

## Ultrasonic Investigations on 1-Bromopropane in Chlorobenzene Mixture at 303.15K, 308.15K, 313.15K and 318.15K at 2 MHz

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### Abstract

*The ultrasonic velocity ( $U$ ), density ( $\rho$ ) and viscosity ( $\eta$ ) have been measured for the binary mixture of 1-Bromopropane with Chlorobenzene at 303.15, 308.15, 313.15 and 318.15 K at frequency 2 MHz. From the experimental data acoustical parameters such as adiabatic compressibility ( $\beta_{ad}$ ), intermolecular free length ( $L_f$ ), acoustic impedance ( $Z$ ), molar volume ( $V_m$ ), free volume ( $V_f$ ), available volume ( $V_a$ ), relaxation time ( $\tau$ ), internal pressure ( $\pi_i$ ) and Gibbs's free energy ( $\Delta G^*$ ) have been computed. These results are interpreted in terms of molecular interaction between the components of the mixtures.*

**Keywords:** *Ultrasonic velocity, 1-Bromopropane, Chlorobenzene and molecular interactions*

### 1. Introduction

This paper is a continuation of our work related to the study of acoustic, transport and thermodynamic properties of binary mixtures of 1-Bromopropane in Chlorobenzene [1]. The variation of ultrasonic velocity and other ultrasonic parameters of binary liquid mixtures have been studied by many researchers and they have shed light upon structural changes associated with liquid mixtures of weakly or strongly interacting compounds [2-7]. In the present study to explore the nature of interactions occurring between the mixing components, ultrasonic velocity ( $U$ ), densities ( $\rho$ ) and viscosity ( $\eta$ ) of binary mixtures of 1-Bromopropane in Chlorobenzene have been measured over the entire range of composition at 303.15, 308.15, 313.15 and 318.15 K at 2 MHz. By using the above experimental values several acoustic parameters such as adiabatic compressibility ( $\beta_{ad}$ ), intermolecular free length ( $L_f$ ), free volume ( $V_f$ ), acoustic impedance ( $Z$ ), molar volume ( $V_m$ ), available volume ( $V_a$ ), relaxation time ( $\tau$ ), internal pressure ( $\pi_i$ ) and Gibbs's free energy ( $\Delta G^*$ ) have been reported.

1-Bromopropane composition is disclosed, which stays stable even under the condition that it is repeatedly used at high temperatures over an extended period of time as in vapour degreasing. 1-Bromopropane based fluids have found widespread use in industry for solvent cleaning, *i.e.*, vapor degreasing, cold cleaning and ultrasonic cleaning of complex metal parts, circuit boards, electronic components, implantable prosthetic devices, optical equipment and others [8]. Chlorobenzene is a polar molecule and is important component in synthetic chemistry (produce latex systems), in medicine and biological processes (fungicides, drugs, flavouring extracts, and antiseptics) and is widely used in preparing industrial solvents [9]. The above characteristic physical nature of 1-Bromopropane and Chlorobenzene motivated the authors to study the molecular associations in binary mixtures of 1-Bromopropane and Chlorobenzene over the whole concentration range.

## 2. Experimental Techniques

Acoustical parameters were calculated using the measure values of velocity, viscosity and density were measured as function of concentration (0.0 to 1.0) of binary liquid mixture 1-Bromopropane in Chlorobenzene at 2 MHz and at different temperatures (303.15K, 308.15K, 313.15K and 318.15K).

The chemicals used were of AR/Merck quality. All were purified by standard procedures discussed in purification of laboratory chemicals before use [10]. Mixtures were prepared by mixing appropriate volumes of liquids in airtight bottles to minimize the evaporation losses and weighed in a single-pan Mettler balance to an accuracy of  $\pm 0.001$ mg. Preferential evaporation losses of solvents from mixtures were kept to a minimum as evidenced by repeated measurement of the physical properties over an interval of 2-3 days, during which no changes in physical properties were observed. The possible error in molefractions is estimated to be around  $\pm 0.0001$ .

The ultrasonic velocities in liquid mixtures have been measured using an ultrasonic interferometer (Model F81) supplied by Mittal Enterprises, New Delhi working at 2 MHz frequency with an accuracy of  $\pm 0.1$  m.sec<sup>-1</sup>. The temperatures were controlled by circulating water around the liquid cell from a thermostatically controlled water bath (accuracy  $\pm 0.1$ °C). The temperature of the cell was measured using a thermocouple (at the crystal) and was found to be accurate to  $\pm 0.25$ °C. The densities of mixtures were measured using specific gravity bottle and Mettler single pan microbalance (E-METTLER, ZURICH), which allows reading the fifth decimal digit. The purity of the sample was checked by comparing the experimental data of density at four temperatures with the values available in the literature [11]. The viscosity at different temperature was measured using Oswald's Viscometer and stop clock with an accuracy of  $\pm 0.0001$  Nsm<sup>-2</sup> and  $\pm 0.1$ s.

### Theory and Calculations:

Using the measured data, the acoustical parameters have been calculated

- 1) Adiabatic compressibility ( $\beta_{ad}$ ) :

$$\beta_{ad} = 1/\rho U^2 \quad \dots\dots (1)$$

- 2) Intermolecular free length ( $L_f$ ) :

$$L_f = K_T \sqrt{\beta_{ad}} \quad \dots\dots (2)$$

- 3) Free volume ( $V_f$ ) :

$$V_f = (M_{eff} U / k \eta) \quad \dots\dots (3)$$

- 4) Acoustic impedance (Z) :

$$Z = \rho \cdot U \quad \dots\dots (4)$$

- 5) Molar Volume ( $V_m$ ) :

$$V_m = M/\rho \quad \dots\dots (5)$$

- 6) Available volume ( $V_a$ ) :

$$V_a = (M/\rho)[1 - (U/U_\infty)] \quad \dots\dots (6)$$

7) Relaxation time ( $\tau$ ) :

$$\tau = \frac{4}{3} \beta_{ad} \eta \quad \dots\dots\dots (7)$$

8) Internal pressure ( $\Pi_i$ ) :

$$\Pi_i = bRT (k\eta/U^2)^{1/2} (\rho^{2/3}/M_{eff}^{7/6}) \quad \dots\dots\dots (8)$$

9) The Gibbs free energy ( $\Delta G^*$ ) :

$$\Delta G^* = k_B T \log(k_B T \tau / h) \quad \dots\dots\dots (9)$$

where  $K_T$  is the temperature-dependent constant known as Jacobson's constant.  $M_{eff}$  is the effective molecular weight of the mixture ( $M_{eff} = \sum m_i X_i$ , where  $m_i$  and  $X_i$  are the molecular weight and mole fraction of individual constituents, respectively),  $k$  is a temperature independent constant which is equal to  $4.281 \times 10^9$  for all liquids.  $b$  stands for cubic packing, which is assumed to be 2 for all liquids.  $T$  is the absolute temperature,  $k_B$  is Boltzmann's constant, and  $h$  is Planck's constant.

### 3. Results and Discussions

The experimental values of ultrasonic velocity, density and viscosity for the binary system 1-Bromopropane in Chlorobenzene with 2 MHz and at different temperatures (303.15K, 308.15K, 313.15K and 318.15K) are given in the Tables 1, 3, 5 and 7. Adiabatic compressibility ( $\beta_{ad}$ ) is a measure of intermolecular association or dissociation or repulsion. It is independent of temperature and pressure. It also determines the orientation of the solvent molecules around the liquid molecules. The structural arrangement of the molecule affects the adiabatic compressibility. From the Tables 1, 3, 5 and 7 it is observed that adiabatic compressibility increases with increase in mole fraction of 1-Bromopropane in the mixture taken up for study which is presented in Figure 4. As adiabatic compressibility is inversely proportional to ultrasonic velocity ( $U$ ), since ultrasonic velocity decreases with mole fraction, so that adiabatic compressibility increases with mole fraction of 1-Bromopropane shown in Figure 1. From Figure 2 the variation of density ( $\rho$ ) of 1-Bromopropane + Chlorobenzene system increases with increases in concentration. It is clearly shows the straight line which is proportional to density. Viscosity ( $\eta$ ) decreases with rise in concentration in system suggesting thereby more association between solute and solvent molecules and given tables and shown in Figure 3.

From Tables 1, 3, 5 and 7 it is also observed that, the values of intermolecular free length ( $L_f$ ) increases with increase in temperatures and also increases with concentration which is plotted in Figure 5, it clearly reveals that interaction become stronger at higher temperatures. The free length is the distance between the surfaces of the neighbouring molecules. Generally, when the ultrasonic velocity decreases, the value of the free length increases. The increase in intermolecular free length indicates the interaction between the solute and solvent molecules due to which the structural arrangement in the neighbourhood of constituent ions or molecules gets affected considerably.

The variation of molar volume ( $V_m$ ) with the increase in concentration of 1-Bromopropane are presented in tables 2, 4, 6 and 8 and shown graphically in Figure 6. The increase in concentration molar volume with of 1-Bromopropane predicts the existence of specific interactions among the components in the binary liquid mixture [12]. The decrease in acoustic

impedance ( $Z$ ) with increase in concentrations of solutes can be explained in terms of inter and intra molecular interactions between the molecules of liquid systems shown in Figure 7 and Tables 1, 3, 5 and 7. This indicates significant of interaction in liquid system. Acoustic impedance is found to be almost inversely proportional to the adiabatic compressibility.

Free volume ( $V_f$ ) is defined as the average volume in which the centre of the molecules can move inside the hypothetical cell due to the repulsion of surrounding molecules [13]. Free volume increases with increase in concentration; this may be due to the decrease in vibration of the molecules about their mean position plotted in Figure 8 and in Tables 2, 4, 6 and 8. The compactness and strength of bonding between the components molecules of the liquid mixture are measured directly by the available volume ( $V_a$ ) [14]. Tables 2, 4, 6 and 8 and Figure 9 shows that the values of available volume ( $V_a$ ) increase with the increase in concentration of 1-Bromopropane. The increase in available volume is caused due to the net packing of molecules inside the shell which may be due to the complexation between like molecules though hydrogen bonding in the binary mixture. The relaxation time ( $\tau$ ) decreases with increasing concentration for all the four temperatures presented in Tables 2, 4, 6 and 8 and Figure 10. The dispersion of the ultrasonic velocity in the system should contain information about the characteristic time  $\tau$  of the relaxation process that causes dispersion. The relaxation time which is in the order of  $10^{-12}$  sec is due to structural relaxation process [15] and in such a situation it is suggested that the molecules get rearranged due to co-operative process.

Internal Pressure ( $\Pi_i$ ) is a fundamental property of a liquid, which provides an excellent basis for examining the solution phenomenon and studying various properties of the liquid state. It is a measure of the change in the internal energy of liquid or liquid mixtures, as it undergoes a very small isothermal change. It is a measure of cohesive or binding forces between the solute and solvent molecules. Also it is observed from Tables 2, 4, 6 and 8 and Figure 11 that internal pressure decreases with increase in mole fraction of 1-Bromopropane and also decreases with temperature. The Gibbs free energy ( $\Delta G^*$ ) increases with the increase in temperature and decrease with increase in concentration of 1-Bromopropane presented in Figure. 12. An increasing value of  $\Delta G^*$  suggests that the more distant of unlike molecules is due to hydrogen bonding and longer time for the rearrangement of molecules in the mixture [16].

**Table 1. Values of Velocity ( $U$ ), Density ( $\rho$ ), Viscosity ( $\eta$ ), Adiabatic Compressibility ( $\beta_{ad}$ ), Intermolecular Free length ( $L_f$ ) and Acoustic Impedance ( $Z$ ) at 303.15K with Frequency of 2MHz for Molefraction 1-Bromopropane with Chlorobenzene**

$X_1$	$U$ ( $m.s^{-1}$ )	$\rho$ ( $Kg.m^{-3}$ )	$\eta$ ( $10^{-3} Ns.m^{-2}$ )	$\beta_{ad}$ ( $10^{-10} m^2.N^{-1}$ )	$L_f$ ( $10^{-10} m$ )	$Z$ ( $10^6 Kg.m^2.s^{-1}$ )
0.0000	1249.20	1095.1	0.7151	5.8517	0.5019	1.3680
0.1836	1198.65	1137.8	0.6747	6.1171	0.5132	1.3638
0.3599	1149.22	1180.1	0.6346	6.4161	0.5256	1.3562
0.5290	1100.10	1222.3	0.5950	6.7602	0.5395	1.3447
0.6921	1051.23	1261.1	0.5547	7.1755	0.5558	1.3257
0.8488	1002.44	1299.5	0.5145	7.6579	0.5742	1.3027
1.0000	954.47	1337.1	0.4745	8.2094	0.5945	1.2762

**Table 2. Values of Molar Volume ( $V_m$ ), Free Volume ( $V_f$ ), Available Volume ( $V_a$ ), Relaxation Time ( $\tau$ ), Internal Pressure ( $\Pi_i$ ) and Gibbs Free Energy ( $\Delta G^*$ ) at 303.15K with Frequency of 2MHz for Molefraction 1-Bromopropane with Chlorobenzene**

$X_1$	$V_m$ ( $10^{-7} \text{ m}^3.\text{mol}^{-1}$ )	$V_f$ ( $10^{-7} \text{ m}^3.\text{mol}^{-1}$ )	$V_a$ ( $10^{-6} \text{ m}^3.\text{mol}^{-1}$ )	$\tau$ ( $10^{-12} \text{ s}$ )	$\pi_i$ ( $10^6 \text{ N.m}^{-2}$ )	$\Delta G^*$ ( $10^{-20} \text{ KJ.mol}^{-1}$ )
0.0000	102.7851	3.1128	2.2536	0.5579	338.98	0.4659
0.1836	100.6037	3.2740	2.5236	0.5503	338.13	0.4657
0.3599	98.5527	3.4508	2.7766	0.5429	336.84	0.4655
0.5290	96.5960	3.6413	3.0180	0.5363	335.32	0.4653
0.6921	94.9746	3.8608	3.2574	0.5307	332.57	0.4651
0.8488	93.4321	4.1077	3.4894	0.5253	329.34	0.4650
1.0000	91.9901	4.3937	3.7114	0.5163	325.39	0.4648

**Table 3. Values of Velocity ( $U$ ), Density ( $\rho$ ), Viscosity ( $\eta$ ), Adiabatic Compressibility ( $\beta_{ad}$ ), Intermolecular Free length ( $L_f$ ) and Acoustic Impedance ( $Z$ ) at 308.15K with Frequency of 2MHz for Molefraction 1-Bromopropane with Chlorobenzene**

$X_1$	$U$ ( $\text{m.s}^{-1}$ )	$\rho$ ( $\text{Kg.m}^{-3}$ )	$\eta$ ( $10^{-3} \text{ Ns.m}^{-2}$ )	$\beta_{ad}$ ( $10^{-10} \text{ m}^2.\text{N}^{-1}$ )	$L_f$ ( $10^{-10} \text{ m}$ )	$Z$ ( $10^6 \text{ Kg.m}^2.\text{s}^{-1}$ )
0.0000	1229.02	1089.9	0.6776	6.0743	0.5163	1.3395
0.1836	1179.13	1132.1	0.6395	6.3532	0.5281	1.3349
0.3599	1130.12	1174.1	0.6020	6.6688	0.5410	1.3269
0.5290	1081.34	1215.7	0.5647	7.0348	0.5557	1.3146
0.6921	1032.73	1254.1	0.5266	7.4764	0.5728	1.2951
0.8488	984.28	1291.9	0.4888	7.9898	0.5922	1.2716
1.0000	936.31	1328.7	0.4511	8.5849	0.6138	1.2441

**Table 4. Values of Molar Volume ( $V_m$ ), Free Volume ( $V_f$ ), Available Volume ( $V_a$ ), Relaxation Time ( $\tau$ ), Internal Pressure ( $\Pi_i$ ) and Gibbs Free Energy ( $\Delta G^*$ ) at 308.15K with Frequency of 2MHz for Molefraction 1-Bromopropane with Chlorobenzene**

$X_1$	$V_m$ ( $10^{-7} \text{ m}^3.\text{mol}^{-1}$ )	$V_f$ ( $10^{-7} \text{ m}^3.\text{mol}^{-1}$ )	$V_a$ ( $10^{-6} \text{ m}^3.\text{mol}^{-1}$ )	$\tau$ ( $10^{-12} \text{ s}$ )	$\pi_i$ ( $10^6 \text{ N.m}^{-2}$ )	$\Delta G^*$ ( $10^{-20} \text{ KJ.mol}^{-1}$ )
0.0000	103.2755	3.2933	2.3946	0.5488	337.09	0.4737
0.1836	101.1081	3.4615	2.6596	0.5417	336.26	0.4734
0.3599	99.0531	3.6419	2.9089	0.5353	335.17	0.4732
0.5290	97.1171	3.8378	3.1482	0.5297	333.73	0.4730
0.6921	95.5020	4.0640	3.3860	0.5249	331.10	0.4729
0.8488	93.9801	4.3158	3.6166	0.5207	328.02	0.4728
1.0000	92.5717	4.6053	3.8399	0.5163	324.25	0.4726

**Table 5. Values of Velocity ( $U$ ), Density ( $\rho$ ), Viscosity ( $\eta$ ), Adiabatic Compressibility ( $\beta_{ad}$ ), Intermolecular Free length ( $L_f$ ) and Acoustic Impedance ( $Z$ ) at 313.15K with Frequency of 2MHz for Molefraction 1-Bromopropane with Chlorobenzene**

$X_1$	$U$ ( $\text{m.s}^{-1}$ )	$\rho$ ( $\text{Kg.m}^{-3}$ )	$\eta$ ( $10^{-3} \text{Ns.m}^{-2}$ )	$\beta_{ad}$ ( $10^{-10} \text{m}^2.\text{N}^{-1}$ )	$L_f$ ( $10^{-10} \text{m}$ )	$Z$ ( $10^6 \text{Kg.m}^2.\text{s}^{-1}$ )
0.0000	1214.29	1084.7	0.6319	6.2524	0.5289	1.3171
0.1836	1164.57	1126.5	0.5976	6.5454	0.5411	1.3119
0.3599	1115.49	1167.9	0.5639	6.8812	0.5548	1.3028
0.5290	1066.49	1208.9	0.5305	7.2727	0.5704	1.2893
0.6921	1017.72	1246.8	0.4959	7.7437	0.5885	1.2689
0.8488	969.04	1283.9	0.4617	8.2944	0.6091	1.2441
1.0000	920.46	1319.9	0.4275	8.9423	0.6325	1.2159

**Table 6. Values of Molar Volume ( $V_m$ ), Free Volume ( $V_f$ ), Available Volume ( $V_a$ ), Relaxation Time ( $\tau$ ), Internal Pressure ( $\pi_i$ ) and Gibbs Free Energy ( $\Delta G^*$ ) at 313.15K with Frequency of 2MHz for Molefraction 1-Bromopropane with Chlorobenzene**

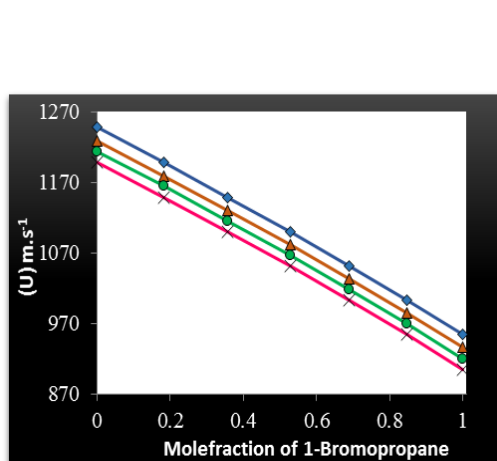
$X_1$	$V_m$ ( $10^{-7} \text{m}^3.\text{mol}^{-1}$ )	$V_f$ ( $10^{-7} \text{m}^3.\text{mol}^{-1}$ )	$V_a$ ( $10^{-6} \text{m}^3.\text{mol}^{-1}$ )	$\tau$ ( $10^{-12} \text{s}$ )	$\pi_i$ ( $10^6 \text{N.m}^{-2}$ )	$\Delta G^*$ ( $10^{-20} \text{KJ.mol}^{-1}$ )
0.0000	103.7706	3.5914	2.5016	0.5393	331.74	0.4809
0.1836	101.6081	3.7610	2.7652	0.5318	331.30	0.4807
0.3599	99.5752	3.9392	3.0153	0.5257	330.65	0.4806
0.5290	97.6594	4.1280	3.2564	0.5209	329.77	0.4805
0.6921	96.0578	4.3504	3.4958	0.5168	327.65	0.4804
0.8488	94.5638	4.5924	3.7291	0.5133	325.17	0.4803
1.0000	93.1889	4.8657	3.9578	0.5101	322.10	0.4801

**Table 7. Values of Velocity ( $U$ ), Density ( $\rho$ ), Viscosity ( $\eta$ ), Adiabatic Compressibility ( $\beta_{ad}$ ), Intermolecular Free length ( $L_f$ ) and Acoustic Impedance ( $Z$ ) at 318.15K with Frequency of 2MHz for Molefraction 1-Bromopropane with Chlorobenzene**

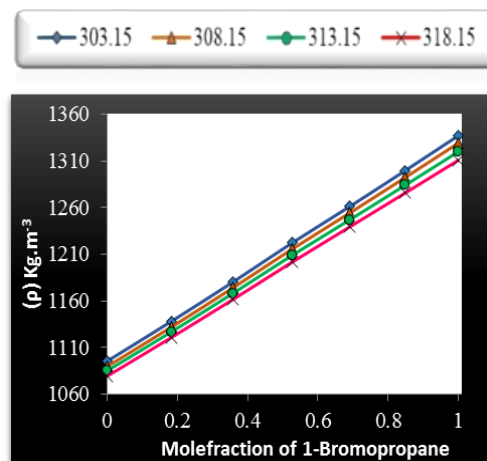
$X_1$	$U$ ( $\text{m.s}^{-1}$ )	$\rho$ ( $\text{Kg.m}^{-3}$ )	$\eta$ ( $10^{-3} \text{Ns.m}^{-2}$ )	$\beta_{ad}$ ( $10^{-10} \text{m}^2.\text{N}^{-1}$ )	$L_f$ ( $10^{-10} \text{m}$ )	$Z$ ( $10^6 \text{Kg.m}^2.\text{s}^{-1}$ )
0.0000	1198.09	1079.5	0.5904	6.4535	0.5424	1.2933
0.1836	1149.06	1120.9	0.5601	6.7569	0.5550	1.2880
0.3599	1100.33	1161.8	0.5307	7.1092	0.5693	1.2784
0.5290	1051.44	1202.1	0.5012	7.5247	0.5857	1.2639
0.6921	1002.67	1239.6	0.4705	8.0242	0.6048	1.2429
0.8488	954.15	1275.9	0.4399	8.6089	0.6264	1.2174
1.0000	905.29	1311.1	0.4095	9.3066	0.6513	1.1869

**Table 8. Values of Molar Volume ( $V_m$ ), Free Volume ( $V_f$ ), Available Volume ( $V_a$ ), Relaxation Time ( $\tau$ ), Internal Pressure ( $\Pi_i$ ) and Gibbs Free Energy ( $\Delta G^*$ ) at 318.15K with Frequency of 2MHz for Molefraction 1-Bromopropane with Chlorobenzene**

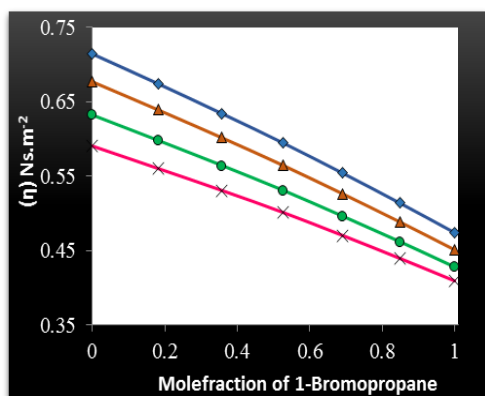
$X_1$	$V_m$ ( $10^{-7} \text{ m}^3 \cdot \text{mol}^{-1}$ )	$V_f$ ( $10^{-7} \text{ m}^3 \cdot \text{mol}^{-1}$ )	$V_a$ ( $10^{-6} \text{ m}^3 \cdot \text{mol}^{-1}$ )	$\tau$ ( $10^{-12} \text{ s}$ )	$\pi_i$ ( $10^6 \text{ N} \cdot \text{m}^{-2}$ )	$\Delta G^*$ ( $10^{-20} \text{ KJ} \cdot \text{mol}^{-1}$ )
0.0000	104.2705	3.8974	2.6192	0.5298	327.00	0.4888
0.1836	102.1132	4.0623	2.8779	0.5229	326.97	0.4886
0.3599	100.0941	4.2267	3.1259	0.5177	326.93	0.4884
0.5290	98.2077	4.4002	3.3670	0.5137	326.76	0.4883
0.6921	96.6124	4.6031	3.6068	0.5102	325.42	0.4882
0.8488	95.1547	4.8244	3.8410	0.5080	323.63	0.4881
1.0000	93.8144	5.0622	4.0734	0.5058	321.51	0.4880



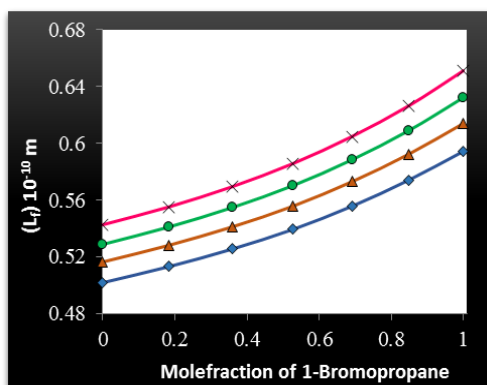
**Figure 1. Variation of Velocity with Molefraction of 1-Bromopropane**



**Figure 2. Variation of Density with Molefraction of 1-Bromopropane**



**Figure 3. Variation of Viscosity with Molefraction of 1-Bromopropane**



**Figure 4. Variation of Adiabatic Compressibility with Molefraction of 1-Bromopropane**

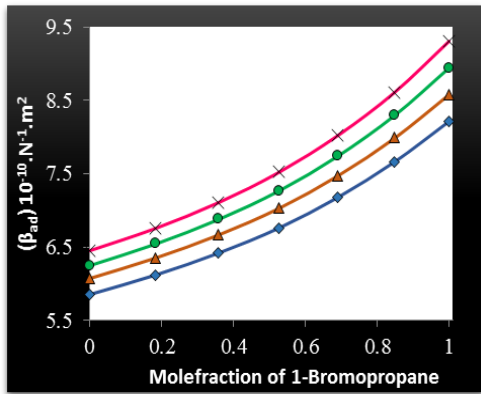


Figure 5. Variation of Intermolecular Free Length with Molefraction of 1-Bromopropane

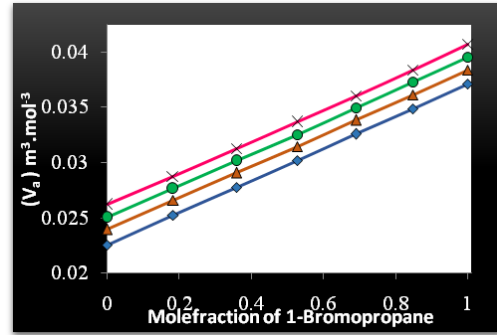


Figure 9. Variation of Available Volume with Molefraction of 1-Bromopropane

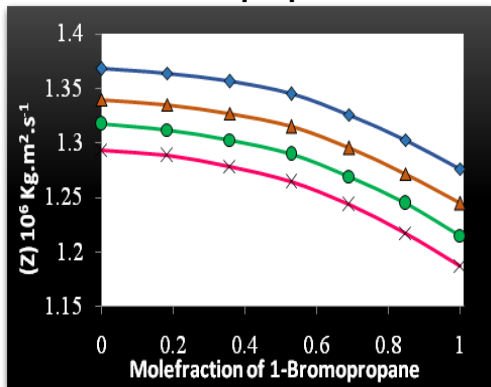


Figure 6. Variation of Molar Volume with Molefraction of 1-Bromopropane

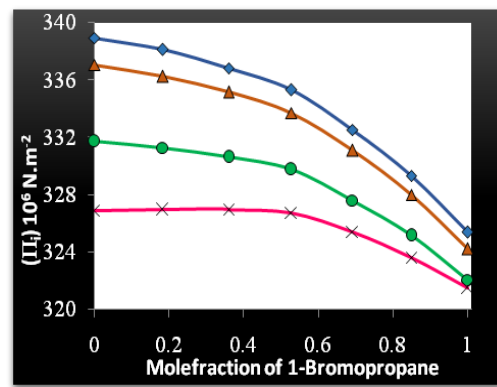


Figure 11. Variation of Internal Pressure with Molefraction of 1-Bromopropane

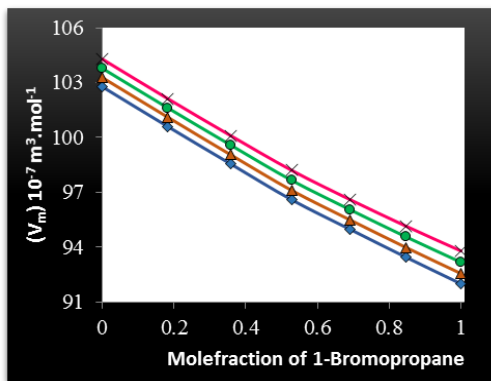


Figure 7. Variation of Acoustic Impedance with Molefraction of 1-Bromopropane

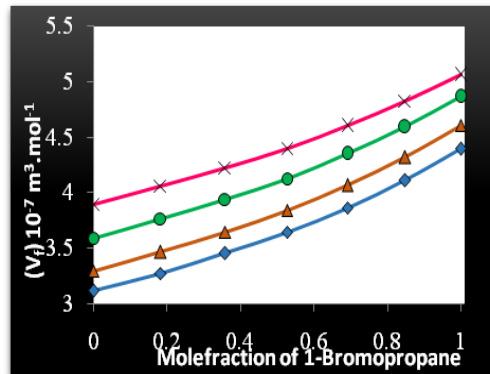
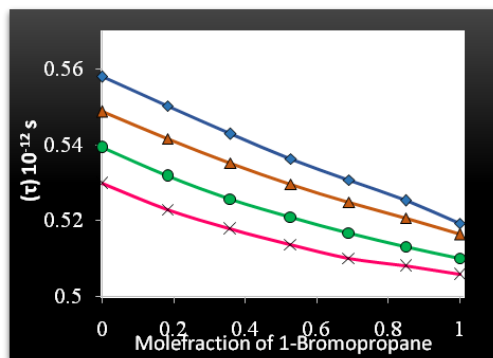
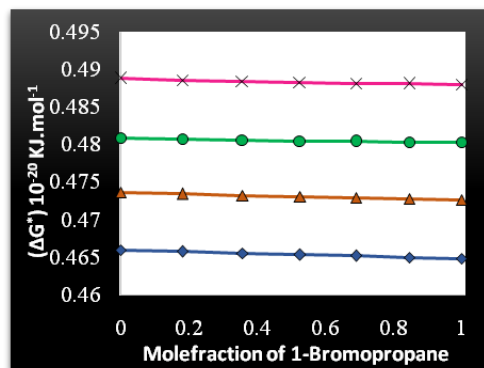


Figure 8. Variation of Free Volume with Molefraction of 1-Bromopropane





**Figure 10. Variation of Relaxation Time with Molefraction of 1-Bromopropane**



**Figure 12. Variation of Gibbs free energy with Molefraction of 1-Bromopropane**

#### 4. Conclusion

From the observed experimental values of ultrasonic velocity, density, viscosity and related acoustical parameters values for the binary liquid mixtures of 1-Bromopropane and Chlorobenzene system at temperatures 303.15K, 308.15K, 313.15K and 318.15K, it is clear that there exists a strong intermolecular association between the component molecules of the liquid mixtures.

#### Acknowledgement

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