# Excess Thermodynamic Studies in Binary Mixture of Ethyl Benzoate with Butan-2-Ol at 293.15 K

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## Abstract

Densities, $\rho$, viscosities, $\eta$, and refractive indices, $n_D$, of the binary mixture of ethyl benzoate (EB) with 2-butanol (2-BuOH), including those of pure liquids were measured over the entire composition range at T= 293.15 K and under atmospheric pressure. From the experimental data, the excess molar volumes, $V^E$ and the other excess thermodynamic properties such as deviations in viscosity, $\Delta\eta$, refractive index, $\Delta n_D$, and excess free energy of activation, $\Delta G^{*E}$, have been evaluated. The computed data $V^E$, $\Delta\eta$, $\Delta G^{*E}$ and $\Delta n_D$ have been correlated with Redlich-Kister polynomial equation to ascertain the fitting parameters and standard deviations. The $V^E$ and $\Delta n_D$ values are positive. The $\Delta\eta$ and $\Delta G^{*E}$ are found to be negative over the entire range of mole fraction. The changes in positive and negative values of excess properties could explain the different kinds of interactions between the component molecules. These results can be used to develop comprehensive theoretical models for designing various technological processes in the chemical and biochemical industries. A good agreement among experimental data and the values estimated by theoretical procedure was obtained. The deviations for excess properties have been explained on the basis of the intermolecular interactions.

## INTRODUCTION

This paper is a continuation of our earlier work concerning the physicochemical studies on binary liquid mixtures [1]. Investigation of binary polar liquid mixtures that contain one associative and a non-associative liquid provides valuable information regarding molecular complex formation in solution. In recent years, measures of the thermodynamic and transport properties have been used adequately to understand the nature and strength of molecular interactions and physicochemical behavior in liquid mixtures. The nonrectilinear behavior of these properties of liquid mixtures with changing mole fractions and temperature are related to the difference in size [2], shape and polarity of the molecules in addition to the strength of the interactions. In the present investigation, alcohol is strongly self-associated through hydrogen bonding and dipole-dipole interaction with degrees of association depending on various variables such as chain length, position of the -OH group, and dilution by other substances. The ethyl benzoate molecules studied here do not exhibit the property of self-associated through hydrogen bonding because they have no groups having that ability.

We report here some new and useful experimental data on density $\rho$, viscosity, $\eta$ and refractive index, $n_D$, for the mixture of ethyl benzoate (EB) with 2-butanol (2-BuOH) at T= 293.15 K for the liquid region and under atmospheric pressure for the whole composition range. From the literature survey, the thermodynamic and transport functions of the binary mixture of EB with 2-BuOH at T= 293.15 K and under atmospheric pressure have not still reported. From the experimental data, the derived properties are determined to explain the strength and the nature of the intermolecular forces existing in the mixing.

Excess and deviation properties have been fitted to the Redlich - Kister equation to determine the correspondent coefficients. The obtained results were discussed in terms of molecular interactions, additionally the investigation of temperature effect on these properties.


## 1. EXPERIMENTAL SECTION

### 1.1. Chemicals

Table 1 regroups the source and purity of ethyl benzoate (EB) and 2-butanol (2-BuOH) used in this work. No further purification of the chemicals was performed. The purity of these samples was checked by comparing the measured densities, viscosities and refractive indices temperature T= 298.15 K with those reported in the literature [3-19]. The obtained values agree well with the literature values as shown in Table 2. This agreement gives a verification of the results obtained by the apparatus.

**Table 1:** Pure component specifications: suppliers, CAS number, specified purity and GC 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% |

**Table 2:** Comparison of experimental density, $\rho$, dynamic viscosities, $\eta$ and refractive indices, $n_D$, of the pure components with the corresponding literature values at T = 298.15 K and $p = 1\times10^5$ Pa.

| Component | T(K) | $\rho$ (g.cm$^{-3}$) | | $\eta$(mPa.s) | | $n_D$ | |
|---|---|---|---|---|---|---|---|
| | | Exp. | Lit. | Exp. | Lit. | Exp. | Lit. |
| Ethyl benzoate | 298.15 | 1.04124 | 1.0413[3] | 1.9430 | 1.945[12] | 1.5025 | 1.5027[15] |
| | | | 1.04142[4] | | 1.936[13] | | 1.5034[16] |
| | | | 1.0423[5] | | 1.9543[5] | | 1.50328[17] |
| | | | 1.04163[6] | | | | 1.5026[6] |
| 2-Butanol | 298.15 | 0.80315 | 0.80253[7] | 3.0972 | 3.0804[14] | 1.3956 | 1.3950[18] |
| | | | 0.8035[8] | | | | 1.3948[19] |
| | | | 0.802528[9] | | | | 1.39544[11] |
| | | | 0.8032[10] | | | | |
| | | | 0.80274[11] | | | | |

The solutions of each composition were prepared fresh and all the properties were measured same day. All mixtures were weighed using an OHAUS Discovery analytical balance with an accuracy of 0.01 mg.

### 1.2. Apparatus and procedure

#### 1.2.1. Density and viscosity measurements

The density and dynamic viscosity of the pure liquids and their binary mixture were measured at temperature T= 293.15 K and under atmospheric pressure by 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 procedure advised by the supplier. Uncertainties arising from the measurement protocol have been taken into account. It was found that the nominal uncertainties in density measurements were u ($\rho$) = 0.05 g·cm$^{-3}$. The instrument can measure simultaneously density in the range of (0 to 3) g·cm$^{-3}$ and viscosity (0.2 to 30000) mm$^2$·s$^{-1}$ in a temperature range of (273.15 to 343.15) K.

#### 1.2.2. Refractive indices measurements

The refractive indices of the pure liquids and their binary mixture were determined at the required temperature and under atmospheric pressure by using a digital refractometer (RFM T

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series, Bellingham & Stanley Ltd, UK).  The calibration of the refractometer was performed by using distilled and deionized water after each measurement. The standard uncertainty of temperature and refractive index are approximated to be ± 0.1 K and ± 2 × 10–4 units, respectively. The refractometer was calibrated by cleaning a prism with doubly distilled water, followed by wiping it with a clean paper towel and measuring a zero sample.

## 2. RESULTS AND DISCUSSION

The experimental densities, viscosities and refractive indices of the binary system {EB (1) + 2-BuOH (2)} at T= 293.15 K and under atmospheric pressure as a function of mole fraction, $x_1$, are recorded in Table 3.

**Table 3:** Experimental densities, $\rho$, dynamic viscosities, $\eta$, refractive indices, $n_D$ and calculated excess molar volume, $V^E$, viscosity deviations, $\Delta\eta$, excess Gibbs energies of activation of viscous flow, $\Delta G^{*E}$, and deviation in refractive indices, $\Delta n_D$ of the binary mixture {EB (1) + 2-BuOH (2)} at T= 293.15 K and under atmospheric pressure.

| $x_1$ | $\rho$ (g.cm$^{-3}$) | $x_1$ | $\eta$ (mPa.s) | $x_1$ | $n_D$ | $V^E$ (cm$^3$.mol$^{-1}$) | $\Delta\eta$ (m.Pa.s) | $\Delta G^{*E}$ ( J.mol$^{-1}$) | $\Delta n_D$ |
|---|---|---|---|---|---|---|---|---|---|
| T= 293.15 K | | | | | | | | | |
| 0.1003 | 0.8423 | 0.1003 | 2.6158 | 0.1001 | 1.4120 | 0.023 | -0.92 | -685.1 | 0.0040 |
| 0.2001 | 0.8735 | 0.2001 | 2.1727 | 0.2000 | 1.4271 | 0.063 | -1.19 | -970.4 | 0.0083 |
| 0.2997 | 0.9017 | 0.2997 | 1.9665 | 0.2999 | 1.4411 | 0.096 | -1.23 | -1049.7 | 0.0115 |
| 0.3989 | 0.9275 | 0.3989 | 1.8643 | 0.3991 | 1.4525 | 0.095 | -1.16 | -1021.4 | 0.0122 |
| 0.4993 | 0.9513 | 0.4993 | 1.8329 | 0.4998 | 1.4628 | 0.095 | -1.01 | -906.2 | 0.0116 |
| 0.5984 | 0.9719 | 0.5984 | 1.8358 | 0.5992 | 1.4731 | 0.212 | -0.84 | -749.4 | 0.0111 |
| 0.7013 | 0.9921 | 0.7013 | 1.8790 | 0.7017 | 1.4819 | 0.253 | -0.62 | -539.3 | 0.0089 |
| 0.8000 | 1.0118 | 0.8000 | 1.9469 | 0.8007 | 1.4909 | 0.044 | -0.38 | -313.9 | 0.0071 |
| 0.9026 | 1.0300 | 0.9026 | 2.0492 | 0.9018 | 1.4983 | -0.064 | -0.10 | -45.6 | 0.0036 |

## 2.1. Volumetric studies

Values of experimental densities (ρ) for {EB (1) + 2-BuOH (2)} binary system at various composition at T = 293.15 K are depicted in Table 3. From Table 3, one can observe that $\rho$ values increase nonlinearly with the composition of the EB. Excess molar volume $V^E$ for various mole fractions of EB at T = 293.15 K are calculated from the experimental data and listed in Table 3.

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

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Where $x_1$, $M_1$, $\rho_1$ and $x_2$, $M_2$, $\rho_2$ are mole fraction, molecular weight and density of the respective pure components 1 and 2, $\rho$ is the mixture density.

Furthermore, the composition dependence of the $V^E$ values of {EB (1) + 2-BuOH (2)} binary system at T = 293.15 K were fitted with the Redlich-Kister type equation:

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

Where $Y^E \equiv V^E$ or $\Delta\eta$ or $\Delta n_D$ or $\Delta G^{*E}$ 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 $\sigma$ with

$$\sigma = \left[ \frac{\sum (Y_{exp} - Y_{cal})^2}{(n - p)} \right]^{1/2} \qquad (3)$$

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 $\sigma$. 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 $\sigma$, for excess molar volumes, $V^E$, deviation in viscosity, $\Delta\eta$, deviation in refractive indices, $\Delta n_D$, and excess Gibb's free energy, $\Delta G^{*E}$, for the liquid mixture {EB (1) + 2-BuOH (2)} at T= 293.15 K and under atmospheric pressure.

| Properties | A1 | A2 | A3 | A4 | A5 | $\sigma$ |
|---|---|---|---|---|---|---|
| T= 293.15 K | | | | | | |
| $V^E$ | 0.532 | 1.293 | 1.828 | -3.203 | -5.043 | 0.043 |
| $\Delta\eta$ | -4.1 | 2.9 | -2.1 | 4.1 | -0.8 | 0.015 |
| $\Delta n_D$ | 0.047 | -0.016 | 0.016 | 0.023 | -0.036 | 0.0003 |
| $\Delta G^{*E}$ | -3619.597 | 2570.207 | -1375.553 | 2687.814 | 985.821 | 12.507 |

$V^E$ values of {EB (1) + 2-BuOH (2)} binary system at T= 293.15 K and under atmospheric pressure are plotted in Fig.1.
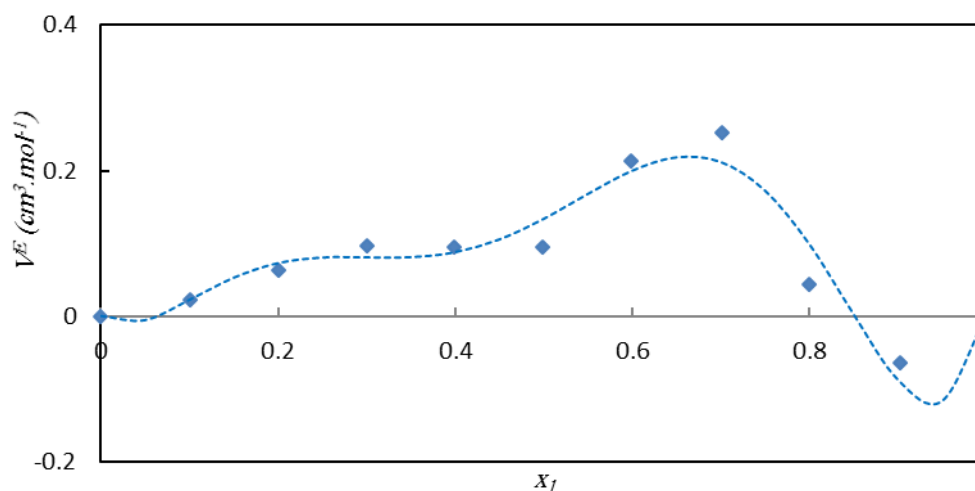
**Figure 1:** Excess molar volume, $V^E$, against the mole fraction of EB, $x_1$, for the binary mixture {EB (1) + 2-BuOH (2)} at T = 293.15 K and under atmospheric pressure. ♦ 293.15K. ---, values derived from the Redlich-Kister equation (Eq. 2) using the coefficients listed in Table 4.

An examination of Fig. 1 illustrates that $V^E$ values have both positive and negative signs. As is widely known, Negative $V^E$ reflects a volume contraction due to disruption of alcohol self-associations while positive contributions to $V^E$ suggest that the hydrogen bond interaction in the mixture is almost destroyed, resulting in the dissociation of 2-BuOH and the increase in the volume of the system. The negative values of $V^E$ beyond $x_1$ = 0.85 with a minimum at $x_1$ = 0.9054, (Fig. 1) may be due to the fitting of smaller molecules in the voids created by the bigger molecules (Less probable due to the steric hindrance in the molecules) in addition to the hydrogen bonding between unlike molecules or Keesom type Van der Waals forces which can happen due to dipole moment of constituents: 2-BuOH is a polar solvent having a dipole moment (μ = 1.66 D) and EB with dipole moment (μ=2.00 D). Up to $x_1$ = 0.85, the added alcohol causes more disruption of the associated structures of the components which seems to dominate over the combined effect of the geometrical fitting of one component into the other and hydrogen bond between unlike molecules or Keesom type interaction, resulting a predominant increase in $V^E$ values. These assumptions were also considered by others to explain the variation of $V^E$ values with the composition of EB with alkanols [17, 20, 21, and 1], toluene, and isomeric chlorotoluenes [22], benzene [23], and hydrocarbons [24].

## 2.2. *Viscometric properties*

Based on the experimental values of dynamic viscosity ($\eta$), the viscosity deviation (⊠$\eta$) can be calculated as:

$$\Delta\eta \Big/ (mPa.s) = \eta - \sum_{i=1}^{n} x_i \eta_i \qquad (4)$$

Where:

$\eta$: Dynamic viscosity of the mixture

$\eta_i$: Dynamic viscosity of pure component $i$

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$x_i$: Mole fraction of the component $i$

$$\Delta G^{*E} \Big/ (Jmol^{-1} = RT[(\ln(vM) - \sum x_i \, \ln(v_i M_i)] \tag{5}$$

$$M = \sum_{i=1}^{n} x_i \, M_i \tag{6}$$

$$\varsigma = v * \rho \tag{7}$$

Where:

$\rho$: Density

$v$: Kinematic viscosity of the mixture.

$v_i$: Kinematic viscosity of pure component $i$.

$\Delta G^{*E}$: Excess Gibbs energy of activation of viscous flow.

$M$ : Molar mass of the mixture.

$R$: Gas constant.

$T$: Absolute temperature.

$x_i$: Mole fraction in component i.

$n$: Number of components in the mixture.

$\Delta \eta$ and $\Delta G^{*E}$ values of the binary system {EB (1) + 2-BuOH (2)} are listed in Table 3 and shown graphically in Fig. 2 and Fig. 3, respectively. Deviation in viscosity and excess Gibbs energy of activation were fitted to Redlich–Kister equation. The adjustable parameters and standard deviations are given in Table 4.
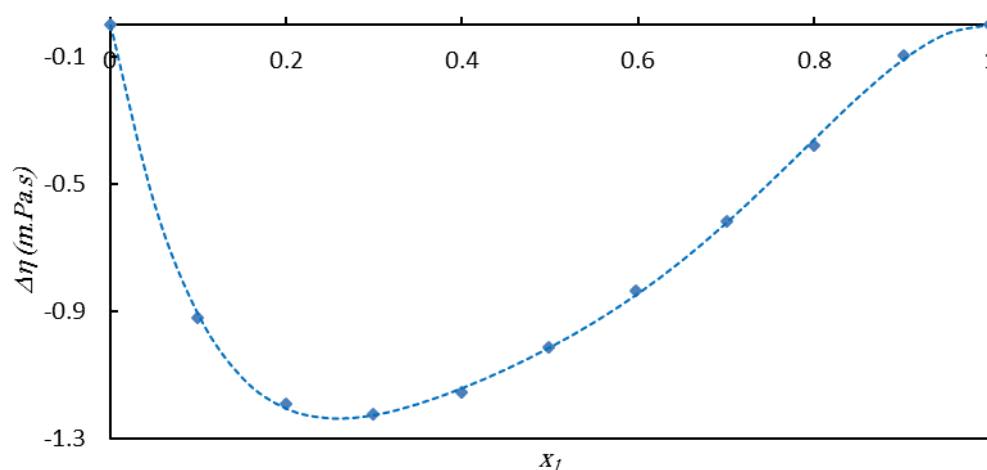


**Figure 2:** Deviation in viscosity, $\Delta \eta$, against the mole fraction of EB, $x_1$, for the binary mixture {EB (1) + 2-BuOH (2)} at T = 293.15 K and under atmospheric pressure. ♦ 293.15K. ---, values derived from the Redlich-Kister equation (Eq. 2) using the coefficients listed in Table 4.

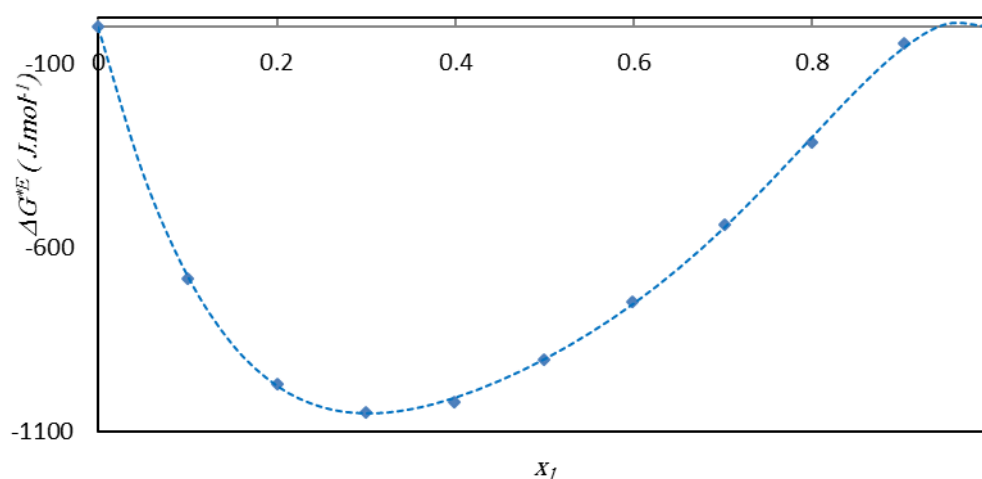**Figure 3:** Excess Gibbs energy of activation of viscous flow, $\Delta G^{*E}$, against the mole fraction of EB, $x_1$, for the binary mixture {EB (1) + 2-BuOH (2)} at T = 293.15 K and under atmospheric pressure. ♦ 293.15K. ---, values derived from the Redlich-Kister equation (Eq. 2) using the coefficients listed in Table 4.

Fort and Moore [25] state that the negative values of $\Delta\eta$ suggest the existence of the dispersion forces and the absence of complex formation. The same conclusions have been attributed by Vogel and Weiss [26]. One can see from Figs. 2 and 3 that $\Delta\eta$ and $\Delta G^{*E}$ show negative deviations from ideality over the entire mole fraction range, and are asymmetrical in shape. It is well-known that 2-BuOH tends to self-association, and the aromatic ester EB is a good hydrogen bonding acceptor. Based on the above-mentioned conclusions, the variation of $\Delta\eta$ values of EB (1) + 2-BuOH (2) binary system with $x_1$ is mostly caused by the dissociation of the associated entities of 2-BuOH. Following Fort and Moore [25] this viscosity behavior corresponds to systems containing an associated component and in which the stability of (solute + solvent) complexes is minimal or nonexistent. Meyer et al. [27] reported that the negative values of $\Delta G^{*E}$ correspond to the existence of (solute + solute) association. Reed and Taylor [28] state that the positive values of $\Delta G^{*E}$ may be attributed to specific interactions between unlike molecules, like hydrogen bonding, dipole-dipole interaction, and charge-transfer complex whereas negative deviations may be ascribed to dispersion forces. The positive values of $\Delta G^{*E}$ at about $x_1 \approx 0.85$ indicate the existence of intermolecular interaction through hydrogen bonding between the component molecules of the liquid mixture under study.

### 2.3. Refractive index

Refractive index deviations $\Delta n_D$, were defined by:

$$\Delta n_D = n_D - (x_1 n_{D1}^* + (1 - x_1)n_{D2}^*) \tag{8}$$

Where $n_D$ denotes the refractive index of the mixture, $n_{D,i}^*$ is that corresponds to the pure

component i, and $x_1$ is the mole fraction of component 1 in the mixture. The experimental refractive indices of the binary system {EB (1) + 2-BuOH (2)} at T = 293.15 K and under atmospheric pressure are listed in Table 3, and the values of $\Delta n_D$ are graphically represented in Fig. 4. The results were fitted by Eq. (2) and the adjustable parameters and standard deviations are gathered in Table 4.
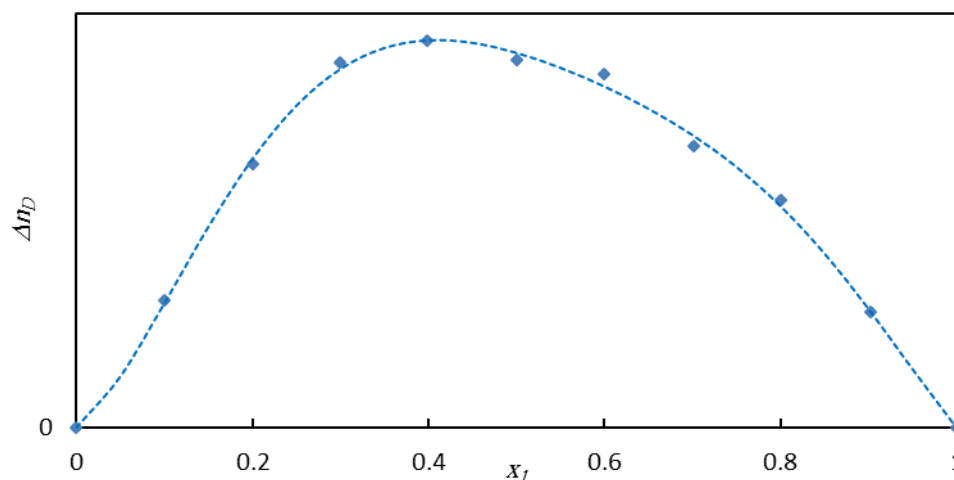


**Figure 4**: Deviation in refractive index, $\Delta n_D$, against the mole fraction of EB, $x_1$, for the binary mixture {EB (1) + 2-BuOH (2)} at T = 293.15 K and under atmospheric pressure. ♦ 293.15K. ---, values derived from the Redlich-Kister equation (Eq. 2) using the coefficients listed in Table 4.

From Fig. 4 it can be noted that the binary system {EB (1) + 2-BuOH (2)} shows the same sign that $V^E$ throughout the entire composition range for each studied temperature. Chaudhary and Kumar [29] have reported that positive values of the change in refractive index are indicative of the formation of significant interactions in the mixtures whereas breaking of associations or weak interactions contributes to negative values. It is obvious that in our case the forces of dispersion are predominant.

## CONCLUSION

The molecular interactions between the components of the binary system {EB (1) + 2-BuOH (2)} are identified through thermodynamic studies. Densities, viscosities, and refractive indices have been measured. The excess properties of this solution were correlated using the Redlich-Kister polynomial equation. The excess molar volume $V^E$ changed from positive to negative. The trend of $V^E$ implies the predominance of the expansion in volume over the contraction. The values excess Gibb's free energy of activation and deviation in viscosity are found to be negative. Deviation in refractive indices is positive over the entire range of composition. This is a clear indication of the presence of special interaction in the mixture. The excess properties were successfully fitted with the Redlich-Kister polynomial.

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## CONFLICTS OF INTEREST STATEMENT

The authors report no conflict of interest.

## NOMENCLATURE

$\rho$: Density (g/cm$^3$).

$\boxtimes n_D$: Deviation in refractive index.

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

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

$\boxtimes G^{*E}$: Excess Gibbs free energy of activation flow (J/mol).

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

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