures at the temperature 313 K. The above experimental data were used to evaluate the excess molar volume (V_m^E), excess intermolecular free length (L_f^E), excess adiabatic compressibility (β^E) and excess acoustic impedance (Z^E) at temperature 313K. The excess values were correlated using the Redlich-Kister polynomial equation (6) to obtain their coefficients and standard deviations [9]. The study of molecular interactions in the liquid mixtures is therefore important in elucidation of the structural properties of the molecules.

II. Experimental:

Toluene was procured from (S.D Fine Chem. Ltd. India) and is further purified by the methods given in Vogel text book of practical organic chemistry [17] [27]. Nitrobenzene, chlorobenzene and Bromobenzene (S.D Fine Chem. Ltd. India) also of A.R grade 99.5% were further purified by the method given in the literature [11]. All the chemicals were stored over 0.4 nm molecular sieves to remove the traces of water, if any, and degassed just before use.

In all systems, the various concentrations of the binary liquid mixtures were prepared in terms of mole fraction. The mole fractions of the first and second component (X_1 and X_2) were varied from 0 to 1. Purities of these chemicals were checked by density determination at the temp. 313 K which showed an accuracy of 0.0001 gm cm^{-3} as compared to reported values. The density, viscosity and velocity were measured as a function of composition of binary liquid mixture at the temp. 313K. The density of sample was measured using digital densitometer (Rudolph) with an accuracy of 0.0001. An Ostwald's viscometer was used for the viscosity measurements. An ultrasonic interferometer having the frequency 2 MHz was used for ultrasonic velocity measurements. An electronically operated constant temperature bath was used to circulate water through measuring cell made up of steel containing experimental solution at 313 K temperature. An ultrasonic interferometer (Model: F81) working at a frequency 3MHz with an overall accuracy of $\pm 2\text{ ms}^{-1}$ has been used for velocity measurement. An electronically digital constant temperature bath has been used to circulate water through the double walled measuring cell made up of steel containing experimental mixtures at the desired temperature. The accuracy in the temperature measurement is $\pm 0.1\text{K}$. Reliability of the experimental data and the purity of the solvents were ascertained by calculating their densities, ultrasonic speeds and viscosities at different temperatures with the values reported as shown in Table 1.

III. Results and discussion:

The experimental densities (ρ), ultrasonic speeds (U) and viscosities (η) of pure toluene, nitrobenzene, chlorobenzene, bromobenzene and their binary mixtures are used to calculate excess thermodynamic properties of mixtures which correspond to the difference between the actual property and the property if the system behaves ideally and thus are useful in the study of molecular interactions and arrangements in the mixtures. In particular, they reflect the interactions that take place between solute-solute, solute-solvent and solvent-solvent species. The effects which are expected to operate between the component molecules under study are (i) structural effect which is due to the differences in shape and size of the component molecules (ii) reorientation effect between component molecules and (iii) energetic effect, i.e., molecular interaction that can be weakened or destroyed or established during the mixing process [22]. Thus, in the present study various acoustical parameters were calculated from measured data by using following equations

$$\text{Adiabatic compressibility } (\beta) = 1/v^2 p \quad \dots\dots (1)$$

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

Where K is temperature dependant constant, value of K 642×10^{-6} at temp. 313 k.

$$\text{Acoustic impedance } (Z) = U\rho \quad \dots\dots (3)$$

$$\text{Molar Volume } (V_m) = M/\rho \quad \dots (4)$$

Where M is mean molecular weight. It is calculated as

$$M = X_1 M_1 + X_2 M_2$$

X_1 and X_2 are mole fractions and M_1 , M_2 are molecular weights of constituent components of binary liquid mixtures.

The values of excess molar volumes (V_m^E), excess intermolecular free length (L_f^E), excess acoustic impedance, (Z^E) and excess adiabatic compressibility (β^E) were calculated with the help of the following standard relations:

$$Y^E = Y_{\text{exp.}} - (X_1 Y_1 + X_2 Y_2) \quad \dots\dots (5)$$

Where,

$Y_{\text{exp.}}$ = experimental values of mixtures

Y_1 & Y_2 = values of parameters for liquids 1 and 2 respectively.

X_1 & X_2 = mole fractions of liquid 1 (NB or CB or BB) and liquid 2 (TOL).

The values of excess adiabatic compressibility (β^E), free length (L_f^E), acoustic impedance (Z^E) and molar volume (V_m^E) for each mixture have been least-squares fitted to Redlich-Kister type polynomial equation given in literature [10] by taking the limits $n=0$ to i .

$$F(X) = X_1 (1-X_1) \sum A_i (1-2X_1)^i, \quad \dots (6)$$

Where $F(x)$ refers to V_m^E or L_f^E . The coefficient A_i is the polynomial coefficient tabulated by using the least square method computed by the MAPLE software has been used. The values of the standard deviation (σ) were obtained from the expression

$$\sigma = \left\{ \sum (F(x)_{\text{exp}} - F(x)_{\text{cal}})^2 / (k/n) \right\}^{1/2} \quad \dots (7)$$

Where k is the number of experimental points excluding the end points and n is order of polynomial equation.

The values of $F(X)_{\text{cal}}$ are obtained from Eq. (6) by using the best fit values of A_i coefficients. The coefficients A_0 , A_1 , A_2 , A_3 and A_4 along with standard deviations σ of fit for all the mixtures are listed in Table 4 and it has been observed that standard deviations are very low.

The sign of V_m^E of a system depends upon the relative magnitude of expansion and contraction of the two liquids due to mixing [8][20]. If the factors causing expansion dominate the contraction factors, the V_m^E becomes positive. On the other hand if the contraction factors dominate the expansion factors, then V_m^E become negative. The factors that are responsible for expansion in volume are as follows, i. Loss of dipolar association, ii. The geometry of molecular structure, which does not allow fitting of one component into other component, iii. Steric hindrance, which opposes the proximity of the constituent molecules. The negative V_m^E values arise due to dominance of the following factors. i. Chemical interaction between constituent chemicals. ii. Accommodation of molecules of one component into the interstitials of the molecules of the other component. iii. Geometry of the molecular structure that favors fitting of the component molecules with each other. The negative V_m^E values in the mixtures under study indicate that interactions between molecules of the mixtures are stronger than interactions between molecules in the pure liquids and that associative force dominate the behavior of the solution [9]. The positive V_m^E values are attributed to weak dipole-dipole interactions between unlike molecules in the mixtures.

Fig. 1(a) reveals that the excess molar volumes, V_m^E is negative at temperature 313K for the investigated mixtures of (Toluene+Nitrobenzene). The observed negative value of V_m^E for the binary mixture indicates the presence of specific interactions between toluene and nitrobenzene molecule. The negative V_m^E values are attributed to strong dipole-dipole interactions between unlike molecules in the mixtures [8]. Fig. 1(b) & Fig. 1(c) reveal that V_m^E are positive for the investigated mixtures of (Toluene+Chlorobenzene) and (Toluene+Bromobenzene) at temperature 313K. The positive V_m^E value is attributed to weak dipole-dipole interactions between unlike molecules in the mixtures. The value for V_m^E is more positive for system III (Toluene+Bromobenzene) than system II (Toluene+Chlorobenzene). The less positive V_m^E for system II attributed to more stronger molecular interactions than system III.

It is clear from fig. 1(a), (b) and (c) that the values of V_m^E shows negative deviation for the mixture of TOL with NB and positive deviations for the mixtures of TOL with CB & BB and follows the sequence: Nitrobenzene > Chlorobenzene > Bromobenzene.

Acoustical parameters such as adiabatic compressibility, intermolecular free length, molar volume and adiabatic compressibility were calculated from the measured ultrasonic velocity and density values at temperature 313K have been tabulated in Table 2 for various mole fractions of pure component (X_1) of investigated binary mixtures System I (TOL +NB), System II (TOL +CB) and System III (TOL +BB). For all the three systems densities and viscosities increases with increase in mole fraction, suggesting thereby more association between solute and solvent molecules [12]. For system I, velocity increases but it decreases for

system II & III. Adiabatic compressibility & intermolecular free length decreases but acoustic impedance increases for all the three systems.

Values of excess adiabatic compressibility (β^E), free length (L_f^E), acoustic impedance (Z^E) and molar volume (V_m^E) at 313 K for all three systems have been tabulated in Table 3. Negative excess values are due to closely packed molecules which accounts for the existence of strong molecular interactions, whereas positive excess values reflect weak interactions between unlike molecules [30].

IV. Conclusions:

The concentration dependencies of ultrasonic velocities (U), viscosities (η) and densities (ρ) of toluene with nitrobenzene, chlorobenzene and bromobenzene binary systems and of pure liquids have been measured at 313K temperature. The nonlinear variation of the related parameters such as adiabatic compressibility (β), intermolecular free length (L_f), molar volume (V_m), and acoustic impedance (Z) were elaborated to understand the molecular interactions that lead to the volume contraction of different binary mixtures which were under investigation. The positive and negative variations of the excess values with concentration and temperature of the same acoustic parameters supported the presence of interaction between unlike molecules. The present work suggest that due to the variation of the excess molar volume and other acoustical parameters, the strength of interaction between toluene and nitrobenzene, chlorobenzene, bromobenzene & molecules follow the sequence: Nitrobenzene > Chlorobenzene > Bromobenzene and also the order of favorable accommodation of the component molecules into each other's structure.

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Symbols and abbreviations

ρ -	Density
η -	Viscosity
U-	Ultrasonic Velocity
β -	Adiabatic Compressibility
L_f -	Intermolecular free length
V_m -	Molar Volume
Z -	Acoustic Impedance
β^E -	Excess Adiabatic compressibility

L_f^E -	Excess Intermolecular Free Length
V_m^E -	Excess Molar Volume
Z^E -	Excess acoustic Impedance
σ -	Standard deviation
X_1 -	Mole fraction for nitrobenzene or chlorobenzene or bromobenzene
X_2 -	Mole fractions for Toluene
M_1 -	Molecular weight of nitrobenzene or chlorobenzene or bromobenzene
M_2 -	Molecular weight of Toluene
K-	Kelvin
TOL-	Toluene
CB-	Chlorobenzene
BB-	Bromobenzene
NB-	Nitrobenzene

Table 1: Values of density (ρ), viscosity (η), ultrasonic velocity (U), adiabatic compressibility (β), free length (L_f), acoustic impedance (Z) and molar volume (V_m) of pure liquids at 313 K.

Component	Temp. T/K	ρ Kg m ³	η x10 ³ Nsm ²	U ms ⁻¹	β x 10 ¹⁰ m ² N ⁻¹	L_f x 10 ¹⁰ m	Z x10 ⁻⁶ Kg m ⁻² s ⁻¹	V_m x10 ⁶ m ³ mol ⁻¹
Toluene	313K	846.2	0.0514	1228.67	7.8281	179.62	1.0397	108.88
Nitrobenzene	313K	1182.7	0.1017	1398.93	4.3205	133.44	1.6545	104.09
Bromobenzene	313K	1466.5	0.0544	1044.80	6.2467	160.46	1.5322	107.07
Chlorobenzene	313K	1084.8	0.0534	1208.93	6.3073	161.23	1.3114	103.76

Table 2: Values of density (ρ), viscosity (η), ultrasonic velocity (U), adiabatic compressibility (β), free length (L_f), acoustic impedance (Z) and molar volume (V_m) at 313 K for all the three binary liquid mixtures.

T/K	Mole fraction X_1	ρ / Kg m ³	$10^3\eta$ /(Nsm ²)	U /ms ⁻¹	$10^{10}\beta$ /(m ² N ⁻¹)	$10^{10}L_f$ / (m)	$10^{-6}Z$ /(Kg m ⁻² s ⁻¹)	10^6V_m /(m ³ mol ⁻¹)
TOL+NB 313K	0.0000	846.20	0.0514	1228.67	7.8281	179.62	1.0397	108.89
	0.2066	948.45	0.0538	1260.73	6.6334	165.35	1.1957	103.89
	0.4098	1029.64	0.0591	1289.87	5.8375	155.11	1.3281	101.81
	0.6097	1082.58	0.0687	1326.27	5.2514	147.12	1.4358	102.55
	0.8064	1129.28	0.0828	1363.47	4.7633	140.12	1.5397	103.71
	1.0000	1182.70	0.1017	1398.93	4.3205	133.44	1.6545	104.09
TOL+BB 313K	0.0000	846.20	0.0514	1228.67	7.8281	179.62	1.0397	108.89
	0.2025	963.10	0.0518	1200.67	7.2025	172.30	1.1564	109.31
	0.4038	1071.60	0.0522	1176.00	6.7477	166.77	1.2602	110.43
	0.6038	1196.50	0.0527	1129.80	6.5476	164.28	1.3518	109.75
	0.8025	1327.60	0.0534	1087.47	6.3694	162.03	1.4437	108.62
	1.0000	1466.50	0.0544	1044.80	6.2467	160.46	1.5322	107.07
TOL+CB 313K	0.0000	846.20	0.0514	1228.67	7.8281	179.62	1.0397	108.89
	0.2076	883.58	0.0518	1222.59	7.5717	176.66	1.0803	109.08
	0.4113	925.96	0.0521	1217.40	7.2868	173.30	1.1273	108.58
	0.6112	974.35	0.0525	1213.67	6.9677	169.46	1.1825	107.38
	0.8074	1028.73	0.0529	1211.47	6.6233	165.22	1.2463	105.59
	1.0000	1084.80	0.0534	1208.93	6.3073	161.23	1.3115	103.76

Table 3: Values of excess adiabatic compressibility (β^E), free length (L_f^E), acoustic impedance (Z^E) and molar volume (V_m^E) at 313 K

Mole fraction(X_1)	$10^{10}\beta^E /(\text{m}^2\text{N}^{-1})$	$10^{10} L_f^E / \text{m}$	$10^{-6} Z^E /$	$10^6 V_m^E$
			($\text{Kg m}^{-2}\text{s}^{-1}$)	/ $(\text{m}^3\text{mol}^{-1})$
SystemI (Toluene + Nitrobenzene)				
0.0000	0.0000	0.0000	0.0000	0.0000
0.1037	-0.4426	-2.7196	0.0139	-2.1283
0.2066	-0.7781	-4.7333	0.0290	-4.0023
0.3086	-0.9704	-5.5228	0.0361	-4.9809
0.4098	-1.0633	-5.5864	0.0364	-5.1078
0.5102	-1.0714	-5.1187	0.0296	-4.3541
0.6097	-1.0083	-4.3460	0.0212	-3.4090
0.7085	-0.8816	-3.4614	0.0138	-2.4296
0.8064	-0.6684	-2.2665	0.0042	-1.3124
0.9036	-0.3862	-1.1998	0.0012	-0.5918
1.0000	0.0000	0.0000	0.0000	0.0000
SystemIII (Toluene + Bromobenzene)				
0.0000	0.0000	0.0000	0.0000	0.0000
0.1014	-0.1015	-1.8432	0.0095	0.2363
0.2025	-0.1957	-3.4456	0.0169	0.7947
0.3033	-0.2707	-4.6804	0.0213	1.6462
0.4038	-0.2915	-5.1176	0.0216	2.2768
0.5039	-0.2473	-4.6126	0.0185	2.2968
0.6038	-0.1871	-3.7749	0.0148	1.9566
0.7033	-0.1477	-3.0768	0.0121	1.6271
0.8025	-0.1040	-2.2167	0.0088	1.1922
0.9014	-0.0570	-1.2163	0.0056	0.5689
1.0000	0.0000	0.0000	0.0000	0.0000
SystemII (Toluene + Chlorobenzene)				
0.0000	0.0000	0.0000	0.0000	0.0000
0.1043	0.0385	0.5289	-0.0093	0.7705
0.2076	0.0602	0.8511	-0.0159	1.2559
0.3100	0.0756	1.0868	-0.0208	1.5955
0.4113	0.0856	1.2427	-0.0242	1.7998
0.5118	0.0914	1.3329	-0.0261	1.8789
0.6112	0.0704	1.0808	-0.0233	1.6221
0.7098	0.0466	0.7703	-0.0188	1.2746
0.8074	0.0240	0.4481	-0.0128	0.8458
0.9042	0.0106	0.2179	-0.0069	0.4437
1.0000	0.0000	0.0000	0.0000	0.0000

Table (4).Redlich-Kister constants for the deviations of Excess parameters of intermolecular free length, acoustic impedance, adiabatic compressibility and molar volume of different binary mixtures of styrene at temperature 313K

Property	T/K	A ₀	A ₁	A ₂	A ₃	A ₄	σ (Y ^E)
TOL+NB							
10 ¹⁰ Lf ^E /(m)	313K	-20.7180	-12.6244	-5.8286	3.8011	7.2416	0.0865570
10 ⁶ Vm ^E /(m ³ mol ⁻¹)		-17.9895	-16.5447	1.3473	10.1212	6.0688	0.0890618
10 ⁻⁶ Z ^E /(Kgm ⁻² S ⁻¹)		0.1232	0.1504	-0.0226	-0.0972	-0.0728	0.0012346
10 ¹⁰ β ^E /(m ² N ⁻¹)		-4.2941	-0.4598	-0.8275	0.3263	0.5647	0.0066810
TOL+BB							
10 ¹⁰ Lf ^E /(m)	313K	-18.6511	-11.7426	1.6079	13.3820	1.9758	0.1564964
10 ⁶ Vm ^E /(m ³ mol ⁻¹)		9.2077	1.1967	-9.6255	-7.0455	3.4963	0.1415438
10 ⁻⁶ Z ^E /(Kgm ⁻² S ⁻¹)		0.7523	0.0659	0.0244	-0.0655	-0.0203	0.2606013
10 ¹⁰ β ^E /(m ² N ⁻¹)		-1.0054	-0.9180	0.0919	1.0865	0.1977	0.0125029
TOL+CB							
10 ¹⁰ Lf ^E /(m)	313K	5.1924	1.3317	-5.4935	1.2786	5.9842	0.0425574
10 ⁶ Vm ^E /(m ³ mol ⁻¹)		7.41542	1.3323	-4.2624	1.1511	4.9918	0.0325190
10 ⁻⁶ Z ^E /(Kgm ⁻² S ⁻¹)		-0.10256	-0.0027	0.0579	-0.0214	-0.0560	0.0005389
10 ¹⁰ β ^E /(m ² N ⁻¹)		0.35461	0.1335	-0.4575	0.0910	0.5201	0.0034876

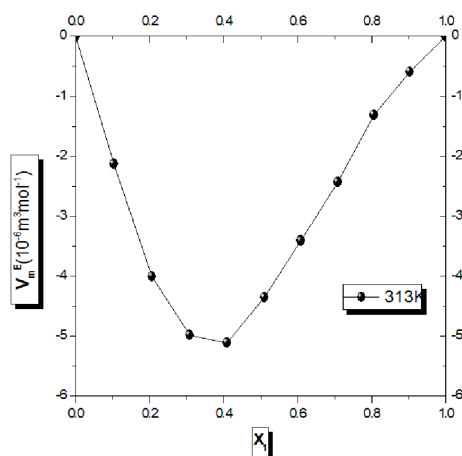


Fig. 1 (a) Plot of excess molar volume (V_m^E) against mole fraction (X_1) of Toluene and Nitrobenzene at temp. 313K.

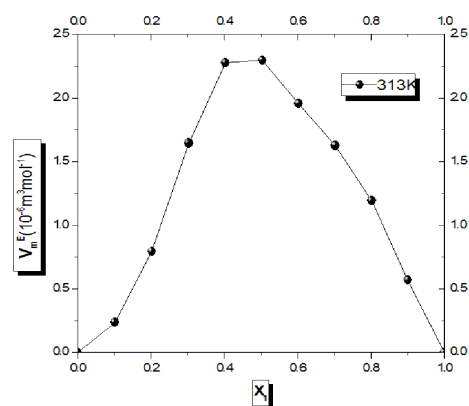


Fig. 1 (c) Plot of excess molar volume (V_m^E) against mole fraction (X_1) of Toluene and Bromobenzene at temp. 313K.

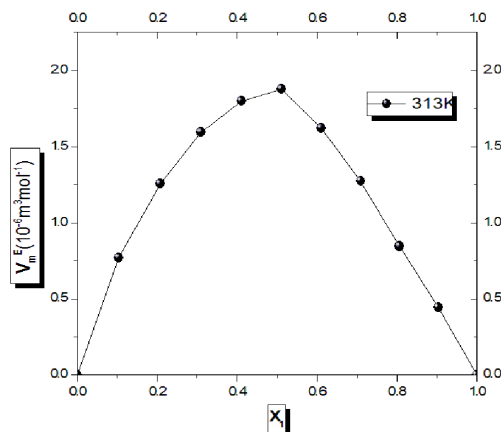


Fig.1 (b) Plot of excess molar volume (V_m^E) against mole fraction (X_1) of Toluene and Chlorobenzene at temp. 313K.