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Refractometric And Excess Parameter Study For Liquid Mixtures Containing High Order Alkanes (C₁₇) And 1-alkanols At Different Temperatures

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*, ** Department of Physics, Lucknow University, Lucknow

ABSTRACT

Refractive index and density have been measured at (293,298,303) K for binary mixtures of high order Alkanes (C₁₇) + 1-Alkanols (C₄-C₆) over the whole mole fraction range. From these data the excess molar volume, excess molar refraction and excess refractive index were calculated. These quantities are discussed in terms of intermolecular interactions as well as role of hydrogen bonding upon refractive indices of liquid mixtures.

Keywords : Refractive index, density, excess parameters, intermolecular interactions,

Introduction

The studies of organic liquids and liquid mixtures provide a deep knowledge into the molecular structures, which includes the molecular interactions between the molecules of liquid mixtures. The investigation of molecular interaction in different types of liquid mixtures with varying temperatures and compositions helps to predict the behavior of molecules after mixing very clearly [1-10]. The excess values of molar volume, molar refraction are quite useful in understanding the solute-solvent interactions in the liquid mixtures. The non-zero value of these parameters gives deep insight into the molecular interactions, arrangements and structural behavior [11]. As a part of our research programme, we present here the experimental results of the refractive index, n_m , and the density, ρ , for the binary mixtures of higher order Alkanes (C₁₇) i.e. Heptadecane (HEP) with 1-Butanol(B), 1-Pentanol(P), and 1-Hexanol(H) at (293,298,303)K over the whole mole fraction range. From these results, the excess molar volume (V^E), excess molar refraction (R^E) and excess refractive index (Δn_m) have been calculated over the whole mole fraction range, from which valuable information about intermolecular interactions between the components can be deduced. These organic species have their wide applicability in chemical analysis and industry. Literature survey shows that study with these organic liquids has been carried out by very few workers [12], but a systematic refractometric and excess parametric study of binary mixtures with above organic liquids, studied here, with temperature range (293,298,303)K is not available in literature.

Experimental Section

The refractive index of mixtures was measured using Abbe refractometer [13,14]. Before measuring refractive index on Abbe refractometer inspection of scale was made using test piece. The refractometer was calibrated with distilled water before each use. The temperature was maintained constant with the help of thermostatically controlled water bath (accuracy ± 0.01 K). Our refractive indices are accurate up to ± 0.0001 . The sample mixtures were directly injected into prism assembly of the instrument with the help of a plastic syringe. An average of three measurements was taken for a sample mixture.

The densities were measured with a bicapillary pycnometer having a bulb volume of about 10 cm³ and an internal

diameter of about 1mm. The pycnometer was calibrated at (293,298,303) K with doubly distilled water. The reproducibility of the density measurements was $\pm 3 \times 10^{-4}$ gcm⁻³. The pycnometer was immersed in water bath whose temperature was maintained constant by circulating the coolant liquid. All measurements were repeated three times, and then average values were calculated. The chemicals used were of AR/BDH grade. All the chemicals were purified by standard procedures discussed by Perrin and Armarego [15] before use.

Theory

The various physical parameters were calculated from the measured values of refractive index (n_m) and density (ρ) using the following formulae:

Excess molar volume

$$V^E = \left(\frac{X_1 M_1 + X_2 M_2}{\rho} \right) - \left(\frac{X_1 M_1}{\rho_1} + \frac{X_2 M_2}{\rho_2} \right) \quad (1)$$

Excess molar refraction

$$R^E = \frac{(n_m^2 - 1)}{(n_m^2 + 2)} \left(\frac{X_1 M_1 + X_2 M_2}{\rho} \right) - \left[\frac{(n_1^2 - 1)}{(n_1^2 + 2)} \frac{M_1}{\rho_1} \phi_1 + \frac{(n_2^2 - 1)}{(n_2^2 + 2)} \frac{M_2}{\rho_2} \phi_2 \right] \quad (2)$$

Excess refractive index

$$\Delta n_m = n_m - (X_1 n_1 + X_2 n_2) \quad (3)$$

Where X_1 and X_2 are mole fractions of respective components; ρ , ρ_1 , and ρ_2 are the densities and n_m , n_1 and n_2 are the measured refractive indices of the mixture and the pure component liquids, respectively. M_1 , M_2 are the molecular weights and ϕ_1 and ϕ_2 are volume fractions of respective components in liquid mixtures

Results

Experimentally find out densities (ρ), refractive indices (n_m) and calculated values of excess molar volume (V^E), excess molar refraction (R^E) and excess refractive index (Δn_m) for binary mixtures mixtures (HEP +B), (HEP +P) and (HEP +H) are given in Table.1 to 3 respectively. Values of V^E and R^E for mixtures (HEP +B), (HEP +P) and (HEP +H) at T=293 K are plotted with respect to mole fraction of Heptadecane (X_1) and shown in Fig.1 and 3, respectively. The variation of V^E and R^E with mole fraction of Heptadecane (X_1) for system (HEP +B) for all the three temperatures

viz. 293K, 298K, 303K are shown graphically in Fig. 2 and 4 respectively. Density is expressed in gm cm^{-3} and excess molar volume and excess molar refraction in $\text{cm}^3 \text{mol}^{-1}$.

Discussion

Excess parameter provides information about intermolecular interactions to the great extent excess molar quantities are properties of mixtures, which characterized the non ideal behavior of real mixtures. Alternatively it is defined as difference between the partial molar property of a component in a mixture and that of pure component. The negative values of excess molar volume V^E indicates about the presence of strong intermolecular interactions in the system[16]. There are mainly three factors viz. physical, chemical and structural which makes positive and negative contributions to the resultant value of excess molar volume and magnitude of contribution depends on mole fraction range. Physical contribution includes non specific physical contribution. Chemical effect appears due to breaking up of the liquid order of associated species. Structural effects appear due to geometrical fittings of molecules having comparatively small size into the voids created by molecules having comparatively large size. The differences in molar and free volumes are also responsible for structural effect[11,16]. From figure (1) it is observed that excess molar volume, V^E , for all systems have negative values over the entire range of mixture composition, for all the selected values of temperature. Negative values of excess molar volume indicates about a closer packing of molecules, because in the present case there is large difference between molar volumes of Heptadecane and 1-Alkanols as molar volume for Heptadecane and 1-Butanol at temperature 293K, is $309.2464 \text{ cm}^3 \text{mol}^{-1}$ and $91.0680 \text{ cm}^3 \text{mol}^{-1}$, respectively.

Thus structural effect becomes predominating factor in this case. Hydrogen bonds plays a role in reducing the experimental values of refractive index in alcohol containing systems, which may also results in negative value of excess quantities[12]. The observed negative deviation of excess molar volume confirms the presence of strong molecular association in the system. Hence it is concluded that there exists strong intermolecular interactions in system. The values of V^E follow the sequence $(\text{HEP} + \text{B}) > (\text{HEP} + \text{P}) > (\text{HEP} + \text{H})$. Almost no effect of temperature increasing is observed for excess molar volume for all systems under consideration. The effect of temperature on excess molar volume for (HEP + B) system is shown in figure (2).

Excess molar refraction represents the complex electronic perturbation between liquid components due to orbital mixing of components. Excess molar refraction, R^E , gives the strength of interaction in a mixture and is sensitive function of mixture composition along with temperature and wavelength[17,18]. From figure (3) it is observed that excess molar refraction R^E have very large negative values for all systems over the entire range of mixture composition, for all the selected values of temperatures. This further supports the existence of strong interaction in these systems. The order of negative magnitude of R^E values is same as existed for excess molar volume V^E i.e. $(\text{HEP} + \text{B}) > (\text{HEP} + \text{P}) > (\text{HEP} + \text{H})$. This again supports the existence of strong interaction between unlike molecules of mixtures. The highest value of R^E for (HEP + B) mixture suggests that interaction between Heptadecane and 1-Butanol is strongest as compared to that between Heptadecane and other 1-Alkanols. It is also seen from fig (3) that for all the binary mixtures containing Heptadecane R^E decreases as a function of mole fraction (X_1) of Heptadecane, attains a minimum and then further increases with X_1 . The minimum of R^E versus X_1 curve remains at same position i.e. for same X_1 values in Heptadecane deficient region. This supports the existence of complex interaction in system containing Alkanes and Alkanols. Figure (4) represents the effect of temperature on R^E for system (HEP + B). It is observed that as temperature changes, almost no effect in the negative value of R^E are obtained, for all the systems under consideration, however there exists very-very small change in R^E . This may

arise due to factor that when temperature increases thermal agitation increases and hence electronic perturbation between orbital increases. For these systems, the excess refractive index (Δn_m) has extremely low values for all the value of temperatures over entire range of composition. Excess refractive index has positive values for all the systems over the entire range of mixture composition and for all the selected values of temperatures. With mole fraction of heptadecane (X_1) the positive values of excess refractive index, first, increases and attains a maximum and thereafter it becomes decreases for all the systems.

Conclusions

Thus from above mentioned discussion, it may concluded that the effects observed for all the systems under investigation show dependence on hydrogen bonding of components of mixture. It may also conclude that refractometric study of liquid mixtures provides a better insight for a comprehensive investigation of molecular association between molecules of mixtures. The variation of excess parameters with concentration and temperature strongly supports the molecular association in these liquid mixtures. This type of result may be used for developing mixing rules for binary liquid mixtures.

Figure1 Excess molar volume V^E as a function of mole fraction of HEP, X_1 , for mixture (HEP + B), (HEP + P) and (HEP + H) at $T = 293 \text{ K}$

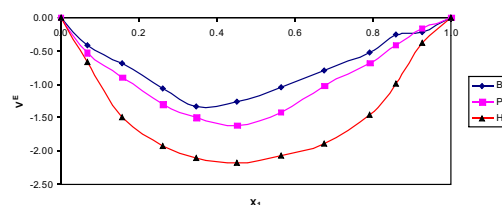


Figure2 Excess molar volumes V^E as a function of mole fraction, X_1 , of Heptadecane (HEP) for mixture (HEP + B) at temperature 293 K, 298 K, 303K

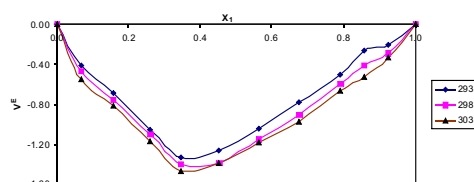


Figure3 Excess molar refraction, R^E , as a function of mole fraction of HEP, X_1 , for mixture (HEP + B), (HEP + P) and (HEP + H) at $T = 293 \text{ K}$

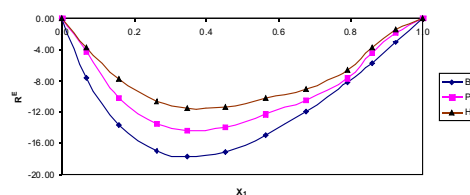


Figure 4. Excess molar refraction, R^E , as a function of mole fraction, X_1 , of Heptadecane (HEP) for mixture (HEP + B) at temperature 293 K, 298 K, 303K.

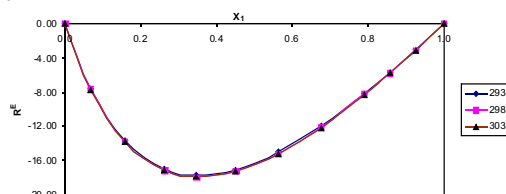


Table 1 Refractive index (n_m), density (ρ), excess molar volume (V^E), excess molar refraction (R^E), excess refractive index (Δn_m) for mixture Heptadecane and 1-Butanol at temperature 293K, 298K, 303K as a function of the mole fraction X_1 of Heptadecane

T	X_1	ρ (g cm ⁻³)	n_m	V^E (cm ³ mol ⁻¹)	R^E (cm ³ mol ⁻¹)	Δn_m
293K	0.0000	0.8091	1.3985	0.0000	0.0000	0.0000
	0.0670	0.8061	1.4060	-0.4145	-7.6451	0.0049
	0.1570	0.8013	1.4134	-0.6843	-13.6514	0.0088
	0.2620	0.7976	1.4195	-1.0569	-16.9368	0.0109
	0.3470	0.7952	1.4233	-1.3294	-17.7064	0.0114
	0.4520	0.7912	1.4269	-1.2669	-17.0283	0.0110
	0.5630	0.7873	1.4299	-1.0366	-14.9850	0.0097
	0.6760	0.7841	1.4323	-0.7866	-11.9578	0.0077
	0.7910	0.7814	1.4343	-0.5109	-8.1625	0.0053
	0.8580	0.7798	1.4353	-0.2598	-5.6749	0.0037
	0.9250	0.7789	1.4362	-0.2110	-3.0885	0.0020
	1.0000	0.7776	1.4371	0.0000	0.0000	0.0000
298K	0.0000	0.8017	1.3978	0.0000	0.0000	0.0000
	0.0670	0.7991	1.4053	-0.4677	-7.7076	0.0049
	0.1570	0.7944	1.4126	-0.7580	-13.7631	0.0088
	0.2620	0.7905	1.4187	-1.1021	-17.0598	0.0109
	0.3470	0.7882	1.4224	-1.3979	-17.8459	0.0113
	0.4520	0.7844	1.4260	-1.3824	-17.1726	0.0109
	0.5630	0.7805	1.4289	-1.1538	-15.1245	0.0095
	0.6760	0.7773	1.4313	-0.9054	-12.0755	0.0076
	0.7910	0.7745	1.4333	-0.5964	-8.2440	0.0052
	0.8580	0.7731	1.4343	-0.4149	-5.7567	0.0036
	0.9250	0.7720	1.4352	-0.2942	-3.1325	0.0020
	1.0000	0.7705	1.4361	0.0000	0.0000	0.0000
303K	0.0000	0.7995	1.3971	0.0000	0.0000	0.0000
	0.0670	0.7975	1.4046	-0.5536	-7.7441	0.0049
	0.1570	0.7925	1.4119	-0.8166	-13.8046	0.0088
	0.2620	0.7886	1.4180	-1.1759	-17.1110	0.0109
	0.3470	0.7862	1.4217	-1.4628	-17.8958	0.0113
	0.4520	0.7821	1.4253	-1.3844	-17.2048	0.0109
	0.5630	0.7783	1.4282	-1.1835	-15.1599	0.0095
	0.6760	0.7752	1.4306	-0.9692	-12.1140	0.0076
	0.7910	0.7724	1.4326	-0.6666	-8.2772	0.0052
	0.8580	0.7711	1.4336	-0.5252	-5.7958	0.0036
	0.9250	0.7698	1.4345	-0.3336	-3.1484	0.0020
	1.0000	0.7682	1.4354	0.0000	0.0000	0.0000

Table 2 Refractive index (n_m), density (ρ), excess molar volume (V^E), excess molar refraction (R^E), excess refractive index (Δn_m) for mixture Heptadecane and 1-Pentanol at temperature 293K, 298K, 303K as a function of the mole fraction X_1 of Heptadecane

T	X_1	ρ (g cm ⁻³)	n_m	V^E (cm ³ mol ⁻¹)	R^E (cm ³ mol ⁻¹)	Δn_m
293K	0.0000	0.8139	1.4100	0.0000	0.0000	0.0000
	0.0463	0.8131	1.4133	-0.5235	-4.2179	0.0020
	0.1478	0.8071	1.4190	-0.8930	-10.1597	0.0050
	0.2638	0.8020	1.4237	-1.2989	-13.4382	0.0066
	0.3583	0.7982	1.4267	-1.4926	-14.2610	0.0070
	0.4592	0.7946	1.4292	-1.6155	-13.8938	0.0068
	0.5734	0.7901	1.4315	-1.4136	-12.2796	0.0060
	0.6583	0.7865	1.4329	-1.0144	-10.4461	0.0051
	0.7423	0.7836	1.4342	-0.6785	-8.2601	0.0041
	0.8718	0.7805	1.4358	-0.4075	-4.3945	0.0022
	0.9479	0.7787	1.4366	-0.1590	-1.8392	0.0009
	1.0000	0.7776	1.4371	0.0000	0.0000	0.0000
298K	0.0000	0.8103	1.4081	0.0000	0.0000	0.0000
	0.0463	0.8093	1.4115	-0.5646	-4.3001	0.0021
	0.1478	0.8028	1.4174	-0.9898	-10.3479	0.0052
	0.2638	0.7969	1.4223	-1.3808	-13.6605	0.0068
	0.3583	0.7926	1.4253	-1.5590	-14.4932	0.0072
	0.4592	0.7886	1.4280	-1.6705	-14.0993	0.0070
	0.5734	0.7838	1.4303	-1.4676	-12.4654	0.0061
	0.6583	0.7801	1.4318	-1.0893	-10.8037	0.0053
	0.7423	0.7769	1.4331	-0.7032	-8.3788	0.0042
	0.8718	0.7737	1.4347	-0.4656	-4.4761	0.0022
	0.9479	0.7718	1.4356	-0.2156	-1.8755	0.0010
	1.0000	0.7705	1.4361	0.0000	0.0000	0.0000
303K	0.0000	0.8067	1.4069	0.0000	0.0000	0.0000
	0.0463	0.8062	1.4104	-0.6185	-4.3078	0.0022
	0.1478	0.8000	1.4164	-1.0592	-10.3584	0.0053
	0.2638	0.7941	1.4213	-1.4169	-13.6718	0.0069
	0.3583	0.7900	1.4244	-1.6127	-14.5074	0.0073
	0.4592	0.7860	1.4271	-1.6990	-14.1139	0.0071
	0.5734	0.7813	1.4295	-1.4941	-12.4748	0.0063
	0.6583	0.7777	1.4310	-1.1249	-10.6192	0.0053
	0.7423	0.7745	1.4323	-0.7194	-8.3925	0.0042
	0.8718	0.7714	1.4340	-0.4910	-4.4812	0.0023
	0.9479	0.7696	1.4349	-0.2653	-1.8879	0.0010
	1.0000	0.7682	1.4354	0.0000	0.0000	0.0000

Table 3 : Refractive index (n_m), density (ρ), excess molar volume (V^E), excess molar refraction (R^E), excess refractive index (Δn_m) for mixture Heptadecane and 1-Hexanol at temperature 293K, 298K, 303K as a function of the mole fraction X_1 of Heptadecane

T	X_1	ρ (g cm ⁻³)	n_m	V^E (cm ³ mol ⁻¹)	R^E (cm ³ mol ⁻¹)	Δn_m	T	X_1	ρ (g cm ⁻³)	n_m	V^E (cm ³ mol ⁻¹)	R^E (cm ³ mol ⁻¹)	Δn_m
293K	0.0000	0.8133	1.4175	0.0000	0.0000	0.0000	303K	0.0000	0.8057	1.4149	0.0000	0.0000	0.0000
	0.0573	0.8126	1.4201	-0.6612	-3.7616	0.0015		0.0573	0.8054	1.4176	-0.7839	-3.8519	0.0015
	0.1438	0.8108	1.4232	-1.4892	-7.7259	0.0029		0.1438	0.8030	1.4209	-1.5857	-7.8628	0.0031
	0.2629	0.8055	1.4267	-1.9193	-10.5944	0.0040		0.2629	0.7975	1.4245	-2.0624	-10.7868	0.0042
	0.3709	0.8008	1.4291	-2.0892	-11.4614	0.0043		0.3709	0.7924	1.4271	-2.2067	-11.6475	0.0046
	0.4538	0.7976	1.4307	-2.1682	-11.3142	0.0043		0.4538	0.7890	1.4287	-2.2787	-11.5010	0.0045
	0.5729	0.7930	1.4325	-2.0670	-10.1552	0.0038		0.5729	0.7844	1.4306	-2.2329	-10.3355	0.0040
	0.6422	0.7903	1.4335	-1.8832	-9.0437	0.0034		0.6422	0.7816	1.4316	-2.0473	-9.2126	0.0035
	0.7584	0.7860	1.4349	-1.4522	-6.6456	0.0025		0.7584	0.7774	1.4331	-1.6941	-6.7962	0.0027
	0.8737	0.7823	1.4360	-0.9945	-3.7603	0.0014		0.8737	0.7734	1.4343	-1.1699	-3.8470	0.0015
	0.9510	0.7793	1.4367	-0.3730	-1.5048	0.0006		0.9510	0.7704	1.4350	-0.5654	-1.5715	0.0006
	1.0000	0.7776	1.4371	0.0000	0.0000	0.0000		1.0000	0.7682	1.4354	0.0000	0.0000	0.0000
298K	0.0000	0.8095	1.4163	0.0000	0.0000	0.0000							
	0.0573	0.8091	1.4189	-0.7943	-3.8603	0.0015							
	0.1438	0.8062	1.4221	-1.5450	-7.8604	0.0030							
	0.2629	0.8003	1.4256	-1.9856	-10.7729	0.0041							
	0.3709	0.7951	1.4280	-2.1433	-11.6462	0.0044							
	0.4538	0.7917	1.4296	-2.2424	-11.4997	0.0043							
	0.5729	0.7868	1.4315	-2.1488	-10.3119	0.0039							
	0.6422	0.7840	1.4325	-1.9823	-9.1883	0.0035							
	0.7584	0.7797	1.4338	-1.6273	-6.7905	0.0025							
	0.8737	0.7756	1.4350	-1.0962	-3.8336	0.0014							
	0.9510	0.7725	1.4357	-0.4722	-1.5494	0.0006							
	1.0000	0.7705	1.4361	0.0000	0.0000	0.0000							

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