

## Thermophysical and Acoustical Properties of Benzylparaben with Benzene at Temperatures of 303.15, 308.15 and 313.15 K

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### Abstract

The Ultrasonic velocity, Densities, viscosities, refractive index and Surface tension have been measured for binary mixture of Benzylparaben with Benzene at temperatures  $T=303.15, 308.15$  and  $313.15$  K at different molefractions such as 0.1 to 1.0. The experimental data have been used to calculate the acoustical and thermodynamical parameters like excess molar volume ( $V^E$ ), viscosity deviation ( $\Delta\eta$ ), refractive index deviation ( $\Delta n_D$ ), deviations in ultrasonic velocity ( $\Delta u$ ), isentropic compressibility ( $\beta$ ), deviations in isentropic compressibility ( $\Delta\beta$ ), intermolecular free length ( $L_f$ ), deviation in intermolecular free length ( $\Delta L_f$ ), Acoustic impedance ( $Z$ ), deviation in acoustic impedance ( $\Delta Z$ ), molar compressibility or Wada's constant ( $W$ ), molar sound velocity ( $R$ ), degree of intermolecular attraction ( $\delta$ ), relaxation time ( $\tau$ ), Free volume ( $V_f$ ) and absorption coefficient ( $\alpha$ ) and deviation in surface tension ( $\Delta\sigma$ ). The viscosity data have been correlated with the equations of Krishnan-Laddha and McAllister. The thermo physical properties under study were fit to the Jouyban-Acree model. The excess values were correlated using Redlich-Kister polynomial equation to obtain their coefficients and standard deviations. It was found that in all cases, the data obtained fitted with the values correlated by the corresponding models very well. The results are interpreted in terms of molecular interactions occurring in the solution.

**Keywords:** Binary mixture; Density; Viscosity; Refractive index; Ultrasonic velocity; Surface tension

### Introduction

Surface tension investigation of liquid mixture containing polar and nonpolar components is of considerable importance in understanding intermolecular interaction between the component molecules as that finds application in several industrial and technological processes [1]. Surface tension, Ultrasonic velocity, Refractive index, Density, Viscosity and the derived acoustical parameters like excess molar volume  $V^E$ , viscosity deviation  $\Delta\eta$ , refractive index deviation ( $\Delta n_D$ ), deviations in ultrasonic velocity ( $\Delta u$ ), deviation in surface tension ( $\Delta\sigma$ ), isentropic compressibility ( $\beta$ ), deviations in isentropic compressibility ( $\Delta\beta$ ), intermolecular free length ( $L_f$ ), deviation in intermolecular free length ( $\Delta L_f$ ), Acoustic impedance ( $Z$ ), deviation in acoustic impedance ( $\Delta Z$ ), molar compressibility or Wada's constant ( $W$ ), molar sound velocity ( $R$ ), degree of intermolecular attraction ( $\delta$ ), relaxation time ( $\tau$ ), Free volume ( $V_f$ ) and absorption coefficient ( $\alpha$ ) with their excess values, provide valuable information about the molecular environments. This has been studied for various binary and ternary mixtures [2-6] with respect to variation in concentration of the liquids and temperatures. The viscosity values have been fitted to McAllister and Krishnan and Laddha model. The Jouyban-Acree model has also been extended to density, viscosity, refractive index and ultrasonic velocity ( $u$ ) of binary mixtures. The deviation values have been fitted to Redlich-Kister type equation. Literature survey showed that no measurements have been previously reported for the mixture studied in this paper. Density, viscosity, refractive index, surface tension and ultrasonic velocity of binary mixtures results are necessary for interpretation of data obtained from thermo chemical, electrochemical, biochemical and kinetic studies [7]. The thermodynamic, acoustic and transport properties of liquids and liquid mixtures [8] are used to study the molecular interactions between the various components of the mixtures and also to understand engineering applications concerning heat transfer, mass transfer, and fluid flow. 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, viscosity, refractive index and ultrasonic velocity

find extensive application in solution theory and molecular dynamics. Parabens are a class of generally utilized additives as a part of restorative and pharmaceutical items. Synthetically, they are a progression of parahydroxybenzoates or esters of parahydroxybenzoic corrosive (otherwise called 4-hydroxybenzoic corrosive). Parabens are compelling additives in numerous sorts of recipes. These mixes, and their salts, are utilized basically for their bactericidal and fungicidal properties. They can be found in shampoos, business lotions, shaving gels, individual oils, topical/parenteral pharmaceuticals, spread tanning arrangement, makeup and toothpaste. They are also used as food additives. Benzene is a natural compound with the synthetic recipe  $C_6H_6$ . Its particle is made out of 6 carbon particles joined in a ring, with 1 hydrogen atom appended to every carbon molecule. Since its atoms contain just carbon and hydrogen particles, benzene is classed as a hydrocarbon. It is a characteristic constituent of raw petroleum, and is a standout amongst the most basic petrochemicals. Benzene is a fragrant hydrocarbon and the second [n]-annulene ([6]-annulene), a cyclic hydrocarbon with a ceaseless pi bond. It is here and there truncated Ph-H. Benzene is a drab and profoundly combustible fluid with a sweet scent. It is for the most part utilized as a forerunner to substantial chemicals, for example, ethyl benzene and cumene, which are created on a billion kilogram scale. Since it has a high octane number, it is a critical segment of fuel, involving a couple of percent of its mass. Most non-modern applications have been constrained by benzene's cancer-causing nature. The objective of the present work is to provide new experimental data on the densities of benzylparaben with benzene mixture, and

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to estimate excess volumes and derived thermodynamic properties such as excess molar volume ( $V^E$ ), viscosity deviation ( $\Delta\eta$ ), refractive index deviation ( $\Delta n_D$ ), deviations in ultrasonic velocity  $\Delta u$ , isentropic compressibility ( $\beta$ ), deviations in isentropic compressibility ( $\Delta\beta$ ), intermolecular free length ( $L_f$ ), deviation in intermolecular free length ( $\Delta L_f$ ), Acoustic impedance ( $Z$ ), deviation in acoustic impedance ( $\Delta Z$ ), molar compressibility or Wada's constant ( $W$ ), molar sound velocity ( $R$ ), degree of intermolecular attraction ( $\delta$ ), relaxation time ( $\tau$ ), Free volume ( $V_f$ ) and absorption coefficient ( $\alpha$ ) using different correlations and a well-known equation of state.

## Materials and Methods

The mixtures of various concentrations in mole fraction were prepared by taking analytical reagent grade and spectroscopic reagent grade chemicals with minimum assay of 99.9 % and obtained from E. Merck Ltd. (India). In all the mixtures, the mole fraction of the mixture is varied from 0.1 to 1.0, so as to have the mixture of different concentration. The density, viscosity, surface tension and ultrasonic velocity were measured as a function of concentration of the binary liquid mixture at temperature  $T=303.15$ ,  $308.15$  and  $313.15$  K. The purity of these experimental chemicals was checked by comparing the observed densities, viscosities, refractive index, surface tension and ultrasonic velocities with those reported in the literature. The measured values are included in Table 1 along with the available literature values. The experimental values of density, dynamic viscosities  $\eta$ , refractive index, ultrasonic velocity and surface tension of pure liquids benzylparaben and benzene at the investigated temperature  $303.15$ ,  $308.15$  and  $313.15$  K and compared with literature values [9-11] are shown in Table 1.

### Density measurement

The densities of the mixture were measured using a 25 ml specific gravity bottle by relative measurement method with an accuracy of  $\pm 0.01$   $\text{kg}\cdot\text{m}^{-3}$ . The specific gravity bottle with the experimental mixture was immersed in the temperature-controlled water bath. The weight of the sample was measured using an electronic digital balance with an accuracy of  $\pm 0.1$  mg (Model: SHIMADZU AX-200, Kyoto, Japan).

### Viscosity measurement

An Oswald viscometer (10 ml) with an accuracy of  $\pm 0.001$  mPa.s was used for the viscosity measurement. The flow time was determined using a digital racer stopwatch with an accuracy of  $\pm 0.1$  s.

### Refractive index

Refractive indexes were measured using thermostatically controlled Abbe refractometer (Atago 3T) with accuracy less than 0.001 units. Water was circulated in to the prism of the refractometer by a circulation pump connected to an external thermo stated water bath. Calibration was performed by measuring the refractive index of doubly distilled water and propyl alcohol at defined temperatures. The

sample mixture was directly injected in to the prism assembly of the instrument using a syringe. The solutions were pre thermo stated at the temperature of the experience before the experiments to achieve a quick thermal equilibrium.

### Velocity measurement

Ultrasonic velocity measurements were made using an ultrasonic interferometer (Model M-84, supplied by M/S Mittal Enterprises, New Delhi), at 313 K with the accuracy of  $\pm 0.1\text{m}\cdot\text{s}^{-1}$ . The measuring cell of interferometer is a specially designed double-walled vessel with provision for temperature constancy. An electronically operated digital constant temperature bath (Model SSI-03 Spl, supplied by M/S Mittal Enterprises, New Delhi), operating in the temperature range of  $-10^\circ\text{C}$  to  $85^\circ\text{C}$  with an accuracy of  $\pm 0.1^\circ\text{C}$  has been used to circulate water through the outer jacket of the double-walled measuring cell containing the experimental liquid.

### Surface tension measurement

Surface tension was determined using drop volume tensiometer described in detail [12] which also discussed procedure and handling of data. The precision capillary is connected to a dosing system. It is located in one of the two liquid phases involved and forces the second liquid phase in to first liquid phase through the capillary. From the flow rate and number of drops, surface tension of each drop is calculated for pure liquid and the binary mixture over the whole composition range. All the samples were equilibrated to 313.15 K under atmospheric pressure. It was calibrated with distilled water. The accuracy of the mixture over the whole composition range. All the samples were equilibrated to 313.15 K under atmospheric pressure. It was calibrated with distilled water. The accuracy of the surface tension measurement was estimated to be  $0.1\text{mNm}^{-1}$ . This can be calculated as

$$\sigma = \frac{V_{\text{drop}}(\rho_H - \rho_L)g}{\pi d} \quad (1)$$

$\sigma$ =Surface tension,  $V$ =Volume of drop,  $d$ =Diameter of capillary,  $g$ =Acceleration due to gravity.

### Theoretical aspect

The following thermodynamic parameters were calculated:

Excess molar volume ( $V^E$ ) has been calculated from the density ( $\rho$ ) of the medium using the following equation

$$V^E = \left( \frac{x_1 M_1 + x_2 M_2}{\rho_m} \right) - \left( \frac{x_1 M_1}{\rho_1} + \frac{x_2 M_2}{\rho_2} \right) \quad (2)$$

Where  $x_1$  and  $x_2$  refer to the mole fraction of components 1 and 2.  $\rho_1$ ,  $\rho_2$  and  $\rho_m$  refer to the density of components 1 and 2 and the density of the mixture, respectively.

Viscosity deviation ( $\Delta\eta$ ) has been determined as follows

$$\Delta\eta = \eta - (x_1 \eta_1 + x_2 \eta_2) \quad (3)$$

Where  $\eta$ ,  $\eta_1$ ,  $\eta_2$  are the viscosity of the mixture and the viscosity

Temperature	Pure liquids	$\rho/\text{g}\cdot\text{cc}$		$\eta/\text{mPa}\cdot\text{s}$		$n_D$		$u/\text{ms}^{-1}$		$\sigma$	
		lit	Exp	lit	Exp	lit	Exp	lit	Exp	lit	Exp
303.15K	Benzylparaben	1.0909	1.0909	0.7501	0.7501	1.5805	1.5805	1725	1725	80.24	80.24
308.15K		1.0169	1.0169	0.7270	0.7270	1.5698	1.5698	1712	1712	79.14	79.14
313.15K		1.0050	1.0050	0.7228	0.7228	1.5601	1.5601	1695	1695	77.05	77.05
303.15K	Benzene	0.8784	0.8784	0.6242	0.6242	1.4981	1.4981	410	410	28.84	28.84
308.15K		0.8651	0.8651	0.6151	0.6151	0.6151	0.6151	405	405	27.04	27.04
313.15K		0.8528	0.8528	0.6071	0.6071	0.6071	0.6071	398	398	26.05	26.05

**Table 1:** Comparison of density, viscosity, refractive index, ultrasonic velocity and surface tension data for liquids at 303.15, 308.15 and 313.15K with the literature.

of pure components 1 and 2, respectively. The uncertainty in the calculation of  $\Delta\eta$  from viscosity measurements was estimated to be  $\pm 0.0001$ .

Refractive index deviation ( $\Delta n_D$ ) from linear additive value of the mole fraction is obtained by

$$\Delta n_D = n_D - (x_1 n_{D1} + x_2 n_{D2}) \quad (4)$$

Where  $n_D, n_{D1}, n_{D2}$  are the refractive index of the mixture and the refractive index of pure components 1 and 2, respectively.

Ultrasonic velocity deviations ( $\Delta u$ ): has been determined as follows:

$$\Delta u = u - (x_1 u_1 + x_2 u_2) \quad (5)$$

Where  $u, u_1, u_2$  are the ultrasonic velocity of the mixture and the ultrasonic velocity of pure components 1 and 2, respectively.

Isentropic compressibility ( $\beta$ ): has been calculated from the ultrasonic velocity ( $u$ ), and the density ( $\rho$ ) of the medium using the Newton-Laplace equation [13] as follows:

$$\beta = \frac{1}{\rho u^2} \quad (6)$$

Isentropic compressibility deviations ( $\Delta\beta$ ) has been determined has follows

$$\Delta\beta = \beta - (x_1\beta_1 - x_2\beta_2) \quad (7)$$

Where  $\beta, \beta_1, \beta_2$  are the isentropic compressibility of the mixture and the isentropic compressibility of pure components 1 and 2, respectively.

Intermolecular free length ( $L_f$ ) has been determined as follows [14]:

$$L_f = K_T \beta^{\frac{1}{2}} \quad (8)$$

Where  $K_T$  is the temperature-dependent constant known as Jacobson's constant ( $K_T=2.131 \times 10^{-6}$  at 313K), and  $\beta$  is the isentropic compressibility.

Deviation in intermolecular free length ( $\Delta L_f$ ) has been determined as follows:

$$\Delta L_f = L_f - (x_1 L_{f1} - x_2 L_{f2}) \quad (9)$$

Where  $L_f, L_{f1}$  and  $L_{f2}$  are the intermolecular free length of the mixture and the intermolecular free length of pure components 1 and 2, respectively.

Acoustic impedance ( $Z$ ) has been determined as follows:

$$Z = \rho u \quad (10)$$

Where  $\rho$  is the density of mixture and  $u$  is the ultrasonic velocity of the mixture.

Deviation in acoustic impedance ( $\Delta Z$ ) has been determined as follows

$$\Delta Z = Z - (x_1 Z_1 - x_2 Z_2) \quad (11)$$

Where  $Z, Z_1$  and  $Z_2$  are the acoustic impedance of the mixture and the acoustic impedance of pure components 1 and 2, respectively.

Molar compressibility or Wada's constant ( $W$ ) was calculated by the equation:

$$W = \left(\frac{M}{\rho}\right) \beta^{-\frac{1}{7}} \quad (12)$$

Where  $M$  is the relative molar mass and  $\beta$  is the isentropic compressibility.

Molar sound velocity( $R$ ) was calculated by the equation:

$$R = \left(\frac{M}{\rho}\right) u^{\frac{1}{7}} \quad (13)$$

Where  $u$  is the ultrasonic velocity of the mixture.

Degree of intermolecular attraction ( $\delta$ ) was calculated by the equation:

$$\delta = \left(\frac{u^2}{u_{im}^2}\right) - 1 \quad (14)$$

Where

$$u_{im}^2 = \frac{1}{(x_1 M_1 + x_2 M_2) \left( \frac{x_1}{M_1 u_1^2} + \frac{x_2}{M_2 u_2^2} \right)}$$

Relaxation time ( $\tau$ ) in terms of adiabatic compressibility ( $\beta$ ) and viscosity ( $\eta$ ) of the liquid is as follows:

$$\tau = \frac{4}{3} (\beta \eta) \quad (15)$$

Where  $\beta$  is the adiabatic compressibility and  $\eta$  is the viscosity of the mixture.

Free Volume ( $V_f$ ) in terms of ultrasonic velocity ( $u$ ) and the viscosity ( $\eta$ ) of liquid is [15] as follows:

$$V_f = \left(\frac{M_{eff} f u}{k \eta}\right)^{\frac{3}{2}} \quad (16)$$

Where  $M_{eff}$  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  $4.281 \times 10^9$  [12] for all liquids.

Absorption coefficient ( $\alpha$ ) or attenuation coefficient is a characteristic of the medium, and it depends on the external condition like temperature, pressure, and frequency of measurement. It is given by the following [16]:

$$\alpha = \frac{8\pi^2 \eta f^2}{3\rho u^2} \quad (17)$$

Where  $f$  is the frequency of ultrasonic wave.

## Results and Discussion

The experimental data relating to density, viscosity, refractive index, ultrasonic velocity and surface tension at 303.15, 308.15 and 313.15 K for different compositions for the given mixtures have been presented in Table 2. Calculated values of excess molar volume  $V^E$ , viscosity deviation  $\Delta\eta$ , refractive index deviation ( $\Delta n_D$ ), deviations in ultrasonic velocity  $\Delta u$ , isentropic compressibility ( $\beta$ ), deviations in isentropic compressibility ( $\Delta\beta$ ), intermolecular free length ( $L_f$ ), deviation in intermolecular free length ( $\Delta L_f$ ), Acoustic impedance ( $Z$ ), deviation in acoustic impedance ( $\Delta Z$ ), molar compressibility or Wada's constant ( $W$ ), molar sound velocity ( $R$ ), degree of intermolecular attraction ( $\delta$ ), relaxation time ( $\tau$ ), Free volume ( $V_f$ ) and absorption coefficient ( $\alpha$ ) are presented in Tables 3, 4 and 5, and their excess values are presented in Tables 6, 7 and 8.

The intermolecular free lengths as well as the relaxation time are properties of liquid mixtures which mainly affect the ultrasonic velocity. The velocity increases with the increase of compositions. This is confirmed by the fact that both free length and relaxation time decreases (Table 5) with the increase of composition. Since the association of the interacting molecules varies with the composition of the ultrasonic wave, cohesive force also varies. Cohesive force as well as free volume decreases (Table 5) with the increase of compositions.

The increase in density (Table 2), suggests minimum interaction between unlike molecules. Density is given by the relation (specific gravity of the mixture X density of the reference substance). Which

$X_1$	Density( $\rho$ )	Viscosity( $\eta$ )	Refractive index( $n_D$ )	Ultrasonic velocity( $u$ )	Surface tension( $\sigma$ )
303.15K					
0	0.8784	0.6242	1.4981	410	28.84
0.0487	0.8856	0.6382	1.4951	569	30.57
0.1033	0.8902	0.6599	1.5162	698	35.74
0.165	0.8968	0.6721	1.5213	752	40.1
0.2351	0.9077	0.6908	1.5369	814	42.67
0.3156	0.916	0.6967	1.5489	964	45.76
0.4089	0.9223	0.7178	1.5578	1058	58.09
0.5183	0.9237	0.7218	1.5607	1247	65.78
0.6485	0.9828	0.7229	1.5725	1498	70.12
0.8058	0.9991	0.7325	1.5799	1605	75.34
1	1.0909	0.7501	1.5805	1725	80.24
308.15K					
0	0.8651	0.615	1.4896	405	27.04
0.0487	0.8714	0.6178	1.4901	554	29.49
0.1033	0.872	0.6447	1.5023	679	34.15
0.165	0.8733	0.6457	1.5189	712	41.18
0.2351	0.8843	0.67	1.5263	855	45.06
0.3156	0.8968	0.6909	1.5397	998	48.01
0.4089	0.9015	0.7013	1.5408	1078	57.58
0.5183	0.9426	0.7161	1.5524	1247	64.08
0.6485	0.9531	0.7218	1.5605	1469	72.19
0.8058	0.9569	0.7235	1.5652	1697	74.74
1	1.0169	0.727	1.5698	1712	79.14
313.15K					
0	0.8528	0.6071	1.4885	398	26.05
0.0487	0.8633	0.6203	1.4892	436	27.57
0.1033	0.867	0.6431	1.4998	638	36.89
0.165	0.8817	0.6473	1.5054	758	40.99
0.2351	0.8955	0.6684	1.5164	891	41.77
0.3156	0.9111	0.6888	1.5287	997	44.66
0.4089	0.9316	0.7065	1.5318	1045	56.07
0.5183	0.9538	0.7081	1.5474	1287	66.17
0.6485	0.9623	0.7098	1.5505	1459	71.89
0.8058	0.9868	0.7165	1.5602	1599	73.31
1	1.005	0.7228	1.5601	1695	77.05

**Table 2:** Experimental Density ( $\rho$ ), Viscosity ( $\eta$ ), refractive index ( $n_D$ ), ultrasonic velocities ( $u$ ) and surface tension ( $\sigma$ ) of Benzylparaben–benzene mixture at 303.15, 308.15 and 313.15 K.

Mole fraction, ( $X_1$ )	Excess molar volume, ( $V^E$ )	Viscosity deviation, ( $\Delta\eta$ )	refractive index deviation, ( $\Delta n_D$ )	Ultrasonic velocity deviation, ( $\Delta u$ )	Surface tension deviation, ( $\Delta\sigma$ )
303.15K					
0	0	0	0	0	0
0.0487	<b>2.6727</b>	0.0185	0.007	95	1.7731
0.1033	<b>4.812</b>	0.0265	0.0115	120	4.5903
0.165	<b>6.9425</b>	0.0325	0.0195	138	6.5214
0.2351	<b>8.7246</b>	0.0345	0.0221	149	8.7458
0.3156	<b>11.0998</b>	0.0367	0.0247	155	10.6981
0.4089	<b>14.1242</b>	0.0354	0.026	150	11.9325
0.5183	<b>15.5104</b>	0.0333	0.0265	140	12.2993
0.6485	<b>16.5924</b>	0.0282	0.0249	125	10.9471
0.8058	<b>13.391</b>	0.0209	0.0154	91	6.0818
1	0	0	0	0	0
308.15K					
0	0	0	0	0	0
0.0487	1.203	0.0096	0.0034	75	1.0872
0.1033	3.2089	0.0181	0.0079	98	3.7281
0.165	5.3762	0.0254	0.0169	118	5.5435
0.2351	7.4067	0.0295	0.0198	131	7.1712
0.3156	9.2951	0.0305	0.0231	146	9.5272

0.4089	11.5839	0.0295	0.0239	142	10.9363
0.5183	13.5891	0.0286	0.0229	131	11.0365
0.6485	14.8068	0.0251	0.0205	115	9.3631
0.8058	9.6486	0.0184	0.0109	81	4.7178
1	0	0	0	0	0
313.15K					
0	0	0	0	0	0
0.0487	0.7551	0.0035	0.0017	51	0.5637
0.1033	2.3869	0.0149	0.0039	81	2.1717
0.165	3.7314	0.0219	0.0124	109	3.5251
0.2351	6.1859	0.0255	0.0175	119	5.7299
0.3156	7.9736	0.0281	0.0204	130	8.5144
0.4089	9.7358	0.0272	0.0224	129	10.0661
0.5183	11.6465	0.0253	0.0217	119	9.6867
0.6485	12.8699	0.0226	0.0191	109	6.7665
0.8058	7.9596	0.0161	0.0098	69	3.7642
1	0	0	0	0	0

**Table 3:** Excess molar volume ( $V^E$ ), Viscosity deviation ( $\Delta\eta$ ), refractive index deviation ( $\Delta n_p$ ), ultrasonic velocity deviation ( $\Delta u$ ) and surface tension deviation ( $\Delta\sigma$ ) of Benzylparaben–benzene mixture at 303.15, 308.15 and 313.15 K.

X	Z	$\Delta Z$	$\beta$	$\Delta\beta$	$L_f \cdot 10^9$	$\Delta L_f$	$\delta$
303.15K							
0	360	0	0.00000677	0	5.1892	0	0
0.0487	503	25	0.00000348	-0.0000029	3.7205	-0.0164	1.0057
0.1033	621	45	0.0000023	-0.0000038	3.0246	-0.0259	2.2251
0.165	674	63	0.00000197	-0.0000045	2.7992	-0.03718	3.9144
0.2351	738	69	0.00000166	-0.0000049	2.5696	-0.0498	4.9041
0.3156	883	74	0.00000117	-0.0000051	2.1572	-0.0552	5.5341
0.4089	975	71	0.00000096	-0.0000049	1.9541	-0.0553	5.1901
0.5183	1151	68	0.00000069	-0.0000045	1.6566	-0.0524	4.5031
0.6485	1472	59	0.00000045	-0.0000036	1.3378	-0.0458	3.1989
0.8058	1603	42	0.00000038	-0.0000021	1.2294	-0.0312	1.9958
1	1881	0	0.0000003	0	1.0923	0	0
308.15K							
0	350	0	0.000007	0	5.3920	0	0
0.0487	482	19	0.000003	-0.0000019	3.5299	-0.0121	0.8486
0.1033	592	35	0.0000024	-0.0000032	3.1572	-0.0216	1.7998
0.165	621	45	0.0000023	-0.0000038	3.0907	-0.0314	2.8124
0.2351	756	55	0.0000015	-0.0000044	2.4960	-0.0415	3.8031
0.3156	895	59	0.0000011	-0.0000047	2.1374	-0.04992	4.5808
0.4089	971	60	0.00000095	-0.0000045	1.9864	-0.0508	4.4361
0.5183	1175	55	0.00000068	-0.0000041	1.6805	-0.0457	3.6021
0.6485	1400	48	0.00000048	-0.0000031	1.4119	-0.0355	2.5516
0.8058	1623	31	0.00000036	-0.0000018	1.2228	-0.0192	1.5041
1	1740	0	0.00000033	0	1.1707	0	0
313.15K							
0	339	0	0.0000074	0	5.5222	0	0
0.0487	376	13	0.000006	-0.0000012	4.9724	-0.0109	0.6684
0.1033	553	30	0.0000028	-0.0000024	3.3968	-0.0176	1.1998
0.165	668	39	0.0000019	-0.0000032	2.7981	-0.0268	2.1521
0.2351	797	44	0.0000014	-0.0000039	2.4019	-0.0359	2.9918
0.3156	908	50	0.0000011	-0.0000042	2.1290	-0.0471	4.1713
0.4089	973	53	0.0000009	-0.0000041	1.9258	-0.0466	3.7352
0.5183	1227	48	0.0000006	-0.0000035	1.5724	-0.0394	3.0198
0.6485	1403	38	0.0000004	-0.0000025	1.2838	-0.0295	2.1007
0.8058	1577	25	0.00000039	-0.0000015	1.2677	-0.0158	1.0141
1	1703	0	0.00000034	0	1.1836	0	0

**Table 4:** Acoustic impedance (Z), Acoustic impedance deviation ( $\Delta Z$ ), isentropic compressibility ( $\beta$ ), isentropic compressibility deviation ( $\Delta\beta$ ), intermolecular free length ( $L_f$ ), intermolecular free length deviation ( $\Delta L_f$ ) and intermolecular attraction ( $\delta$ ) of Benzylparaben–benzene mixture at 303.15, 308.15 and 313.15 K.

X	W	R	$\tau \times 10^{-7}$	$V_f \times 10^{-19}$	$\alpha$
303.15K					
0	953	71413	5.6344	4.1285	0.000445033
0.0487	1039	98302	2.9612	7.4672	0.000234327
0.1033	1097	119966	2.0236	1.1071	0.00016018
0.165	1113	128296	1.7653	1.3877	0.000139518
0.2351	1127	137206	1.5289	1.7358	0.000120918
0.3156	1174	161017	1.0868	2.5712	8.61637E-05
0.4089	1197	175511	9.1878	3.3134	7.31961E-05
0.5183	1199	206550	6.6405	4.9689	5.29031E-05
0.6485	1254	233205	4.3374	7.7928	3.45078E-05
0.8058	1255	245786	3.7113	1.0240	2.99624E-05
1	1264	241933	3.0004	1.3516	2.43268E-05
308.15K					
0	963	100756	0.5764	4.1561	0.023901025
0.0487	1079	137199	2.4712	7.553	0.012828232
0.1033	1113	161140	2.0630	1.1031	0.008847559
0.165	1118	168709	1.9801	1.3615	0.008056478
0.2351	1174	195246	1.3452	1.9618	0.005729064
0.3156	1210	221007	1.0133	2.7503	0.004281316
0.4089	1229	235183	8.8831	3.5387	0.003698611
0.5183	1233	266430	6.4926	5.0424	0.002793507
0.6485	1267	311384	4.6195	7.6062	0.002020535
0.8058	1281	358868	3.4728	1.1373	0.001515718
1	1330	360297	3.1988	1.4045	0.001492331
313.15K					
0	969	100303	5.9900	4.1281	0.024434085
0.0487	986	107541	4.9624	5.2416	0.020787923
0.1033	1095	151786	2.4009	1.0085	0.010003547
0.165	1138	179165	1.6398	1.4900	0.007120186
0.2351	1171	203954	1.2476	2.0945	0.005266663
0.3156	1191	221459	1.0102	2.7588	0.004282845
0.4089	1198	226305	8.4781	3.3402	0.003950864
0.5183	1240	278083	5.6648	5.3768	0.002608029
0.6485	1275	314492	3.7856	7.7204	0.002031536
0.8058	1277	341447	3.7258	1.0555	0.001699114
1	1303	358792	3.2766	1.3957	0.001518386

**Table 5:** Molar compressibility or Wada's constant (W), molar sound velocity (R), relaxation time ( $\tau$ ), Free volume ( $V_f$ ) and absorption coefficient ( $\alpha$ ).

T/K	$v_{12}$	$v_{21}$	SD
303.15	0.8986	0.8551	0.0043
308.15	0.9435	0.9583	0.0014
313.15	0.9754	0.9796	0.0012

**Table 6:** Parameters and Standard Deviation of the of the McAllister model for benzyl paraben with benzene.

T/K	$A_1$	$A_2$	$A_3$	SD
303.15	-0.5166	0.0802	0.0982	0.1857
308.15	0.0747	0.0925	-0.0991	0.2236
313.15	-0.8234	0.1204	-0.1047	0.7164

**Table 7:** Parameters and Standard Deviation of the Krishnan – Laddha model for benzyl paraben with benzene.

shows that as composition increases, the excess molar volumes with the mole fraction ( $x_1$ ) of Benzylparaben and Benzene at (303.15, 308.15 and 313.15) K are represented in Table 3 and Figure 1. This shows that the excess molar volumes are always positive for all the studied temperatures. The sign of excess volume of a system depends on the

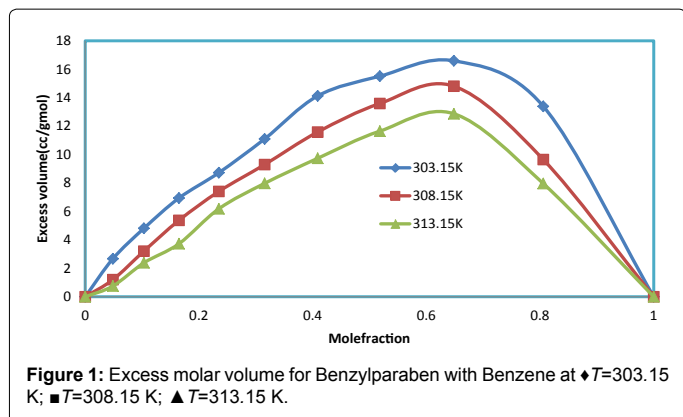
Fuction	$A_1$	$A_2$	$A_3$	$A_4$	$A_5$	SD
303.15K						
$V^E/\text{cm}^3 \text{ mol}^{-1}$	-0.9617	0.1609	-0.8743	-0.5976	0.5607	0.7025
$\Delta\eta / \text{mPa}\cdot\text{s}$	0.6959	-0.8938	-0.7729	0.6578	0.6059	0.0987
$\Delta n_D$	0.864	0.7301	-0.7625	0.7023	-0.9075	0.9319
$\Delta Z$	-0.7916	0.8916	-0.7423	-0.6961	0.7701	1.2655
$\Delta u / \text{m s}^{-1}$	0.864	0.7301	-0.7625	0.7023	-0.9075	0.9319
$\Delta\beta / \text{m}^2 \text{ N}^{-1}$	-0.9443	0.1035	-0.9573	0.5639	1.3039	0.1753
$\bar{\delta}$	-6.0634	2.2555	-3.2008	-1.7502	8.6928	1.0029
$\Delta L_f / \text{m}$	-0.9617	0.1609	-0.8743	-0.5976	0.5607	0.7025
$\Delta\sigma / \text{N m}^{-1}$	0.464	0.2255	-0.2576	0.8132	-0.7095	0.9987
308.15K						
$V^E/\text{cm}^3 \text{ mol}^{-1}$	-0.9461	-0.5205	0.3579	0.9812	-0.8637	0.0852
$\Delta\eta / \text{mPa}\cdot\text{s}$	-0.46517	0.3869	-0.4587	-0.3678	0.8116	0.895
$\Delta n_D$	0.864	0.7301	-0.7625	0.7023	-0.9075	0.9319
$\Delta Z$	-0.4169	-0.2505	0.5614	0.2918	-0.3687	0.6138
$\Delta u / \text{m s}^{-1}$	2.1796	1.8961	-2.7342	-1.0669	3.8154	0.0422
$\Delta\beta / \text{m}^2 \text{ N}^{-1}$	1.7517	1.388	-0.9378	-0.8378	0.9029	0.0069
$\bar{\delta}$	9.6517	-6.1198	-3.2991	0.6017	-2.384	0.2054
$\Delta L_f / \text{m}$	12.146	-8.3465	6.7449	4.5275	-6.771	0.3527
$\Delta\sigma / \text{N m}^{-1}$	-0.7916	0.8916	-0.7423	-0.6961	0.7701	1.2655
313.15K						
$V^E/\text{cm}^3 \text{ mol}^{-1}$	-0.8079	0.1402	0.3452	0.9786	-0.5392	0.9991
$\Delta\eta / \text{mPa}\cdot\text{s}$	-0.8315	0.6016	-0.2801	-0.4811	0.4969	0.9876
$\Delta n_D$	-0.7416	0.8601	-0.4801	-0.5011	0.5976	1.1405
$\Delta Z$	-0.7571	0.9738	-0.5387	-0.6378	0.6811	1.1738
$\Delta u / \text{m s}^{-1}$	1.9146	1.86	-1.6801	-1.71	2.7891	0.0488
$\Delta\beta / \text{m}^2 \text{ N}^{-1}$	-0.4078	0.2014	0.4325	0.7968	-0.3529	0.6333
$\bar{\delta}$	8.8687	-5.6724	-4.7875	2.6406	-1.6153	0.1259
$\Delta L_f / \text{m}$	-7.851	3.805	2.7249	-5.444	-9.135	0.4424
$\Delta\sigma / \text{N m}^{-1}$	-0.6731	0.3222	0.1366	-0.5789	0.1675	0.1106

**Table 8:** Parameters and Standard Deviation of the Redlich–Kister Equation for benzyl paraben with benzene  $T = (303.15, 308.15, \text{ and } 313.15) \text{ K}$ .

Properties	Temperature	$A_1$	$A_2$	$A_3$	$A_4$	SD
$\rho/\text{g}\cdot\text{cm}^{-3}$	303.15	64.1453	-87.2501	25.0169	98.091	0.0062
	308.15	-94.0562	-49.0284	200.7956	-20.0913	0.0087
	313.15	161.0849	10.6982	-82.6541	-40.8531	0.0079
$\eta / \text{mPa}\cdot\text{s}$	303.15	58.7405	-98.1325	10.3174	78.89	0.2586
	308.15	-84.0256	-11.8428	200.6957	-20.1093	0.5142
	313.15	185.6549	9.8682	-67.1405	-40.3052	0.321
$n_D$	303.15	63.0354	-78.3152	0.5137	68.98	0.7776
	308.15	-74.652	-4.6248	188.5796	-17.9301	0.54772
	313.15	151.8479	8.9628	-76.2514	-36.5813	0.06879
$u \text{ m/s}$	303.15	220.78	-200.2	69.411	204.23	0.0241
	308.15	166.32	-165.2	-8.8144	162.36	0.0092
	313.15	66.835	-180.24	211.71	170.87	0.0238
$\sigma \text{ n/m}$	303.15	69.6453	-89.2054	29.0168	99.291	0.0162
	308.15	-94.056	-59.028	101.796	-30.091	0.0287
	313.15	171.085	12.6982	-89.654	-49.853	0.0089

**Table 9:** Parameters and Standard Deviation of the Jouyban–Acree Model for benzyl paraben with benzene  $T = (303.15, 308.15, \text{ and } 313.15) \text{ K}$ .

relative magnitude of expansion/contraction on mixing of two liquids. If the factors causing expansion dominate the factors causing contraction, then  $V^E$  becomes positive [17,18]. The factors that are responsible for expansion in volume are as follows. (1) Loss of dipolar association (2) The geometry of molecules structure, which does not allow fitting of



one component in to other component (3) Steric hindrance, which opposes the proximity of the constituent molecules. The experiment in present investigation indicates that the factors which are responsible for expansion in volume are dominant over the entire composition range in all the studied temperatures of Benzylparaben with Benzene.

The viscosity increases with the increase in composition (Table 2). An increasing value of viscosity suggests that the closer approach of unlike molecules is due to hydrogen bonding. The increase in viscosity suggests shorter time for the rearrangement of molecules in the mixture. When composition increases, the viscosity imparted to the molecules obviously expedites the rearrangement procedure. Viscosity deviation is given by the relation,  $\Delta\eta = \eta - (x_1\eta_1 + x_2\eta_2)$  which shows Viscosity deviations are positive over the entire range of composition (Table 3 and Figure 2) for the binary mixture of Benzylparaben with Benzene, which indicates that the interaction between binary mixtures is strong.

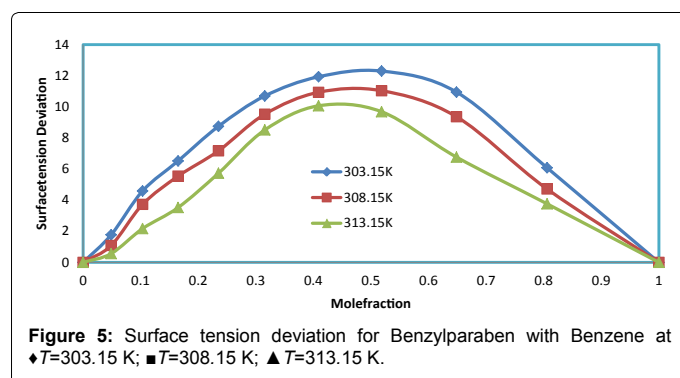
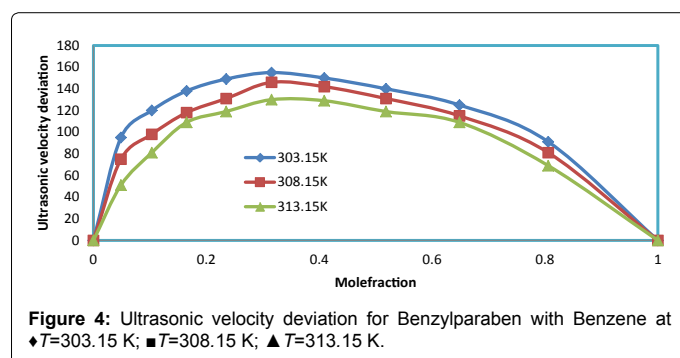
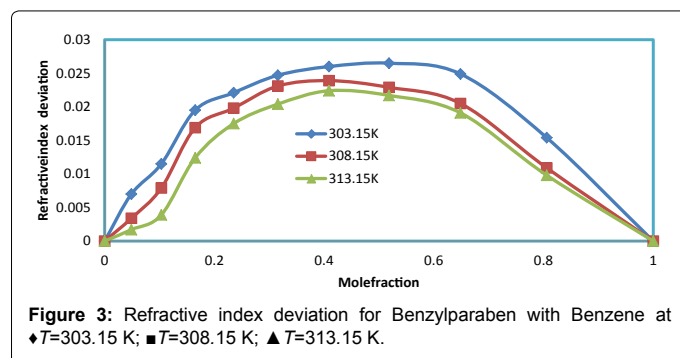
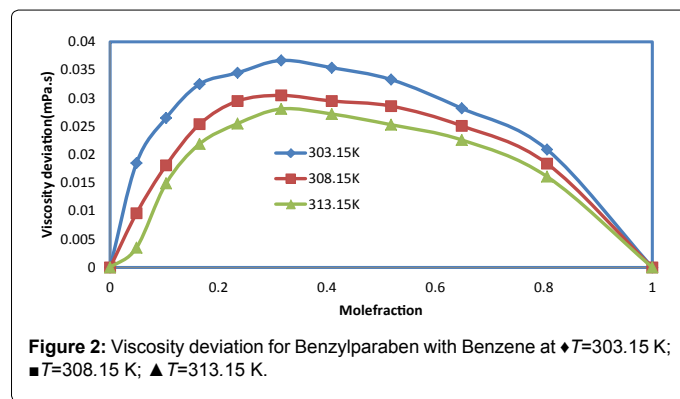
The refractive index increases with the increase in composition (Table 2). An increasing refractive index suggests that the weak interaction between unlike due to hydrogen bonding between unlike molecules. Refractive index deviation is given by the relation  $\Delta n_D = n_D - (x_1n_{D1} + x_2n_{D2})$  which shows refractive index deviation are positive over the entire range of composition (Table 3 and Figure 3) for the binary mixture of Benzylparaben with Benzene. This may be attributed to the weakening of structure making interactions at elevated temperatures due to enhanced thermal motion.

Table 3 and Figure 4 show the positive deviations over the entire range of mole fraction. The ultrasonic velocity of the mixture strongly not depends on the entropy of mixture, which is related with liquid's structure and enthalpy. It will consequently with molecular interactions between the components of the mixtures. Therefore the ultrasonic velocity between benzylparaben with benzene not depends on molecular interactions as well as on the size and shape of the molecules.

Temperature of the mixture increases ultrasonic velocity deviation also increases. The Ultrasonic velocity increases with the increase in composition (Table 2). An increasing Ultrasonic velocity suggests that the liquid structure and enthalpy between unlike due to hydrogen bonding between unlike molecules.

Surface tension ( $\sigma$ ) for the binary liquid mixture of benzylparaben with benzene over the entire composition range at 303.15, 308.15 and 313.15 K. A detailed observation of the (Table 2) shows that the surface tension of the mixture increases with the increases in mole fraction and decreases with increases in temperature. According to Karla Granados [19] strong interaction in the liquid mixture increases the  $\sigma$  value of

the mixture. This means that interactions in the mixture are strong and hence rise in the  $\sigma$  value was observed when mole fraction increases. It has also been observed that the surface tension deviation (Table 3 and Figure 5) shows positive values for the entire mole fraction which indicates the bulk of a liquid; each benzylparaben molecule can make



an optimal number of H-bonds to other benzene molecules. On the surface, however, the interactions with the neighboring molecules are limited and weaker, resulting in a higher free energy and reduced intermolecular hydrogen bonding of the molecules.

The decrease in isentropic compressibility (Table 4), suggests minimum interaction between unlike molecules. Isentropic compressibility is given by the relation,  $\beta = \frac{1}{\rho v^2}$  which shows that as composition increases, ultrasonic velocity and density increases and hence adiabatic compressibility decreases. The values of  $\Delta\beta$  are negative at all the temperatures and the values of  $\Delta\beta$  become less negative as temperature increased (Figure 6). This may be attributed to the weakening of structure making interactions at elevated temperatures due to enhanced thermal motion [20].

Intermolecular free length ( $L_f$ ) also confirms the fact that free length decreases (Table 4), with the increase in composition. The intermolecular free length is positive over the whole mole fraction range at different temperatures. Figure 7. intermolecular free length deviation indicates structural readjustment in the liquid mixtures towards less compressible phase of fluid and closer packing of molecules [21,22].

Acoustic impedance increases with the increase in composition (Table 4). This supports the possibility of strong interaction between like molecules. Acoustic impedance is also given by the product of the ultrasonic velocity and density  $Z = U\rho$  and is used for assessing the absorption of sound in a medium. As composition increases, absorption coefficient decreases (Table 5) as it is proportional to the square of the frequency. The values of  $\Delta Z$  were positive (Figure 8) over the entire range of mole fraction and the curves are asymmetrical in nature over the entire range of mole fraction for benzylparaben with benzene mixture at 303.15, 308.15 and 313.15 K.

The degree of intermolecular attraction ( $\alpha$ ) has also been evaluated to study the structural variations and the nature of interaction occurring in the system (Table 4 and Figure 9). It has been observed that the

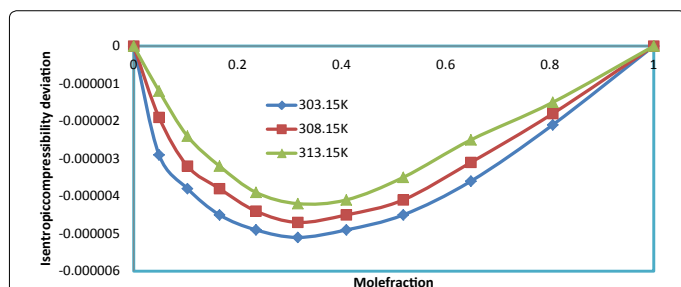


Figure 6: Isentropic compressibility deviation for Benzylparaben with Benzene at  $\blacklozenge T=303.15$  K;  $\blacksquare T=308.15$  K;  $\blacktriangle T=313.15$  K.

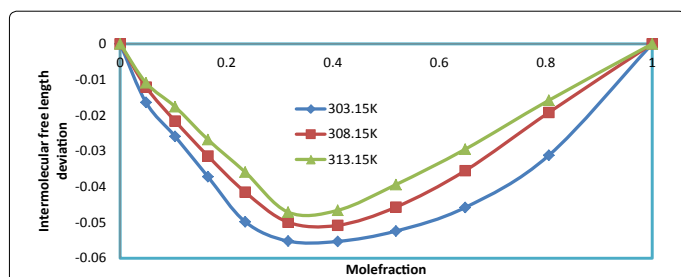


Figure 7: Intermolecular free length deviation for Benzylparaben with Benzene at  $\blacklozenge T=303.15$  K;  $\blacksquare T=308.15$  K;  $\blacktriangle T=313.15$  K.

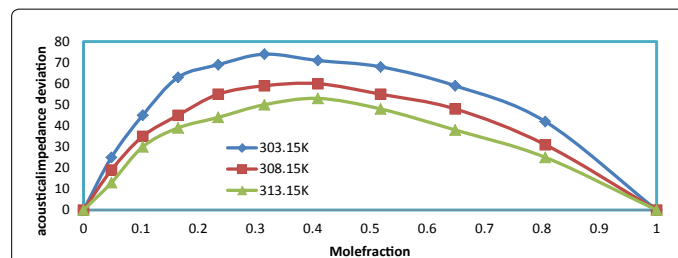


Figure 8: Acoustic impedance deviation for Benzylparaben with Benzene at  $\blacklozenge T=303.15$  K;  $\blacksquare T=308.15$  K;  $\blacktriangle T=313.15$  K.

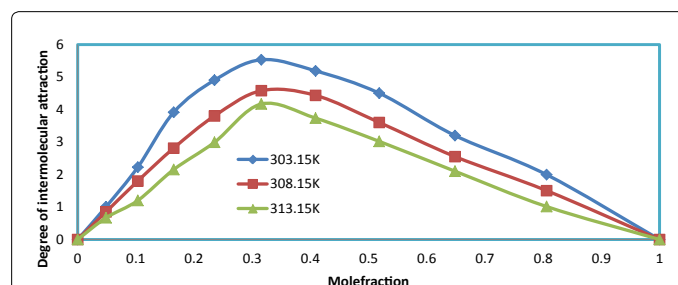


Figure 9: Degree of intermolecular attraction for Benzylparaben with Benzene at  $\blacklozenge T=303.15$  K;  $\blacksquare T=308.15$  K;  $\blacktriangle T=313.15$  K.

maximum value of  $\alpha$  occurs at nearly mole fraction of 0.3. This suggests the presence of intermolecular interaction in the system.

The molar sound velocity ( $R$ ) and molar compressibility ( $W$ ) were increasing linearly with mole fraction indicating solute-solvent interactions [23] may occur in the system as Seen in Table 5 which indicates that the interaction between the molecules does not seem to vary very much in strength with changing composition in the case of benzylparaben with benzene mixture due to the fact that benzene has a large dipole moment. Hence, intermolecular interaction in the case of the mixture containing benzene is strong.

The viscosities of mixtures of benzylparaben with benzene have been correlated with the model proposed by McAllister [24] for a two-component mixture considering three body interactions.

$$\ln v = x_1^2 \ln v_1 + 3x_1^2 x_2 \ln v_{12} + 3x_1 x_2^2 \ln v_{21} + x_2^2 \ln v_2 - \ln \left( x_1 + \frac{x_2 M_2}{M_1} \right) + 3x_1^2 x_2 \ln \left( \frac{1 + \frac{2x_2}{M_1}}{3} \right) + 3x_1 x_2^2 \ln \left( \frac{1 + \frac{2x_1}{M_2}}{3} \right) + x_2^2 \ln \left( \frac{M_2}{M_1} \right) \quad (18)$$

In equation,  $v_1$  and  $v_2$  refer to the kinematic viscosity of pure liquids 1 and 2 having mole fractions  $X_1$  and  $X_2$ , respectively. The parameters  $v_{12}$  and  $v_{21}$  represent the interaction parameters obtained by multiple regression analysis, while  $M_1$  and  $M_2$  are the molar masses of the components.

The viscosity was correlated by means of the Krishnan and Laddha [25] model for a two-component mixture, which gives

$$\ln v_{mix} = x_1 \ln v_1 + x_2 \ln v_2 - 2.303 x_1 x_2 (A + B(x_1 - x_2)) - \ln(x_1 M_1 + x_2 M_2) + x_1 \ln M_1 + x_2 \ln M_2 \quad (19)$$

where  $A$  and  $B$  are interaction parameters. Jouyban et al. [9] proposed a model for correlating the thermal properties of liquid mixtures at various temperatures.

$$\ln v_{mix} = x_1 \ln v_1 + x_2 \ln v_2 + \left( \frac{x_1 x_2}{T} \right) \sum a_i (x_1 - x_2)^i \quad (20)$$

The excess molar volumes, deviations of viscosity, deviations of refractive index, deviation in acoustic impedance, deviation in



ultrasonic velocity, degree of intermolecular attraction, surface tension deviation, isentropic compressibility deviation and change of intermolecular free length were fitted to a Redlich-Kister [10] equation of the type

$$Y = x_1 x_2 \sum A_{i-1} (x_1 - x_2)^{i-1} \quad (21)$$

Where, Y refers to  $V^E$ ,  $\Delta\eta$ ,  $\Delta n_D$ ,  $\Delta Z$ ,  $\Delta u$ ,  $\Delta\beta$ ,  $\delta$ ,  $\Delta L_f$  and  $\Delta\sigma$  and n is the degree of polynomial. Coefficients  $A_i$  was obtained by fitting equation (21) to experimental results using a least squares regression method. In each case, the optimum number of coefficients was ascertained from an examination of the variation in standard deviation (SD).

SD was calculated using the relation:

$$SD = \left[ \frac{\sum (A_{exp} - A_{cal})^2}{(N-n)} \right]^{\frac{1}{2}} \quad (22)$$

Where N is the number of data points and n is the number of coefficients

The measured viscosity data tailored to the McAllister model to derive the binary coefficients. Standard deviations obtained from the fitted outcomes and the calculated data is helpful deliberate mixing behavior of the binary mixtures. It can conclude that in our case, the data found with the values correlated by the corresponding model is very well (Table 6).

Table 7 presents the values of the coefficients optimized to the experimental viscosity over the entire composition range (therefore, they are composition-independent coefficients) for different temperatures and pressures. The values of the standard deviations are also reported in this table. It is observed that they have good agreement with the experimental data. The outcome of the above Table 7 is interaction between benzylparaben and benzene mixture leads to strong dispersive type interaction.

The intermolecular interactions between benzylparaben and benzene mixture leads to strong dispersive type. It is clear that Redlich kister polynomial equation can represent the excess molar volume ( $V^E$ ), viscosity deviation ( $\Delta\eta$ ), refractive index deviation ( $\Delta n_D$ ), ultrasonic velocity deviation ( $\Delta u$ ) intermolecular free length deviation ( $\Delta L_f$ ), acoustical impedance deviation ( $\Delta Z$ ), intermolecular attraction ( $\delta$ ), surface tension deviation ( $\Delta\sigma$ ), and isentropic compressibility deviation ( $\Delta\beta$ ) very well by standard deviation values (Table 8).

It has been concluded that the Jouyban Acree model is very well suited for correlating the thermo physical properties of the studied binary mixture (Table 9). It can conclude that physical intermolecular forces, including electrostatic forces between charged particles and between a permanent dipole and so on induction forces between a permanent dipole and an induced dipole and forces of attraction and repulsion between non polar molecules.

## Conclusion

Experimental data of the density, viscosity, refractive index, ultrasonic velocity and surface tension of benzylparaben and benzene mixture have been measured at 303.15, 308.15 and 313.15 K. These data have been used to compute the excess properties of the system. Negative deviations were observed for  $\Delta L_f$  and  $\delta\beta$ . The positive deviations were observed for  $V^E$ ,  $\Delta\eta$ ,  $\Delta u$ ,  $\Delta\sigma$ ,  $\Delta Z$ ,  $\delta$  and  $\Delta u$ . It may be concluded that the interactions resulting in the interstitial accommodation of benzene in to benzylparaben are the predominant factor over dipole-dipole and dipole induced-dipole interaction. The intermolecular interactions between benzylparaben and benzene mixture leads to strong dispersive type. It is clear that Redlich kister polynomial equation can represent

the excess molar volume ( $V^E$ ), viscosity deviation ( $\Delta\eta$ ), refractive index deviation ( $\Delta n_D$ ), ultrasonic velocity deviation ( $\Delta u$ ) intermolecular free length deviation ( $\Delta L_f$ ), acoustical impedance deviation ( $\Delta Z$ ), intermolecular attraction ( $\delta$ ), surface tension deviation ( $\Delta\sigma$ ), and isentropic compressibility deviation ( $\Delta\beta$ ) very well by standard deviation values. The viscosity data are correlated with the Krishnan-Laddha and McAllister. It has been concluded that the Jouyban Acree model is very well suited for correlating the thermo physical properties of the studied binary mixture.

## Acknowledgment

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