

Refractive index studies of binary liquid mixtures containing 1, 4-butanediol + o- cresol/m-cresol/p-cresol

J.V.Srinivasu¹, K. Narendra^{2*}, K. Shobha Rani³ and B. Subba Rao⁴

¹Department of Basic Science, Sri Vishnu Engineering College for Women, Bhimavaram-534 202,
Andhra Pradesh, India

²Department of Physics, VR Siddhartha Engineering College, Vijayawada-520 007, Andhra Pradesh, India

³Department of Chemistry, VSR & NVR College, Tenali, Andhra Pradesh, India

⁴Department of Physics, VSR & NVR College, Tenali, Andhra Pradesh, India

*Corresponding author: E-Mail: narenk75@gmail.com, Tel: 9885038074

ABSTRACT

Refractive index (n_D) and related properties such as molar refraction (R) have been investigated for binary system (1, 4-Butanediol with o-cresol/m-cresol/p-cresol) at temperatures $T = (303.15, 308.15, 313.15$ and $318.15)$ K. Some hypothetical and observational connections have been utilized to think about the reliance of the deliberate and inferred amounts on twofold creation and on temperature. Refractive index deviation, Δn_D , molar refraction deviation, ΔR , have also been calculated. Redlich-Kister polynomial equation is used to get the flexible parameters and standard deviations between the deliberate and fitted qualities, separately. The results obtained have been discussed based on specific intermolecular interactions between different species.

KEY WORDS: Refractive Index, Binary Mixtures, 1, 4-Butanediol, Cresol.

1. INTRODUCTION

Refractive index of pure liquids and liquid mixtures constitutes an important property for different engineering applications. The data related to refractive indices of binary liquids gives the knowledge to comprehend the sub-atomic communications between the liquids. For continuing study of thermodynamic and transport properties of binary liquids containing 1, 4-butanediol (1, 4-BD) with o-cresol/m-cresol/p-cresol, we present some studies about refractive properties of binary liquids of 1,4-butanediol (1, 4-BD) with o-cresol/m-cresol/p-cresol at different temperatures.

The chemicals chosen for this study are important in industrial and scientific context because of their applications. 1, 4-BD is a reasonable goeey fluid, which is miscible with water and most polar natural solvents. It is utilized as a part of the produce of a few kinds of plastics and flexible strands. Also it is suitable as a useful chemical intermediate in the manufacture of many chemical products. Cresols can be used as second generation bio-oils/fuels, emerging as an alternative to fossil fuels that contribute to the reduction of global warming, minimizing the generation of greenhouse gases and those harmful to the ozone layer.

In this paper we report the refractive index, n_D at $T = (303.15, 308.15, 313.15$ and $318.15)$ K for 1, 4-BD with o-cresol/m-cresol/p-cresol over the entire composition range. Some empirical and theoretical relationships (Arago, 1806; Eykman, 1895; Eyring, 1969; Gladstone, 1858; Heller, 1945; Lorentz, 1880; Kurtz, 1936; Oster, 1948; Weiner, 1910) have been connected to ponder the reliance of the deliberate and determined amounts on temperature and on paired structure. Several researchers (Bhatia, 2002; Lal, 2000; Narendra, 2011; Srinivasu, 2016) has been tested the validity of these mixing rules. Refractive index deviation, Δn_D , molar refraction deviation, ΔR , have also been calculated. To acquire the movable parameters and standard deviations between the deliberate and fitted qualities, results have been fitted to Redlich-Kister polynomial equation. The results obtained have been discussed based on specific intermolecular interactions between different species.

2. EXPERIMENTAL

1, 4-BD and o-cresol, m-cresol and p-cresol (Sigma-Aldrich, USA, mass fraction purity 0.99) used in the present investigation were purified by standard methods (Aminabhavi, 1984; Mosterio, 2001). Before utilize, all synthetic substances were put away more than 0.4nm sub-atomic strainers for 72 hrs to evacuate the water content, assuming any, and were degassed at low weight. The blends were set up by mass and were kept in extraordinary water/air proof plug glass jugs to maintain a strategic distance from dissipation. All examples were arranged promptly preceding estimations utilizing an electronic adjust (CPA-225D, Sartorius, Germany) accurately within $\pm 1 \times 10^{-5}$ g. The uncertainty in the mole part was evaluated to be within $\pm 1 \times 10^{-4}$. The refractive indices in pure liquids and in their binary mixtures was measured using an Abbe Refractometer (Sravankumar, 2011). The temperature was regulated by using a circulation pump connected with a temperature water bath. Adjustment of the instrument was finished by estimating the refractive indices of doubled refined water and toluene at known temperatures. The example blends were specifically infused into the crystal gathering of the instrument by methods for a sealed shut hypodermic syringe. The refractive indices were measured, when the liquid mixture attained constant temperature. A normal of three to four estimations was taken for each sample blend. The density and refractive index data of the liquid mixtures have been included in table.1.

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Theory: The 'mixing rules' tested are,
Arago-Biot (A-B):

$$n_m = \phi_1 n_1 + \phi_2 n_2 \quad (1)$$

Eykman's (E):

$$\left(\frac{n_m^2 - 1}{n_m + 0.4} \right) V_m = \left(\frac{n_1^2 - 1}{n_1 + 0.4} \right) \frac{M_1 x_1}{\rho_1} + \left(\frac{n_2^2 - 1}{n_2 + 0.4} \right) \frac{M_2 x_2}{\rho_2} \quad (2)$$

Where, symbols have their usual meaning.

Eyring and John (E-J):

$$n = n_1 \phi_1^2 + 2(n_1 n_2)^{1/2} \phi_1 \phi_2 + n_2 \phi_2^2 \quad (3)$$

Gladstone-dale (G-D):

$$(n_m - 1) = \phi_1 (n_1 - 1) + \phi_2 (n_2 - 1) \quad (4)$$

Heller's (H):

$$\left(\frac{n_m - n_1}{n_1} \right) = \frac{3}{2} \left(\frac{m^2 - 1}{m^2 + 2} \right) \phi_2 \quad (5)$$

Where $m = \frac{n_2}{n_1}$

Lorentz-Lorenz (L-L):

$$\left(\frac{n_m^2 - 1}{n_m^2 + 2} \right) \frac{1}{\rho_m} = \left(\frac{n_1^2 - 1}{n_1^2 + 2} \right) \frac{w_1}{\rho_1} + \left(\frac{n_2^2 - 1}{n_2^2 + 2} \right) \frac{w_2}{\rho_2} \quad (6)$$

Newton (N):

$$(n_m^2 - 1) = \phi_1 (n_1^2 - 1) + \phi_2 (n_2^2 - 1) \quad (7)$$

Oster (OS):

$$\frac{(n^2 - 1)(2n^2 + 1)}{n^2} = \frac{(n_1^2 - 1)(2n_1^2 + 1)}{n_1^2} \phi_1 + \frac{(n_2^2 - 1)(2n_2^2 + 1)}{n_2^2} \phi_2 \quad (8)$$

Weiner's (W):

$$\left(\frac{n_m^2 - n_1^2}{n_m + 2n_1^2} \right) = \left(\frac{n_2^2 - n_1^2}{n_2^2 + 2n_1^2} \right) \phi_2 \quad (9)$$

In the above equations, n_m is the refractive index of the mixture; n_1 and n_2 are the refractive indices of the pure components 1 and 2 respectively. ϕ_i is volume fraction of component i. $\phi_i = x_i V_i / \sum x_i V_i$, V_i is the molar volume of component i and x_i is the mole fraction of component i.

The average percentage deviations (APD) by different approaches have been computed using the expression:

$$APD = \frac{1}{n} \sum \frac{A_{\text{exp}} - A_{\text{theoretical}}}{A_{\text{exp}}} \times 100 \quad (10)$$

Where A is refractive index and n is the number of data points.

Molar refraction (R) obtained by using Lorentz-Lorenz equation

$$R = \left(\frac{n_m^2 - 1}{n_m^2 + 2} \right) V_m \quad (11)$$

The deviation in molar refraction ΔR gives the strength of interaction in the mixture and is a sensitive function of wavelength, temperature and mixture composition.

ΔR and Δn_D are calculated using the following equations:

$$\Delta R = R - (x_1 R_1 + x_2 R_2) \quad (12)$$

$$\Delta n_D = n_D - (x_1 n_{D1} + x_2 n_{D2}) \quad (13)$$

Where, x_i is the mole fraction, and R_i is the molar refraction of component i.

3. RESULTS AND DISCUSSION

The values of density and refractive index of pure and binary liquid mixtures with mole fraction of 1, 4-BD are listed in Table 1. Variation of refractive index deviation and molar refraction deviation with mole fraction of 1, 4-BD have been depicted in Figs.1 and 2 respectively. The deviation parameters are fitted to Redlich-Kister equation (Redlich, 1979), the adjustable parameters and standard deviations have been given in Table.2. Table.3, summarizes the APD values between the experimental n_D values and those calculated for each mixing rule. It is evident that all selected relations can be profitably used if only a rough approximation in predictive calculations is required.

Table.1. Comparison of refractive index n_D and viscosity η of pure liquids with their literature values at $T = 303.15, 308.15, 313.15$ and $318.15K$

Compounds	T(K)	$\rho/kg.m^{-3}$		n_D	
		(Expt.)	(Lit.)	(Expt.)	(Lit.)
1, 4-butanediol	303.15	1009.4	1009.67 ^a	1.4435	
	308.15	1006.8	1006.70 ^a	1.4413	
	313.15	1003.8	1003.73 ^a	1.4393	
	318.15	1000.4	1000.76 ^a	1.4379	
o-cresol	303.15	1036.6	1036.90 ^b	1.5395	
	308.15	1030.9	1031.60 ^b	1.5375	
	313.15	1026.3	1026.00 ^b	1.5358	
	318.15	1021.4	1021.10 ^c	1.5340	
m-cresol	303.15	1025.7	1026.10 ^b	1.5347	1.535 ^d
	308.15	1021.9	1022.10 ^b	1.5331	
	313.15	1017.4	1017.00 ^c	1.5324	
	318.15	1013.6	1013.50 ^c	1.5306	
p-cresol	303.15	1026.6	1026.30 ^b	1.5313	
	308.15	1022.4	1022.40 ^b	1.5297	
	313.15	1019.0	1018.10 ^c	1.5285	
	318.15	1014.3	1013.90 ^c	1.5267	

^aNain (2007), ^bBhatia (2011), ^cNarendra (2011), ^dMaimoonan (2011)

Table.2. Densities, ρ , and refractive indices, n_D , as a function of mole fraction 1, 4-BD at $T = (303.15 - 318.15) K$ at atmospheric pressure 0.1MPa

x_1	ρ ($kg.m^{-3}$)	n_D	ρ ($kg.m^{-3}$)	n_D	ρ ($kg.m^{-3}$)	n_D	ρ ($kg.m^{-3}$)	n_D
1,4-Butanediol + o-cresol								
	T=303.15K		T=308.15K		T=313.15K		T=318.15K	
0.0000	1036.6	1.5395	1030.9	1.5375	1026.3	1.5358	1021.4	1.5340
0.1001	1035.7	1.5334	1030.0	1.5319	1025.3	1.5305	1020.3	1.5292
0.2001	1034.2	1.5263	1028.7	1.5248	1023.9	1.5236	1019.1	1.5223
0.3002	1032.3	1.5185	1027.0	1.5169	1022.4	1.5156	1017.7	1.5143
0.4002	1030.0	1.5100	1025.1	1.5083	1020.7	1.5069	1016.0	1.5056
0.5002	1027.5	1.5007	1022.8	1.4990	1018.6	1.4975	1014.2	1.4962
0.6002	1024.6	1.4904	1020.4	1.4887	1016.3	1.4872	1012.0	1.4860
0.7002	1021.6	1.4794	1017.6	1.4777	1013.7	1.4762	1009.5	1.4750
0.8001	1018.2	1.4677	1014.4	1.4659	1010.8	1.4643	1006.7	1.4633
0.9001	1014.3	1.4556	1010.9	1.4537	1007.4	1.4521	1003.7	1.4508
1.0000	1009.4	1.4435	1006.8	1.4413	1003.8	1.4393	1000.4	1.4379
1, 4-Butanediol + m-cresol								
	T=303.15K		T=308.15K		T=313.15K		T=318.15K	
0.0000	1025.7	1.5347	1021.9	1.5331	1017.4	1.5324	1013.6	1.5306
0.0992	1025.6	1.5286	1021.6	1.5274	1017.0	1.5271	1013.0	1.5257
0.1986	1024.9	1.5215	1020.9	1.5204	1016.3	1.5200	1012.2	1.5187
0.2982	1024.1	1.5137	1020.1	1.5124	1015.4	1.5118	1011.3	1.5105
0.3979	1022.9	1.5052	1019.0	1.5038	1014.4	1.5031	1010.2	1.5018
0.4979	1021.4	1.4962	1017.6	1.4947	1013.1	1.4938	1009.0	1.4926
0.5979	1019.7	1.4865	1016.0	1.4850	1011.7	1.4841	1007.6	1.4830

0.6982	1017.8	1.4760	1014.2	1.4745	1010.1	1.4735	1006.1	1.4726
0.7986	1015.5	1.4652	1012.1	1.4636	1008.3	1.4624	1004.4	1.4615
0.8992	1012.7	1.4544	1009.6	1.4525	1006.2	1.4510	1002.6	1.4500
1.0000	1009.4	1.4435	1006.8	1.4413	1003.8	1.4393	1000.4	1.4379
1, 4-Butanediol + p-cresol								
	T=303.15K		T=308.15K		T=313.15K		T=318.15K	
0.0000	1026.6	1.5313	1022.4	1.5297	1019.0	1.5285	1014.3	1.5267
0.0961	1026.3	1.5252	1022.0	1.5239	1018.3	1.5230	1013.5	1.5215
0.1931	1025.7	1.5181	1021.3	1.5167	1017.5	1.5157	1012.7	1.5143
0.2909	1024.7	1.5105	1020.4	1.5090	1016.6	1.5079	1011.7	1.5065
0.3895	1023.5	1.5023	1019.3	1.5008	1015.4	1.4996	1010.6	1.4982
0.4890	1022.0	1.4936	1017.9	1.4920	1014.0	1.4908	1009.3	1.4895
0.5894	1020.2	1.4843	1016.3	1.4826	1012.5	1.4812	1007.9	1.4800
0.6907	1018.2	1.4741	1014.4	1.4724	1010.7	1.4711	1006.3	1.4700
0.7929	1015.8	1.4638	1012.2	1.4621	1008.6	1.4607	1004.5	1.4598
0.8960	1012.8	1.4536	1009.7	1.4518	1006.4	1.4503	1002.6	1.4492
1.0000	1009.4	1.4435	1006.8	1.4413	1003.8	1.4393	1000.4	1.4379

Table.3. Redlich–Kister coefficients, A_j along with standard deviation σ for binary mixture of 1, 4-BD with cresols

Parameter	T/K	a	b	c	d	e	σ
1, 4-Butanediol + o-cresol							
Δn_D	303.15	0.0367	-0.0034	-0.0082	-0.0052	0.0048	0.00003
	308.15	0.0004	-0.0032	-0.0034	-0.0080	0.0043	0.00004
	313.15	0.0398	-0.0039	-0.0020	-0.0075	0.0086	0.00007
	318.15	0.0412	-0.0030	0.0014	-0.0111	0.0075	0.00004
ΔR	303.15	0.0842	-0.0198	-0.0534	-0.0312	0.0194	0.00008
	308.15	0.0948	-0.0215	-0.0339	-0.0397	0.0403	0.00021
	313.15	0.1060	-0.0292	-0.0042	-0.0338	0.0327	0.00035
	318.15	0.1140	-0.0246	0.0206	-0.0560	0.0286	0.00020
1, 4-Butanediol + m-cresol							
Δn_D	303.15	0.0276	-0.0067	-0.0076	-0.0038	0.0068	0.00006
	308.15	0.0292	-0.0057	-0.0015	-0.0085	0.0031	0.00005
	313.15	0.0313	-0.0047	0.0013	-0.0115	0.0057	0.00007
	318.15	0.0328	-0.0034	0.0042	-0.0124	0.0094	0.00005
ΔR	303.15	0.0405	-0.0371	-0.0464	-0.0185	0.0412	0.00038
	308.15	0.0540	-0.0341	-0.0119	-0.0384	0.0255	0.00034
	313.15	0.0703	-0.0299	0.0077	-0.0558	0.0282	0.00027
	318.15	0.0842	-0.0221	0.0321	-0.0706	0.0265	0.00025
1, 4-Butanediol + p-cresol							
Δn_D	303.15	0.0209	-0.0080	-0.0128	-0.0035	0.0143	0.00006
	308.15	0.0220	-0.0071	-0.0097	-0.0056	0.0160	0.00003
	313.15	0.0234	-0.0065	-0.0103	-0.0063	0.0243	0.00002
	318.15	0.0247	-0.0054	-0.0048	-0.0066	0.0218	0.00006
ΔR	303.15	0.0113	-0.0430	-0.0703	-0.0161	0.0767	0.00026
	308.15	0.0225	-0.0407	-0.0508	-0.0215	0.0838	0.00019
	313.15	0.0361	-0.0379	-0.0458	-0.0313	0.1190	0.00015
	318.15	0.0486	-0.0324	-0.0194	-0.0374	0.1050	0.00030

Table.4. Calculated APD for different "Mixing rules" applied to the selected binary systems at T = 303.15 to 318.15 K

T / K	A-B	E	E-J	G-D	H	L-L	N	Os	W
1, 4-BD + o-cresol									
303.15	0.206	0.280	0.221	0.206	0.239	0.240	0.175	0.299	0.218
308.15	0.226	0.300	0.242	0.226	0.259	0.260	0.195	0.319	0.238
313.15	0.246	0.321	0.262	0.246	0.280	0.281	0.215	0.341	0.259
318.15	0.265	0.340	0.281	0.265	0.299	0.300	0.234	0.359	0.278
1, 4-BD + m-cresol									
303.15	0.116	0.183	0.130	0.116	0.146	0.147	0.088	0.200	0.127
308.15	0.139	0.207	0.153	0.139	0.169	0.170	0.110	0.224	0.150
313.15	0.163	0.233	0.178	0.163	0.195	0.196	0.134	0.251	0.175
318.15	0.188	0.258	0.203	0.188	0.220	0.220	0.159	0.276	0.200
1, 4-BD + p-cresol									
303.15	0.052	0.114	0.065	0.052	0.080	0.080	0.026	0.130	0.062
308.15	0.067	0.130	0.080	0.067	0.096	0.096	0.041	0.146	0.077
313.15	0.086	0.150	0.100	0.086	0.115	0.116	0.059	0.167	0.097
318.15	0.106	0.170	0.119	0.106	0.135	0.135	0.079	0.186	0.116

The premise of these blending tenets of refractive index rules is e. m. hypothesis of light, which regards the atoms as dipoles or congregations of dipoles by an outer field. Refractive lists rely upon nature of fluid, weight and temperature and are straightforwardly identified with various physical parameters of the fluid, for example, dielectric consistent, thickness, molar refractivity and so on. An endeavor has been made to consider the legitimacy of nine blending rules for anticipating the refractivity of parallel fluid framework, over the whole mole portion.

From Table.3, it can be observed that the APD values are smaller for Newton relation and larger for Oster relation, even if the relative merits of the checked equations are comparable in magnitude to each other. The equations used in this paper for investigation is limited to some literature models which can be applied when preliminary knowledge of liquid mixture density is available. Some other equations are also available for prediction of refractive indices but the applicability of those models needs a priori knowledge of one or more empirical parameters other than density for each mixed liquid.

Figure.1, shows the profiles of Δn_D against the mole fraction of 1, 4-BD at 303.15K for all the studied mixtures. The trends are always positive for the mixtures studied, with a sharp maximum at $x_1 = 0.5$ which becomes higher as temperature increases. At all studied temperatures the same trends have been observed. The maximum values may indicate the presence of a complex adduct.

The sign and size of Δn_D likewise changes with the basic attributes of the fluid parts emerging from the geometrical fitting of one segment into the structure of other segment because of the distinction in the sub-atomic size and state of the segments. In the event that the not at all like particles have nearly a similar molar volumes, this impact ought to be critical as proposed by Aminabhavi (1979). In any case, regardless of whether slight contrast in the free volume between various species could encourage the infiltration of one part into the other and as the distinction of the free volumes of the two unadulterated species builds, the more positive ought to be the commitment to Δn_D .

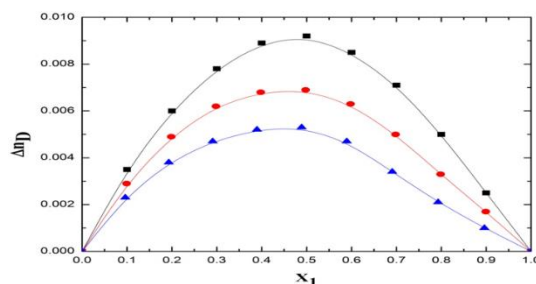


Figure.1. Deviation in refractive index Δn_D vs mole fraction (x_1) of 1,4-Butanediol in the binary mixture of 1, 4-Butanediol + o-cresol (■), + m-cresol (●) and + p-cresol (▲) at 303.15K

With the point of social affair assist informations about the specific intermolecular interactions in these binary mixtures, we have investigated the molar refraction R. R being affected by any variation of the chemical environment of a real species, its functional dependecne on temeptrature and binary composition provides information

about the intermolecular interactions and driving forces acting on the liquid structure of the binary mixtures. According to Ciocirlan (2008), the molar refraction values depends only on the λ of light used for the measurements.

Starting from the R data, it is possible to study the related deviation ΔR , obtained by applying the relation (12). The results of this analysis are plotted in Fig. 2 for the systems at temperature 303.15K. Increasing ΔR values are observed for increase in temperature.

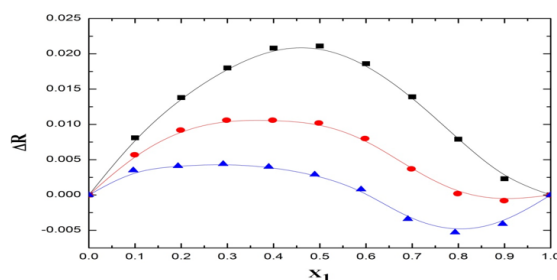


Figure.2. Deviation in molar refraction ΔR vs mole fraction (x_1) of 1,4-Butanediol in the binary mixture of 1, 4-Butanediol + o-cresol (■), + m-cresol (●) and + p-cresol (▲) at 303.15K

The deviations of hypothetical qualities from exploratory ones are temperature-autonomous. This might be ascribed to the way that variety in refractive list with temperature is repaid by the adjustment in thickness of the fluid blend. Nonetheless, in situations where the variety is huge with change in temperature, it can be utilized for deciphering the structure and interactions in the liquid by computing other dielectric, optical and acoustical properties using experimental data. If the concept of excess volume (which is an indirect measure of interaction) is taken into consideration in various mixing rules, the deviations of hypothetical qualities from exploratory ones are reduced.

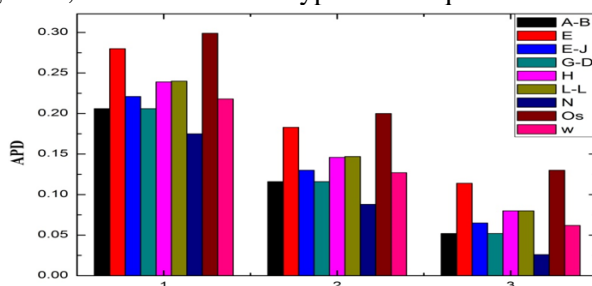


Figure.3. The APD values for different mixing relations in the binary liquid mixtures 1, 4-BD + o-cresol (1), 1, 4-BD + m-cresol (2) and 1, 4-BD + p-cresol (3) at temperature T = 303.15 K

4. CONCLUSION

Various mixing rules of refractive index were used to test their legitimacy for binary mixtures by using the measured values of density and refractive index. It may be observed that all the blending rules talked about are interrelated in a straightforward quantitative way and perform well inside the points of confinement of exploratory mistake. Negative and positive deviations are seen amongst trial and hypothetical qualities calculated by using various theories/models. The lowest deviations for the calculated refractive indices have been obtained by using Newton's equation, whereas the Oster equation gives the highest deviations at all temperatures for the mixtures studied. Deviations in refractive index and molar refraction are observed to be positive for all the temperatures studied.

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