

Refractive Properties of Liquid Binary Mixture at Temperature Range T=298.15-313.15 K

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Abstract

Densities and refractive indices were measured for the binary liquid mixtures formed by benzyl alcohol with benzene at (T=298.15, 303.15, 308.15 and 313.15) K and atmospheric pressure over the whole concentration range. Lorenz-Lorenz mixing rule, Ramaswamy and Anbananthan models and model devised by Gelenski were used to study the associational behaviour and molar refractivity of two weakly interacting liquids. These results have been discussed to study the type of mixing behaviour between the molecules. The measured data were fitted to the Redlich-Kister polynomial relation to estimate the binary coefficients and standard errors. Furthermore, McAllister multi body interaction model is used to correlate the binary refractive index with the experimental findings.

Keywords: Binary liquid, refractive index, Lorenz-Lorenz, molar refractivity, theoretical models, Redlich-Kister

1. INTRODUCTION

Refractive index at different temperatures of liquid mixtures is an important step for their structure and characterization. Along with other thermodynamic data, refractive index values are also useful in engineering calculations. Refractive index is useful in many practical purposes such as, to assess purity of substances, to calculate the molecular electronic polarizability [1], to estimate the boiling point with Meissner's [2] method or to estimate the other thermodynamic properties. In recent past, several workers [3-7] have applied various mixing rules in binary and ternary liquid mixtures.

The studied systems have industrial utility; the mixing behaviour of benzyle alcohol are interesting due to presence of hydroxyl group coupled with benzene. In this work, we present the experimental data on density and refractive index of binary liquid mixtures for benzyl alcohol with benzene at (T=298.15, 303.15 308.15and 313.15) K and atmospheric pressure over the entire composition range. These data were analyzed in terms of Lorenz-Lorenz mixing rule [8], model of Ramaswami and Anbananthan [9] and model suggested by Gelenski [10]. Models [9, 10] (associated) are based on the association constant as an adjustable parameter where as model [8] (non-associated) is based on the additivity of liquids. From these experimental data, deviation in molar refraction (ΔR) have been studied and fitted to a Redlich-Kister type polynomial equation [11] to derive binary coefficients and estimated the standard errors. McAllister multi body interaction model [12] has also been used to study the nature of interactions involved in liquid mixture.

2. EXPERIMENTAL SECTION

2.1 Materials

High purity and AR grade samples of benzyl alcohol and benzene used in this experiment were obtained from Merck Co. Inc., Germany and purified by distillation in which the middle fraction was collected. The liquids were stored in dark bottles over 0.4nm molecular sieves to reduce water content and were partially degassed with a vacuum pump. The purity of each compound was checked by gas chromatography and the results indicated that the mole fraction purity was higher than 0.99. All the materials were used without further purification. The purity of chemicals used was confirmed by comparing the densities and viscosities with those reported in the literature as shown in table 1.

Table 1 Comparison of Density and Refractive index with literature data for pure components at 298.15, 303.15, 308.15 and 313.15 K

Compound	T/K	V/ cm ³ mole ⁻¹	$\rho_{\text{exp}}/\text{g.cm}^3$	* $\rho_{\text{lit}}/\text{g.cm}^3$	n_{exp}	* n_{lit}
Benzene	298.15	89.3196	0.8732	0.8736	1.4984	1.49792
	303.15	89.9366	0.8680	0.8683	1.4941	1.49486
	308.15	90.7306	0.8653	-	1.4801	-
	313.15	91.1329	0.8575	0.8576	1.4785	-
Benzyl alcohol	298.15	103.8210	1.0412	1.0413	1.5378	1.53837
	303.15	104.2413	1.0376	1.0376	1.5362	-
	308.15	105.4509	1.0372	-	1.5340	-
	313.15	107.9780	1.0366	-	1.5317	-

* Ref.18

2.2 Apparatus and Procedure

Before each series of experiments, we calibrated the instrument at atmospheric pressure with doubly distilled water. The calibration was accepted if the measurements were within $\pm 7.9 \cdot 10^{-4} \text{g.cm}^{-3}$ of the published values. The densities of the pure components and their mixtures were measured with the bi capillary pycnometer. The uncertainty in the density measurements was better than $\pm 7.9 \cdot 10^{-4} \text{g.cm}^{-3}$ and reproducible to $\pm 5 \cdot 10^{-4} \text{g.cm}^{-3}$. The liquid mixtures were prepared by mass in an air tight stopped bottle using an electronic balance model SHIMADZU AX-200 accurate to within $\pm 0.1 \text{mg}$. The average uncertainty in the composition of the mixtures was estimated to be less than ± 0.0001 . All molar quantities were based on the IUPAC relative atomic mass table.

Refractive index for sodium D-line was measured using a thermostatically controlled Abbe refractometer (Agato 3T, Japan). Calibration of the instrument was performed with double distilled water. A minimum of three readings were taken for each composition and the average value was considered in all calculations. Refractive index data are accurate to ± 0.0001 units.

3. THEORETICAL

Ramswami and Anbananthan [9] proposed the model based on the assumption of linearity of acoustic impedance with the mole fraction of components. Further Glinski [10] assumed that when solute is added to solvent, the molecules interact according to the equilibrium as;



and the association constant K_{as} can be defined as;

$$K_{as} = \frac{[AB]}{[A][B]} \quad (2)$$

where A is amount of solvent and B is amount of solute in the liquid mixture.

By applying the condition of linearity with composition

$$\eta_{obs} = x_A n_A + x_{AB} n_{AB} \quad (3)$$

where x_A , x_{AB} , n_A and n_{AB} are the mole fraction of A, mole fraction of associate AB, refractive index of A and refractive index of associate AB respectively. The component AB can not be obtained in its pure form. Following simplifications have been made, firstly, concentration term should be replaced by activities for concentrated solution and second, there are also molecules of non associated components in the liquid mixture. The eq (3) takes the form,

$$n_{obs} = [x_A n_A + x_B n_B + x_{AB} n_{AB}] \quad (4)$$

On changing both the adjustable parameters K_{as} and n_{AB} gradually, one can get

different values of the sum of squares of deviations,

$$S = \sum (n_{\text{obs}} - n_{\text{cal}})^2 \quad (8)$$

The minimum value of S can be obtained theoretically by a pair of the fitted parameters. But we found that for some K_{as} and n_{as} , the value of S is high and changes rapidly, and for others, it is low and changes slowly when changing the fitted parameters. In such case, the value of n_{AB} should not be much lower than the lowest observed refractive index of the system or much higher than the highest one. Quantitatively, it should be reasonable to accept the pair of adjustable parameters K_{as} and n_{ab} which has the physical sense and which reproduces the experimental refractive indices satisfactorily.

Gelenski [10] suggested that experimental results with significantly good accuracy can be produced from the following equation as:

$$n_{\text{cal}} = \frac{n_1 n_2}{\phi_1 n_2 + \phi_2 n_1} \quad (9)$$

where n_{cal} is the calculated refractive index, ϕ_1 , ϕ_2 are the volume fraction of component 1 and 2 and n_1 , n_2 are the refractive index of pure components.

Lorentz-Lorenz (L-L) relation has widest application during the evaluation of refractive indices of mixture and density of pure components as well as density of the mixture and represented in terms of specific refraction as:

$$\left[\frac{n_m^2 - 1}{n_m^2 + 2} \right] = \left[\frac{n_1^2 - 1}{n_1^2 + 2} \right] \phi_1 + \left[\frac{n_2^2 - 1}{n_2^2 + 2} \right] \phi_2 \quad (10)$$

here n_m , n_1 , n_2 are the refractive indices of mixture and pure components, 1,2 respectively and ϕ_1, ϕ_2 are the volume fractions of pure components.

4. RESULTS AND DISCUSSION

Table 1 presents the comparison of experimental densities and refractive index of benzene, and benzyl alcohol with literature values at 298.15, 303.15, 308.15 and 313.15 K. Coefficients of the Redlich-Kister polynomials and their standard deviations (σ) are presented in Table 2. Parameters of McAllister three body and four body interaction model and standard deviations for refractive indices are presented in Table 3. Table 4 presents the comparison of average refractive index deviation (Δn_{AV}) and percent average deviation ($\% \Delta n_{\text{AV}}$) obtained from Lorentz-Lorenz mixing rule, Ramaswami and Anbananthan model and model suggested by Gelenski. Mixing data were recorded in Table 5.

Table 2 Coefficients of the Redlich-Kister Equation and Standard Deviations (σ) for Molar Refractivity of Binary Liquid Mixtures at Various Temperatures**benzene+benzylalcohol**

	T	A0	A1	A2	A3	$\sigma \times 10^4$
ΔR	298.15	0.033578	0.068232	0.025245	-0.134	20.59
	303.15	0.035781	0.053327	-0.0058	-0.13114	28.94
	308.15	0.049181	0.09133	0.045692	-0.15151	54.28
	313.15	0.06321	0.090003	0.027498	-0.12614	41.63

Table 3 Parameters of McAllister Three body and Four body Interaction Model and Standard Deviations (σ) for Refractive index of Binary Liquid Mixtures at Various Temperatures**benzene + benzylalcohol**

	298.15	1.538348	1.523292	18.17	1.529790	1.522560	1.524593	17.53
n	303.15	1.528057	1.526193	22.65	1.516193	1.528753	1.520016	22.46
	308.15	1.539904	1.517244	28.52	1.534321	1.505753	1.529106	23.44
	313.15	1.542262	1.514626	35.82	1.529325	1.516063	1.519763	34.89

Table 4 Comparison of Average and Average Percent Refractive Index Deviation (Δn_{av} , % Δn_{av}) computed from all the models at Various Temperatures**benzene + benzylalcohol**

Temperature	K_{as}	n_{ab}	$\Delta n_{av} \times 10^4$ (eq.4)	$\Delta n_{av} \times 10^4$ (eq.10)	$\Delta n_{av} \times 10^4$ (eq.9)	% Δn_{av} (eq.10)	% Δn_{av} (eq.4)	% Δn_{av} (eq.9)
298.15	0.000135	1.5179	69.0930	65.0193	64.8058	0.42	0.46	0.42
303.15	0.000136	1.5159	61.9600	56.4439	56.3326	0.37	0.40	0.37
308.15	0.000137	1.5155	108.5830	100.4258	10.0511	0.66	0.71	0.66
313.15	0.000119	1.5140	128.9990	118.8113	119.1049	0.78	0.85	0.78

Table 5 Volume fraction (Φ), Experimental Densities (ρ), Observed Molar Refractivity (R_{Exp}), Theoretical Molar Refractivity(R_{Theo}) and Percent Refractive Index Deviation ($\% \Delta n$) obtained from different Models for Binary Liquid Mixtures at various temperatures.

benzene + benzylalcohol

T=298.15K

ϕ_1	$\rho/g.cc$	n_{exp}	R_{exp}	$R_{Eq.10}$	$R_{Eq.4}$	$R_{Eq.9}$	$\% \Delta n$ Eq.10	$\% \Delta n$ Eq.4	$\% \Delta n$ Eq.9
0.1542	1.0008	1.5199	31.2487	31.0279	31.0092	31.0287	0.2873	0.3116	0.2863
0.2909	0.9935	1.5189	30.0699	29.7905	29.7626	29.7917	0.3770	0.4146	0.3753
0.4129	0.9825	1.5172	29.1490	28.8579	28.8266	28.8593	0.4039	0.4473	0.4020
0.5224	0.9601	1.5163	28.7457	28.4117	28.3804	28.4131	0.4691	0.5129	0.4671
0.6214	0.9445	1.5200	28.4655	27.8876	27.8590	27.8889	0.8233	0.8638	0.8214
0.7110	0.9236	1.5145	28.0189	27.6256	27.6011	27.6267	0.5647	0.5998	0.5631
0.7929	0.9001	1.5120	27.8759	27.5328	27.5135	27.5337	0.4932	0.5208	0.4919
0.8677	0.8935	1.5099	27.3063	27.0057	26.9926	27.0063	0.4396	0.4587	0.4387
0.9366	0.8801	1.5021	26.7500	26.7492	26.7426	26.7495	0.0011	0.0109	0.0006

T=303.15K

ϕ_1	$\rho/g.cc$	n_{exp}	R_{exp}	$R_{Eq.10}$	$R_{Eq.4}$	$R_{Eq.9}$	$\% \Delta n$ Eq.10	$\% \Delta n$ Eq.4	$\% \Delta n$ Eq.9
0.1546	0.9998	1.5178	31.1738	30.9700	30.9483	30.9705	0.2649	0.2931	0.2642
0.2915	0.9874	1.517	30.1625	29.8642	29.8316	29.8650	0.3999	0.4435	0.3988
0.4136	0.9754	1.5162	29.3134	28.9398	28.9032	28.9407	0.5144	0.5646	0.5132
0.5231	0.9564	1.5145	28.7722	28.3771	28.3407	28.3780	0.5526	0.6033	0.5513
0.6220	0.9354	1.5101	28.2813	27.9994	27.9660	28.0003	0.3982	0.4454	0.3970
0.7116	0.9002	1.5099	28.5305	28.1678	28.1387	28.1686	0.5076	0.5482	0.5065
0.7934	0.8897	1.5102	28.1181	27.6680	27.6454	27.6686	0.6390	0.6710	0.6381
0.8680	0.8845	1.4989	27.0802	27.0853	27.0700	27.0857	-0.0074	0.0147	-0.0080
0.9368	0.8789	1.4965	26.5332	26.5829	26.5752	26.5831	-0.0734	-0.0620	-0.0737

T=308.15K

ϕ_1	$\rho/g.cc$	n_{exp}	R_{exp}	$R_{Eq.10}$	$R_{Eq.4}$	$R_{Eq.9}$	% Δn Eq.10	% Δn Eq.4	% Δn Eq.9
0.1542	0.9875	1.5170	31.5211	31.2338	31.2031	31.2335	0.3686	0.4079	0.3690
0.2909	0.9756	1.5165	30.5025	30.0465	30.0005	30.0461	0.6033	0.6640	0.6039
0.4129	0.9632	1.5152	29.6363	29.0792	29.0275	29.0787	0.7563	0.8262	0.7571
0.5224	0.9426	1.5100	28.9782	28.5216	28.4700	28.5211	0.6287	0.6996	0.6295
0.6214	0.9254	1.5099	28.5775	27.9930	27.9456	27.9924	0.8151	0.8809	0.8159
0.7111	0.8992	1.5089	28.5150	27.8523	27.8115	27.8518	0.9241	0.9807	0.9248
0.7930	0.8745	1.5099	28.5927	27.7670	27.7347	27.7666	1.1488	1.1934	1.1493
0.8677	0.8645	1.4998	27.7490	27.3036	27.2816	27.3034	0.6291	0.6602	0.6295
0.9366	0.8566	1.4900	26.9208	26.8436	26.8325	26.8435	0.1107	0.1267	0.1109

T=313.15K

ϕ_1	$\rho/g.cc$	n_{exp}	R_{exp}	$R_{Eq.10}$	$R_{Eq.4}$	$R_{Eq.9}$	% Δn Eq.10	% Δn Eq.4	% Δn Eq.9
0.1517	0.9745	1.5150	31.8376	31.5232	31.4835	31.5221	0.3980	0.4481	0.3994
0.2869	0.9683	1.5142	30.6173	30.1205	30.0612	30.1188	0.6521	0.7297	0.6543
0.4082	0.9512	1.5132	29.9121	29.2696	29.2024	29.2676	0.8608	0.9504	0.8635
0.5176	0.9354	1.5101	29.2061	28.5431	28.4762	28.5411	0.9046	0.9954	0.9073
0.6168	0.9102	1.5098	29.0499	28.2404	28.1783	28.2385	1.1086	1.1930	1.1111
0.7071	0.8754	1.5076	29.2271	28.3658	28.3116	28.3641	1.1676	1.2405	1.1698
0.7898	0.8563	1.5089	29.1521	28.0945	28.0518	28.0932	1.4384	1.4959	1.4402
0.8655	0.8365	1.4887	28.1361	27.9365	27.9070	27.9356	0.2730	0.3134	0.2743
0.9354	0.8236	1.4865	27.8291	27.6228	27.6077	27.6223	0.2842	0.3049	0.2849

Ramaswami and Abanathan model has been extended and corrected for the prediction of refractive index of weak interacting binary mixtures which was originally derived for the prediction of acoustical impedance. The results of fittings obtained from the model were utilized properly. The basic doubt regarding this model except the assumption of linearity of refractive index with mole fraction is that these liquids have poor tendency to form dimmers. Calculations were performed using a computer program which allows easily both the adjustable parameters simultaneously or the parameters were changed, manually.

We constructed the data sheet in a computer program, with association constant K_{as} and C_{AB} as the fitted parameters (C_{AB} is the refractive index in the pure component

AB means a hypothetical liquid having only the associate A-B. On changing these parameters, the equilibrium concentrations of species [A], [B] and [AB] will change and the refractive index can be computed. The difference between experimental and theoretical refractive index is used to obtain the sum of squares of deviations. It is assumed that in solution three associates are formed instead of two (pure A, pure B and AB). The values of refractive index in pure associate can be treated as a fitted one with the value of K_{as} . The mixing function, ΔR was represented mathematically by the Redlich-Kister equation [11] for correlating the experimental data as:

$$y = x_1(1-x_1) \sum_{i=0}^p A_i (2x_1 - 1)^i \quad (11)$$

where y refers to ΔR , x_1 is the mole fraction and A_i is the coefficients. The values of coefficients A_i were determined by a multiple regression analysis based on the least squares method and are summarized along with the standard deviations between the experimental and fitted values of the respective function in Table 2. The standard deviation is defined by.

$$\sigma = \left[\sum_{i=1}^m (y_{exp_i} - y_{cal_i})^2 / (m - p) \right]^{1/2} \quad (12)$$

where m is the number of experimental points and p is the number of adjustable parameters. For the case the σ values lie between 3.35×10^{-4} to 54.28×10^{-4} and the largest σ value corresponds to benzene + benzyl alcohol mixture at 308.15 K.

Molar refractivity was obtained from refractive index data according to following expression:

$$R = [n^2 - 1 / n^2 + 2] M / \rho \quad (13)$$

where M is the mean molecular weight of the mixture and ρ is the mixture density.

The molar refractivity deviation function has been calculated by the following expression:

$$\Delta R = R - \Phi_1 R_1 - \Phi_2 R_2 \quad (14)$$

where Φ_1 and Φ_2 are volume fraction $\Phi_i = (\sum x_i v_i / \sum^2 x_i v_i)$ was used [13-15]. There is no general rule that states how to calculate a refractivity function. Konti et al [16] reported deviations in molar refractivity with volume fraction referring to the Lorenz-Lorenz mixing rules.

McAllister multibody interaction model [12] which is based on Eyring's theory of absolute reaction rates and for liquids the free energy of activation are additive on a number fraction and that interactions of like and unlike molecules. The three body model is defined as:

$$\begin{aligned} \ln n = & x_1^3 \ln n_1 + 3x_1^3 x_2 \ln a + 3x_1^3 x_2^2 \ln b + x_1^3 \ln n_2 - \\ & \ln(x_1 + x_2 M_2 / M_1) + 3x_1^2 x_2 \ln(2 + M_2 / M_1) / 3 + \\ & 3x_1 x_2^2 \ln[(1 + 2M_2 / M_1) / 3] + x_2^3 \ln(M_2 / M_1) \end{aligned} \quad (15)$$

and the four body model is given by

$$\begin{aligned} \ln n = & x_1^4 \ln n_1 + 4x_1^3 x_2 \ln a + 6x_1^2 x_2^2 \ln b \\ & 4x_1 x_2^3 \ln c + x_2^3 \ln \eta_2 - \ln(x_1 + x_2 M_2 / M_1)] + \\ & 4x_1^3 x_2 \ln[(3 + M_2 / M_1) / 4] + 6x_1^2 x_2^2 \ln(1 + M_2 / M_1) / 2] + \\ & 4x_1 x_2^3 \ln[(1 + 3M_2 / M_1) / 4] + x_2^4 \ln(M_2 / M_1) \end{aligned} \quad (16)$$

where n is the refractive index of the mixture and x_1 , n_1 , M_1 , x_2 , n_2 and M_2 are the mole fractions, refractive index, and molecular weights of pure components 1 and 2 respectively; a , b and c are adjustable parameters that are characteristic of the system. In the above eqs. (15) and (16) the coefficient a , b and c have been calculated using the least squares procedure. The estimated parameters of the refractive index equations and the standard deviations, σ , between the calculated and experimental values are given in Table 3. It is observed that the four body model of the McAllister equation correlated the refractive index of the mixture to a significantly higher degree of accuracy for the systems than does the three body model. Furthermore, the values of the McAllister parameters have shown a decreasing tendency with rise in temperature. Generally, McAllister's models are adequate in correlation for those systems as evidenced by small deviations.

With the increase of volume fraction (Φ), the values of refractive index obtained from all the models decrease at all temperatures except in few places. Results of molar refractivity computed from eq 13 for the systems show regular trend except in few places as shown in Table 5. The minimum and maximum percent deviation ($\% \Delta n_{AV}$) in refractive index are 0.001 at 298.15 K for benzene + benzyle alcohol respectively. The results of deviation in molar refraction, ΔR , plotted as a function of Φ_i of benzene + benzyle alcohol mixtures at 298.15, 303.15, 308.15 and 313.15 K were displayed in the Figure 1. In all the cases, theoretical molar refractivity computed from all the models agree well within the experimental precision. The trend in all the figures is almost similar and negative. Molar refraction increases with molecular weight for all the systems. Density and refractive index depend on molecular weight and nature of solution and values decrease with increase of temperature as displayed in Table 5.

Finally, it can be concluded that the expressions used for the interpolating the experimental data measured in this work gave good results as can be seen by inspecting the σ values obtained. All the models based on associated processes and non-associated processes give more reliable results and helpful in deducing the internal structure of associates through the fitted values of refractive index.

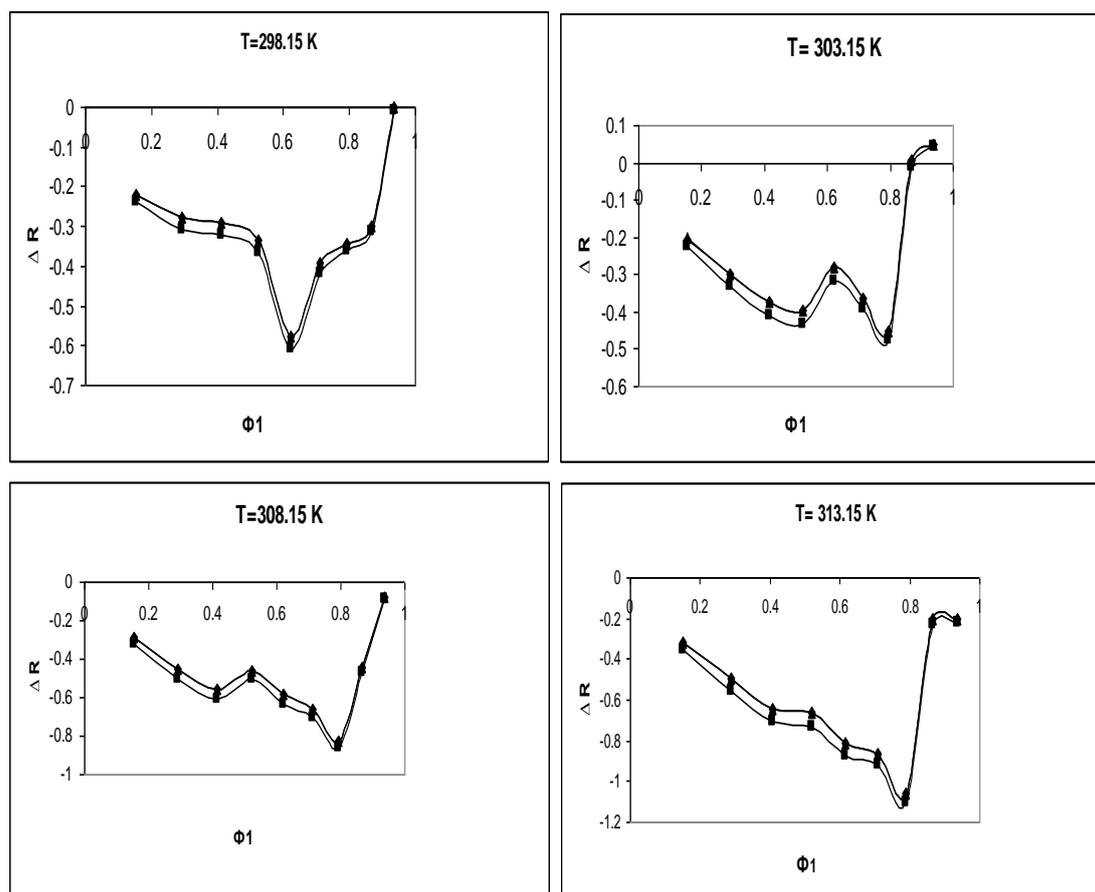


Figure 1 Plot of molar refractivity deviation, ΔR , with volume fraction, Φ_1 for Φ benzene+ (1- Φ) benzylalcohol at 298.15, 303.15, 308.15 and 313.15 K: \blacklozenge Lorenz-Lorenz mixing rule (Eq 10), \blacksquare , Ramaswami Anbananthan model (Eq. 4), \blacktriangle , model devised by Gelenski (Eq.9)

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