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Refractive Indices and Related Excess Properties of Binary Mixtures of Ricinoleic Acid with some Halobenzenes

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ABSTRACT

Refractive indices, (n_D) of binary liquid mixtures of ricinoleic acid with some halobenzenes (Fluorobenzene, Chlorobenzene and Bromobenzene) at 303.15, 308.15 and 313.15 K have been measured over the entire composition range. From the experimentally measured values, deviation in refractive index, (Δn_D), and molar refraction, (ΔR_m), have been computed and fitted to the Redlich-Kister polynomial equation to obtain the coefficients and standard errors. Using nine mixing rules namely Arago-Boit, Dale-Gladstone, Lorentz-Lorentz, Eykman, Weiner, Heller, Newton, Oster and Eyring-John refractive indices of the binary liquid mixtures have been calculated theoretically and the results are discussed in terms of the molecular interactions between the mixing components. Results reveal that strong interactions are observed between these selected binaries.

Keywords: Refractive index, molar refraction, ricinoleic acid, halobenzenes.

INTRODUCTION

Refractive index measurements in binary and multicomponent liquid mixtures have been reported by several workers[1-4]. The refractive index, (n_D), of a material is defined as the ratio of the velocity of light in a vacuum to the velocity of light in the material, and therefore, for a liquid it is greater than unity. The refractive index is a thermodynamic property and is a state function, which is for a pure liquid depends on temperature and pressure[5]. Also, where measurement of other bulk thermodynamic properties is time consuming, it is convenient, and accurately measurable quantity and a good tool for the study of molecular interactions occurring between the component molecules of a liquid mixture[6]. Ricinoleic acid (R12-Hydroxy-9-*cis*-octadecenoic acid) is a fatty acid. It is an unsaturated omega-9 fatty acid. It is a major component of the seed oil obtained from mature castor plant seeds. About 90% of the fatty acid content in castor oil is the triglyceride formed from ricinoleic acid. Literature survey indicates that there are no information is available regarding behavior of ricinoleic acid with organic solvents. So, it was decided to study refractometric behaviour of the binary mixtures of ricinoleic acid with halobenzenes.

Chemical Name	T/K	n_D	
		exp.	lit.
Ricinoleic acid	303.15	1.4693	-
	308.15	1.4685	-
	313.15	1.4667	-
Fluorobenzene	303.15	1.4617	1.4617[22]
	308.15	1.4596	1.4594[22]
	313.15	1.4569	1.4563[22]
Chlorobenzene	303.15	1.5167	1.5210[20]
	308.15	1.5158	1.5166[21]
	313.15	1.5151	1.5150[20]
Bromobenzene	303.15	1.5561	1.5542[21]
	308.15	1.5531	1.5514[21]
	313.15	1.5500	1.5500[20]

MATERIALS AND METHODS

In this study, all organic components used were of analytical grade and were supplied by the reputed companies. Ricinoleic acid (95% pure, supplies by Royal castor products limited, India) and halobenzenes namely Fluorobenzene, Chlorobenzene and Bromobenzene (99.5% pure, supplied by S.D. Fine chemicals, India) were used after purification using standard methods. The experimental values and literature values of refractive indices of used compounds are presented in table. 1 and all these values are found in close proximity.

Procedure: Binary mixtures are prepared by mixing appropriate amount of two components. The uncertainty in mole fraction of the mixtures was estimated to be ± 0.002. To avoid losses of solvents due to evaporation specially designed ground glass ampoules were used and were placed in dark place to avoid any photolytic effect on binary mixtures. Electronic balance (Reptech RA-2012) with accuracy of weight ± 0.001 g was used for measurements. For measurement of refractive indexes of pure components and binary mixtures for sodium D-line were measured by thermostated Abbe's Refractometer SER. No. 995033. The Refractometer was calibrated using 1-bromonaphthalene, methanol and double distilled water. A water pump was attached to Refractometer for circulating water through Refractometer to maintain the temperature. A thermostatic water bath (Model No. 14L-SS supplied by Equiptron, India) having accuracy of ± 0.01°C with range of 20°C to 90°C has been used for this study.

Table 1. Comparison of the refractive indices (n_D) data of ricinoleic acid + halobenzenes (Fluorobenzene, Chlorobenzene and Bromobenzene)

Chemical Name	T/K	n_D	
		exp.	lit.
Ricinoleic acid	303.15	1.4693	-
	308.15	1.4685	-
	313.15	1.4667	-
Fluorobenzene	303.15	1.4617	1.4617[22]
	308.15	1.4596	1.4594[22]
	313.15	1.4569	1.4563[22]
Chlorobenzene	303.15	1.5167	1.5210[20]
	308.15	1.5158	1.5166[21]
	313.15	1.5151	1.5150[20]
Bromobenzene	303.15	1.5561	1.5542[21]
	308.15	1.5531	1.5514[21]
	313.15	1.5500	1.5500[20]

RESULTS AND DISCUSSION

Mixing rules of refractive index: Following nine equations were used for quantitative determination of refractive index of binary solution by different scientists.

Arago-biot (A-B) [7]

$$n_D = n_{D_1}\phi_1 + n_{D_2}\phi_2 \quad (1)$$

Dale-Glastone (D-G) [8]

$$n_D - 1 = (n_{D_1} - 1)\phi_1 + (n_{D_2} - 1)\phi_2 \quad (2)$$

Lorentz-lorenz (L-L) [9]

$$\frac{n_D^2-1}{n_D^2+2} = \left(\frac{n_{D_1}^2-1}{n_{D_1}^2+2}\right)\phi_1 + \left(\frac{n_{D_2}^2-1}{n_{D_2}^2+2}\right)\phi_2 \quad (3)$$

Eykman (Eyk) [10]

$$\frac{n_D^2-1}{n_D^2+0.4} = \left(\frac{n_{D_1}^2-1}{n_{D_1}^2+0.4}\right)\phi_1 + \left(\frac{n_{D_2}^2-1}{n_{D_2}^2+0.4}\right)\phi_2 \quad (4)$$

Weiner (w) [11]

$$\frac{n_D^2-n_{D_1}^2}{n_D^2+2n_{D_1}^2} = \left(\frac{n_{D_2}^2-n_{D_1}^2}{n_{D_2}^2+2n_{D_1}^2}\right)\phi_2 \quad (5)$$

Heller (Hr) [12]

$$\frac{n_D-n_{D_1}}{n_{D_1}} = \left(\frac{(n_{D_2}-n_{D_1})^2-1}{(n_{D_2}-n_{D_1})^2+2}\right)\phi_2 \quad (6)$$

Newton (Nw) [13]

$$n_{D_1}^2 - 1 = (n_{D_1}^2 - 1)\phi_1 + (n_{D_2}^2 - 1)\phi_2 \quad (7)$$

Oster (Os) [14]

$$\left[\frac{(n_D^2-1)(2n_D^2+1)}{n_D^2}\right]V = \left[\frac{(n_{D_1}^2-1)(2n_{D_1}^2+1)}{n_{D_1}^2}\right]x_1v_1 + \left[\frac{(n_{D_2}^2-1)(2n_{D_2}^2+1)}{n_{D_2}^2}\right]x_2v_2 \quad (8)$$

Eyring and John (EJ) [15]

$$n_D = n_{D_1}\phi_1^2 + 2(n_{D_1}n_{D_2})^{1/2}\phi_1\phi_2 + n_{D_2}\phi_2^2 \quad (9)$$

In all these equations, ϕ_1, ϕ_2 and n_{D_1}, n_{D_2} are volume fraction and refractive index of pure components 1 and 2 respectively.

The values of average deviation in refractive index (n_D) from nine mixing rules for all binaries at three temperatures are listed in table 2. All mixing relations reveal deviation up to third or fourth digit after decimal for all four binary mixtures at three temperatures. It should be noted that the calculated average deviation between the Arago-Biot and Dale-Gladstone relations are exactly same for all four binary mixtures at three temperatures. All nine relations show good agreement as for the deviation values are concerned for all these binaries.

Table 2. Average deviation (σ) in the refractive index from nine different mixing relations for ricinoleic acid + halobenzenes (Fluorobenzene, Chlorobenzene and Bromobenzene)

Parameters	*T*/(K)		
	303.15	308.15	313.15
	Ricinoleic acid + Fluorobenzene		
A–B	0.001470909	0.001103636	0.000254545
G–D	0.001470909	0.001103636	0.000254545
L–L	0.001474076	0.001107975	0.000259799
WR	0.001445687	0.00106903	0.000212535

Heller	0.001474162	0.001108095	0.000259953
Newton	0.001467953	0.001099579	0.000249618
Eyring–John	0.001472387	0.001105665	0.000257009
Eykman	0.001477921	0.001113257	0.00026622
Oster	0.001469109	0.001101172	0.000251563
Ricinoleic acid + Chlorobenzene			
A–B	0.001145455	0.000713636	-0.000281818
G–D	0.001145455	0.000713636	-0.000281818
L–L	0.001270527	0.000838125	-0.000151561
WR	0.000164073	-0.000264142	-0.001306853
Heller	0.001278203	0.000845889	-0.00014304
Newton	0.00103259	0.000601184	-0.000399661
Eyring–John	0.00120189	0.000769866	-0.000222893
Eykman	0.001415347	0.000982478	-0.000000193
Oster	0.001073886	0.000642414	-0.000356322
Ricinoleic acid + Bromobenzene			
A–B	0.000118182	-0.000427273	-0.002040909
G–D	0.000118182	-0.000427273	-0.002040909
L–L	0.000541758	-0.0000252	-0.001651644
WR	-0.003171169	-0.003553826	-0.0050759
Heller	0.000577366	0.00000845	-0.001618335
Newton	-0.000255364	-0.00078257	-0.00238593
Eyring–John	0.000304995	-0.000249588	-0.001868364
Eykman	0.001016351	0.000426571	-0.001212326
Oster	-0.000124929	-0.000657945	-0.002264196

Deviation in refractive index (Δn_D), and deviation in molar refraction (ΔR_m): Table 3 represents the data of refractive indexes (n_D) of pure components and their binary mixtures at 303.15, 308.15 and 313.15 K. Deviation in refractive index (Δn_D) and deviation in molar refraction (ΔR_m) are calculated using following equations and their values are also reported in tables 3and 4 respectively.

$$\Delta n_D = n_{D_{12}} - \sum_{i=1}^{2} \phi_i \, n_{D_i} \quad (10)$$

$$\Delta R_m = R_{m_{12}} - \sum_{i=1}^{2} R_{m_i} n_{D_i} \quad (11)$$

where, ϕ_i, n_{D_i} and R_{m_i} are volume fraction, refractive index and molar refraction of i[th] component. $n_{D_{12}}$ and $R_{m_{12}}$ are refractive index and molar refraction of the mixture. ϕ_i and R_{m_i} can be calculated using following equations:

$$\phi_i = \frac{x_i v_i}{\sum_{i=1}^{2} x_i v_i} \quad (12)$$

$$R_{m_i} = \frac{n_{D_i}^2 - 1}{n_{D_i}^2 + 2} v_i \quad (13)$$

$$v_i = \frac{M_i}{\rho_i} \quad (14)$$

where, x_i, v_i and M_i are mole fraction, molar volume and molecular mass of i[th] component.

Table 3. Refractive index, (n_D), and deviation in refractive index, (Δn_D) for ricinoleic acid + halobenzenes (Fluorobenzene, Chlorobenzene and Bromobenzene)

ϕ_1	n_D			Δn_D		
	303.15 K	**308.15 K**	**313.15 K**	**303.15 K**	**308.15 K**	**313.15 K**
Ricinoleic acid + Fluorobenzene						
0.0000	1.4617	1.4596	1.4569	0.00000	0.00000	0.00000
0.1000	1.4635	1.4613	1.4580	0.00104	0.00076	0.00012

0.2000	1.4650	1.4627	1.4591	0.00174	0.00131	0.00025
0.3000	1.4661	1.4639	1.4602	0.00212	0.00159	0.00036
0.4000	1.4670	1.4649	1.4612	0.00227	0.00174	0.00042
0.5000	1.4679	1.4659	1.4623	0.00240	0.00189	0.00047
0.6000	1.4685	1.4667	1.4632	0.00227	0.00171	0.00043
0.7000	1.4689	1.4673	1.4641	0.00192	0.00147	0.00034
0.8000	1.4693	1.4678	1.4650	0.00152	0.00108	0.00026
0.9000	1.4694	1.4682	1.4659	0.00090	0.00059	0.00015
1.0000	1.4693	1.4685	1.4667	0.00000	0.00000	0.00000
			Ricinoleic acid + Chlorobenzene			
0.0000	1.5167	1.5158	1.5151	0.00000	0.00000	0.00000
0.1000	1.5128	1.5115	1.5104	0.00084	0.00043	0.00014
0.2000	1.5085	1.5071	1.5053	0.00128	0.00076	-0.00012
0.3000	1.5041	1.5026	1.5001	0.00162	0.00099	-0.00048
0.4000	1.4995	1.4980	1.4950	0.00176	0.00112	-0.00074
0.5000	1.4949	1.4934	1.4902	0.00190	0.00125	-0.00070
0.6000	1.4900	1.4886	1.4856	0.00174	0.00118	-0.00046
0.7000	1.4850	1.4837	1.4809	0.00148	0.00101	-0.00032
0.8000	1.4800	1.4787	1.4761	0.00122	0.00074	-0.00028
0.9000	1.4748	1.4736	1.4714	0.00076	0.00037	-0.00014
1.0000	1.4693	1.4685	1.4667	0.00000	0.00000	0.00000
			Ricinoleic acid + Bromobenzene			
0.0000	1.5561	1.5531	1.5500	0.00000	0.00000	0.00000
0.1000	1.5474	1.5441	1.5391	-0.00002	-0.00054	-0.00257
0.2000	1.5386	1.5352	1.5293	-0.00014	-0.00098	-0.00404
0.3000	1.5298	1.5269	1.5205	-0.00026	-0.00082	-0.00451
0.4000	1.5213	1.5186	1.5128	-0.00008	-0.00066	-0.00388
0.5000	1.5127	1.5103	1.5054	0.00000	-0.00050	-0.00295
0.6000	1.5042	1.5020	1.4980	0.00018	-0.00034	-0.00202
0.7000	1.4959	1.4935	1.4904	0.00056	-0.00038	-0.00129
0.8000	1.4872	1.4851	1.4826	0.00054	-0.00032	-0.00076
0.9000	1.4785	1.4768	1.4746	0.00052	-0.00016	-0.00043
1.0000	1.4693	1.4685	1.4667	0.00000	0.00000	0.00000

Table 4. Molar refraction (R_m) and deviation in molar refraction, (ΔR_m) for ricinoleic acid + halobenzenes (Fluorobenzene, Chlorobenzene and Bromobenzene)

ϕ_1	R_m			ΔR_m / (cm^3 mol^{-1})		
	303.15 K	**308.15 K**	**313.15 K**	**303.15 K**	**308.15 K**	**313.15 K**
			Ricinoleic acid + Fluorobenzene			
0.0000	1.4617	1.4596	1.4569	0.00000	0.00000	0.00000
0.1000	1.4635	1.4613	1.4580	-4.21263	-4.23737	-4.27267
0.2000	1.4650	1.4627	1.4591	-8.12834	-8.17146	-8.23393
0.3000	1.4661	1.4639	1.4602	-11.64057	-11.70031	-11.77956
0.4000	1.4670	1.4649	1.4612	-14.61937	-14.68950	-14.78327
0.5000	1.4679	1.4659	1.4623	-16.88578	-16.96381	-17.07545
0.6000	1.4685	1.4667	1.4632	-18.19698	-18.28702	-18.39997
0.7000	1.4689	1.4673	1.4641	-18.10960	-18.19624	-18.31084
0.8000	1.4693	1.4678	1.4650	-15.97842	-16.06660	-16.16415
0.9000	1.4694	1.4682	1.4659	-10.69375	-10.76049	-10.82258
1.0000	1.4693	1.4685	1.4667	0.00000	0.00000	0.00000
			Ricinoleic acid + Chlorobenzene			
0.0000	1.5167	1.5158	1.5151	0.00000	0.00000	0.00000

0.1000	1.5128	1.5115	1.5104	-3.75963	-3.78676	-3.79622
0.2000	1.5085	1.5071	1.5053	-7.17794	-7.21852	-7.25899
0.3000	1.5041	1.5026	1.5001	-10.21736	-10.27242	-10.35126
0.4000	1.4995	1.4980	1.4950	-12.80699	-12.87040	-12.98178
0.5000	1.4949	1.4934	1.4902	-14.72618	-14.79800	-14.92746
0.6000	1.4900	1.4886	1.4856	-15.76175	-15.83337	-15.95235
0.7000	1.4850	1.4837	1.4809	-15.58290	-15.65174	-15.75931
0.8000	1.4800	1.4787	1.4761	-13.63193	-13.70604	-13.80135
0.9000	1.4748	1.4736	1.4714	-9.00485	-9.06954	-9.12396
1.0000	1.4693	1.4685	1.4667	0.00000	0.00000	0.00000
			Ricinoleic acid + Bromobenzene			
0.0000	1.5561	1.5531	1.5500	0.00000	0.00000	0.00000
0.1000	1.5474	1.5441	1.5391	-3.46475	-3.50530	-3.61957
0.2000	1.5386	1.5352	1.5293	-6.74471	-6.81874	-7.00704
0.3000	1.5298	1.5269	1.5205	-9.62191	-9.69251	-9.94032
0.4000	1.5213	1.5186	1.5128	-11.97483	-12.05896	-12.29823
0.5000	1.5127	1.5103	1.5054	-13.74167	-13.83022	-14.03414
0.6000	1.5042	1.5020	1.4980	-14.62788	-14.72617	-14.88539
0.7000	1.4959	1.4935	1.4904	-14.36400	-14.50949	-14.61100
0.8000	1.4872	1.4851	1.4826	-12.59112	-12.73456	-12.79421
0.9000	1.4785	1.4768	1.4746	-8.25454	-8.37473	-8.41684
1.0000	1.4693	1.4685	1.4667	0.00000	0.00000	0.00000

The experimentally determined values of Δn_D, and ΔR_m for binary mixtures were fitted to a following Redlich-Kisters' equation[16],

$$\Delta_{12} = x_1 x_2 \sum_{i=0}^{i-n} A_i \, (2x_1 - 1)^i \qquad (15)$$

Where, Δ_{12} is given excess or Deviation function, X_1 and X_2 are the mole fraction of component 1 and 2 respectively. The coefficients A_i were determined by a multiple regression analysis based on least square method. The number of coefficient for a given set of experimental point of any function were optimized by keeping the ratio between experimental points and number of coefficient to a maximum values which in case was not less than 3.

The standard deviation (σ) was calculated using following expression,

$$\sigma = [\sum(\Delta_{obs} - \Delta_{cal})^2/(N-n)]^{1/2} \qquad (16)$$

Where, N is the number of measurement and n is the number of parameters. Fitting coefficients, A_i, and the standard deviations, σ, are represented in Table 5.

It can be observed from table 3 and fig. 1, deviation in refractive index (Δn_D) values are positive for ricinoleic acid + halobenzenes binaries over the entire composition range except for bromobenzene binaries where small negative values are obtained in bromobenzene rich region at 303.15, 308.15 and 313.15K. Δn_Dvalues decrease with increase in temperature. This trend can be understood by the fact that (i) the kinetic energy of molecules increase due to increase in temperature, the molecules will try to orient at large distances from each other (ii) the declustering of component is enhanced with increase of temperature (iii) also weakening of dipole-dipole interactions leads to decrease in polarizability due to increase of temperature. The maxima for Δn_D occur at 0.50 volume fraction of Fluro and chloro binaries of ricinoleic acid at all considered temperatures. According to Campos et. al. and brocos et. al., the positive values of Δn_D indicate for the presence of strong intermolecular interaction between components of binary mixtures [17,18]. From data it can be concluded that interaction become weaker when binary mixtures change from fluorobenzene to bromobenzene. The order of intermolecular interactions of ricinoleic acid

with halobenznes on the basis of Δn_D study is as under: Fluorobenzene > + Chlorobenzene > + Bromobenzene

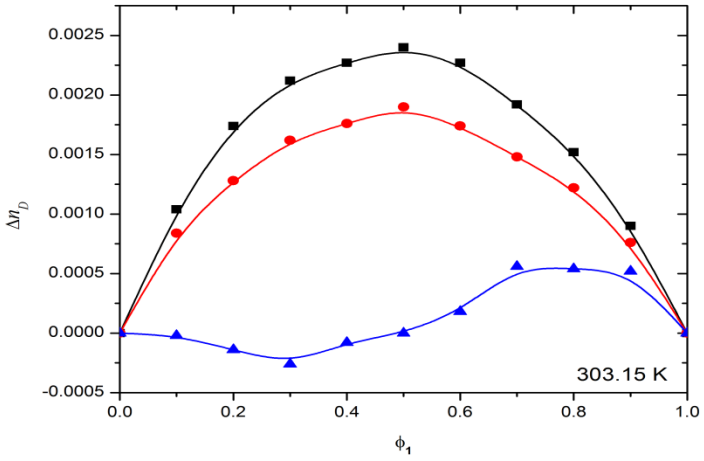

Figure. 1 Deviation in Refractive Index, Δn_D, as a function of volume fraction for the system Ricinoleic acid + Fluorobenzene, (■); Chlorobenzene, (●); Bromobenzene, (▲); at T = 303.15KC

Moreover, deviation in molar refraction (ΔR_m) values are found to be negative to small negative from fluorobenzene to bromobenzene and with rise of temperature negative values becomes more negative. According to Baragi et. al and Brocos et al., the negative value of deviation in molar refraction (ΔR_m) indicates for the strong intermolecular interaction between binaries components [18,19]. ΔR_m values support the idea that interactions become weaker from fluorobenzene to bromobenzene binaries. The values of molar refraction (R_m) and deviation in molar refraction (ΔR_m) is presented in Table 4. The graphical representation of ΔR_m is shown in fig. 2.

Table 5. Fitting coefficients, A_i, and the standard deviations, σ, determined for Δn_D *and* ΔR_m for ricinoleic acid + halobenzenes

Excess Properties	*T*/K	A_0	A_1	A_2	A_3	A_4	σ
				Ricinoleic acid + Fluorobenzene			
Δn_D	303.15	0.0094452200	-0.0010323300	0.0014908300	0.0000240083	0.0009713230	0.000037
	308.15	0.0073064000	-0.0007058770	0.0001586040	-0.0008236920	0.0002698470	0.000037
	313.15	0.0018282800	-0.0001385630	-0.0009833610	0.0005358020	0.0007486220	0.000010
ΔR_m (cm^3 mol^{-1})	303.15	-67.5926	-36.1842	-18.7805	-13.6914	-7.7975	0.0219
	308.15	-67.9116	-36.3318	-18.9669	-13.9163	-7.9693	0.0226
	313.15	-68.3435	-36.5341	-19.1614	-13.8877	-7.9338	0.0225
				Ricinoleic acid + Chlorobenzene			
Δn_D	303.15	0.007410950	-0.000488099	-0.000751462	-0.000042380	0.004813690	0.00003
	308.15	0.004876860	0.000376533	-0.000552616	-0.001256990	-0.000171631	0.00002
	313.15	-0.002687610	0.001766010	0.004285120	-0.005993300	-0.000181242	0.00005
ΔR_m (cm^3 mol^{-1})	303.15	-58.9130	-30.2957	-14.7341	-9.5331	-6.2754	0.0091
	308.15	-59.1949	-30.3415	-14.7440	-9.8704	-6.8196	0.0085

	313.15	-59.6911	-30.4924	-14.4920	-10.1398	-6.8756	0.0063
				Ricinoleic acid + Bromobenzene			
	303.15	0.0001047560	0.0041145600	0.0025881200	-0.0006588700	0.0024245100	0.000064
Δn_D	308.15	-0.0017755700	0.0033460400	-0.0093099200	-0.0009471780	0.0093045700	0.00005
	313.15	-0.0119103000	0.0202102000	-0.0112718000	-0.0083853600	0.0060993700	0.00004
ΔR_m	303.15	-54.8628	-26.7797	-14.1979	-10.1273	-2.8705	0.0233
(cm^3 mol^{-1})	308.15	-55.2100	-27.0400	-15.2408	-10.5667	-2.5723	0.0199
	313.15	-56.0511	-26.1070	-14.9129	-11.2461	-3.1398	0.0176

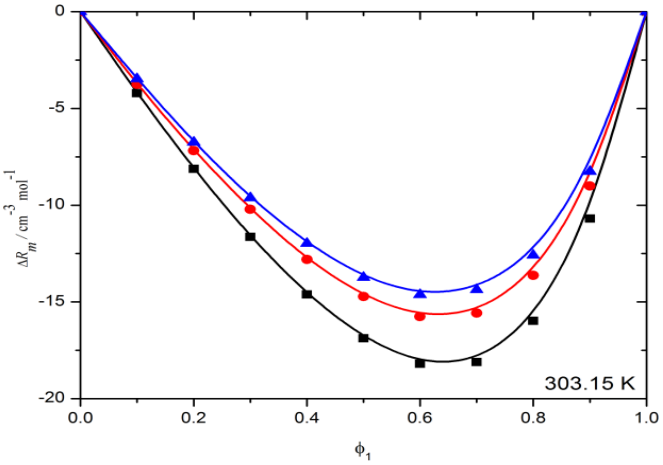

Figure. 2 Deviation in Molar refraction, ΔR_m, as a function of volume fraction for the system Ricinoleic acid + Fluorobenzene, (■); Chlorobenzene, (●); Bromobenzene, (▲); at T = 303.15K.

CONCLUSION

Positive value of deviation in Δn_D and negative values of molar refraction ΔR_m show that the strong interactions present in ricinoleic acid + haobenzene binaries. For better interpretation, theoretical refractive indices are calculated using nine relations namely Arago-Biot (A-B), Dale-Glastone (D-G), Lorentz-Lorentz (L-L), Weiner (WR), Heller (Hr), Newton (Nw), Eyring-John (E-J) Eykman (Eyk), Oster (Os). All these nine relations show good agreement in results with experimentally obtained values of refractive indices. Nine mixing relations for refractive index studies also supports that the interaction became weaker when moving from fluorobenzene to bromobenzene in these binaries. Because of the electronegative character of halobenzenes, the interactions may either decrease or increase depending on the structure of the components in the binaries. In present study fluorobenzene is a highly electronegative and polar than the other two halobenzenes, so strong interactions are observed in its binaries.

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