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Research Article

Experimental Investigation of Density & Refractive Index of Binary Liquid Mixtures of Tetrahydrofuran with Benzene, Methyl Benzene and Ethylbenzene at 308.15 K

K. Vijaya Lakshmi*

Department of Chemistry, Government College (Autonomous), Ananthapuramu-515001. Andhra Pradesh, India.

*Corresponding author:kvlakshmikontham@gmail.com

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ABSTRACT

Densities and refractive index have been measured at 308.15 K over the entire composition range of binary mixtures of tetrahydrofuran with benzene, methyl benzene, and ethyl benzene at 308.15 K. The measured data have been used to calculate the molar volume, molar refraction, excess refractive index, andexcess molar refraction, which are fitted to the Redlich-Kister polynomial equation. The results indicate the presence of strong interactions between tetrahydrofuran with benzene, methyl benzene, and ethyl benzene. The deviations in excess refractive indices values the follow the order THF + B < THF + MB < THF + EB. The results were discussed in terms of the existence of intermolecular interactions between the components in the liquid mixtures.

Keywords: Density, Refractive index, Tetrahydrofuran, Benzene, Methyl Benzenes and ethyl benzene, Excess refractive indices, Excess molar refraction.

INTRODUCTION

Refractive index measurement is an essential part of the thermodynamic studies of liquid-liquid mixtures, used to explain the intermolecular interactions present amongst the mixing components. Refractive index, along with density, is important for the determination of the composition of binary mixtures, usually non-ideal mixtures, where direct experimental measurement, along with other parameters such as density, melting point, boiling point, and other analytical data are very useful for common substances, which include oils, waxes, sugar syrups, etc. Deviation in refractive index is used to explain the nature of solute-solvent interactions.

The refractive index is an important physical property of liquids and liquid mixtures, which affects the solution of different problems in chemical engineering in order to develop industrial processes. Knowledge of the refractive index of a multicomponent mixture provides information regarding the interactions in these mixtures [1-3]. The prediction of the refractive index of multicomponent liquid mixtures is essential for many physico-chemical calculations, which include correlation of the refractive index with density, excess molar volumes,[4] and surface tension [5]. Based on the refractive index study, the reports on intermolecular interactions on binary liquids are very few [6-11]. A survey of the literature indicates that there were no reports on the study of the refractive index with regard to the present systems, except the work of Giner *et al*.[1], Oswal *et al*.[12], and Nain [13].

Tetrahydrofuran is a polar (μ = 1.75D) aprotic solvent. Its main use is as a precursor to polymers. It is also used as an industrial solvent for PVC and in varnishes.

This research aims to investigate the density and refractive index of binary liquid mixtures of tetrahydrofuran with benzene, methyl benzene and ethyl benzene at 308.15 K and intends to view molecular-level information from macroscopic properties on the molecular interactions between benzene, methyl benzene, and ethyl benzene with tetrahydrofuran.

MATERIALS AND METHODS

Tetrahydrofuran, benzene, methyl benzene, and ethyl benzene with greater than 98% purity were used in the investigation. Tetrahydrofuran is used as a solvent & benzene, methyl benzene, and ethyl benzene were used as solutes. Binary liquid mixtures were prepared by mixing weighed amounts of the pure liquids, adopting the method of a closed system.

The weighing was done by using a Mettler balance with a precision of ± 0.1 mg. The uncertainty in the mole function was estimated to be less than $\pm 1\times10^{-4}$. Mixtures were allowed to stand for some time before every measurement so as to avoid air bubbles. The densities were measured by using a single-capillary pycnometer with a bulb capacity of ≈ 10 mL. The capillary, with graduated marks, had a uniform bore and could be closed by a well-fitting glass cap. The marks on the capillary were calibrated by using triply distilled water. The uncertainty in density measurements was within $\pm 2\times10^{-5}$ g cm^{-3}.

The refractive indices of the pure liquids and their binary mixture were measured by using a thermostated Dioriter Abbe refractometer.

The refractometer was calibrated by measuring the refractive indices of triply distilled water and toluene at desired temperatures. The values of refractive index were obtained using sodium D light. The reproducibility of refractive index measurements was within ± 0.0001. The temperature of the test liquids during the measurements was maintained to an accuracy of ± 0.02 K.

The temperature of the test liquids during the measurements was maintained within an uncertainty of ± 0.01 K in an electronically controlled thermostatic water bath. The measurements were made with proper care in an AC room to avoid evaporation loss.

The purities of the liquids were checked by comparing the values of densities and refractive indices with literature data (Table 1) and found good in general.

RESULTS AND DISCUSSION

The experimental results of the measurement of densities and refractive indices of binary mixtures are presented in Table 2. The molar volume, molar refraction, excess refractive indices, and excess molar refraction were calculated from the measured data using the following equations 1 to 4, respectively.

$$V = (X_1M_1 + X_2M_2) / \rho \quad (1)$$

$$R_m = [(n^2-1) / (n^2+2)] V \quad (2)$$

$$n^E = n_m - (X_1n_1 + X_2n_2) \quad (3)$$

$$R_m^E = R_m - [X_1(R_m)_1 + X_2(R_m)_2] \quad (4)$$

Where ρ, n, V, and R_m are the density, refractive index, molar volume, and molar refraction of the mixture. n^E and R_m^E are the excess refractive indices, excess molar refraction. M_1 and M_2 are the molar masses, n_1 and n_2 are the refractive indices, V_1 and V_2 are the molar volumes of the benzenes, and $(R_m)_1$ and $(R_m)_2$ are the molar refraction indices of the benzene, methyl benzene, ethyl benzene, and tetrahydrofuran, respectively. 'X' is the mole fraction of THF, and suffixes 1 & 2 denote the components 1 & 2 in binary mixtures, and the values are given in Table 2.

The dependence of n^E, V^E, and R_m^E on the mole fraction of tetrahydrofuran (X_{THF}) for all the systems was fitted to the following Redlich-Kister equation by the least-squares method, and the values are given in Table 3.

$$Y^E = x(1-X)\sum_i A_i(2x-1)^i \quad (5)$$

Table 1: Comparison of experimental density and refractive index of pure liquids with literature values at 308.15 K.

Liquid	*Density x 10^{-3} (Kg m^{-3})*		*Refractive index x 10^3 (Kg m^{-1} s^{-1})*	
	Exptl.	*Lit.*	*Exptl.*	*Lit.*
Benzene	0.8629	0.86291 [14]	1.4919	1.4918[14]
Methyl benzene	0.8526	0.8529 [14]	1.4888	1.4887[17]
Ethyl benzene	0.8387	0.8390 [15]	1.4873	1.4874[18]
Tetrahydrofuran	0.8714	0.8701 [16]	1.4001	1.4008[19]

Where Y^E is n^E and R_m^E parameters.

The parameters A_i, obtained by a non-linear least squares polynomial fitting procedure, are also given in Table 3 together with the standard deviations (σ) values.

The variation of the parameters n^E and R_m^E, with mole fraction of tetrahydrofuran (X_{THF}) for the systems under study, is shown graphically in Figs 1 to 2, respectively.

The density and the refractive index have been measured over the whole composition range for the binary liquid mixtures of tetrahydrofuran (THF) with benzene (B), + methyl benzene (MB), and + ethyl benzene (EB) at 308.15 K. The experimental values of density and the refractive index are used to calculate the molar volume (V), molar refraction (R_m), excess refractive indices (n^E), and excess molar refraction (R_m^E) using the relations given above. The results are given in Table 2.

The variation of the parameters, of n^E and R_m^E, with mole fraction of THF (X_{THF}) for the systems under study is graphically shown in Figs 1 to 2, respectively.

Benzene (MB, −●−) and ethyl benzene (EB, −▲−) at 308.15 K.

It is evident from Fig. 1 that n^E values are positive for all the systems under study. From Fig.1 shows the graphical variations of n^E values with mole fractions of THF (X_{THF}) of the binary liquid mixtures at 308.15 K. These show positive deviation over the entire composition range. This indicates the existence of strong intermolecular interactions between the unlike molecules in the binary liquid mixtures. The positive n^E Vs X_{THF} plots were formed to be large and unsymmetrical with maxima between 0.5 to 0.6 mole fractions of THF (X_{THF}). The excess refractive index values fall in the following order.

THF + B < THF + MB < THF + EB

Tetrahydrofuran (THF) with benzene (B, −♦−), methyl benzene (MB, −●−) and ethyl benzene (EB, −▲−) at 308.15 K.

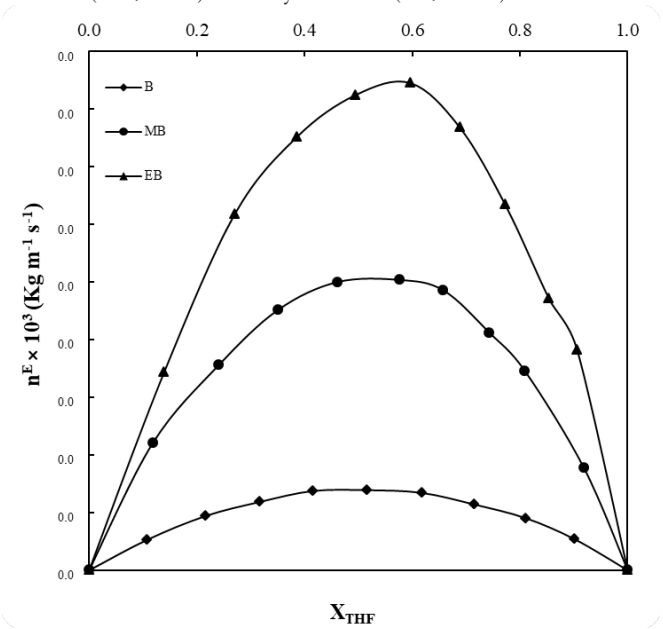

Fig 1: Plots of deviation in excess refractive indices (n^E) vs mole fraction of tetrahydrofuran (X_{THF}) for binary mixtures of tetrahydrofuran (THF) with benzene (B, −♦−), methyl

Table 2: Values of density (ρ), refractive index (n), excess refractive indices (n^E), excess molar refraction (R_m^E) for the binary mixtures tetrahydrofuran (THF) + benzene, methyl benzene and ethyl benzene at 308.15 K

Mole fraction of THF (X_{THF})	*ρ x 10^{-3} Kg m^{-3}*	*Refractive index (n)*	*n^E x10^5 m^3 mol*	*R_m^E x 10^5 m^3 mol^{-1}*
Tetrahydrofuran (THF) + Benzene (B)				
0.0000	0.8629	1.4919	0.0000	0.0000
0.1084	0.8638	1.4825	0.0005	0.0005
0.2157	0.8647	1.4730	0.0009	0.0009
0.3162	0.8655	1.4641	0.0012	0.0014
0.4163	0.8664	1.4551	0.0014	0.0015
0.5165	0.8672	1.4459	0.0014	0.0014
0.6173	0.8681	1.4366	0.0013	0.0012
0.7154	0.8689	1.4274	0.0011	0.0010
0.8103	0.8698	1.4184	0.0009	0.0007
0.9012	0.8706	1.4097	0.0005	0.0002
1.0000	0.8714	1.4001	0.0000	0.0000
Tetrahydrofuran (THF) + Methyl Benzene (MB)				
0.0000	0.8526	1.4888	0.0000	0.0000
0.1185	0.8546	1.4805	0.0022	0.0013
0.2408	0.8568	1.4710	0.0036	0.0022
0.3508	0.8588	1.4622	0.0045	0.0028
0.4611	0.8609	1.4529	0.0050	0.0031
0.5765	0.8632	1.4427	0.0050	0.0031
0.6579	0.8648	1.4353	0.0049	0.0029
0.7432	0.8664	1.4270	0.0041	0.0025
0.8090	0.8676	1.4205	0.0035	0.0021
0.9186	0.8697	1.4091	0.0018	0.0011
1.0000	0.8714	1.4001	0.0000	0.0000
Tetrahydrofuran (THF) + Ethyl Benzene (EB)				
0.0000	0.8387	1.4873	0.0000	0.0000
0.1394	0.8430	1.4786	0.0034	0.0020
0.2709	0.8471	1.4699	0.0062	0.0036
0.3862	0.8507	1.4611	0.0075	0.0044
0.4945	0.8544	1.4524	0.0082	0.0048
0.5969	0.8580	1.4437	0.0084	0.0048
0.6881	0.8611	1.4350	0.0077	0.0044
0.7727	0.8642	1.4263	0.0063	0.0035
0.8540	0.8670	1.4175	0.0047	0.0026
0.9062	0.8687	1.4121	0.0038	0.0020
1.0000	0.8714	1.4001	0.0000	0.0000

From Fig. 2, the R_m^E values give more information about the mixture phenomenon because it takes into account the electronic perturbation of the molecular orbital during the liquid mixing process [20]. In general, R_m is directly related to the dispersion forces, and the

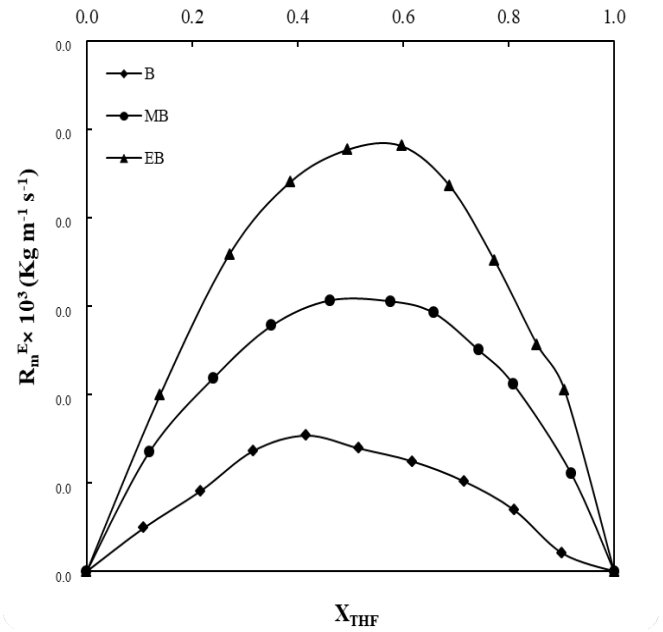

Fig. 2: Plots of deviation in excess molar refraction (R_m^E) vs mole fraction of tetrahydrofuran (X_{THF}) for binary mixtures of

positive values of R_m^E indicate that the dispersion forces are higher in the mixtures than in the pure liquids[21] whereas negative R_m^E values indicate that the dispersion forces are weaker in the mixtures than in the pure liquids, i.e., the presence of interactions amongst the mixture components. In the present study, we observed that the positive R_m^E values for the binary liquid mixtures follow the trend shown below.

THF + B < THF + MB < THF + EB

The observed trends (Fig. 2) of values indicate the presence of significant interactions in THF + benzene, methyl benzene,and ethyl benzenes.

This further reinforces conclusions regarding the intermolecular interactions from the variations of Excess volume (V^E)[22] and excess isentropic compressibility (K_S^E) values [23-25] of these mixtures. Also, the deviations in n^E values are found opposite to the sign of excess volumes (V^E) for all three binary mixtures, which are in agreement with the view proposed by Brocos *et al.*, [6, 26] and Jyoti N. Nayak ***et al.*** [27] These trends for these systems again support our view that the interactions between unlike molecules are quite obvious and these values are in the following order:

THF + B < THF + MB < THF + EB

The dependence of the above excess properties ($n^E$and R_m^E) on the mole fraction of tetrahydrofuran (X_{THF}) for all three systems was fitted to the Redlich-Kister equation by the non-linear least-squares polynomial methods using the relation given in equation5. The parameters A_i, obtained by a non-linear least square fitting procedure, are given in Table 3 together with the standard deviations (σ) values.

Table 3: Parameters of Eq. (5) and standard deviations

Excess Property	A_0	A_1	A_2	A_3	A_4	σ
Tetrahydrofuran (THF) + Benzene (B)						
n^E	-0.000007	0.00521	-0.00368	-0.00288	0.00136	0.00003
$R_m{}^E \times 10^5$ ($m^3\ mol^{-1}$)	-0.000011	0.00467	0.00304	-0.01816	0.01046	0.00008
Tetrahydrofuran (THF) + Methyl benzene (MB)						
n^E	0.00002	0.02095	-0.02845	0.01931	-0.01183	0.00007
$R_m{}^E \times 10^5$ ($m^3\ mol^{-1}$)	0.000002	0.01247	-0.01461	0.00639	-0.00424	0.00002
Tetrahydrofuran (THF) + Ethyl benzene (EB)						
n^E	-0.00003	0.02896	-0.02487	0.0024	-0.00713	0.00022
$R_m{}^E \times 10^5$ ($m^3\ mol^{-1}$)	-0.000013	0.01626	-0.00937	-0.00844	0.00161	0.00010

CONCLUSION

Experimental investigation of density & refractive index of binary liquid mixtures of tetrahydrofuran with benzene, methyl benzene, and ethyl benzene at 308.15 K and different compositions is measured. Refractive index and density measured values are useful for the evaluation of excess molar refraction, and deviations of refractive indices were correlated using the Redlich-Kister polynomial equation. These evaluations give information about strong molecular interactions between the components of the mixture. The order of increasing positive values of excess molar refraction followed:THF + B < THF + MB < THF + EB.

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