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THEORETICAL EVALUATION OF REFRACTIVE INDEX IN BINARY LIQUID MIXTURES AT DIFFERENT TEMPERATURES

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Abstract:-Refractive indices and densities of the binary mixture pyrrolidinone with propane-1,3-diol have been measured at 303.15, 308.15, 313.15 and 318.15 K over the entire mole fraction range. A comparative study of Arago-Biot (A-B), Eykman (E), Eyring and John (E-J), Gladstone-Dale (G-D), Heller (H), Lorentz-Lorentz (L-L), Newton (N), Oster (Os) and Weiner (W) relations for predicting the refractive index of a liquid has been carried out to test the validity for the binary mixtures over the entire composition range of pyrrolidinone at four temperatures. Comparison of various mixing rules has been expressed in terms of average percentage deviation.

Keywords: Pyrrolidinone, refractive index, propane-1,3-diol, Goldstone-Dale relation.

1. INTRODUCTION

Refractive index measurements in combination with density and other analytical data are very useful in industry and also for common substances like oils, waxes, syrup etc. For the determination of composition of binary liquid mixtures it is essential for the prediction of refractive indices. Literature survey reveals that there is no such data on these mixtures. Here we report the experimental values of density and refractive indices for the binary mixture pyrrolidinone with propane-1,3-diol at temperatures 303.15, 308.15, 313.15 and 318.15 K over the entire composition range. The experimental values of refractive indices are compared with the values of refractive indices calculated from various theories. The most widely used theoretical rules for predicting the refractivity of binary liquid mixtures are Arago-Biot [1], Eykman [2], Eyring and John [3], Gladstone-Dale [4], Heller [5], Lorentz-Lorentz [6], Newton [7], Oster [8] and Weiner [9]. The relative applicability of these mixing rules has been tested by many researchers [10-14]. The major drawback of these mixing rules is their inability to account for change in volume and refractivity during mixing as they are based only on volume additivity [15].

2. EXPERIMENTAL PROCEDURE:

Densities of pure liquids and mixtures were measured using Anton Paar DSA 5000 density and sound analyser provided with two Pt 100 platinum thermometer. The density is extremely sensitive to temperature, so the apparatus was controlled to 0.001 K by a built in solid state thermostat.

The mixtures were prepared by mass and were kept in special airtight stoppered glass bottles to avoid evaporation. The weighings were done with an electronic balance with a precision of 0.01 mg.

Refractive indices of the mixtures at the sodium D-line were determined with an Abbe refractometer equipped with a circulating water bath permitting to maintain the sample at constant temperature to within 0.1 K. The instrument is calibrated by measuring the refractive index of deionised water. The sample support was rinsed with acetone and dried with a paper towel.

3.THEORY:

Mixing rules for refractive index of liquid mixtures are listed below:

Arago-Biot (A-B), assuming volume additivity, proposed the following relation for refractive index of binary mixtures:

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

Eykman's (E) relation is represented as:

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

where, symbols have their usual meaning.

Eyring and John (E-J) relation:

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

Gladstone-dale (G-D) equation for predicting the refractive index of a binary mixture is as follows:

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

Heller's (H) relation is given by:

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

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

The Lorentz-Lorentz (L-L) relation for refractive index is based on the change in the molecular polarizability with volume fraction:

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

Newton (N) gave the following equation:

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

Oster (Os) relation:

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

Weiner's (W) relation is represented as:

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

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.

$$\phi_i = x_i V_i / \sum x_i V_i, \quad V_i$$

is the molar volume of component i and x_i is the mole fraction of component i.

The n_m data of the mixtures have been used to calculate the deviation of refractive index from ideality, Δn , using:

$$\Delta n = n_m - (x_1 n_1 + x_2 n_2) \quad (10)$$

The refractive index is related to molar refraction, R_m , by:

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

where, $V_m = (x_1M_1 + x_2M_2)/$ is the molar volume.

4. RESULTS AND DISCUSSION:

The experimental data on refractive index and density of the binary system chosen at 303.15, 308.15, 313.15 and 318.15 K has been given in table 1. The average percentage deviations determined to assess their validity are shown in table 2. A close look of the table 1 reveals that the refractive indices of the pure components and mixtures decrease with rise in temperature.

TABLE-1: Experimental values of density, ρ and refractive index, n for binary mixtures at different temperatures.

x_1	ρ (Kg m ⁻³)	n	ρ (Kg m ⁻³)	n
	T = 303.15 K		T = 308.15 K	
0.0000	1046.789	1.436314	1043.649	1.434887
0.1108	1053.210	1.441986	1049.907	1.440545
0.2172	1057.343	1.445379	1053.957	1.443848
0.3044	1064.500	1.451898	1061.034	1.450297
0.4174	1070.951	1.457411	1067.297	1.455741
0.5076	1076.083	1.461630	1072.339	1.459993
0.6177	1081.390	1.465858	1077.566	1.464124
0.6909	1086.228	1.470591	1082.378	1.468811
0.8139	1092.129	1.475129	1088.173	1.473287
0.8736	1096.224	1.478547	1092.207	1.476821
1.0000	1103.073	1.484032	1098.989	1.482177
	T = 313.15 K		T = 318.15 K	
0.0000	1040.494	1.433374	1037.322	1.431895
0.1108	1046.553	1.439076	1043.227	1.437555
0.2172	1050.555	1.442348	1047.136	1.441854
0.3044	1057.548	1.448681	1054.029	1.447094
0.4174	1063.631	1.454058	1059.949	1.451343
0.5076	1068.586	1.458312	1064.823	1.456576
0.6177	1073.738	1.462338	1069.902	1.461505
0.6909	1078.541	1.466986	1074.661	1.464952
0.8139	1084.206	1.471487	1080.242	1.469675
0.8736	1088.198	1.475026	1084.191	1.473142
1.0000	1094.903	1.480330	1090.823	1.478412

TABLE-2: Average percentage deviation between theoretical and experimental values of refractive index of binary liquid mixtures at different temperatures.

Relation	Temperature (K)			
	303.15	308.15	313.15	318.15
A-B	0.00326	-0.00417	0.00401	-0.00008
E	0.02261	0.01486	-0.00440	0.01841
E-J	0.00734	-0.00016	-0.00440	0.00383
G-D	0.00326	-0.00418	-0.00198	-0.00008
H	0.00989	0.00236	0.00356	0.00628
L-L	0.01195	0.00436	-0.01234	0.00818
N	-0.0049	-0.01221	-0.00043	-0.00789
Os	0.02777	0.01996	0.01439	0.02338
W	0.00575	-0.00173	0.01945	0.00231

Electromagnetic theory of light is the basis of these mixing rules of refractive index which treats the molecules as dipoles or assemblies of dipoles by an external field. Here an attempt has been made to study the validity of nine mixing rules for predicting the refractivity of binary liquid system, over the entire mole fraction range.

The deviations of theoretical values from experimental ones are temperature-independent. This may be attributed to the fact that variation in refractive index with temperature is compensated by the change in density of the liquid mixture. However, in cases where the variation is significant with change in temperature, it can be used for interpreting the structure and interactions in the liquid by computing other dielectric, optical and acoustical properties using experimental data. The deviation between the theoretical and observed values of refractive index for the system chosen for investigation may be reduced if the concept of excess volume which is an indirect measure of interaction is taken into consideration in various mixing rules [16].

5. CONCLUSIONS:

From the above investigations it may be concluded that all the mixing rules discussed are interrelated in a simple quantitative manner and perform well within the limits of experimental error. Positive and negative deviations are observed between experimental and theoretical values calculated from various theories/models.

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