



Thermodynamic and Acoustic Study on Molecular Interactions in Certain Binary Liquid Systems Involving Ethylbenzene at Temperature 313K

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Abstract: This paper presents experimental data for densities (ρ), viscosities (η) and ultrasonic speeds (U) of pure ethylbenzene (ETB), 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 ethylbenzene, chlorobenzene, bromobenzene and nitrobenzene molecules follow the sequence: Chlorobenzene > Bromo benzene > Nitrobenzene. 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: Density, Viscosity, Ultrasonic speed, Molar volume, Binary 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 ethylbenzene (ETB) with chlorobenzene (CB), bromobenzene (BB) and nitrobenzene (NB). The aromatic hydrocarbon ethylbenzene is non-polar compounds with no measurable dipole moment. Thus it involves weak intermolecular interactions. The choice of this solvent was done because of its opposite nature of polarity and their wide range of applicability [13]. Ethyl benzene has been used as a solvent in paints. Ethyl benzene is important in petrochemical industry in the production of styrene which is in turn used for making polystyrene [6]. Para anisaldehyde and ethyl benzene mixture is used to prepare allyl anisole analog repellent pesticides [15]. Chlorine atom in chlorobenzene is an electron- withdrawing atom, tends to attract to π electrons of the benzene ring thereby and decreases the electron density of the benzene ring. As a result, the benzene ring in chlorobenzene becomes relatively poor electron-donor towards the electron seeking proton of any groups. Chlorobenzene is more reactive because chlorine atom is bonded with SP^3 hybridized carbon atom and consequently can be removed easily [6]. Bromobenzene and Nitrobenzene are occasionally used as a solvent for electrophilic substitution reactions. Therefore, a better understanding of the physicochemical properties of mixed solvent System I (Ethylbenzene & Chlorobenzene), System II

(Ethylbenzene & Bromobenzene) and System III (Ethylbenzene& Nitrobenzene) 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 ETB, CB, BB, NB 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:

Ethylbenzene (S.D. Fine chem., Pvt. Ltd.) were distilled at atmospheric pressure. Ethylbenzene was dried over phosphorus pentoxide for several days, distilled, stored over 4 Å molecular sieve and used immediately [7]. Chlorobenzene, Nitrobenzene 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 ± 2 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 ± 0.1 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 ethylbenzene, chlorobenzene, bromobenzene, nitrobenzene 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

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

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

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

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

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

Where M is mean molecular weight. It is calculated as

$$M = X_1M_1 + X_2M_2$$

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:

$$Y^E = Y_{\text{exp.}} - (X_1Y_1 + X_2Y_2) \quad \dots\dots (5)$$

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(CB or BB or NB) and liquid 2 (ETB).

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

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

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), Fig.1(b) & Fig.1(c) reveal that the excess molar volumes, V_m^E are negative at temperature 313K for all the investigated mixtures of (ethylbenzene+chlorobenzene), (ethylbenzene+bromobenzene) and (ethylbenzene+nitrobenzene). The observed negative values of V_m^E for the binary mixtures indicate the presence of specific interactions between ethylbenzene and chlorobenzene or bromobenzene or nitrobenzene molecules. The negative V_m^E values are attributed to strong dipole-dipole interactions between unlike molecules in the mixtures [8]. The value for V_m^E is more negative for system I (ethylbenzene+chlorobenzene) than system II (ethylbenzene+bromobenzene) & system III (ethylbenzene+nitrobenzene). The more negative V_m^E for system I attributed to strongest molecular interactions than system II & III.

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

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 (ETB +CB), System II (ETB +BB) and System III (ETB +NB). For all the three systems densities increase with increase in mole fraction. For the system III velocities and viscosities increases with increase in concentration, suggesting thereby more association between solute and solvent molecules [12], but for systems I & II both are decreasing. Acoustical parameters like adiabatic compressibility (β), intermolecular free length (L_f) & molar volume (V_m) decreases for all the three systems but acoustic impedance (Z) increases for all the three systems with increase in mole fraction (X_1). 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 [17].

IV. Conclusions:

The concentration dependencies of ultrasonic velocities (U), viscosities (η) and densities (ρ) of ethylbenzene with chlorobenzene, bromobenzene and nitrobenzene 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 ethylbenzene and chlorobenzene, bromobenzene & nitrobenzene molecules follow the

sequence: Chlorobenzene > Bromobenzene > Nitrobenzene 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 chlorobenzene or bromobenzene or nitrobenzene
X_2 -	Mole fractions for Ethylbenzene
M_1 -	Molecular weight of chlorobenzene or bromobenzene or nitrobenzene
M_2 -	Molecular weight of Ethylbenzene
K-	Kelvin
ETB-	Ethylbenzene
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.

	Temp.	ρ	η	U	β	L_f	Z	V_m
		Kg m ³	x10 ³ Nsm ²	ms ⁻¹	x 10 ¹⁰ m ² N ⁻¹	x 10 ¹⁰ m	x10 ⁻⁶ Kg m ² s ⁻¹	x10 ⁶ m ³ mol ⁻¹
Component	T/K							
Ethylbenzene	313K	848.6	0.0569	1252.00	7.5178	176.03	1.0624	125.11
Chlorobenzene	313K	1084.8	0.0534	1208.93	6.3073	161.23	1.3114	103.76
Bromobenzene	313K	1466.5	0.0544	1044.80	6.2467	160.46	1.5322	107.07
Nitrobenzene	313K	1182.7	0.1017	1398.93	4.3205	133.44	1.6545	104.09

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)
ETB+CB	0.0000	848.60	0.0569	1252.00	7.5178	176.03	1.0624	125.11
313K	0.2313	899.79	0.0556	1237.80	7.2537	172.91	1.1138	119.64
	0.4452	947.70	0.0549	1227.50	7.0030	169.89	1.1633	115.03
	0.6435	997.30	0.0538	1214.50	6.7980	167.39	1.2112	110.58
	0.8280	1038.00	0.0538	1213.73	6.5397	164.18	1.2599	107.38
	1.0000	1084.80	0.0534	1208.93	6.3074	161.23	1.3114	103.76
ETB+BB								
313K	0.0000	848.60	0.0569	1252.00	7.5178	176.03	1.0624	125.11
	0.2258	974.00	0.0557	1203.47	7.0888	170.93	1.1722	120.79
	0.4375	1101.20	0.0553	1172.00	6.6112	165.07	1.2906	116.61
	0.6363	1224.50	0.0551	1133.00	6.3618	161.93	1.3874	113.13
	0.8235	1347.20	0.0547	1089.00	6.2591	160.62	1.4671	109.89
	1.0000	1466.50	0.0544	1044.80	6.2467	160.46	1.5322	107.07
ETB+NB								
313K	0.0000	848.60	0.0569	1252.00	7.5178	176.03	1.0624	125.11
	0.2302	916.80	0.0620	1278.13	6.6769	165.89	1.1718	120.06
	0.4436	985.00	0.0688	1310.67	5.9099	156.07	1.2910	115.42
	0.6421	1051.60	0.0770	1335.87	5.3287	148.20	1.4048	111.30
	0.8271	1117.10	0.0887	1366.60	4.7932	140.56	1.5266	107.58
	1.0000	1182.70	0.1017	1398.93	4.3205	133.44	1.6545	104.09

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)	$10^{10}\beta^E$ /(m ² N ⁻¹)	$10^{10}L_f^E$ /m	$10^{-6}Z^E$ / (Kg m ⁻² s ⁻¹)	$10^6V_m^E$ /(m ³ mol ⁻¹)
System I (ethylbenzene + chlorobenzene)				
0.0000	0.0000	0.0000	0.0000	0.0000
0.1180	0.0009	0.1781	-0.0027	-0.5345
0.2313	0.0010	0.3019	-0.0063	-0.5369
0.3403	0.0024	0.4001	-0.0087	-0.5653
0.4452	0.0031	0.4521	-0.0100	-0.5764
0.5462	0.0203	0.6689	-0.0116	-0.5965
0.6435	0.0394	0.8803	-0.0115	-0.7912
0.7374	0.0211	0.5994	-0.0100	-0.4460
0.8280	0.0117	0.3983	-0.0088	-0.0528
0.9155	0.0067	0.2235	-0.0048	-0.0457
1.0000	0.0000	0.0000	0.0000	0.0000
System II (ethylbenzene + bromobenzene)				

0.0000	0.0000	0.0000	0.0000	0.0000
0.1147	-0.0356	-0.8882	0.0019	-0.1897
0.2258	-0.0670	-1.5805	0.0037	-0.2473
0.3333	-0.1618	-2.9292	0.0122	-0.3736
0.4375	-0.2520	-4.1437	0.0227	-0.6062
0.5384	-0.2996	-4.7567	0.0287	-0.5671
0.6363	-0.2599	-4.1905	0.0260	-0.5025
0.7313	-0.2414	-3.8229	0.0259	-0.4866
0.8235	-0.1601	-2.5888	0.0178	-0.3642
0.9130	-0.0783	-1.2957	0.0093	-0.2381
1.0000	0.0000	0.0000	0.0000	0.0000
System III (ethylbenzene + nitrobenzene)				
0.0000	0.0000	0.0000	0.0000	0.0000
0.1173	-0.2307	-0.0257	-0.0162	-0.1720
0.2302	-0.4391	-0.3350	-0.0269	-0.2156
0.3389	-0.6058	-0.7140	-0.0322	-0.3617
0.4436	-0.7220	-1.0651	-0.0341	-0.3710
0.5446	-0.7367	-0.7219	-0.0382	-0.3307
0.6421	-0.7085	-0.4859	-0.0378	-0.3117
0.7362	-0.6310	-0.3355	-0.0335	-0.2504
0.8271	-0.4962	-0.2514	-0.0255	-0.1432
0.9150	-0.2895	-0.1393	-0.0144	-0.0515
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)
ETB+CB							
10 ¹⁰ Lf ^E /(m)	313K	2.47885	-3.08342	0.22616	4.54968	-0.94650	0.1278431
10 ⁶ V _m ^E /(m ³ mol ⁻¹)		-2.64342	1.52280	1.08842	-8.30780	-2.13711	0.1576142
10 ⁻⁶ Z ^E /(Kgm ⁻² S ⁻¹)		-0.04370	0.01695	-0.00925	0.01038	0.01382	0.0005477
10 ¹⁰ β ^E /(m ² N ⁻¹)		0.06458	-0.24118	0.01402	0.37239	-0.07471	0.0104003
ETB+BB							
10 ¹⁰ Lf ^E /(m)	313K	-18.11647	9.80981	19.59542	-6.70524	-17.61694	0.2445042
10 ⁶ V _m ^E /(m ³ mol ⁻¹)		-2.33929	0.78436	2.97298	-0.75078	-4.95702	0.0757134
10 ⁻⁶ Z ^E /(Kgm ⁻² S ⁻¹)		0.10493	-0.10953	-0.15047	0.06711	0.14992	0.0022020
10 ¹⁰ β ^E /(m ² N ⁻¹)		-1.12566	0.78238	1.60345	-0.49798	-1.43506	0.0195878
ETB+NB							
10 ¹⁰ Lf ^E /(m)	313K	-3.53527	-2.61626	7.90088	6.65231	-5.89980	0.1386764
10 ⁶ V _m ^E /(m ³ mol ⁻¹)		-1.45881	-0.15043	0.61004	-0.45017	-0.18773	0.0434188
10 ⁻⁶ Z ^E /(Kgm ⁻² S ⁻¹)		-0.14674	0.03929	-0.07404	-0.04317	0.05728	0.0012355
10 ¹⁰ β ^E /(m ² N ⁻¹)		-2.94818	0.55961	0.30551	0.63885	-0.44010	0.0104532

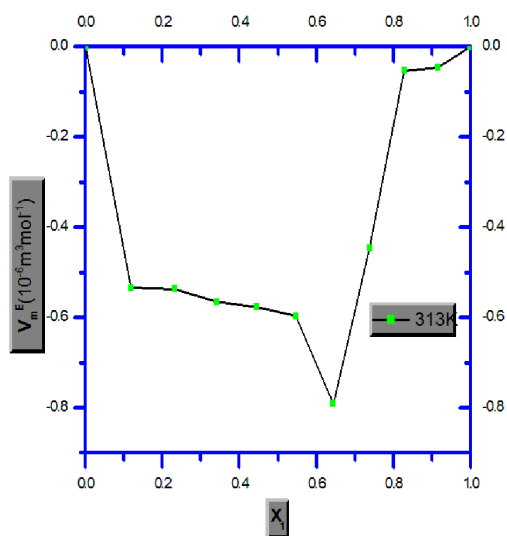


Fig. 1 (a) Plot of excess molar volume (V_m^E) against mole fraction (X_1) of Ethylbenzene and Chlorobenzene at temp. 313K.

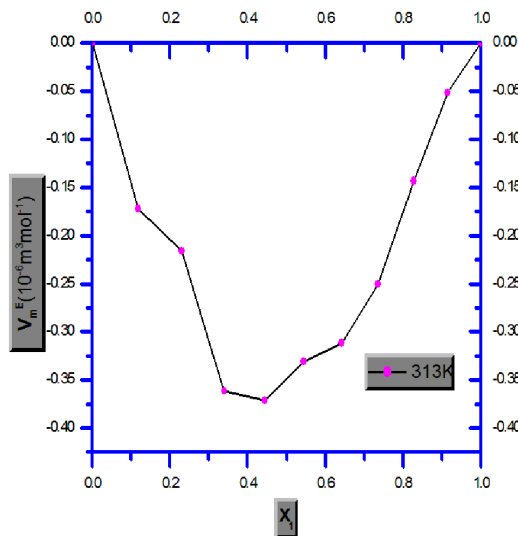


Fig. 1 (c) Plot of excess molar volume (V_m^E) against mole fraction (X_1) of Ethylbenzene and Nitrobenzene at temp. 313K.

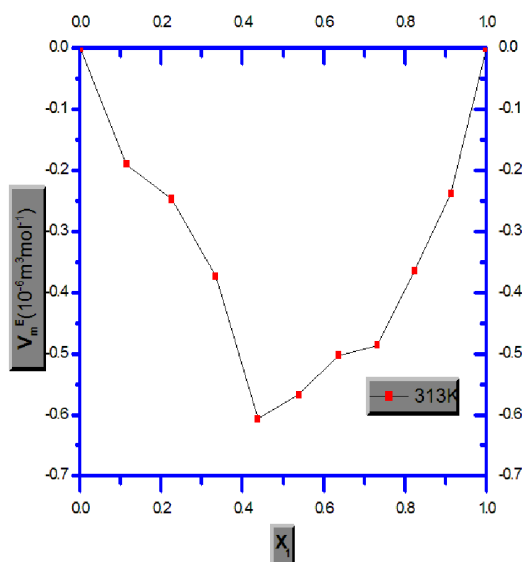


Fig.1 (b) Plot of excess molar volume (V_m^E) against mole fraction (X_1) of Ethylbenzene and Bromobenzene at temp. 313K.