Viscometric and Refractometric Studies on Molecular Interactions in Binary Liquid Mixture containing non-associative component

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Received: 05/2023, Published: 06/2023

Abstract

In this study, Density (ρ), viscosity (η), and refractive index (n_D) including those of pure liquids were measured over the entire mole fraction range at 298.15 K, for the binary mixture of ethyl benzoate (EB) and 1-Butanol (1-BuOH). Densities and viscosities were determined using a viscosity meter X sample 530 with SVM 3001 (Anton Paar). Refractive indices were measured with a digital refractometer (RFM 81, Bellingham & Stanley Ltd). From these experimentally data, various thermophysical properties such as excess molar volume, V^E, deviations in viscosity ($\Delta\eta$), refractive index (Δn_D) and excess free energy of activation (ΔG^{*E}) have been calculated then fitted to the Redlich- Kister polynomial equation. In the chemical and biochemical industries, these results can be used to develop comprehensive theoretical models for the design of various technological processes. The values estimated by theoretical procedure and the experimental results demonstrated good agreement. The deviations for excess properties have been explained on the basis of the intermolecular interactions. The structural characteristics of the liquid components influence the trend of excess properties. They are hence results of molecular interactions.

Keywords: Ethyl benzoate, 1-Butanol, thermophysical properties, excess properties, Redlich-Kister equation.

Tob Regul Sci. ™ 2023;9(1): 2512-2523

DOI: doi.org/10.18001/TRS.9.1.173

INTRODUCTION

In order to understand the types of interactions that exist between components of binary mixture, it is important to understand the physicochemical properties of pure and binary mixes. Understanding thermodynamic and thermophysical qualities is important for developing models and planning processes in the chemical, pharmaceutical, and petrochemical industries. The results that have been achieved can be changed with the use of theoretical and numerical investigations of excess and deviation functions. The fundamental thermophysical properties of the molecular interaction system include density, viscosity, dielectric constant, and refractive index [01, 02]. By revealing information on the forces influencing the mixing up of species to form the binary mixture, the variation of these features with composition provides us with important information on intermolecular interactions. There are usually two types of associations in the binary mixture. Self-association occurs when the binary mixture's components engage with one another, and mutual association occurs when they interact with the other component. Due to charge transfer, dipole-dipole interactions, and dipole-induced dipole interactions, the polar-polar and polar-non-polar components of a binary mixture are likely to interact with one another. [03]. Alkanol molecules are polar and self-associated through hydrogen bonding of their hydroxyl groups[04].Many studies have measured the density, viscosity, and sound velocity for a variety of binary mixes that include alcohol as one of the components[05,06]. Ethyl benzoate, an aromatic ester and one of the various types of esters, is a polar solvent (= 1.8 D), a strongly associated liquid, and a component of perfume. It serves as a flavour agent for food. It is a key ingredient in synthetic fruit flavors. Additionally, it serves as a preservative as an ingredient in fragrances for cosmetics and Personal care products. [07,08] Since molecules with -OH groups form associative liquids as a result of hydrogen bonding, it is crucial to understand how the presence of molecules with other functional groups affects these molecules. The aim of the current investigation is Reporting the impact of non-associative molecules on associative molecules in liquid state by measuring densities, viscosities and refractive indices of the binary system {ethyl benzoate (1) + 1-Butanol (2)} at atmospheric pressure and temperature at 298.15K to obtain a better understanding of the nature of molecule interaction. These data have been used to calculate the excess properties, excess molar volume (VE), deviation in viscosity ($\Delta\eta$), excess Gibb's free energy of activation for viscous flow (ΔG^{*E}) and deviation in refractive index (ΔnD). The Redlich-Kister polynomial equation has been employed to correlate the excess properties.

1. EXPERIMENTAL SECTION

1.1. Chemicals

Table 1 indicates the sources, purities and chemical composition of the liquids used in the

present study.

Table 1: Pure component specifications: suppliers, CAS number, specified purity.

Chemical name	Supplier	CAS N°	Mass fraction purity (Supplier)
Ethyl benzoate (EB)	Acrōs organics	93-89-0	99+%
2-Butanol (2-BuOH)	Sigma Aldrich	78-92-2	≥ 99 %

The chemicals weren't subjected to any additional purification. The purity of these samples was checked by comparing the measured densities, viscosities and refractive indices at 298.15 K with those previously reported by Nain et al. (2008) [12],] Nikam et al. (1998) [13], Zaoui–Djelloul Daouadji Manel et al. . (2022) [14], Lien et al. (2003) [15]. As demonstrated in Table 2, the obtained values are in good agreement with the literature values. This agreement can be considered a confirmation of the results of our apparatus.

Table 2: Comparison of experimental density, ρ, dynamic viscosities, η and refractive indices, *nD*, of the pure components with the corresponding literature values at T= 298.15 K and at p = 1×10^5 Pa.

		ρ (g.cm^{-3})		**η (mPa.s)**		**nD**	
Component	**Temperature**	**Exp.**	**Lit.**	**Exp.**	**Lit.**	**Exp.**	**Lit.**
1-BuOH	298.15	0.8066	0.8057 [09] 0.8055 [10] 0.80587[11]	2.5946	2.5416 [12] 2.571 [13] 2.547[11]	1.389	1.3954 [18]
EB	298.15	1.0412	1.04124[14] 1.0413[15] 1.04142[16] 1.0423[17] 1.04163[18]	1.9430	1.9430[14] 1.9543[17]	1.5025	1.5025 [14] 1.5027[19] 1.5034[20] 1.50328 [21]

The binary liquid mixtures were carefully prepared through combining known masses of pure liquids in glass vials (12 mL) in order to decrease evaporation losses. Both pure liquids and mixtures were degassed before readings to avoid difficulties. Fresh solutions for each composition were used same day, to measure each mixture. OHAUS Discovery analytical balance was used to weigh all of the mixes with an accuracy of 0.01 mg. The values were measured as soon as the

samples with various compositions were produced [22].

1.2. Apparatus and procedure

1.2.1. Density and viscosity measurements

The density and dynamic viscosity of both pure liquids and their binary mixture were measured at temperature T=298.15 K and under atmospheric pressure using an SVM 3001 Stabinger viscometer (Anton Paar) with density and viscosity repeatability of 0.00005 g•cm^{-3} and 0.1%, respectively, and temperature stability of 0.005 K. The instrument was calibrated in accordance with the supplier's recommended process. The measuring protocol's inherent uncertainties have been taken into consideration. The nominal precision of density measurements were discovered to be u (ρ) = 0.05 g•cm3. The instrument can measure density in the range of 0 to 3 g/cm3 and viscosity in the range of 0.2 to 30000 mm2/s-1 at a temperature of 298 K. The measured densities and viscosities were compared with values found in the literature.

1.2.2. **Refractive indices measurements**

Using a digital refractometer (RFM T series, Bellingham & Stanley Ltd., UK), the refractive indices of the pure liquids and their binary mixture were determined at the required temperatures and atmospheric pressure. The refractometer was calibrated with distilled deionized water before each measurement session. The temperature and refractive index standard errors are approximately ± 0.1 K and ± 2 × 10^{-4} units, respectively. The prism was cleaned with doubly distilled water and dried with a clean paper towel before a zero sample was measured to calibrate the refractometer.

2. RESULTS AND DISCUSSION

Table 3 lists the directly measured properties of density, viscosity and refractive index, vs. the mole fraction of 1-BuOH, x_1, for the mixture {ethyl benzoate (EB) (1) + 1-Butanol (BuOH) (2)} at T = 298.15 K. As the mole fraction of alcohol x_1 increased from 0 to 1.0, it was observed that the density and refractive index increased, in contrast to the viscosity values.

The experimental data of ρ, η and n_D were used to calculate the excess molar volume (V^E), deviations in viscosity ($\Delta\eta$), excess Gibbs free energy (ΔG^{*E}) of activation of viscous flow, and deviations in refractive index Δn_D.

Table 3: Experimental densities, ρ, dynamic viscosities, η, refractive indices, n_D and calculated excess molar volume, V^E, viscosity deviations, $\Delta\eta$, excess Gibbs energies of activation of viscous flow, ΔG^{*E}, and deviation in refractive indices, Δn_D of the binary mixture {EB (1) + BuOH(2)} at T= 298.15K and under atmospheric pressure.

x_1	ρ (g.cm^{-3})	η (mPa.s)	n_D	V^E ($cm^3.mol^{-1}$)	$\Delta\eta$ (m.Pa.s)	ΔG^{*E} (J.mol^{-1})	Δn_D
T = 298.15 K							
0.0000	0.8066	2.5946	1.3897	0	0	0	0
0.1054	0.8412	2.1740	1.3977	0.231	-0.352	-330.3	-0.003
0.2095	0.8707	1.8690	1.4143	0.575	-0.589	-603.9	0.002
0.3122	0.8991	1.7595	1.4268	0.619	-0.632	-668.0	0.002
0.4139	0.9257	1.7164	1.4397	0.514	-0.609	-654.3	0.004
0.5142	0.9496	1.7089	1.4516	0.440	-0.551	-595.2	0.004
0.6137	0.9706	1.7156	1.4719	0.441	-0.479	-519.5	0.013
0.7120	0.9910	1.7290	1.4808	0.285	-0.402	-442.1	0.011
0.8089	1.0091	1.7790	1.4889	0.193	-0.288	-316.6	0.008
0.9054	1.0260	1.8617	1.4964	0.078	-0.143	-153.3	0.005
1.0000	1.0412	1.9430	1.5025	0.000	0	0	0

The values of V^E were computed from the experimental values of density of the mixture and their

pure components according to the following equation:

$$V^E = \frac{(x_1M_1 + x_2M_2)}{\rho} - \frac{x_1M_1}{\rho_1} - \frac{x_2M_2}{\rho_2} \quad (1)$$

Where x_1, M_1 and ρ_1 are mole fraction, molar masse and density respectively of pure components 1 and 2. ρ is the density of the binary mixture.

The $\Delta\eta$ values of the binary mixtures were determined using the mole fraction and viscosity of the pure components and the mixture, as given in Eq. (2):

$$\Delta\eta \Big/ (mPa.s) = \eta - \sum_{i=1}^{n} x_i\eta_i \quad (2)$$

The ΔG^{*E} values of activation of viscous flow were determined for the binary mixture by the following relationship:

$$\Delta G^{*E} \Big/ (Jmol^{-1} = RT[(\ln(\upsilon M) - \sum x_i \ln(\upsilon_i M_i)] \quad (3)$$

Where ν is the kinematic viscosity of the mixture, ν_i is the kinematic viscosity of pure component i, M is the molar mass of the mixture. The terms T and R denote the total temperature and universal gas constant.

Refractive index deviations Δn_D can be calculated as:

$$\Delta n_D = n_D - (x_1 n_{D1}^* + (1 - x_1) n_{D2}^*) \quad (4)$$

Where n_D the refractive index of the mixture, $n^*_{D,i}$ is that corresponding to the pure component i and x_1 is the mole fraction of component 1 in the mixture.

Furthermore, the composition dependence of the excess property values of {EB (1) + 2-BuOH (2)} binary system at T = 298.15 K were fitted with the Redlich-Kister type equation:

$$Y^E = x_1 (1 - x_1) \sum_{k=0}^{m} A_k (1 - 2x_1)^k \quad (5)$$

Where $Y^E \equiv V^E$ or $\Delta\eta$ or Δn_D or ΔG^{*E} and x_1 is the mole fraction of EB, A_k are adjustable parameters obtained by least-squares method, and k is the degree of the polynomials. In each

case, the optimum number of coefficients was ascertained from an examination of the variation of standard deviation σ with

$$\sigma = \left[\frac{\sum(Y_{exp} - Y_{cal})^2}{(n-p)}\right]^{1/2} \quad (6)$$

Where $Y_{exp.}$ and $Y_{cal.}$ are the experimental and calculated values of the property Y, respectively, and n and p denote the number of experimental points and number of parameters retained in the respective equations. Table 4 presents the values of the parameters A_k together with the standard deviation σ. The number of coefficients reported was chosen to achieve the best correlation obtained.

Table 4: Coefficients of Redlich–Kister equation A_k, and standard deviations σ, for excess molar volumes, V^E, deviation in viscosity, $\Delta\eta$, deviation in refractive indices, Δn_D, and excess Gibb's free energy, ΔG^{*E}, for the liquid mixture {EB (1) + 1-BuOH (2)} at T= 298.15 K and under atmospheric pressure.

Properties	A1	A2	A3	A4	A5	σ
V^E	2.0	-1.9	0.3	1.0	-	0.085
$\Delta\eta$	-3.5	1.8	-1.4	2.6	-1.6	0.010
ΔG^{*E}	-2399.172	1403.771	-2247.649	-452.070	2856.106	10.617
Δn_D	0.024	0.061	0.088	-0.026	-0.180	0.002

The different behavior of V^E, $\Delta\eta$, ΔG^{*E}, and Δn_D, and their magnitude as observed in this work reflect the type of interactions (intermolecular forces) taking place in the mixture as well as the difference in size and shape of the components. The estimated values of V^E, $\Delta\eta$, ΔG^{*E} and Δn_D are included in Table 3 for the entire composition range and depicted in Figs. 1- 4. The curves of V^E were found to be positive over the whole composition range and asymmetrical. The excess molar volume first increases then decreases over the whole composition range and reaches the maximum value at approximately 0.4139 mole fraction of EB. The positive values of V^E suggest the presence of weak interactions i.e. dispersion forces between EB and 1-BuOH, rupture of hydrogen bonded chains, resulting in the dissociation of 1-BuOH with a result of an increasing in the volume of the system.

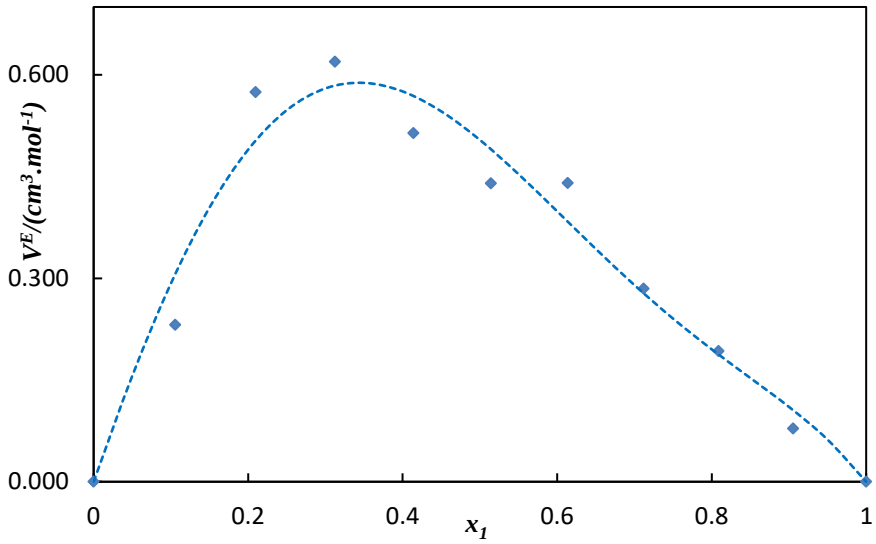

Figure 1: Excess molar volume, V^E, against the mole fraction of EB, x_1, for the binary mixture {EB (1) +1-BuOH (2)} at T = 298.15K and under atmospheric pressure. ◆ 298.15 K. ---, values derived from the Redlich-Kister equation (Eq. 5) using the coefficients listed in Table 4.

Dikio *et al.* [23] rapported that positive V^E and negative Δη causes a disruption in associated molecules while a negative V^E and positive Δη is a result of an association or complex formation between components. According to Vogel and Weiss[24] strong specific interactions between unlike molecules are the result of positive values of $\Delta\eta$ *whereas* the dispersion forces and absence of complex formation causes negative deviation of $\Delta\eta$. In our case, dispersion forces and disruption in hydrogen bonding are dominant. The negative deviation in $\Delta\eta$ implies that the mixture is less viscous than the corresponding ideal mixture.

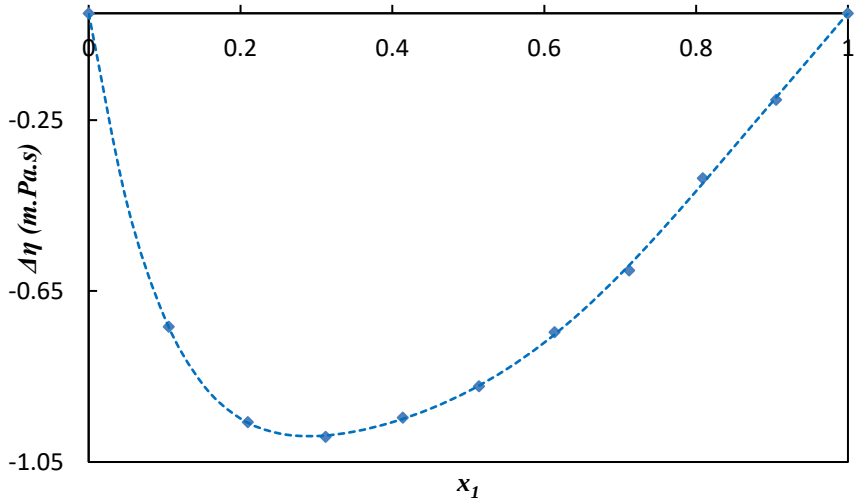

Fig. 5: Deviation in viscosity, $\Delta\eta$, against the mole fraction of EB, x_1, for the binary mixture {EB (1) +1-BuOH (2)} at T = 298.15K and under atmospheric pressure. ◆ 298.15 K. ---, values derived from the Redlich-Kister equation (Eq. 5) using the coefficients listed in Table 4.

The magnitude of ΔG^{*E} represents the strength of interaction between unlike molecules [25]. Deviation in Excess Gibbs free energy of activation of viscous flow are negative, that reflect the dominance of dispersion forces in the mixture under study.

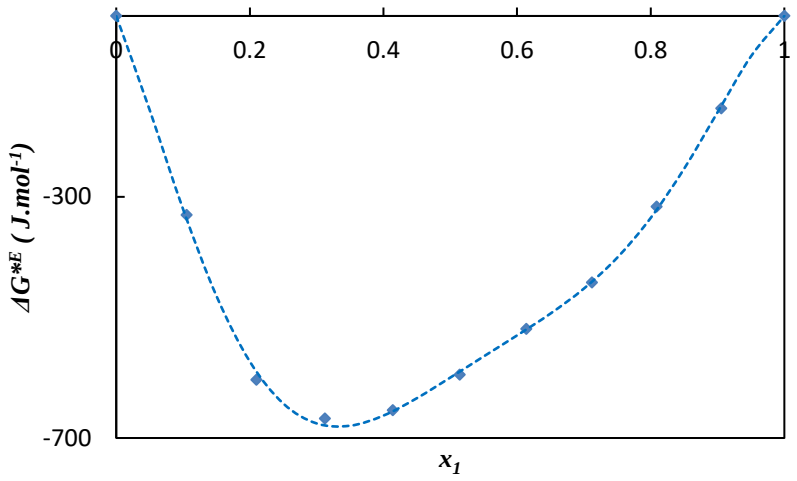

Fig. 6: Excess Gibbs energy of activation of viscous flow, ΔG^{*E}, against the mole fraction of EB, x_1, for the binary mixture {EB (1) +1-BuOH (2)} at T = 298.15K and under atmospheric pressure. ◆ 298.15 K. ---, values derived from the Redlich-Kister equation (Eq. 5) using the coefficients listed in Table 4.

A perusal of Fig.6 shows that Deviation in refractive index, Δn_D, exhibit a S-shape with both negative and positive values. Chaudhary and Kumar [26] state that positive values are sign of formation of significant interactions while negative values are indicative of weak forces or breaking association of the components of the mixture.

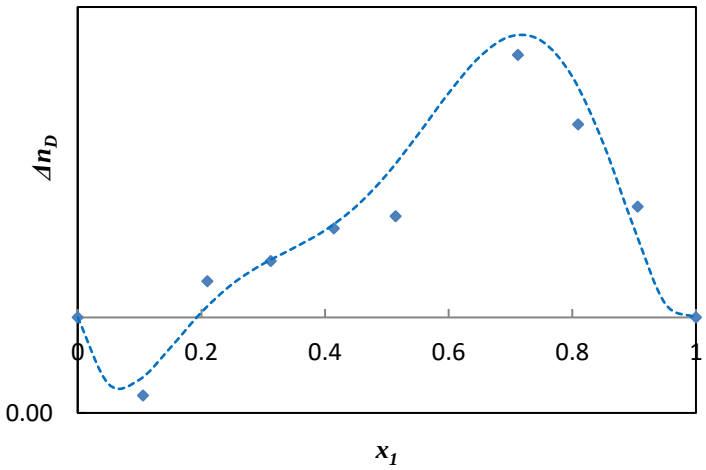

Fig. 8: Deviation in refractive index, Δn_D, against the mole fraction of EB, x_1, for the binary mixture {EB (1) +1-BuOH (2)} at T = 298.15K and under atmospheric pressure. ◆ 298.15 K. ---, values derived from the Redlich-Kister equation (Eq. 5) using the coefficients listed in Table

CONCLUSION

Understanding the thermophysical characteristics of particular binary liquid mixture allowed researchers to explore the molecular interactions especially between associative molecules and non-associative ones. In this work, density (ρ) viscosity (η) and Refractive indices (n_D) for the binary system {ethyl benzoate (EB) (1) + 1-butanol (1-BuOH)} were measured at T = 298.15 K at 0.1 MPa. The excess properties V^E (excess molar volume), $\Delta\eta$ (deviation in viscosity), ΔG^{*E} (excess Gibbs free energy of activation of viscous flow), and Δn_D (Deviation in refractive index) were evaluated using experimental data. The sign and magnitude of these parameters with composition emphasized dominance of dispersion forces and disruption in hydrogen bonding. The Redlich-Kister polynomial equation was used to correlate the excess properties of this mixture.

ACKNOWLEDGMENTS

The authors express their sincere gratitude to the DGRSDT (**D**irection **G**énérale de la **R**echerche **S**cientifique et du **D**éveloppement **T**echnologique), the entire staff at the Scientific and Technical Research Center in Physico-Chemical Analyses (CRAPC) in Ouargla, Algeria.

DECLARATION OF INTEREST

There is no conflict of interest.

NOMENCLATURE

ρ: Density (g/cm^3).

ΔnD: Deviation in refractive index.

η: Dynamic viscosity (mPa. s).

$\Delta\eta$: Dynamic viscosity deviation (mPa. s).

ΔG^{*E}: Excess Gibbs free energy of activation flow (J/mol).

V^E: Excess molar volume (cm^3/mol).

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