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STUDIES ON ULTRASONIC VELOCITIES OF BINARY LIQUID MIXTURE OF METHYL ACRYLATE AND BENZENE AND SUBSTITUTED HALO BENZENES AT 313.15K

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ABSTRACT- In this study, binary liquid mixtures of Methyl Acrylate (MA) with Benzene (B), as well as Substituted Halo Benzene molecules like Chloro Benzene (CB) and Bromo Benzene (BB), were investigated at 313.15K. The initial measurements encompassed the determination of Density (ρ) and Ultrasonic Velocity (u) for the mixtures. Subsequently, the acquired data was employed to compute isentropic compressibility (K_S), Intermolecular free length (L_f), acoustic impedance (Z), and relative association (R_A). Utilizing these Ultrasonic parameters, excess properties, including excess ultrasonic Velocity (u^E), excess acoustic impedance (Z^E), excess isentropic compressibility (K_S^E), and excess inter-molecular free-length (LfE), were determined. These excess parameter values were fitted into the Redlich–Kister Polynomial equation, leading to a comprehensive discussion on the intermolecular interactions involved in the studied mixtures.

Keywords: Ultrasonic Velocity, Density, Methyl Acrylate, Benzene, Chloro Benzene, Bromo Benzene, excess ultrasonic velocity, excess isentropic compressibility, excess inter molecular free-length, excess acoustic impedance, Redlich-Kister equation.

1. INTRODUCTION

Ultrasonic studies serve as highly effective techniques for comprehending the molecular behavior and intermolecular interactions within mixtures of liquid solvents. The properties of the Ultrasonic Velocity in binary liquid mixtures play a crucial role in the design of process simulation equipment, contribute to solution theory, and serve as a key parameter in calculating the thermodynamic properties of substances as well as molecular dynamics.

The excess or deviation properties of liquid-liquid mixtures play a crucial role in various industrial applications due to their impact on the efficiency of operations. In contemporary times, the assessment of ultrasonic velocity and its associated acoustical parameters has garnered significant interest from numerous researchers [1-4].

This research is specifically concentrated on Methyl Acrylate (MA) with Benzene (B) and Substituted Halo Benzene molecules like Chloro Benzene (CB) and Bromo Benzene (BB) at 313.15K.

MA holds a crucial role as an industrial solvent, significantly contributing to the commercial synthesis of technologically important high polymeric and latex compounds. With a relative polarity characterized by a dipole moment (μ) of 1.77 D (where 1 D = 3.334×10^{-3} Cm) [5], MA is classified as an aprotic and associated liquid. In contrast, aromatic hydrocarbons possess substantial quadrupole moments, inducing orientational order among molecules through the partial alignment of neighboring segments or entire molecules [6].

Acrylates, including MA, have diverse applications in sectors such as leather, textiles, adhesives, paints, antioxidant agents, inks, amphoteric surfactants, paper, detergents, surface coatings, and more. Liquid mixtures containing B and halo benzenes are utilized to investigate polymer phase diagrams and preferential interactions in mixed media. Consequently, combinations of MA with B and substituted halo benzene prove highly valuable in various chemical and industrial applications [7].

MA's electron-donor ability, attributed to lone electron pairs, interacts with aromatic rings that function as electron acceptors. This interaction may involve charge-transfer interactions, potentially influenced by the presence of halo groups on the ring. CB finds major applications as an intermediate in the production of commodities such as herbicides, dyestuffs, and rubber, while BB can be employed to prepare the corresponding Grignard reagent and serves as an ingredient in the manufacture of phenylcyclidine [8].

In the current scientific research, there has been a notable emphasis on both speculative and experimental investigations into the thermo-physical properties of solutions. The Redlich–Kister polynomial equation is employed to discern the standard deviation

between speculative and untested values of liquid solutions. These assessments incorporate various theoretical mixing rules. Following a comprehensive examination, the interaction behavior of MA with B and Substituted Halo Benzenes like CB and BB at 313.15K was studied. The objective of this study was to understand the impact of the addition of Chloro and Bromo groups to the benzene molecule, potentially influencing both the sign and magnitude of excess properties when these components are mixed with MA at 313.15K [9].

2. EXPERIMENTAL METHODS

2.1. Materials and procedure:

MA, B, and mono-substituted halo Benzenes like CB, and BB were purchased from E-Merck and used as purchased. Mixtures were prepared by mixing weighed amounts of the pure liquids adopting the method of the closed system by using Mettler balance with the precision of ± 0.1 mg. Mixtures were allowed to stand for some time before every measurement, to avoid air bubbles. The purity of the liquids was checked by comparing the values of densities and ultrasonic velocities with literature data (Table 1). The measurements were made with proper care in an AC room to avoid evaporation loss [5-7].

Table 1: Comparison of experimental density and ultrasonic velocity of pure liquids with literature values at 313.15 K.

Liquid	Density (ρ) x 10^{-3} Kg m^{-3}		Ultrasonic Velocity (u) m s^{-1}	
	Exptl.	Lit.	Exptl.	Lit.
Benzene	0.8574	0. 8576 [10]	1236.2	1236.3 [13]
Chloro Benzene	1.0847	1.0848 [11]	1208.9	1208.93 [11]
Bromo Benzene	1.4666	1.4665 [11]	1044.8	1044.80 [11]
Methyl Acrylate	0.9292	0.9292 [12]	1117.80	1117.81 [14]

The densities (ρ) of both individual liquids and their mixtures were determined utilizing a bi-capillary pycnometer with a capillary diameter of 0.85 mm, calibrated with double-distilled water. Buoyancy corrections were applied with precision, and the recorded Density values exhibited consistency within an error margin of ± 0.2 Kg m^{-3}. Ultrasonic Velocity (u) measurements were carried out using a single-frequency (2 MHz) variable path interferometer, maintaining an accuracy of ± 0.03%. The experiments took place in a thermostatically controlled water bath, utilizing a Schott Gerate (Model CT 050/2), ensuring a well-stirred and constant temperature environment controlled to within ± 0.02 K for all measurements [15, 16].

3. RESULTS AND DISCUSSIONS

The results obtained from experimental measurements of densities and Ultrasonic Velocity of binary mixtures are outlined in Table 2. The various acoustical parameters such as K_S, Z, L_f, and R_A were calculated using the following equations.

From the measured density (ρ) and Ultrasonic Velocity (u), the

$K_s = 1/u^2 \rho$ … (1)

$Z = \rho u$ … (2)

$L_f = K(K_s)^{1/2}$ … (3)

Where 'K' is Jacobson's constant [17].

The excess functions Y^E have been calculated using the relation:

$Y^E = Y_{mix} - (X_1Y_1 + X_2Y_2)$ … (4)

Where Y denotes u, Z, K_S, and L_f, respectively, X is the mole fraction and suffixes 1 & 2 denote the components 1 & 2 in the binary mixture, and the values are given in Table 2.

The dependence of u^E, Z^E, K_S^E, and L_f^E on the mole fraction of MA (X_{MA}) for all three systems were fitted to the following Redlich-Kister equation by the least-squares method and the values are given in Table 2.

$$Y^E = x(1-X)\sum_i A_i(2x-1)^i \quad \ldots \quad (5)$$

When Y^E is considered, the parameters include u^E, Z^E, K_S^E, and L_f^E. The values for the parameters Ai, which are obtained through a procedure of fitting a linear least squares polynomial, are also provided in Table 3 along with their corresponding standard deviations (σ) values.

As shown in Table 2, it is noted that the values of u, Z, K_s, and L_f exhibit changes corresponding to the mole fraction of MA (X_{MA}). This observation strongly suggests the presence of interactions between the components within these binary liquid mixtures. The excess functions, which serve as indicators of deviations from ideal behavior, are found to be relatively more responsive to the intermolecular interactions among the dissimilar molecules in the mixture compared to the pure acoustical parameters [18].

With this perspective in mind, the variations in excess ultrasonic parameters such as excess ultrasonic velocity, excess acoustic impedance, excess isentropic compressibility, and excess intermolecular free length are carefully examined concerning the mole fraction of MA (X_{MA}), as illustrated in Figures 1 to 4 respectively.

Table 2: Values of Density (ρ), Ultrasonic Velocity (u), excess ultrasonic velocity (u^E), excess acoustic impedance (Z^E), excess isentropic compressibility (K_S^E), and excess intermolecular free-length (L_f^E) for the binary liquid mixtures of MA with B and Substituted Halo Benzenes like CB and BB at 313. 15 K.

Mole fraction of MA (X_{MA})	ρ x 10^{-3} Kg m^{-3}	u m s^{-1}	u^E m s^{-1}	Z^E X 10^{-4} Kg m^{-2} s^{-1}	K_S^E x 10^{11} m^2 N^{-1}	L_f^E x 10^{12} m
MA + B						
0.0000	0.8574	1236.2	0.0000	0.0000	0.0000	0.0000
0.0916	0.8646	1224.4	-0.9969	0.0586	-0.0614	-0.0139
0.1849	0.8718	1212.5	-1.7890	0.1039	-0.1107	-0.0247
0.2800	0.8789	1200.7	-2.3705	0.1360	-0.1472	-0.0326
0.3769	0.8861	1188.8	-2.7352	0.1549	-0.1706	-0.0374
0.4757	0.8933	1177.0	-2.8768	0.1609	-0.1803	-0.0392
0.5764	0.9005	1165.2	-2.7889	0.1540	-0.1756	-0.0378
0.6792	0.9077	1153.3	-2.4644	0.1343	-0.1559	-0.0332
0.7840	0.9148	1141.5	-1.8965	0.1020	-0.1206	-0.0254
0.8909	0.9220	1129.6	-1.0776	0.0572	-0.0689	-0.0144
1.0000	0.9292	1117.8	0.0000	0.0000	0.0000	0.0000
MA + CB						
0.0000	1.0847	1208.9	0.0000	0.0000	0.0000	0.0000
0.1268	1.0692	1199.8	2.4463	0.6045	-1.0307	-0.3408
0.2463	1.0536	1190.7	4.2230	1.0369	-1.8134	-0.5960
0.3591	1.0381	1181.6	5.3866	1.3143	-2.3580	-0.7703
0.4657	1.0225	1172.5	5.9876	1.4516	-2.6727	-0.8676
0.5666	1.0070	1163.4	6.0708	1.4623	-2.7642	-0.8915
0.6623	0.9914	1154.2	5.6767	1.3586	-2.6373	-0.8450
0.7531	0.9759	1145.1	4.8415	1.1511	-2.2958	-0.7307
0.8395	0.9603	1136.0	3.5977	0.8498	-1.7418	-0.5506
0.9217	0.9448	1126.9	1.9750	0.4634	-0.9766	-0.3065
1.0000	0.9292	1117.8	0.0000	0.0000	0.0000	0.0000
MA + BB						
0.0000	1.4666	1044.8	0.0000	0.0000	0.0000	0.0000
0.1685	1.4129	1052.1	4.9997	3.7345	-2.5090	-0.9209
0.3132	1.3591	1059.4	8.2597	6.2139	-4.3177	-1.5758
0.4387	1.3054	1066.7	10.1248	7.6715	-5.5216	-2.0029
0.5487	1.2516	1074.0	10.8542	8.2825	-6.1856	-2.2292
0.6459	1.1979	1081.3	10.6468	8.1814	-6.3518	-2.2730
0.7323	1.1442	1088.6	9.6574	7.4731	-6.0435	-2.1464
0.8097	1.0904	1095.9	8.0087	6.2403	-5.2685	-1.8559
0.8794	1.0367	1103.2	5.7989	4.5496	-4.0198	-1.4036
0.9426	0.9829	1110.5	3.1076	2.4548	-2.2760	-0.7872
1.0000	0.9292	1117.8	0.0000	0.0000	0.0000	0.0000

Table 3: Parameters of Eq. (5) and Standard deviations						
Excess Property	**A_0**	**A_1**	**A_2**	**A_3**	**A_4**	**σ**
MA + B						
U^E m s^{-1}	0.00003	-12.09339	13.31966	-1.33494	0.10863	0.00006
Z^E X 10^{-4} Kg m^{-2} s^{-1}	-0.000002	0.71952	-0.88663	0.18963	-0.02252	0.00001
K_S^E x 10^{11} m^2 N^{-1}	0.000002	-0.74217	0.78667	-0.05241	0.00791	0.00003
L_f^E x 10^{12} m	-0.00001	-0.16866	0.19452	-0.02861	0.00277	0.00003
MA + CB						
U^E m s^{-1}	-0.00041	21.48132	-16.77026	-2.92431	-1.78552	0.00100
Z^E X 10^{-4} Kg m^{-2} s^{-1}	-0.00001	5.33382	-4.36644	-0.73976	-0.22754	0.00009
K_S^E x 10^{11} m^2 N^{-1}	0.00093	-8.9552	6.2243	0.36386	2.36443	0.00213
L_f^E x 10^{12} m	0.00021	-2.96863	2.12574	0.26472	0.57761	0.00047
MA + BB						
U^E m s^{-1}	-0.00496	33.75071	-23.90876	5.76375	-15.58449	0.01765
Z^E X 10^{-4} Kg m^{-2} s^{-1}	0.00511	25.24156	-18.30559	7.73265	-14.64674	0.01823
K_S^E x 10^{11} m^2 N^{-1}	0.01509	-18.02893	20.40302	-29.8807	27.43756	0.03634
L_f^E x 10^{12} m	0.00446	-6.50515	6.67616	-8.80688	8.61569	0.01652

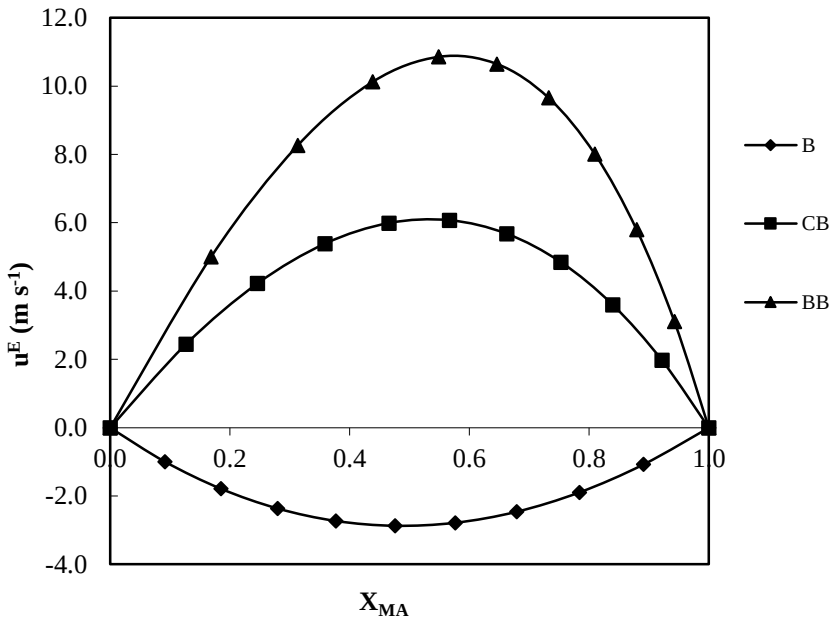

Fig:1: Plots of excess ultrasonic velocities (u^E) for various B (-♦-) and Substituted Halo Benzenes like CB (-■-), and BB (-▲-) vs mole fraction of MA (X_{MA}) at 313.15 K.

It is observed from Fig. 1 that excess ultrasonic velocity is positive for MA+ CB, and MA+BB liquid mixtures whereas the MA+B liquid mixture is slightly negative. In general, if the media is dense, the ultrasonic velocity value will be higher and if the media is less dense, the ultrasonic velocity value will be less. When we mix two liquids if they condense or compress more ultrasonic velocity will be more. For these mixtures, since the excess volume (V^E) values are negative [19], this indicates the mixtures compressed more and it is natural to get positive excess ultrasonic velocities for these mixtures.

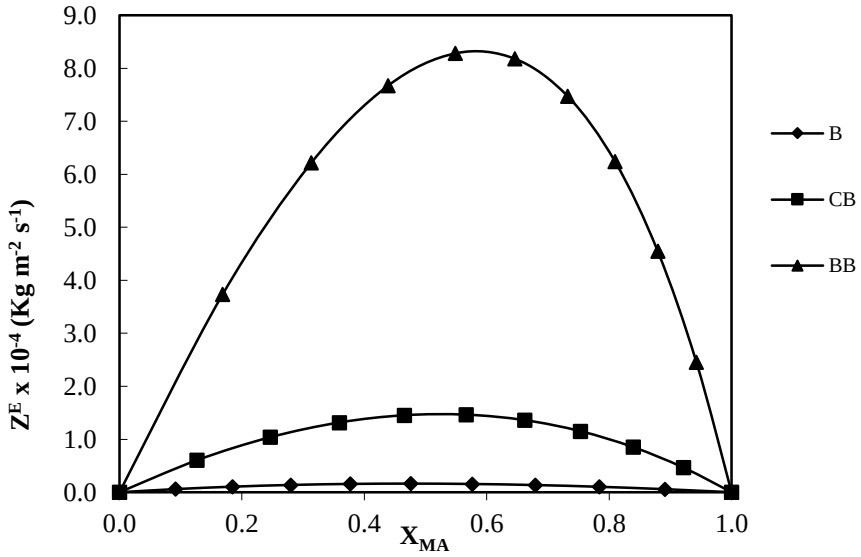

Fig: 2: Plots of excess acoustic impedance (Z^E) for various B (-♦-) and Substituted Halo Benzenes like CB (-■-), and BB (-▲-) vs mole fraction of MA (X_{MA}) at 313.15 K.

Fig. 2 shows a variation of Z^E with a composition of liquid mixtures which exhibit positive deviations as expected as per equation (2) for Z^E calculation.

The positive deviations in u^E and Z^E (Figs. 1 and 2) for all the systems under study are observed over the entire range of composition. These trends for these systems again support our view that the interactions between unlike molecules are quite possible and these values are in the following order:

[MA + BB] > [MA + CB] > [MA + B]

Similar observation was reported by Eswari Bai et al., [20] from their ultrasonic velocity studies of binary liquid mixtures of butoxy ethanol with some amines. Also, same observations were reported by Vijaya Lakshmi et al. [21] from their Intermolecular interactions in binary mixtures of methyl acrylate with benzene and mono-substituted benzenes at 313.15K.

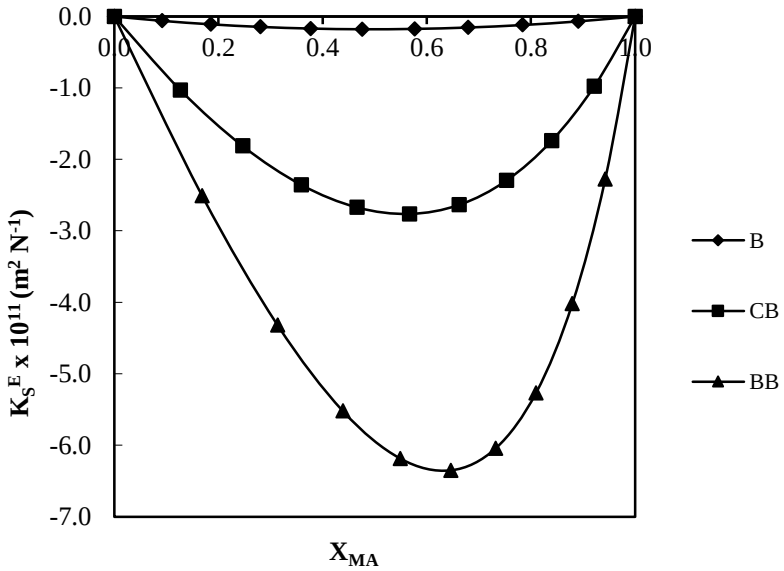

Fig: 3: Plots of Excess Isentropic Compressibility (K_s^E) for various B (-♦-) and Substituted Halo Benzenes like CB (-■-), and BB (-▲-) vs mole fraction of MA (X_{MA}) at 313.15 K.

Figures 3 and 4 show that both K_s^E and L_f^E consistently exhibit negative values across the entire mole fraction range for all systems. Particularly, these negative values reach their maximum at 0.56 mole fraction of MA. This observation indicates that the negative excess isentropic compressibility and intermolecular free length are associated with the existence of molecular interactions within the systems. These interactions are likely facilitated through electron donor-acceptor interactions, ultimately resulting in the formation of complexes between molecules of different natures.

Previous studies by Ali et al. [22] and Vijaya Lakshmi et al. [2] have emphasized that the negative values of excess intermolecular free length (L_f^E) signify that sound waves cover longer distances. This phenomenon arises from a reduction in intermolecular free length, which is a consequence of stronger donor-acceptor interactions between MA, B, and Halo-substituted benzene molecules. Consequently, this leads to the manifestation of large positive values for both K_s^E and L_f^E.

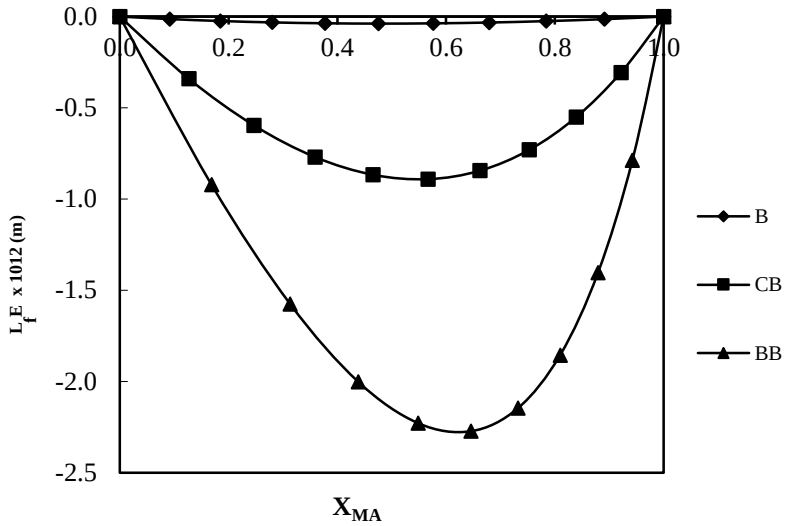

Fig: 4: Plots of excess inter-molecular free-length (L_f^E) for various B (-♦-) and Substituted Halo Benzenes like CB (-■-), and BB (-▲-) vs mole fraction of MA (X_{MA}) at 313.15 K.

In general, the values of K_s^E are influenced by two primary factors. Firstly, an increase in free length, as defined by Jacobson [17], occurs due to the loss of dipolar association, the breaking up of hydrogen bonding [23], and variations in the size and shapes of the component molecules. Secondly, a decrease in free length results from dipole-dipole interactions, hydrogen bonding association [23], and the formation of complexes between the component molecules [24, 25]. The first effect contributes to an expansion of the intermolecular space within mixtures, causing sound waves to cover shorter distances compared to pure components. Consequently, this leads to a negative deviation in the Ultrasonic Velocity and a positive deviation in isentropic compressibility. On the other hand, the second effect results in a reduction of intermolecular space within mixtures, causing sound waves to cover longer distances than in pure components. This leads to a positive deviation in Ultrasonic Velocity and a negative deviation in isentropic compressibility. The actual values of K_s^E depend on the delicate balance between these two opposing effects. Experimental findings suggest that, in these mixtures, the negative contributions tend to predominate. The negative values obtained for the K_s^E and L_f^E are in the following order:

[MA + BB] > [MA + CB] > [MA + B]

Similar behavior has been reported for binary mixtures of cyclohexane with benzene, containing different halogen atoms [26, 27].

4. CONCLUSION:

From the experimentation and above discussions, we can draw the following conclusions,

1) The ultrasonic velocity method is a strong tool for studying the physical and chemical properties and molecular interactions in mixtures. Density, Ultrasonic Velocity, and related parameters confirm these interactions.
2) The properties of the studied mixtures indicate that interactions between unlike molecules are predominant, characterized by positive u^E and Z^E, and negative K_s^E and L_f^E for all systems, following a specific order.
   [MA + BB] > [MA + CB] > [MA + B]
3) Hydrogen bonding between unlike molecules of MA and B, as well as substituted halo benzene molecules, leads to the formation of complex structures between these components.
4) MA molecules fit into B and substituted halo benzene molecules, owing to differences in their size and shape.

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