

# **RESEARCH ARTICLE**

## ULTRASONIC STUDIES ON BINARY LIQUID MIXTURES OF METHYL ACRYLATE WITH 2-ALKOXY ETHANOLS AT 308.15 K.

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Manuscript Info	Abstract
Manuscript History	
	Densities and ultrasonic velocities of binary liquid mixtures of Methyl
Received: 12 June 2016	Acrylate with 2-Alkoxy Ethanols [2-Methoxy Ethanol, 2-Ethoxy
Final Accepted: 19 July 2016	Ethanol, 2-Butoxy Ethanol] have been measured at 308.15 K. The
Published: August 2016	observed data have been utilised to calculate various acoustical
	parameters like Isentropic compressibility, Intermolecular free length,
Key words:-	Acoustic impedance and Relative association. The various excess

Ultrasonic velocity, Methyl acrylate, 2-Alkoxy Ethanols, Excess isentropic com pressibility, Excess acoustic impedance and Redlich Kister equation.

properties like Excess ultrasonic velocity, Excess acoustic impedance, Excess isentropic compressibility and Excess Inter molecular freelength were calculated and fitted to the Redlich Kister equation.

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#### Introduction:-

In recent years, density and ultrasonic velocity measurements in pure liquids and liquid mixtures have been successfully employed in understanding the nature of intermolecular interactions. Physico chemical and thermodynamic investigations play an important role to understand the nature and extent of the aggregation of molecules that exists in the binary liquid mixtures and their sensitivities to the variations in composition and the molecular structure of the pure components [1-2].

The ultrasonic velocity measurement is an excellent tool to investigate inter and intermolecular interactions between liquid mixtures of non-electrolytes. Density and ultrasonic velocity properties of binary liquid mixtures are essential in process simulation, equipment design, solution theory, important parameter in calculations of the thermodynamic properties of matter and molecular dynamics. Acoustic properties are useful in understanding the molecular interactions. The propagation of sound in real liquids is accompanied by irreversible loss of energy resulting in the absorption of sound by medium. The irreversibility of the process of propagation of sound waves in a liquid is associated primarily with its viscosity and thermal conductivity.

The present research aims to measure densities and ultrasonic velocity of binary liquid mixtures of methyl acrylate (MA) with 2-Alkoxy Ethanols [2-Methoxy Ethanol (2ME), 2-Ethoxy Ethanol (2EE), 2-Butoxy Ethanol (2BE)] at 308.15 K, by using this data various acoustical parameters like Acoustic impedance (Z), Isentropic compressibility  $(K_s)$ , Inter molecular free-length  $(L_f)$  and Relative association  $(R_A)$  and also various excess properties like excess ultrasonic velocity  $(u^{E})$ , excess acoustic impedance  $(Z^{E})$ , excess isentropic compressibility  $(K_{s}^{E})$  and excess inter molecular free-length  $(L_f^E)$  were calculated and fitted to the Redlich Kister equation.

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## Material and methods:-

Methyl acrylate, 2-Alkoxy Ethanols [2-Methoxy Ethanol (2ME), 2-Ethoxy Ethanol (2EE), 2-Butoxy Ethanol (2BE)] of E-Merck make were used in the present investigation. Mixtures were prepared by mixing weighed amounts of the pure liquids adopting the method of closed system by using Mettler balance with the precision of  $\pm 0.1$  mg. Mixtures were allowed to stand for some time before every measurement so as to avoid air bubbles. The purities of the liquids were checked by comparing the values of densities and ultrasonic velocities with literature data and are given in Table 1.

The measurements were made with proper care in an Air Conditioned room to avoid evaporation loss. The densities of liquids and their mixtures were measured using bicapillary pycnometer having a capillary diameter of 0.85 mm, which was calibrated using double distilled water. The necessary buoyancy corrections were applied. The density (p) values were reproducible within  $\pm$  0.2 Kg m<sup>-3</sup>. The ultrasonic velocity (u) measurements were made by a single frequency (2 MHz) variable path interferometer with an accuracy of  $\pm$  0.03%. A thermostatically controlled, well stirred constant temperature water bath, Schott Gerrate (Model CT 050/2 made in Germany) whose temperature was controlled to  $\pm 0.02$  K was used for all the measurements.

**Table 1:** Comparison of Experimental densities ( $\rho$ ) and ultrasonic velocities (u) of pure liquids with literature at 308.15 K

Liquid	Density K	y(ρ) x 10 <sup>-3</sup> g m <sup>-3</sup>	Ultrasonic Velocity (u) ms <sup>-1</sup>		
	EXP	LIT	EXP	LIT	
Methyl Acrylate	0.9367	0.93565[3]	1141.0	1140.0 [5 & 6]	
2-methoxy ethanol	0.9529	0.9511[4]	1342.0	1318.8(4)	
2-ethoxy ethanol	0.9163	0.9164[4]	1295.0	1272.4(4)	
2-butoxy ethanol	0.8887	0.8887[4]	1279.0	1260.8(4)	

#### **Results and discussions:-**

From the measured density ( $\rho$ ) and ultrasonic velocity (u) the various acoustical parameters such as  $K_s$ , Z,  $L_f$  and  $R_A$ were calculated using the following equations 1, 2, 3 & 4 respectively and are incorporated in Table 2 for the binary systems under study.

$$K_s = 1/u^2 \rho \qquad \dots (1)$$
  
$$Z = \rho u \qquad \dots (2)$$

$$= K(K_s)^{1/2}$$
 ... (3)

$$L_{f=}K(K_{s})^{1/2} \qquad \dots (3)$$
  
Where 'K' is Jacobson's constant [7].  
$$R_{A} = \rho / \rho_{o} (u_{o}/u)^{1/3} \qquad \dots (4)$$

The excess functions  $Y^E$  have being calculated using the relation:  $Y^E = Y_{mix} - (X_1Y_1 + X_2Y_2)$ ... (5)

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 binary mixture and the values are given in Table 3.

The dependence of  $u^{E}$ ,  $Z^{E}$ ,  $K_{S}^{E}$  and  $L_{f}^{E}$  on the mole fraction of methyl acrylate (X<sub>MA</sub>) for all the three systems was fitted to the following Redlich-Kister equation by the least-squares polynomial method and the values are given in Table 4.

$$Y^{E} = x(1-X)\sum_{i} A_{i}(2x-1)^{i} \qquad \dots (6)$$

Where  $Y^E$  is  $u^E$ ,  $Z^E$ ,  $K_S^E$  and  $L_f^E$  excess parameters.

The parameters A<sub>i</sub>, obtained by a linear least squares polynomial fitting procedure, are also given in Table 3 together with the standard deviations ( $\sigma$ ) values.

From the Table 2, it is clear that the values of u, Z,  $K_s$ ,  $L_f$  and  $R_A$  varied with the mole fraction of MA (X<sub>MA</sub>). This indicates the presence of interactions between the components in these binary liquid mixtures. The variations of ultrasonic velocity for the mixtures depend on the values of  $L_{\rm f}$ . It is observed that, decrease in ultrasonic velocity and the corresponding increase in  $L_f$  with mole fraction of MA (Table 2) for all the systems are in accordance with the view proposed by Eyring and Kincaid [8]. However, the excess functions which are a measure of the deviations from the ideal behaviour are relatively more sensitive to the intermolecular interactions between the unlike molecules of the mixture than the pure acoustical parameters as explained above.

The positive deviations in  $u^E$  and  $Z^E$  (Figs. 1 to 2) for all the systems of MA and 2-Alkoxy Ethanols are observed over the entire range of composition except 2-methoxy ethanol. These trends for these systems again support the view that, the interactions between unlike molecules are quite possible and these values are in the following order.

#### $MA{+}2BE > MA{+}2EE > MA{+}2ME$

In general if the media is dense, the ultrasonic velocity value will be more and if the media is less dense, the ultrasonic velocity value will be less. When two liquids are mixed, if they condense or compress more, the ultrasonic velocity value will be positive [9]. From Fig. 1 it is clear that  $u^E$  values are positive for all the systems except for 2-Methoxy Ethanol. This indicates that the mixtures are compressed more and it is natural to get positive excess ultrasonic velocities. In Fig 2, Excess acoustic impedance ( $Z^E$ ) with composition of liquid mixtures exhibited positive deviations as per the equation 2. A similar observation was reported by Eswari Bai et al [10], Vijaya lakshmi et al [11] and Sastry et al [12].

From the Figs. 3 & 4 that  $K_s^E$  and  $L_f^E$  are negative for all the systems over the whole mole fraction range. In general,  $K_s^E$  and  $L_f^E$  values depend upon three factors.

(i) Mutual disruption of associated present in the pure liquids, defined by Jacobson [7]

(ii) Formation of weak H- bonding by dipole-induced dipole interaction between unlike molecules and

(iii) Geometrical complex fitting formation between the component molecules [13].

First factor contributes to positive  $K_S^E$  values, whereas the remaining two factors lead to negative  $K_S^E$  values. The resultant values for the present mixtures are due to the net effect of above all the three factors [14].

The negative deviations of  $K_S^E$  and  $L_f^E$  values indicated that the presence of strong interactions between unlike molecules, which may be resulted in complex formation. The polar nature of the two unlike molecules leads to formation of H-bonding between the oxygen atom of >C=O group of MA with hydrogen atom of –OH group of 2-Alkoxy Ethanols [>C=O......H-O] and also expected intermolecular H-bonding between 2-Alkoxy Ethanol molecules which may be due to complex formation.

2-Alkoxy Ehanol with the interstitial accommodation of smaller sized MA molecules into interstitial space of bigger size 2-Alkoxy Ethanol molecules. Thus, the complex formation between the two component molecules lead to a decrease in the intermolecular distance thereby decreasing the isentropic compressibilities of the mixtures. The negative values obtained for the  $K_s^E$  and  $L_f^E$  are in the following order:

MA + 2BE > MA + 2EE > MA + 2ME

Further, it is also observed from the experimental results that the negative contributions increase with increase in chain length of 2-Alkoxy ethanols. The change in magnitude of negative values of  $K_S^E$  and  $L_f^E$  from 2-Methoxy Ethanol (2ME), 2-Ethoxy Ethanol (2EE), 2-Butoxy Ethanol (2BE) shows that, increase in chain length of 2-Alkoxy ethanols lead to less compressibility in homologous series. Further the data suggests that the strength of complex formation between unlike molecules increase from 2-Methoxy Ethanol (2ME) to 2-Butoxy Ethanol (2BE). This conclusion is supported by Fort and Moore [15], Mahendranath Roy [16], Douhéreta [17] and Sastry et al [18].

# **Conclusions:-**

From the above discussion, following conclusions are drawn.

- 1. The observed variation of the properties of the mixtures studied support the view that the interactions between unlike molecules is predominant and characterised by the positive  $u^E$  and  $Z^E$  for all mixtures except for 2-methoxy ethanol.
- 2. The Excess isentropic compressibility and Excess Inter molecular free-length values are negative for all the mixtures. The negative contributions increase with increase in chain length of 2-Alkoxy ethanols.
- 3. The possible intermolecular interactions of hydrogen bonding between unlike molecules of Methyl Acrylate and 2-Alkoxy Ethanols and inter H-bonding between 2-Alkoxy Ethanols molecules which leads to complex formations between the component molecules.

- 4. Interstitial accommodation of MA molecules into the 2-Alkoxy Ethanol molecules is due to their differences in size and shape.
- **Table 2:-** Values of density ( $\rho$ ), ultrasonic velocity (u), acoustic impedance (Z), isentropic compressibility (K<sub>S</sub>),Inter molecular free-length (L<sub>f</sub>) and relative association (R<sub>A</sub>) for the binary liquid mixtures of MethylAcrylate (MA) with 2-Alkoxy Ethanols at 308.15 K.

Mole fraction of	ρ x 10 <sup>-3</sup> Kg m <sup>-3</sup>	u m s <sup>-1</sup>	Z x 10 <sup>-4</sup> Kg m <sup>-2</sup> s <sup>-1</sup>	$\frac{K_{\rm S}  x  10^{11}}{m^2  {\rm N}^{-1}}$	L <sub>f</sub> x 10 <sup>12</sup> m	Relative association	
MA (X <sub>MA</sub> )	0					( <b>R</b> <sub>A</sub> )	
Methyl Acrylate (MA) + 2-Methoxy Ethanol (2ME)							
0.0000	0.9529	1342.0	1.2788	58.2704	5.0555	1.0000	
0.0894	0.9513	1322.0	1.2576	60.1490	5.1364	1.0033	
0.1810	0.9497	1302.0	1.2365	62.1169	5.2197	1.0067	
0.2747	0.9480	1282.0	1.2154	64.1796	5.3057	1.0102	
0.3708	0.9464	1262.0	1.1944	66.3433	5.3944	1.0138	
0.4692	0.9448	1242.0	1.1734	68.6147	5.4859	1.0174	
0.5700	0.9432	1222.0	1.1526	71.0008	5.5805	1.0212	
0.6734	0.9416	1202.0	1.1318	73.5094	5.6782	1.0251	
0.7795	0.9399	1182.0	1.1110	76.1491	5.7793	1.0290	
0.8883	0.9383	1162.0	1.0903	78.9290	5.8838	1.0331	
1.0000	0.9367	1142.0	1.0697	81.8592	5.9920	1.0373	
	Met	hyl Acryla	te (MA) + 2-Et	hoxy Ethanol	(2EE)		
0.0000	0.9163	1295.0	1.1866	65.0763	5.3426	1.0000	
0.1042	0.9183	1279.7	1.1752	66.4936	5.4005	1.0062	
0.2074	0.9204	1264.4	1.1637	67.9616	5.4598	1.0125	
0.3097	0.9224	1249.1	1.1522	69.4827	5.5205	1.0189	
0.4110	0.9245	1233.8	1.1406	71.0595	5.5828	1.0253	
0.5114	0.9265	1218.5	1.1289	72.6948	5.6467	1.0319	
0.6109	0.9285	1203.2	1.1172	74.3916	5.7122	1.0385	
0.7095	0.9306	1187.9	1.1054	76.1529	5.7794	1.0452	
0.8072	0.9326	1172.6	1.0936	77.9822	5.8484	1.0521	
0.9040	0.9347	1157.3	1.0817	79.8830	5.9193	1.0590	
1.0000	0.9367	1142.0	1.0697	81.8592	5.9920	1.0660	
	Met	hyl Acryla	te (MA) + 2-Bu	itoxy Ethanol	(2BE)		
0.0000	0.8887	1279.0	1.1366	68.7866	5.4928	1.0000	
0.1323	0.8935	1265.3	1.1305	69.9066	5.5373	1.0090	
0.2555	0.8983	1251.6	1.1243	71.0636	5.5830	1.0181	
0.3704	0.9031	1237.9	1.1179	72.2592	5.6297	1.0273	
0.4779	0.9079	1224.2	1.1115	73.4949	5.6777	1.0366	
0.5785	0.9127	1210.5	1.1048	74.7726	5.7268	1.0460	
0.6731	0.9175	1196.8	1.0981	76.0941	5.7772	1.0555	
0.7621	0.9223	1183.1	1.0912	77.4613	5.8289	1.0651	
0.8459	0.9271	1169.4	1.0842	78.8764	5.8819	1.0748	
0.9251	0.9319	1155.7	1.0770	80.3416	5.9362	1.0846	
1.0000	0.9367	1142.0	1.0697	81.8592	5.9920	1.0946	

Mole fraction of	u <sup>E</sup> m s <sup>-1</sup>	Z <sup>E</sup> X 10 <sup>-4</sup> Kg m <sup>-2</sup> s <sup>-1</sup>	$\frac{{{K_S}^E} x{10^{11}}}{{m^2}{N^{-1}}}$	L <sub>f</sub> <sup>E</sup> x 10 <sup>12</sup> m			
MA (X <sub>MA</sub> )	$\frac{MA(A_{MA})}{Methyl Acrylate (MA) + 2-Methoxy Ethanol (2ME)}$						
0.0000	-2.1154		-0.2307	0.0000			
0.0894		-0.2503		-0.0290			
0.1810	-3.8054	-0.4497	-0.4224	-0.0529			
0.2747	-5.0548	-0.5965	-0.5712	-0.0713			
0.3708	-5.8472	-0.6890	-0.6729	-0.0838			
0.4692	-6.1660	-0.7256	-0.7229	-0.0898			
0.5700	-5.9933	-0.7043	-0.7160	-0.0887			
0.6734	-5.3104	-0.6232	-0.6468	-0.0799			
0.7795	-4.0978	-0.4802	-0.5090	-0.0627			
0.8883	-2.3349	-0.2733	-0.2959	-0.0363			
1.0000	0.0000	0.0000	0.0000	0.0000			
		MA) + 2-Etho					
0.0000	0.0000	0.0000	0.0000	0.0000			
0.1042	0.6416	0.0771	-0.3313	-0.0980			
0.2074	1.1353	0.1367	-0.5958	-0.1755			
0.3097	1.4832	0.1789	-0.7911	-0.2321			
0.4110	1.6873	0.2038	-0.9150	-0.2673			
0.5114	1.7496	0.2117	-0.9648	-0.2807			
0.6109	1.6720	0.2027	-0.9378	-0.2717			
0.7095	1.4563	0.1768	-0.8311	-0.2397			
0.8072	1.1046	0.1343	-0.6415	-0.1842			
0.9040	0.6185	0.0753	-0.3657	-0.1045			
1.0000	0.0000	0.0000	0.0000	0.0000			
		MA) + 2-Buto	xy Ethanol (2				
0.0000	0.0000	0.0000	0.0000	0.0000			
0.1323	4.4308	0.2757	-0.6100	-0.2153			
0.2555	7.6038	0.4767	-1.0630	-0.3739			
0.3704	9.6454	0.6094	-1.3695	-0.4798			
0.4779	10.6657	0.6789	-1.5384	-0.5369			
0.5785	10.7611	0.6902	-1.5771	-0.5483			
0.6731	10.0159	0.6472	-1.4918	-0.5165			
0.7621	8.5049	0.5536	-1.2876	-0.4440			
0.8459	6.2938	0.4127	-0.9688	-0.3327			
0.9251	3.4415	0.2273	-0.5387	-0.1842			
1.0000	0.0000	0.0000	0.0000	0.0000			

**Table 3:-** Values of excess ultrasonic velocity ( $u^E$ ), excess acoustic impedance ( $Z^E$ ), excess isentropiccompressibility ( $K_S^E$ ) and excess intermolecular free-length ( $L_f^E$ ) for the binary liquid mixturesof Methyl Acrylate (MA) + 2-Alkoxy Ethanols at 308.15 K.

**Table 4:-** Parameters of Eq. (6) and Standard deviations

Excess Property	$\mathbf{A_0}$	$A_1$	$A_2$	$A_3$	$A_4$	σ		
Methyl Acrylate (MA) + 2-Methoxy Ethanol (2ME)								
$U^E m s^{-1}$	0.000194	-26.2787	29.69434	-3.78305	0.36772	0.000377		
Z <sup>E</sup> X 10 <sup>-4</sup> Kg m <sup>-2</sup> s <sup>-1</sup>	0.000017	-3.11401	3.56742	-0.50819	0.05479	0.000043		
$K_{s}^{E} x 10^{11} m^{2} N^{-1}$	-0.000009	-2.81752	2.64154	0.16524	0.01075	0.000036		
$L_{f}^{E} x 10^{12} m$	0.000018	-0.35443	0.34199	0.01154	0.00092	0.000026		
Methyl Acrylate (MA) + 2-Ethoxy Ethanol (2EE)								
$U^E m s^{-1}$	0.000011	6.84109	-6.53079	-0.30159	-0.00877	0.000078		

$Z^{E} X 10^{-4} \text{Kg m}^{-2} \text{ s}^{-1}$	0.000001	0.82070	-0.76919	-0.05043	-0.00109	0.000036		
$K_{\rm S}^{E} \ge 10^{11} {\rm m}^2 {\rm N}^{-1}$	0.000041	-3.48037	2.82118	0.49136	0.16777	0.000070		
$L_{f}^{E} x 10^{12} m$	0.0000003	-1.03289	0.87263	0.12763	0.03263	0.000016		
Methyl Acrylate (MA) + 2-Butoxy Ethanol (2BE)								
$U^E m s^{-1}$	0.000933	37.32526	-28.05933	-4.9196	-4.34371	0.00228		
$Z^{E} X 10^{-4} \text{Kg m}^{-2} \text{ s}^{-1}$	-0.000103	2.30973	-1.64395	-0.25808	-0.40741	0.00025		
$K_{\rm S}^{\rm E} \ge 10^{11} {\rm m}^2 {\rm N}^{-1}$	0.000510	-5.10003	3.54555	0.14277	1.41024	0.00124		
$L_{f}^{E} x 10^{12} m$	0.000145	-1.80274	1.27136	0.11476	0.41621	0.00034		

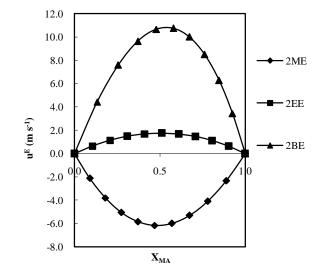
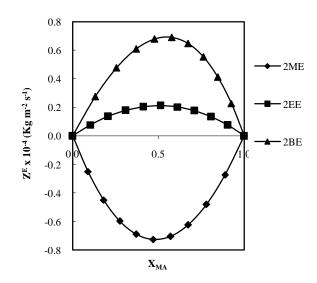


Fig: 1:- Plots of Excess Ultrasonic velocities (u<sup>E</sup>) vs mole fraction of Methyl Acrylate (X<sub>MA</sub>) for binary mixtures of Methyl Acrylate (X<sub>MA</sub>) with 2-Methoxy Ethanol (2ME, -♦-), 2-Ethoxy Ethanol (2EE, -■-), 2-Butoxy Ethanol (2BE, -▲-), at 308.15 K.



**Fig: 2:-** Plots of Excess acoustic impedance ( $Z^E$ ) vs mole fraction of Methyl Acrylate ( $X_{MA}$ ) for binary mixtures of Methyl Acrylate ( $X_{MA}$ ) with 2-Methoxy Ethanol (2ME,  $\neg \bullet \neg$ ), 2-Ethoxy Ethanol (2EE,  $\neg \bullet \neg$ ), 2-Butoxy Ethanol (2BE,  $\neg \bullet \neg$ ) at 308.15 K.

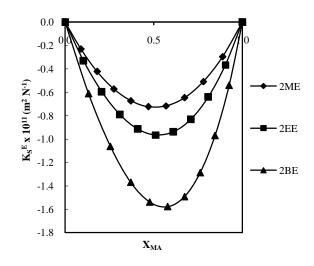


Fig: 3:- Plots of Excess isentropic compressibility (K<sup>E</sup><sub>s</sub>) vs mole fraction of Methyl Acrylate (X<sub>MA</sub>) for binary mixtures of Methyl Acrylate (X<sub>MA</sub>) with 2-Methoxy Ethanol 2ME, -♦-), 2-Ethoxy Ethanol (2EE, -■-), 2-Butoxy Ethanol (2BE, -▲-) at 308.15 K.

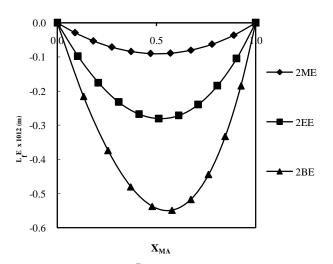


Fig: 4:- Plots of Excess intermolecular free-length (L<sub>f</sub><sup>E</sup>) vs mole fraction of Methyl Acrylate (X<sub>MA</sub>) for binary mixtures of Methyl Acrylate (X<sub>MA</sub>) with 2-Methoxy Ethanol (2ME, -♦-), 2-Ethoxy Ethanol (2EE, -■-), 2-Butoxy Ethanol (2BE, -▲-) at 308.15 K.

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