

## Viscosity Studies of Binary Liquid Mixture of Methyl Acrylate and Benzene & Substituted Halo Benzenes at 313.15K

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### Abstract

Studied the binary liquid mixtures of Methyl Acrylate with Benzene (B), and Substituted Halo Benzene molecules like Chloro Benzene (CB) and Bromo Benzene (BB) at 313.15K. Initially, the density and viscosities of the mixtures are measured and this data is utilized to calculate excess properties like excess molar volume (VE), excess viscosity ( $\eta E$ ), and excess Gibbs free energy ( $\Delta G^*E$ ). These excess parameter values are fitted in the Redlich–Kister Polynomial equation and the intermolecular interaction is discussed.

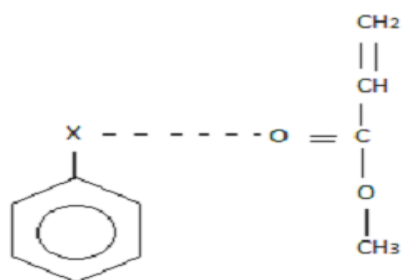
**Keywords:** Methyl acrylate, Benzene, Chloro Benzene, Bromo Benzene, density, viscosity, Thermochemical and Excess properties, Redlich–Kister Polynomial equation, Molecular interaction.

### 1. Introduction

Comprehending the volumetric properties of non-electrolyte solutions is essential for exploring heat transfer, mass transfer, chemical separation, and fluid dynamics. Similarly, understanding thermo-physical properties is crucial for understanding the characteristics of the [1-3] liquid state and the intermolecular bonds within liquid structures. Carbon plays a pivotal role in advancing our scientific community. Our investigation focuses on Methyl Acrylate (MA) with Benzene (B), as well as Substituted Halo Benzene molecules such as Chloro Benzene (CB) and Bromo Benzene (BB) at [4] a temperature of 313.15K. Methyl Acrylate serves as a crucial industrial solvent, playing a significant role in the industrial manufacturing of technologically significant high-polymer and latex compounds. Characterized by its relative polarity (with a dipole moment,  $\mu$ , of 1.77 D at 298.15 K, where 1 D = 3.334  $\times 10^{-30}$  Cm) [5], MA is an aprotic and associated liquid. On the other hand, aromatic hydrocarbons exhibit substantial quadrupole moments [6], inducing orientational order among molecules due to the partial alignment of neighbouring segments or entire molecules [6]. Acrylates, including Methyl Acrylate (MA), are extensively utilized across various industries, including leather, textiles, adhesives, paints, antioxidant agents, inks, amphoteric surfactants, paper, detergents, surface coatings, and

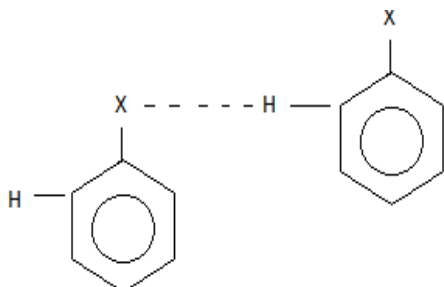
other sectors. A Schematic Representation of H-bond Formation between Halo benzenes and MA Shown in Figure 1 [7]. Liquid mixtures containing aromatic hydrocarbons are employed in studying polymer phase diagrams and preferential interactions in mixed media [8]. Consequently, mixtures of MA with aromatic hydrocarbons prove highly valuable in numerous chemical and industrial applications. The electron-donor ability of MA, attributed to lone electron pairs [9], interacts with aromatic rings functioning as electron acceptors [10]. This interaction could entail charge-transfer interactions, which might be affected by the presence of halo groups on the ring. [11-13]. Chloro Benzene is crucial in the production of various products such as herbicides, dyestuffs, and rubber, while Bromo Benzene is used to create the corresponding Grignard reagent and is a vital ingredient in phencyclidine manufacturing. [12, 13]. In the current scientific landscape, there has been notable attention towards speculative and experimental examinations of the thermo-physical properties of solutions. The Redlich–Kister polynomial equation is employed to scrutinize the variability in speculative and untested liquid solution values, incorporating diverse theoretical mixing rules. This investigation delves into the interaction behavior of Methyl Acrylate (MA) with Benzene (B) and Substituted Halo

Benzenes like Chloro benzene (CB) and Bromo benzene (BB) at 313.15K. A Schematic Representation of H-bond Formation between Halo Benzenes Shown in Figure.2. The primary objective is to comprehend the effects of introducing Chlorine and Bromo groups to the benzene molecule, potentially impacting both the direction and magnitude of excess properties when combined with Methyl Acrylate at 313.15K. [14-17].



Where X=Cl or Br

**Figure 1** A Schematic Representation of H-bond Formation between Halo benzenes and MA



Where X=Cl or Br

**Figure 2** A Schematic Representation of H-bond Formation between Halo Benzenes

## 2. Experimental Procedures

### 2.1 Materials

The experimental liquids employed in this study were of analytical grade, obtained from Merck, and used without further purification. The experimental liquid purities were as follows [18]. Methyl acrylate (99.5%), Benzene (99%), Chloro Benzene (99%), and Bromo Benzene (99%). The purities of the liquids were verified by comparing their density and viscosity values with those reported in the literature. (Table 1).

**Table 1** Comparing the experimental density and viscosity of pure liquids with literature values at 313.15 K.

Liquid	Density ( $\rho$ ) x $10^{-3}$ Kg m $^{-3}$		Viscosity ( $\eta$ ) x $10^{-3}$ Kg m $^{-1}$ s $^{-1}$	
	Exptl.	Lit.	Exptl.	Lit.
Benzene	0.8574	0.8576 [18]	0.4981	0.4980 [19]
Chloro Benzene	1.0847	1.0848 [12]	0.6340	0.6341 [12]
Bromo Benzene	1.4666	1.4665 [12]	0.8736	0.8736 [12]
Methyl Acrylate	0.9292	0.9292 [19]	0.3551	0.3550 [19]

### 2.2 Apparatus and procedure

Experimental liquid mixtures were prepared by accurately combining measured quantities of these pure liquids within a closed system [19]. The measurements were conducted using a Mettler balance, providing a precision of  $\pm 0.1$  mg, and the precision in the mole fraction was determined to be within  $\pm 1 \times 10^{-4}$ . To ensure accuracy, mixtures were allowed to settle before each measurement to eliminate any potential air bubbles. The densities of both individual pure liquids and their binary mixtures were determined using a Borosilicate glass single-capillary pycnometer, featuring a bulb capacity of approximately 10 mL. The capillary, marked with graduations, was calibrated using triply distilled water. The precision of density measurements was within  $\pm 2 \times 10^{-5}$  g cm $^{-3}$ . Viscosity measurements were conducted with a precision of  $\pm 0.2\%$  using a calibrated Schott-Gerate AVS 400 viscometer. The flow time of doubly distilled water through the instrument was recorded as 375 seconds at 25°C. Throughout the experiments, the temperature of the test liquids was maintained with a precision of  $\pm 0.01$  K using an electronically regulated thermostatic water bath. All measurements were carried out meticulously in a controlled environment to minimize any potential evaporation loss [20, 21].

### 3. Results and Discussions

Table 2 displays the experimental results of density and viscosity measurements for binary mixtures. The excess volume (VE), excess viscosity ( $\eta_E$ ), and excess Gibbs free energy of activation of flow ( $G^*E$ ) were computed from the collected data using the following equations:

$$V = (X_1M_1 + X_2M_2) / \rho \quad (1)$$

$$VE = V - (X_1 V_1 + X_2 V_2) \quad (2)$$

$$\eta_E = \eta - (X_1 \eta_1 + X_2 \eta_2) \quad (3)$$

$$G^*E = RT[\ln \eta V - (X_1 \ln \eta_1 V_1 + X_2 \ln \eta_2 V_2)] \quad (4)$$

In the equations,  $\rho$ ,  $\eta$ , and  $V$  represent the density, viscosity, and molar volume of the mixture, respectively.  $M_1$  and  $M_2$  denote the molar masses,  $\eta_1$  and  $\eta_2$  are the viscosities, and  $V_1$  and  $V_2$  are the molar volumes of the benzene and methyl acrylate, respectively. The variable  $x$  represents the mole fraction of methyl acrylate. The relationship between  $\eta_E$ ,  $VE$ , and  $G^*E$  with the mole fraction of methyl acrylate (XMA) for all three systems was modeled using the Redlich-Kister equation via the least-squares method.

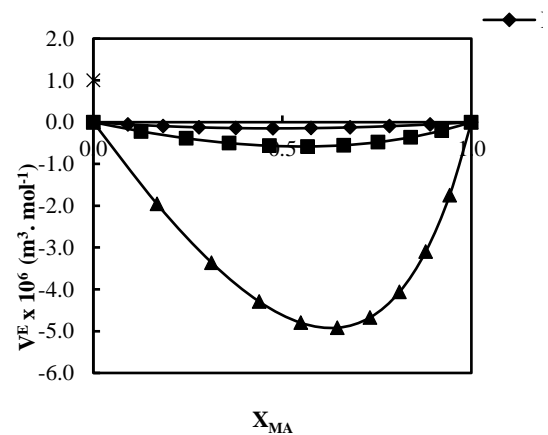
$$Y^E = x(1-X) \sum_i A_i (2x-1)^i \quad (6)$$

Where  $Y^E$  is  $\eta_E$ ,  $VE$ , and  $G^*E$  parameters.

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

Here,  $k$  represents the number of experimental points excluding the endpoints, while  $n$  denotes the order of the polynomial equation. The values of  $F(x)$  calculated are derived from Eq. (6) using the best-fit values of  $A_i$  coefficients. Table 3 lists the coefficients  $A_0$ ,  $A_1$ ,  $A_2$ ,  $A_3$ , and  $A_4$ , along with their corresponding standard deviations ( $\sigma$ ) for all three mixtures. Notably, the observed standard deviations are very small. Experimental densities and viscosities of pure Methyl Acrylate, Benzene, Chloro Benzene, and Bromo Benzene, as well as their binary mixtures, are utilized to calculate excess thermodynamic properties. These properties represent the deviation from ideal behavior and provide insights into molecular interactions within the mixtures.

Specifically, they depict interactions among solute-solute, solute-solvent, and solvent-solvent species. The anticipated effects between the component molecules include (I) disruption of hydrogen bonds and dipolar interactions in Methyl Acrylate and intermolecular hydrogen bonding in Benzene, (II) accommodation of one component molecule within another due to differences in size, and (III) potential intermolecular interactions, such as hydrogen bonding, between unlike molecules, which may weaken, strengthen, or establish during mixing. [22]. The measured density ( $\rho$ ) and viscosity ( $\eta$ ) at 313.15 K for mixtures of Methyl Acrylate (MA) with Benzene (B), Chloro Benzene (CB), and Bromo Benzene (BB) are used to calculate excess parameters  $VE$ ,  $\eta_E$ , and  $G^*E$ , as presented in Tables 2. The variations of these excess parameters with the mole fraction of Methyl Acrylate (XMA) for the three systems are depicted graphically in Figures 3, respectively. From Figure 3, it is evident that the negative values of excess volume ( $VE$ ) for all three systems indicate the presence of strong molecular interactions between the unlike components of the mixtures. These negative  $VE$  values follow a specific sequence.  $(MA + B) < (MA + CB) < (MA + BB)$



**Figure 3** Plots of excess volumes ( $VE$ ) for Benzene and Substituted Halo Benzene molecules like Benzene (-.-), Chloro Benzene (-■-), and Bromo Benzene (-▲-) vs mole fraction of Methyl acrylate (XMA) at 313.15 K

**Table 2 Values of density ( $\rho$ ), viscosity ( $\eta$ ), excess viscosity ( $\eta^E$ ), molar volume ( $V$ ), excess volume ( $V^E$ ), and excess Gibbs free energy of activation of viscous flow ( $G^{*E}$ ) for the binary liquid mixtures of Methyl Acrylate (MA) + Benzene, Chloro Benzene, and Bromo Benzene at 313.15 K**

Mole fraction of MA ( $X_{MA}$ )	$\rho \times 10^{-3}$ Kg m <sup>-3</sup>	$\eta \times 10^3$ Kg m <sup>-1</sup> s <sup>-1</sup>	$\eta^E \times 10^3$ Kg m <sup>-1</sup> s <sup>-1</sup>	$V \times 10^6$ m <sup>3</sup> mol <sup>-1</sup>	$V^E \times 10^6$ m <sup>3</sup> mol <sup>-1</sup>	$G^{*E} \times 10^3$ N mol <sup>-1</sup>
<b>Methyl Acrylate (MA) + Benzene (B)</b>						
0.0000	0.8574	0.4981	0.0000	91.1010	0.0000	0.0000
0.0916	0.8646	0.4838	-0.0012	91.1897	-0.0531	0.7907
0.1849	0.8718	0.4695	-0.0022	91.2928	-0.0945	1.4827
0.2800	0.8789	0.4552	-0.0029	91.4104	-0.1242	2.0527
0.3769	0.8861	0.4409	-0.0033	91.5425	-0.1422	2.4752
0.4757	0.8933	0.4266	-0.0035	91.6893	-0.1483	2.7210
0.5764	0.9005	0.4123	-0.0034	91.8510	-0.1426	2.7576
0.6792	0.9077	0.3980	-0.0030	92.0277	-0.1251	2.5484
0.7840	0.9148	0.3837	-0.0023	92.2196	-0.0955	2.0516
0.8909	0.9220	0.3694	-0.0013	92.4268	-0.0538	1.2200
1.0000	0.9292	0.3551	0.0000	92.6496	0.0000	0.0000
<b>Methyl Acrylate (MA) + Chloro Benzene (CB)</b>						
0.0000	1.0847	0.6340	0.0000	103.7706	0.0000	0.0000
0.1268	1.0692	0.6061	0.0075	102.1394	-0.2205	16.5820
0.2463	1.0536	0.5782	0.0129	100.6447	-0.3863	29.4216
0.3591	1.0381	0.5503	0.0165	99.2767	-0.5002	38.6266
0.4657	1.0225	0.5224	0.0183	98.0270	-0.5644	44.2585
0.5666	1.0070	0.4946	0.0186	96.8881	-0.5811	46.3319
0.6623	0.9914	0.4667	0.0174	95.8533	-0.5519	44.8119
0.7531	0.9759	0.4388	0.0148	94.9168	-0.4782	39.6090
0.8395	0.9603	0.4109	0.0110	94.0736	-0.3611	30.5714
0.9217	0.9448	0.3830	0.0060	93.3192	-0.2015	17.4737
1.0000	0.9292	0.3551	0.0000	92.6496	0.0000	0.0000
<b>Methyl Acrylate (MA) + Bromo Benzene (BB)</b>						
0.0000	1.4666	0.8736	0.0000	107.0571	0.0000	0.0000
0.1685	1.4129	0.8218	0.0355	102.6713	-1.9582	44.7244
0.3132	1.3591	0.7699	0.0587	99.1822	-3.3630	76.1992
0.4387	1.3054	0.7181	0.0719	96.4442	-4.2921	96.6904
0.5487	1.2516	0.6662	0.0771	94.3527	-4.7989	107.7410
0.6459	1.1979	0.6144	0.0756	92.8335	-4.9183	110.3432
0.7323	1.1442	0.5625	0.0686	91.8355	-4.6709	105.0349
0.8097	1.0904	0.5107	0.0569	91.3267	-4.0644	91.9370
0.8794	1.0367	0.4588	0.0412	91.2910	-3.0954	70.7364
0.9426	0.9829	0.4070	0.0221	91.7274	-1.7495	40.6020
1.0000	0.9292	0.3551	0.0000	92.6496	0.0000	0.0000

**Table 3 Parameters of Eq. (6) and Standard deviations**

Excess Property	A <sub>0</sub>	A <sub>1</sub>	A <sub>2</sub>	A <sub>3</sub>	A <sub>4</sub>	σ
<b>Methyl Acrylate (MA) + Benzene (B)</b>						
$\eta^E \times 10^3 \text{ Kg m}^{-1} \text{ s}^{-1}$	0.000003	-0.0148	0.01653	-0.0020	0.0002	0.00001
$V^E \times 10^6 \text{ m}^3 \text{ mol}^{-1}$	-0.000003	-0.6492	0.77283	-0.1422	0.01869	0.00003
$G^{*E} \times 10^3 \text{ N mol}^{-1}$	-0.00001	9.19117	-5.75584	-3.0605	-0.37453	0.00027
<b>Methyl Acrylate (MA) + Chloro Benzene (CB)</b>						
$\eta^E \times 10^3 \text{ Kg m}^{-1} \text{ s}^{-1}$	0.000006	0.06546	-0.04992	-0.01113	-0.00442	0.00002
$V^E \times 10^6 \text{ m}^3 \text{ mol}^{-1}$	0.00015	-1.91952	1.36386	0.14118	0.41403	0.00037
$G^{*E} \times 10^3 \text{ N mol}^{-1}$	-0.04223	145.38777	-109.27053	35.78226	-71.77812	0.10153
<b>Methyl Acrylate (MA) + Bromo Benzene (BB)</b>						
$\eta^E \times 10^3 \text{ Kg m}^{-1} \text{ s}^{-1}$	0.00001	1.3942	-0.54136	0.18392	-0.0368	0.00005
$V^E \times 10^6 \text{ m}^3 \text{ mol}^{-1}$	-0.000002	0.32663	-0.37684	0.08055	-0.03033	0.00001
$G^{*E} \times 10^3 \text{ N mol}^{-1}$	-0.17164	415.02372	-490.24978	317.88792	-242.1405	0.43351

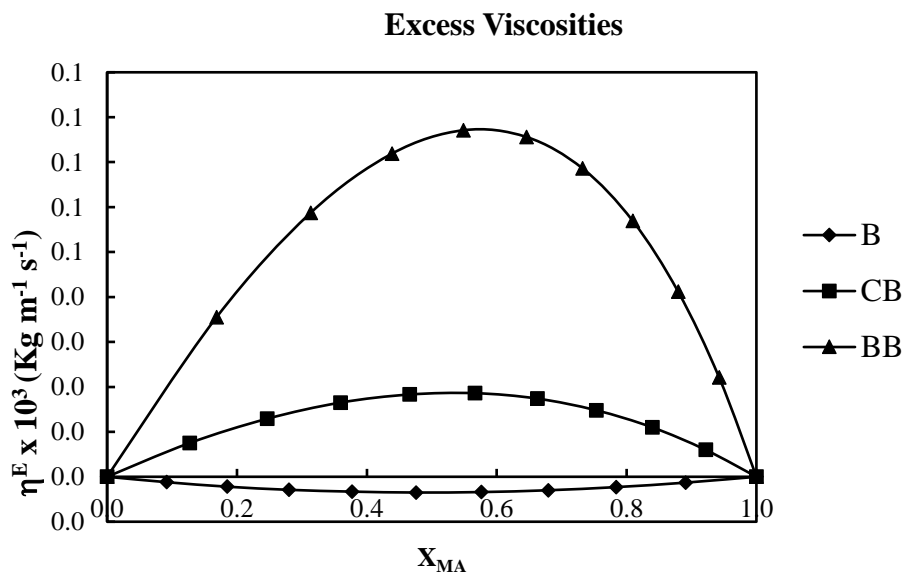
S. Parthasarathi and workers [23] have reported negative excess volumes for the mixtures of Methyl Acrylate with polar components; this observation is supported by the present work where VE values are negative for all the Methyl Acrylate with Benzene & substituted halo Benzene molecules. A similar study was supported by S. L. Dahire et al. [13], a study of intermolecular interactions in binary mixtures of DMSO with Chloro Benzene, Bromo Benzene, and Nitro Benzene at 313.15K. The interactions observed between Methyl Acrylate and Benzene, as well as Substituted halobenzene molecules, could result from the formation of robust intermolecular hydrogen-bonded complexes. These complexes are likely responsible for the observed negative excess volume. The negative excess volumes of MA + Benzene & Substituted Halo Benzene molecules over the whole range of composition suggest that the 2nd and 3rd factors are responsible for this situation i.e. formation of

hydrogen bond interaction between >C=O group of MA and X-C (or) H-C- groups of Benzenes [ $>C=O \cdots H-C-$  (OR)  $>C=O \cdots X-C-$ ] and the

accommodation of smaller size Methyl Acrylate molecules in the bigger size Benzene & Substituted Halo Benzene molecules i.e. interstitial accommodation. These two effects lead to the negative VE values. And also these two effects outweigh the 1st effect. Contraction which accompanies the formation of strong hydrogen-bonded complexes is sufficient to compensate for the physical effect caused during mixing. Another effect of interaction between the two components becomes more and more predominant as the increased electron-withdrawing ability of Halo Benzenes to MA. Also, the size of Halo benzene molecules increases from benzene, Chloro benzene, and Bromo benzene, this in turn makes a larger difference in the size of MA and Benzene and Substituted Halo Benzene molecules. This gives more possibilities for

more interstitial accommodation resulting in negative VE to confirm the negative excess volumes as the following order. (MA + B) < (MA + CB) < (MA + BB) Figure 4, it is noted that the  $\eta^E$  values are consistently positive across the entire composition range for all three systems. A relationship has been observed between the signs of  $\eta^E$  and VE in various binary solvent systems, where  $\eta^E$  tends to be positive when VE is negative, and vice versa. Typically, in systems where dispersion and dipolar interactions are dominant,  $\eta^E$  values are negative, whereas positive  $\eta^E$  values are indicative of charge transfer and hydrogen bonding interactions, leading to the formation of complex species between, unlike molecules. The algebraic values of  $\eta^E$  for all three mixtures of MA + Benzene, Chloro Benzene, and Bromo Benzene follow a specific order. (MA + B) < (MA + CB) < (MA + BB). This sequence implies that the degree of charge transfers and hydrogen bonding between dissimilar molecules rises alongside the enhanced withdrawing ability in Halo Benzene molecules. A comparable finding was documented by

Manapragada et al. [24] in their viscosity examination of binary liquid mixtures. They assessed the excess volumes of methyl formate, ethyl formate, propyl formate, and benzyl acetate with Bromo-, chloro-, and nitrobenzenes at temperatures of 303.15 K, 308.15 K, and 313.15 K. Figure 4, the variation of excess Gibbs free energy of activation of viscous flow (GE) with XMA is depicted for all three systems. The magnitude of the positive values serves as a reliable indicator of the strength of specific interactions. The order of GE values for the systems examined suggests the following sequence. (MA + B) < (MA + CB) < (MA + BB) Reed and Taylor [25] and Meyer et al., [26] reported that the  $G^*E$  -parameter can as a reliable criterion to detect or exclude the presence of interactions between unlike molecules. Similar behavior was reported by Manapragada et al., [27] in the case of  $G^*E$  values for mixtures of methyl formate, ethyl formate, propyl formate, and benzyl acetate with bromo-, chloro-, nitro benzenes at 303.15, 308.15 and 313.15 K.



**Figure 4** Plots of excess viscosities ( $\eta^E$ ) for Benzene and Substituted Halo Benzene molecules like Benzene (-•-), Chloro Benzene (-■-), and Bromo Benzene (-▲-) vs mole fraction of Methyl acrylate (XMA) at 313.15 K

### Conclusion

This study concludes that the densities and viscosities of binary mixtures of Methyl Acrylate with Benzene

(B), Chloro Benzene (CB), and Bromo Benzene (BB) at 313.15K were measured, and the values of VE,  $\eta^E$ ,

and GE were calculated across the entire composition range [28 and 29]. A comparison with literature data was conducted for pure liquids and binary liquid mixtures, revealing good agreement between experimental and literature values. The VE values were consistently negative for all three mixtures, while  $\eta E$  and GE values were consistently positive, indicating the presence of specific interactions between Methyl Acrylate, Benzene (B), and Substituted haloene molecules. The magnitude of these interactions followed a specific order. (MA + B) < (MA + CB) < (MA + BB).

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