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Refractive Indices of Binary Liquid Mixture at Different Temperatures

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ABSTRACT

Densities and refractive indices have been experimentally determined for binary liquid mixture cyclohexane with o-xylene at 303.15 to 318.15 K over the entire composition range. The refractive indices were measures at the required temperatures with the thermostated highly precised Abbe's refractometer. A comparative study of Lorentz-Lorentz (L-L), Gladstone-dale (G-D), Weiner (W) and Heller (H) relations for predicting the refractive index of a liquid has been carried out to test their validity for the binary system over the entire mole fraction range at the four temperatures. The experimental values of refractive indices are in good agreement with all the above relations except Gladstone-Dale relation.

Key words: Binary mixtures, cyclohexane, o-xylene, refractive indices, lorentz-lorentz relation, weiner relation

INTRODUCTION

Refractive index measurements in combination with density, boiling point, melting point and other analytical data are very useful industrially also for common substances which include oils, waxes, sugar syrups etc. Literature survey reveals that its general applicability in chemical analysis and industry. Number of mixing rules has been proposed in the literature for the refractive index measurements. Some of them are not suitable when there is a large change of volume on mixing. The most widely used theoretical rules are presented by Lorentz (1916) and Weiner (1910). Various empirical and semi-empirical relations have been formulated earlier and tested by Heller (1945) and Dale and Gladstone (1858). The validity of these mixing rules has been tested by researchers (Tasic *et al*., 1992; Oster, 1948; Dubey *et al*., 2005; Bhatia *et al*., 2002; El-Sayed, 2003; Oswal *et al*., 2003). Relative merits of these mixing rules have been discussed by Pandey *et al*. (1992), Oswal *et al*. (2005), Francesconi and Ottani (2007), Ali *et al*. (2007), Ali and Tariq (2008), Roy *et al*. (2010) and Gupta *et al*. (2010). Aminabhavi (1984) pointed out the usefulness of mixing rules in determining binary refractive indices and density data. In the present study, experimental values of density and refractive index which are measured at different temperatures for the entire molefraction range is given and an attempt has been made to study four refractive index mixing rules for the binary mixture cyclohexane+o-xylene covering the entire range of composition at 303.15 to 318.15 K. At the end the experimental data is compared with the theoretical mixing rules of refractive indices.

Table 1: Experimental and literature values of density (ρ) and refractive index (n) of pure liquid components at 303.15 K

	ρ		n	
Liquid	Exptl.*	Lit*	Exptl.*	Lit*
Cyclohexane	0.7678	0.7686[a]	1.4200	1.4239[b]
o-xylene	0.8707	0.8718[c]	1.4950	1.5002[d]

*Exptl: Experimental value, *Lit: Literature value, [a]Singh *et al*. (2005), [b]Bhatia *et al*. (2002), [c]Vargaftic *et al*. (1996), [c]Sharma *et al*. (2007)

MATERIALS AND METHODS

The chemicals were purchased from Ranbaxy and were of LR grade. These products are further purified by distillation. The close agreement of experimental values with those reported in literature ascertains the reliability of the present results. The mixtures were prepared by mixing measured volumes of the components in airtight stoppered bottles to minimize evaporation losses. The weighing was done on an electronic balance with a readability of 0.0001 g. Density (Saravanakumar *et al*., 2010; Ziyada *et al*., 2011) measurements were made using 25 mL specific gravity bottle. The refractive indices of the liquids and of the mixtures were determined using Abbe's refractometer which has the least number of moving parts; thus the chances of error arising due to strain are also minimal. Before measuring the refractive index on Abbe's refractometer an inspection of the scale was made with the help of a test piece supplied by the suppliers. A drop of mono bromo-naphthalene was placed on the polished surface of the test piece and then it is placed on the main prism surface of Abbe's refractometer. Measuring knob is turned to find the demarcation line of the brightness and the darkness in the field of view. Using dispersion knob the demarcation line is cleared. Once again the measuring knob is turned in such a way the demarcation line fallen just on the centre of the cross-line in the field of view. The reading on scale was noted, which was shown on the below of cross line in the same field of view. There is a provision for connecting to thermo stated temperature bath. The circulated water will keep the prism hose in fixed temperature. The temperature of prism hose can be read with the help of built in digital thermometer. The same procedure is adopted first to measure the refractive indices of pure liquids such as benzene and n-hexane to ascertain the accuracy of the results and then for the liquid mixtures chosen for the present work. Good agreement was found between measured and literature values. Uncertainty in refractive index measurements was found to be 0.06%. The experimental work is carried in the year 2007. Its values are given in Table 1.

RESULTS AND DISCUSSION

In the present study, an attempt has been made to study the validity of 4 mixing rules for predicting the refractive indices of binary mixtures of cyclohexane+o-xylene at four different temperatures for the entire molefraction range. Various equations for determining refractive index are given below:

Lorentz-Lorentz (L-L): This is the most frequently used mixing rule in the analysis of refractive index data:

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

where, n is the refractive index of the mixture of x_1 and x_2, n_1 and n_2 are the refractive indices of the pure components respectively, ϕ_1 and ϕ_2 are volume fractions. The volume fractions are obtained from $\phi_1 = x_1v_1/(x_1v_1+x_2v_2)$, where x is the mole fraction and v is the molar volume of component 1.

Gladstone-Dale (G-D): This can be referred to as specific refraction and can be formulated as:

$$n-1 = (n_1-1)\,\phi_1+(n_2-1)\,\phi_2 \tag{2}$$

Weiner relation: It applies to isotropic bodies of spherically symmetrical shape and proposes volume additivity and is given by:

$$\frac{n^2-n_1^2}{n^2+2n_2^2} = \left(\frac{n_2^2-n_1^2}{n_2^2+2n_1^2}\right)\phi_2 \tag{3}$$

Heller relation: Heller assumed equivalence of light scattering of Debye and Rayleigh and derived the following equation:

$$\frac{n-n_1}{n_1} = \frac{3}{2}\left[\frac{(n_2/n_1)^2-1}{(n_2/n_1)^2+2}\right]\phi_2 \tag{4}$$

The experimental data on refractive index and density of the binary system at different temperatures has been given in Table 2.

Table 3 contains the experimental and theoretical data of refractive indices at different temperatures.

A close look of the data presented in Table 3 at different temperatures reveals that for the binary mixture cyclohexane+o-xylene, L- L relation, Weiner relation and Heller relations show good agreement with experimental values. Further it is observed that for lower molefractions at all temperatures L-L relation shows excellent agreement with experimental values. Whereas deviations are more pronounced in case G-D relation. It is interesting to report that at all temperatures the values obtained from Weiner and Heller relations are found to be nearer. The L-L relation performs best of all since the variation of deviation with the increase of molefraction of o-xylene has less interaction with cyclohexane on mixing the components. During mixing excess volume is the measurement of molecular interaction in liquid mixtures. The structural property of liquid and liquid mixtures can be integrated through refractive indices employing molefractions. In general refraction increases with molecular weight for symmetric and asymmetric molecules. Density and refractive index depend on molecular weight and nature of liquids. It is observed from Table 2 that density and refractive index values decreases with increase of temperature from 303.15 to 318.15 K. The deviations may be used for interpreting the structure and interactions in the liquid and liquid mixtures as reported by Mehra (2003), Tripathi (2005) and Ali *et al.* (2005). The excess volumes of the present mixture are found to be of small magnitudes at all temperatures as reported by Narendra *et al.* (2010). That means the intermolecular interactions among the components are very weak. It also leads to say no change in molecular polarisability on mixing the compounds. Aminabhavi and Banerjee (1998) suggested that deviations of the theoretical values from experimental ones can be reduced if excess volumes are taken into consideration in various mixing rules. It was supported by several researchers (Romero *et al.*, 2006; Rathnam *et al.*, 2005).

Table 2: Refractive indices and densities of binary system cyclohexane + o-xylene at different temperatures

x_1	n_{exp}	$\rho\times10^3$ (kg m^{-3})	x_1	n_{exp}	$\rho\times10^{-3}$ (kg m^{-3})
at 303.15 K			**at 308.15 K**		
0.0000	1.4200	0.7678	0.0000	1.4188	0.7625
0.0908	1.4333	0.7768	0.0908	1.4292	0.7731
0.1835	1.4422	0.7865	0.1835	1.4381	0.7831
0.2781	1.4495	0.7975	0.2781	1.4450	0.7946
0.3747	1.4556	0.8096	0.3747	1.4510	0.8070
0.4734	1.4606	0.8222	0.4734	1.4569	0.8194
0.5742	1.4654	0.8304	0.5742	1.4617	0.8280
0.6772	1.4702	0.8368	0.6772	1.4674	0.8349
0.7824	1.4792	0.8486	0.7824	1.4777	0.8466
0.8900	1.4912	0.8614	0.8900	1.4877	0.8600
1.0000	1.4950	0.8707	1.0000	1.4918	0.8694
at 313.15 K			**at 318.15 K**		
0.0000	1.4170	0.7587	0.0000	1.4145	0.7531
0.0908	1.4256	0.7711	0.0908	1.4217	0.7668
0.1835	1.4338	0.7811	0.1835	1.4287	0.7787
0.2781	1.4419	0.7930	0.2781	1.4365	0.7907
0.3747	1.4478	0.8054	0.3747	1.4433	0.8034
0.4734	1.4536	0.8181	0.4734	1.4492	0.8158
0.5742	1.4588	0.8270	0.5742	1.4565	0.8254
0.6772	1.4660	0.8337	0.6772	1.4637	0.8329
0.7824	1.4758	0.8453	0.7824	1.4747	0.8440
0.8900	1.4858	0.8585	0.8900	1.4847	0.8567
1.0000	1.4908	0.8677	1.0000	1.4887	0.8659

Table 3: Experimental and theoretical values of refractive indices in cyclohexane+o-xylene system at different temperatures

Molefraction	n_{exp}	n_{L-L}	n_{G-D}	n_W	n_H
at 303.15 K					
0.0000	1.4200	1.4200	1.4200	1.4200	1.4200
0.0908	1.4333	1.4265	1.4409	1.4287	1.4284
0.1835	1.4422	1.4335	1.4606	1.4373	1.4368
0.2781	1.4495	1.4413	1.4793	1.4458	1.4452
0.3747	1.4556	1.4499	1.4968	1.4544	1.4536
0.4734	1.4606	1.4589	1.5127	1.4631	1.4622
0.5742	1.4654	1.4651	1.5261	1.4715	1.4705
0.6772	1.4702	1.4733	1.5387	1.4799	1.4790
0.7824	1.4792	1.4787	1.5496	1.4880	1.4872
0.8900	1.4919	1.4880	1.5598	1.4966	1.4959
1.0000	1.4950	1.4950	1.5685	1.5048	1.5043
at 308.15 K					
0.0000	1.4188	1.4188	1.4188	1.4188	1.4188
0.0908	1.4292	1.4259	1.4404	1.4273	1.4270
0.1835	1.4381	1.4326	1.4600	1.4357	1.4352
0.2781	1.4450	1.4403	1.4786	1.4440	1.4434
0.3747	1.4510	1.4487	1.4958	1.4525	1.4517
0.4734	1.4569	1.4571	1.5114	1.4609	1.4600
0.5742	1.4617	1.4630	1.5245	1.4691	1.4682
0.6772	1.4674	1.4679	1.5362	1.4771	1.4762

Table 3: Continued

Molefraction	n_{exp}	n_{L-L}	n_{G-D}	n_W	n_H
0.7824	1.4777	1.4760	1.5475	1.4853	1.4846
0.8900	1.4877	1.4852	1.5574	1.4937	1.4931
1.0000	1.4918	1.4918	1.5657	1.5018	1.5013
at 313.15 K					
0.0000	1.4170	1.4170	1.4170	1.4170	1.4170
0.0908	1.4256	1.4252	1.4396	1.4256	1.4253
0.1835	1.4338	1.4318	1.4593	1.4341	1.4336
0.2781	1.4419	1.4398	1.4782	1.4426	1.4419
0.3747	1.4478	1.4480	1.4955	1.4511	1.4503
0.4734	1.4536	1.4566	1.5112	1.4597	1.4588
0.5742	1.4588	1.4627	1.5244	1.4680	1.4671
0.6772	1.4660	1.4674	1.5361	1.4761	1.4753
0.7824	1.4758	1.4753	1.5473	1.4845	1.4837
0.8900	1.4858	1.4844	1.5573	1.4930	1.4924
1.0000	1.4908	1.4908	1.5657	1.5011	1.5011
at 318.15 K					
0.0000	1.4145	1.4145	1.4145	1.4145	1.4145
0.0908	1.4217	1.4240	1.4385	1.4231	1.4228
0.1835	1.4287	1.4309	1.4585	1.4317	1.4312
0.2781	1.4365	1.4387	1.4773	1.4403	1.4396
0.3747	1.4433	1.4470	1.4946	1.4489	1.4481
0.4734	1.4492	1.4551	1.5101	1.4576	1.4567
0.5742	1.4565	1.4615	1.5235	1.466	1.4651
0.6772	1.4637	1.4665	1.5352	1.4742	1.4734
0.7824	1.4747	1.4739	1.5462	1.4827	1.4819
0.8900	1.4847	1.4824	1.5560	1.4912	1.4906
1.0000	1.4887	1.4887	1.5643	1.4995	1.4990

CONCLUSION

All mixing rules could be successfully applied at lower concentrations of o-xylene with cyclohexane omitting other factors such as volume reduction, volume addition and temperature. G-D relation could not give better results. All four theoretical mixing rules discussed are interrelated in a simple quantitative manner and perform well within the limits of experimental error.

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