Densities, Viscosities and Refractive Indices of n- Butanol + Allyl Chloride Mixture at 298K

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Abstract

The density ρ , viscosity η , refractive index *n* and dielectric constant ε_s of binary mixture of Allyl Chloride (AC) with n-Butanol including those of the pure liquids, were measured over the complete composition range at 298K. The experimental data is used to calculate excess molar volumes (v^E), excess permittivity (ε_s^{E}), excess viscosity (η^{E}), excess refractive index, molar refraction, excess molar polarization and Dunstan's constant. The variations of these parameters with composition and the effect of bonding in the binary mixture are discussed from the point of view of intermolecular interactions in these mixtures. The positive value of excess molar volume (v^E) is attributed to the breaking of H-bonded associated species formed between unlike molecule at 298K.

Keywords: Excess density, Intermolecular interactions, Excess viscosity

1. Introduction

The particular manner whereby solute and solvent molecules are associated with one another in a liquid mixture brings about a distinct effect on the properties of the resulting system. Although physical properties of pure liquids abound in the literature, such properties measured for binary systems have been rather scarcely investigated up to now. Previously we described the intermolecular interactions between the components of Alcohols with other groups of liquids. In this work we extend the study by investigating properties like density, refractive index & viscosity. Ordinary liquids are made up of large molecules bound by weakly directional van der Waals forces; n-Butanol on the contrary, is made up of small molecules strongly bound by hydrogen bonds, in order of magnitude greater than van der Waals forces.

Hydrogen bonds in butanol are responsible for the striking behavior, and in some instances provide evidence for the role of butanol in the sequence of chemical reactions. Hydrogen bonding in butanol is believed to be highly cooperative, i.e., interaction of butanol molecule with a cluster of hydrogen bonded molecules is more likely than interaction with a single molecule to give a dimmer. Binary & ternary representations and prediction of intermolecular interactions with liquid dynamics by using reliable models intermolecular interactions is an essential task of considerable importance to gain some insights into the interactions among the groups of molecules or in important technological applications such as mass transfer, fluid flow heat transfer, design calculation or separation processes. To predict nature of interactions within the binary system excess properties were used.

In AC (CH2=CHCH2Cl) molecules strong dipole - dipole interaction exists. Strong hydrogen bonding is present in n-Butanol (C_4H_9OH) molecules. Because of presence of different type of association in both molecules, it is interesting to study the nature of intermolecular interactions between n-Butanol + Allyl Chloride. Allyl chloride is a common alkylating agent, useful in the manufacture of pharmaceuticals and pesticides. n-Butanol is used in the manufacture of pharmaceuticals. n-Butanol is also used as an extractant in the manufacture of antibiotics, hormones, and vitamins (Mellan, I., 1950; Doolittle, 1954); industrial uses of n-Butanol are as polymers, pyroxylin plastics, herbicide esters; n-Butanol is used as an ingredient in perfumes and as solvents for the extraction of essential oils (Doolittle, 1954), a solvent for paints, coatings, natural resins, gums, synthetic resins, dyes, alkaloids, and camphor. Alcohols play an important role in many chemical reactions due to its ability to undergo self-association with manifold internal structures and are in wide use in industries and science as reagents, solvents and fuels (Jui, 2005).

2. Experimental

2.1 Chemicals

The chemicals used in the present paper (n-Butanol & AC) are of spectroscopic grade and were used without further purification. The solutions were prepared at eleven different volume percentage of AC 0% to 100% in steps of 10% within 0.02% error limit.

2.2 Measurements

Viscosities were measured using Brookfield Viscometer with an accuracy of $\pm 1\%$ for small sample size of 1 ml. Refractive indices (at Sodium D line) were measured by using Abbe Refractometer thermostated with constant water bath with precision of ± 0.001 and the values presented here are the mean values from five independent readings for each sample.

Density is measured using specific gravity bottle having an internal volume 3 ml and was calibrated with distilled water at 298K. Three specific gravity bottles were immersed in a thermostatic bath which was controlled at 298 K. A precision digital thermometer with a thermister probe was used to read the temperature with an accuracy of ± 0.10 K. The mixture densities were obtained by averaging the results from these three specific gravity bottles.

Dielectric constant is measured by using self developed setup in our laboratory.

3. Result & Discussion

The measured and estimated values of refractive index, density, viscosity and dielectric constant at 298 K are given in table 1. The density of the binary mixture of AC and n-Butanol is increasing as a mole fraction of AC in the mixture is increasing. This is expected because density of pure AC is more than that of pure n-Butanol. This variation of density is shown in table 1. From table 1 it can also be seen that the refractive index (n) is increasing with increase in mole fraction of AC in the mixture. This increase in n is further supported by the increase in density of the mixture with increase in mole fraction of AC.

Viscosity decreases as the mole fraction of AC in the binary mixture increases, as shown in table 1. Decrease in viscosity of the binary mixture can be attributed to decrease in the effect of hydrogen bonding with increase in mole fraction of allyl chloride. This is because n-Butanol is more viscous than AC. It is due to presence of strong hydrogen bonding in n-Butanol and more number of carbon atom and long straight chain compound. Figure 1 shows that the excess viscosity is positive.

The values of dielectric constant decrease with increase in mole fraction of AC in the mixture, as can be seen from the fifth column of table 1. This can be attributed to the decrease in hydrogen bonding in the mixture. This is because there is dipole – dipole interaction in pure AC and strong hydrogen bonding as in pure n-Butanol.

From figure 2 and 3, we see that the excess refractive index and excess density are negative, indicating the existence of intermolecular interaction and that these two excess parameters are supporting each other.

The Excess molar volumes v_m^E were calculated by using the following relation (El Hefnawy M, 2005; Yaw,

2005)

$$v_{m}^{E} = x_{1}M_{1}\left(\frac{1}{\rho} - \frac{1}{\rho_{1}}\right) + x_{2}M_{2}\left(\frac{1}{\rho} - \frac{1}{\rho_{2}}\right)$$
(1)

where M is the molecular mass; subscripts 1 and 2 stand for the pure components, AC and n-Butanol respectively.

The variation of v_m as mole fraction of AC is shown in figure 4. In pure n-Butanol, molecules are hydrogen bonded. On adding AC, these molecules get in between the n-Butanol molecules breaking the hydrogen bonds of n-Butanol molecules, thus leading to multimer formation. This changes the intermolecular interaction pattern and becomes the cause for decrease in molar volume of the mixture with increase in mole fraction of AC in the mixture as shown in figure 4. The excess molar volume is positive, as shown in figure 5. This fact is further supported by the negative excess density, shown in figure 3.

Excess permittivity is given by (25-26)

$$\varepsilon_{s}^{E} = (\varepsilon_{s})_{m} - [(\varepsilon_{s})_{A} x_{A} + (\varepsilon_{s})_{B} x_{B}]$$
⁽²⁾

The variation of ε_s^{E} for increase in mole fraction of AC in the binary mixture is shown in figure 6. The positive values of ε_s^{E} indicate that the solute and solvent interact in such a way that the effective dipole moment increases. There is formation of monomers and dimers.

Molar refraction is given by the formula

$$R = v_{m}[(n^{2}-1)/(n^{2}+2)]$$
(3)

Figure 7 shows that the molar refraction decreases with increase in mole fraction of AC in the mixture. As molar refraction is a measure of total polarizability of a mole of a substance and is dependent on index of refraction and temperature, decrease in molar refraction indicates the decrease in molar polarization and decrease in dielectric constant with increase in the mole fraction of AC increases in the mixture.

Molar polarization is given by

$$P_{\rm m} = \left[(\varepsilon - 1) / (\varepsilon + 2) \right] (M / \rho) \tag{4}$$

The estimated P_m is shown in figure 8. Decrease in P_m with increase in mole fraction of AC in the mixture also indicates the decrease in the effect of hydrogen bonding in the mixture. Figure 9 shows the positive excess molar polarization for different mole fraction of AC in the mixture.

Dunstan's constant is given by

$(\rho/M) \eta . 10^6$.

The calculated values of Dunstan's constant are shown in figure 10.

Dunstan (1909) showed that for 'associated liquids' the value of this constant is much higher. From figure 10, it can be seen that higher values of Dunstan's constant are in the range of 30590 - 3000 indicating the association of the two liquids. The value decreases as the concentration of AC increases. This suggests breaking of hydrogen bonded associated species formed by unlike molecules at same temperature.

The excess properties follow a Redlich –Kister type polynomial equation (23-24)

$$f(x_1) = x_1 \cdot (1 - x_1) \cdot \sum_{j} B_j \cdot (1 - 2 \cdot x_1)^j$$

This equation is fitted by using method of least squares to all the excess properties. The estimated B_j coefficients are given in table 2. The standard error (σ) between the data and the Redlich Kister model for the excess properties is given in the last column of the table. σ is calculated by using the relation

 $\sigma = (\Sigma$ square of error between data and model/ (n-1)) (5)

where n is the number of experimental data points. The graphs of excess properties and the corresponding Redlich-Kister model are shown the figures.

The standard errors in the B_j coefficients of the Redlich-Kister model along with the 95% confidence intervals for all excess properties are given in table 3. Comparison of estimated values refractive index, density, viscosity and dielectric constants for pure liquids (i.e. allyl chloride and n-Butanol) at 298K with the literature values is given in table 4.

4. Conclusions

The summary of conclusions of intermolecular interaction between allyl chloride and n-Butanol is as follows:

• The values of refractive index and density are increasing with increase in mole fraction of allyl chloride. These two parameters are supporting each other.

• Decrease in viscosity of the binary mixture with increase in mole fraction of allyl chloride indicates that effect of hydrogen bonding decreases as mole fraction of allyl chloride increases

• The values of $_{\rm s}$ decrease with the addition of allyl chloride in n-Butanol. This is due to decrease in effect of hydrogen bonding.

• Variations in refractive index, density, viscosity and dielectric constant with mole fraction of AC in the binary mixture of AC + n-Butanol indicate the existence of intermolecular interaction between n-Butanol and allyl chloride.

• There is monomer and dimer formation in the mixture which is supported by positive excess viscosity, positive excess molar volume, positive excess permittivity, decrease in molar refraction, decrease in molar polarization and decrease in dielectric constant.

• The estimated values of refractive index, density, viscosity for pure liquids at 298 K, are in good agreement with those reported in the literature.

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Mole fraction of AC	Refractive index	Density of AC + n-Butanol	Viscosity	Dielectric constant
0	1.397	0.8055	2.578	15.40
0.111	1.398	0.8155	2.350	14.58
0.220	1.400	0.8255	2.122	13.77
0.326	1.401	0.8365	1.894	12.96
0.429	1.403	0.8485	1.666	12.13
0.530	1.403	0.8606	1.439	11.32
0.629	1.404	0.8732	1.210	10.49
0.725	1.405	0.8851	0.984	9.68
0.819	1.407	0.9002	0.756	8.85
0.910	1.409	0.9119	0.528	8.06
1	1.412	0.9390	0.301	7.27

Table 1. Refractive index, density, viscosity and dielectric constant of allyl chloride + n-Butanol mixture at 298K

Excess property		Standard			
Ļ	B ₀	\mathbf{B}_1	B ₂	B ₃	Error
Excess density	-3.633E-02	-0.0331	1.349E-02	0.044403455	1.5223E-03
Excess viscosity	1.921E+00	6.824E-01	-3.464E-01	-2.140E+00	4.053E-02
Excess refractive index	-2.031E-02	-8.026E-03	7.014E-03	-6.525E-03	3.524E-04
Excess molar volume	2.095E+00	3.724E+00	-1.495E+00	-5.355E+00	1.458E-01

Table 2. B_j coefficients and standard error for excess density, excess viscosity, excess refractive index, and excess molar volume

Table 3. Standard errors and 95% confidence intervals of the estimated B_j coefficients for excess density, excess viscosity, excess refractive index and excess molar volume

Excess property		B _j coefficients				
Ļ		B ₀	B ₁	B ₂	B ₃	
Excess Density	standard error of parameter	0.0029	0.0115	0.0135	0.0299	
	95 % confidence interval	0.0069	0.0271	0.0318	0.0707	
Excess Viscosity	standard error of parameter	0.0004	0.0017	0.0020	0.0045	
	95 % confidence interval	0.0010	0.0041	0.0048	0.0107	
Excess Refractive Index	standard error of parameter	0.0007	0.0026	0.0031	0.0069	
	95 % confidence interval	0.0016	0.0062	0.0073	0.0163	
Excess Molar Volume	standard error of parameter	0.2598	1.0201	1.1975	2.6600	
	95 % confidence interval	0.6143	2.4121	2.8315	6.2898	
Excess	standard error of parameter	0.0117	0.0460	0.0540	0.1199	
permittivity	95 % confidence interval	0.0277	0.1087	0.1276	0.2835	
Excess molar polarization	standard error of parameter	0.1946	0.7641	0.8969	1.9924	
	95 % confidence interval	0.4601	1.8068	2.1209	4.7113	

Property ↓	$Liquid \rightarrow$	n-Butanol			Allyl chloride				
	This work	1.397			1.412				
Refractive index Lite v (temp Re Thi Lite Density v (temp Re Thi	Literature value (temperature)	1.3993 (20)	1.39741 (25)	1.39716 (25)	1.39719 (25)	1.4157(20)	1.413 ()	1.4157 (20)	1.414 (20)
	Ref. No.	[28]	[35]	[39]	[40]	[28]	[18]	[17]	[19]
	This work		0.80)554			0.939	yl chloride 1.412 .413 1.4157 () (20) [18] [17] 0.939 9376 0.9392 (20) (20) [28] [43] 0.301 0.354 0.32 (20) (22) 46] [47] 7.27 8.2 8.2 (20) () 41] [42]	
Density	Literature value (temperature)	0.8098 (20)	0.80556 (25)	0.80567 (25)	0.80554 (25)	0.81	0.9376 (20)	0.9392 (20)	0.939 (25)
	Ref. No.	[28]	[29]	[30]	[31]	[36]	[28]	[43]	[45]
	This work		2.5	578			$\begin{array}{c ccccccccccccccccccccccccccccccccccc$		
Viscosity	Literature value (temperature)	2.948 (20)	2.578 (25)	2.2601 (30)	2.2601 (30)	0.347 (15)	0.354 (20)	0.32 (22)	0.34 (20)
	Ref. No.	[28]	[32]	[33]	[34]	[28]	[46]	[47]	[48]
	This work	15.4				7.27			
Dielectric constant	Literature value (temperature)	17.8 (20)	17.5 (20)	17.15 (25)	1 7.5 (25)	8.2 (20)	8.2 (20)	8.2 ()	
	Ref. No.	[28]	[35]	[37]	[38]	[44]	[41]	[42]	

Table 4. Comparison of estimated values refractive index, density, viscosity and dielectric constants for pure liquids(i.e. allyl chloride and n-Butanol) at 298K with the literature values







Figure 2. Variation of excess refractive index of AC + n-Butanol at 298 K



Figure 3. Variation of excess density of AC + n-Butanol at 298 K



Figure 4. Variation of molar volume of AC+ n-Butanol at 298 K



Figure 5. Variation of excess molar volume of AC + n-Butanol at 298 K



Figure 6. Variation of excess permitivity of AC +n-Butanol at 298 K



Figure 7. Variation of molar refraction of AC + n-Butanol at 298 K



Figure 8. Variation of molar polarization of AC + n-Butanol at 298 K







Figure 10. Variation of Dunstan's constant of AC + n-Butanol at 298 K