

## NOVEL THERMODYNAMIC PROPERTIES OF BINARY MIXTURES OF TETRAHYDROFURFURYL ALCOHOL WITH SOME ALCOHOLS AT 298.15 K

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### ABSTRACT

To understand the nature of molecular aggregation between two solvents, excess properties (e.g. excess molar volume ( $V_m^E$ ), excess Gibb's free energy of activation viscous flow ( $\Delta G^{*E}$ ) and viscosity deviation of binary mixtures) are therefore needed. Thus, in the current study, the refractive index ( $n_D$ ), viscosity ( $\eta$ ), and density ( $\rho$ ) of binary mixtures were measured for tetrahydrofurfuryl alcohol (THFA) with ethanol, 2-propanol and 2-butanol at 298.15K. Moreover and based on experimentally obtained results, the  $V_m^E$ , the refractive index deviation ( $\Delta n_D$ ), excess molar viscosity ( $\eta^E$ ) and  $\Delta G^{*E}$  were calculated. To correct and fit the experimental  $V_m^E$ ,  $\eta^E$  and  $\Delta n_D$  deviations, Redlich-Kister equation was used. The molecular interaction in binary mixtures was found to explain the variability of physical properties with the composition of the mixtures. In light of obtained results, factors influencing the molecular interactions (e.g. the length of the carbon chain and the position of hydroxyl groups in targeted alcohol) have been also discussed. In all cases the obtained  $V_m^E$ ,  $\eta^E$ ,  $\Delta G^{*E}$  and  $\Delta n_D$  values were negative at 298.15K except for the binary system (THFA + 2-butanol) where the results indicate the positive deviation in refractive indices.

**Keywords:** Binary system, tetrahydrofurfuryl alcohol, refractive index, density, viscosity, thermodynamic properties.

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### INTRODUCTION

In binary liquid solvents mixtures, the excess thermodynamic functions are the key tool for understanding the molecular interactions behavior, they were highly used in the chemical engineering industry for chemical reaction cleaning, extraction and purification<sup>1</sup>. Additionally, physical properties of binary mixtures are essential when heat transfer and mass transfer phenomena involved in industrial processes<sup>2</sup>. A considerable amount of research studying the properties of binary solvent mixtures has been published in the recent decade<sup>3-8</sup>. Hydrogen bond formation, dipole-dipole, and charge transfer interactions have been suggested as the mechanisms of interactions between of solvents<sup>9-11</sup>. For example, Nain<sup>3</sup> studied the densities and volumetric properties of binary mixtures of formamide with alcohols at two temperatures and found that negative  $V_m^E$  is attributed to the formation of hydrogen bond interaction. This was confirmed by Kemeakegha<sup>12</sup> when studied the behavior of binary mixtures of organic solvents and amines. Moreover,  $\rho$ ,  $\eta$  and  $n_D$  measurement is expected to shed some light on both solvent-solute interactions<sup>13</sup>. On the other hand, and up to our knowledge, no study was done to investigate the  $V^E$ ,  $\eta^E$ ,  $\Delta n_D$  and  $\Delta G^{*E}$  at any temperature. Therefore, the objective of this research is to study excess properties including  $\rho$ ,  $\eta$  and  $n_D$  for a range of binary mixtures including (THFA+ Ethanol), (THFA+ 2-propanol) and (THFA + 2-butanol) at 298.15K. From obtained results, another parameter such as  $V_m^E$ , viscosity deviation, refractive index deviation and Gibbs free energy (excess molar) were also determined.

### EXPERIMENTAL

#### Chemicals

All solvents used in the study are of high purity (>99%) Tetrahydrofurfuryl alcohol (THFA), 2-propanol, 2-butanol, and ethanol were purchased from Sigma-Aldrich (UK). The purities of this experiment used

solvents were checked and results showed high compatibility to those in previous studies (Table-1). Densities, viscosities, and refractive indices were listed for all solvents except THFA where no data available in the literature.

### Density and Viscosity Measurements

The average  $\rho$  value of three replicates was determined with a precision of  $\pm 10^{-5}$  g.cm<sup>-3</sup> using a digital density meter (Anton paar, Model DMA 60/602). For viscosity ( $\eta$ ) measurements, a controlled temperature suspended-level ubbelohde viscometer was used under atmospheric pressure at  $298.15 \pm 0.01$  K. The final values represent the average of three measurements.

### Refractive Index Measurements

A digital Abbe refractometer (Model:BOE 32400) was used to measure the  $n_D$  values for both pure and their binary mixtures solvents. Pure toluene and double distilled water were used for refractometer calibration, at a temperature of 298.15 K.

Table-1: This study and literature obtained density ( $\rho$ ), viscosity ( $\eta$ ) and refractive index ( $n_D$ ) values for solvents at 298.15 K.

Compound	$\rho$ (g.cm <sup>-1</sup> )		$\eta$ (mpa.s)		$n_D$	
	Measured	Literature	Measured	Literature	Measured	Literature
THFA	1.04761		4.89883		1.45322	
Ethanol	0.78522	0.78730 <sup>14</sup> 0.78493 <sup>15</sup> 0.78510 <sup>16</sup>	1.10221	1.0826 <sup>17</sup>	1.36003	1.3593 <sup>14</sup> 1.35941 <sup>15</sup> 1.3592 <sup>16</sup>
2-Propanol	0.78084	0.78088 <sup>18</sup> 0.78089 <sup>19</sup> 0.78126 <sup>20</sup>	2.12112	2.0436 <sup>22</sup>	1.37521	1.37444 <sup>19</sup> 1.3752 <sup>[20,21]</sup>
2-Butanol	0.80252	0.80248 <sup>23</sup> 0.80237 <sup>24</sup> 0.80241 <sup>20</sup>	3.51542	3.1150 <sup>25</sup>	1.39523	1.39488 <sup>24</sup> 1.39530 <sup>20</sup>

## RESULTS AND DISCUSSION

In the current study, measured density ( $\rho$ ) values were used to estimate the  $V_m^E$  of the binary mixtures following the equation (1)<sup>26</sup>:

$$V_m^E = \frac{X_1 M_1 + X_2 M_2}{\rho_m} - \frac{X_1 M_1}{\rho_1} - \frac{X_2 M_2}{\rho_2} \quad (1)$$

Where  $M_1$ ,  $X_1$ , and  $\rho_1$  are the molecular weight ( $M_{wt}$ ), mole fraction, and density of the pure THFA, respectively; while  $M_2$ ,  $X_2$  and  $\rho_2$  are the  $M_{wt}$ , mole fraction and density of alcohols,  $\rho_m$  is the density of the binary mixtures, respectively. Table -2 lists the density and  $V_m^E$  of the binary mixtures at the various mole fractions of THFA and (ethanol, 2-propanol and 2-butanol). The  $V_m^E$  plotted against ( $X_1$ ) mole fraction for THFA is shown in Fig. -1.

Table-2: Experimentally obtained density, viscosity and refractive index values and their corresponding parameters values in binary solvents Mixtures.

X <sub>1</sub> THFA + X <sub>2</sub> Ethanol at 298.15 K							
X <sub>1</sub>	$\rho$ (g cm <sup>3</sup> )	$V^E$ cm <sup>3</sup> mol <sup>-1</sup>	$n_D$	$\Delta n_D$	$\eta$	$\eta^E$ mpa.s	$\Delta G^{*E}$ J.mol <sup>-1</sup>
0.0000	0.78522	0.0000	1.36003	0.0000	1.10221	0.0000	0.0000
0.0556	0.81012	-0.11407	1.36505	-0.55088	1.10534	-0.08011	-178.99553
0.2003	0.86480	-0.18979	1.37553	-1.66105	1.31196	-0.12458	-252.46773

0.2717	0.88824	-0.20756	1.38417	-1.89185	1.43335	-0.14260	-285.66860
0.4115	0.92906	-0.22680	1.39508	-2.25869	1.72669	-0.16493	-329.93131
0.5054	0.95301	-0.21833	1.40398	-2.24400	1.99616	-0.16551	-351.24693
0.6219	0.97940	-0.17580	1.41302	-2.11791	2.41119	-0.14481	-287.69246
0.7334	1.00195	-0.12520	1.42197	-1.81120	2.93841	-0.11345	-223.83736
0.8055	1.01378	-0.09323	1.42880	-1.48260	3.33886	-0.09324	-186.08495
0.9278	1.036461	-0.05379	1.43914	-0.84900	4.16206	-0.05529	-118.46276
1.0000	1.04761	0.0000	1.45322	0.0000	4.89883	0.0000	0.0000

X<sub>1</sub> THFA + X<sub>2</sub> 2- propanol at 298.15

X <sub>1</sub>	$\rho(\text{g. cm}^{-3})$	$V^E \text{cm}^3 \text{mol}^{-1}$	$n_D$	$\Delta n_D$	$\eta$	$\eta^E \text{ mpa.s}$	$\Delta G^{*E} \text{ J.mol}^{-1}$
0.0000	0.78084	0.0000	1.37521	0.0000	2.12112	0.0000	0.0000
0.1101	0.82262	-0.54309	1.38772	-0.12789	2.18949	-0.06044	-142.59655
0.2240	0.86442	-1.14715	1.40052	-0.17011	2.35137	-0.08445	-196.76866
0.3408	0.90885	-2.07105	1.41189	-0.19689	2.54123	-0.10456	-243.27626
0.4539	0.94311	-2.32896	1.42105	-0.22405	2.74769	-0.12113	-282.99355
0.5534	0.96779	-2.19084	1.42811	-0.23356	2.96293	-0.12899	-302.81608
0.6497	0.98765	-1.80049	1.43439	-0.21909	3.24407	-0.11895	-279.48010
0.7472	1.00770	-1.49545	1.43999	-0.19451	3.59524	-0.09778	-229.82004
0.8353	1.02229	-0.97714	1.44435	-0.16946	3.96494	-0.07448	-173.51734
0.9249	1.03612	-0.42426	1.44555	-0.11083	4.36632	-0.05221	-124.93248
1.0000	1.04761	0.0000	1.45322	0.0000	4.89883	0.0000	0.0000

X<sub>1</sub> THFA + X<sub>2</sub> 2- butanol at 298.15 K

X <sub>1</sub>	$\rho(\text{g. cm}^{-3})$	$V^E \text{cm}^3 \text{mol}^{-1}$	$n_D$	$\Delta n_D$	$\eta$	$\eta^E \text{ mpa.s}$	$\Delta G^{*E} \text{ J.mol}^{-1}$
0.0000	0.80252	0.0000	1.39523	0.0000	3.51542	0.0000	0.0000
0.0940	0.83401	-0.81245	1.40329	0.10142	3.52793	-0.02765	-68.213775
0.2027	0.86405	-1.04580	1.41112	0.21055	3.57309	-0.05099	-115.61921
0.2978	0.88871	-1.09891	1.42019	0.31392	3.60972	-0.07235	-178.57860
0.4044	0.91574	-1.12394	1.42833	0.40165	0.68238	-0.08779	-216.76150
0.4993	0.93852	-1.04328	1.43429	0.42228	3.79225	-0.0889	-221.91290
0.5946	0.96038	-0.88946	1.43745	0.36444	3.94820	-0.08121	-200.4288
0.6958	0.98330	-0.72903	1.44266	0.28581	4.15021	-0.06489	-160.08807
0.8040	1.00733	-0.54637	1.44738	0.22631	4.39545	-0.04338	-101.70914
0.9059	1.02899	-0.31478	1.45081	0.12573	4.64281	-0.02245	-55.34835
1.0000	1.04761	0.0000	1.45322	0.0000	4.89883	0.0000	0.0000

Excess molar volume ( $V^E$ ), refractive index deviation ( $\Delta n_D$ ), excess molar viscosities ( $\eta^E$ ) and excess molar Gibbs free energy ( $\Delta G^{*E}$ ).

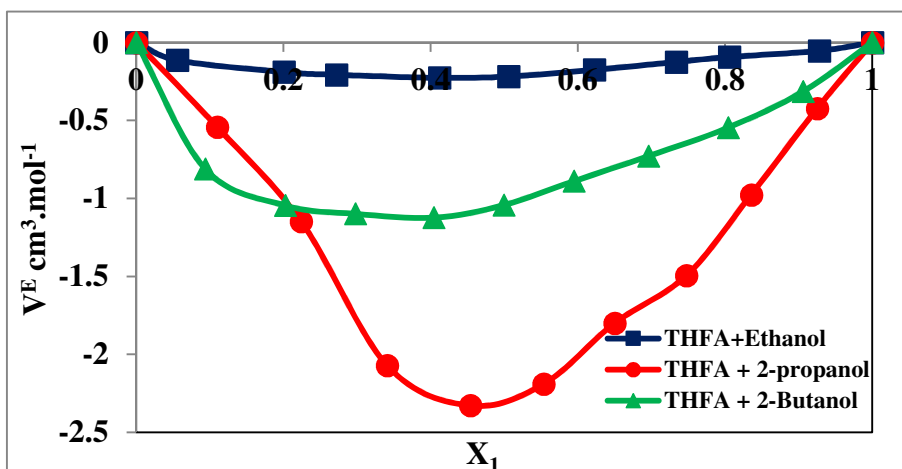


Fig.-1: Plots ( $V^E$ ) excess molar volume against  $X_1$  for ( $X_1$ THFA+  $X_2$ -Mono alcohols) at 298.15 K.

### Refractive index deviation

Refractive index deviation ( $\Delta n_D$ ) was calculated using equation (2)<sup>27</sup>:

$$\Delta n_D = n_D - X_1 n_{D1} - X_2 n_{D2} \quad (2)$$

Where  $n_D$  is the refractive index of the mixture and ( $n_{D1}$ ,  $n_{D2}$ ) are pure component refractive index.

Lorentz-Lorenz equation (3,4, and 5)<sup>28</sup> were used to calculate ( $n_{D1}$ ,  $n_{D2}$  and  $n_D$ ) as follows:

$$n_D = \left( \frac{n_m^2 - 1}{n_m^2 + 2} \right) \frac{X_1 M_1 + X_2 M_2}{\rho_m} \quad (3)$$

$$n_{D1} = \left( \frac{n_1^2 - 1}{n_1^2 + 2} \right) \frac{X_1 M_1}{\rho_1} \quad (4)$$

$$n_{D2} = \left( \frac{n_2^2 - 1}{n_2^2 + 2} \right) \frac{X_2 M_2}{\rho_2} \quad (5)$$

Table -2 lists the experimental ( $n_D$ , and  $\Delta n_D$ ) values of the binary mixtures, while Fig-2 illustrates the relation between  $\Delta n_D$  against mole fraction ( $X_1$ ) for the THFA .

### Excess molar viscosities ( $\eta^E$ )

Excess molar viscosities ( $\eta^E$ ) were obtained by using equation (6)<sup>29</sup>:

$$(\eta^E) = (\eta_m) - (X_1 \eta_1 + X_2 \eta_2) \quad (6)$$

Where  $X_1$ ,  $X_2$ ,  $\eta_1$ ,  $\eta_2$  are the mole fraction and the viscosity of the components 1 and 2; while  $\eta_m$  is the viscosity of the mixture, as listed in Table -2 . The relation between the  $\eta^E$  against mole fraction ( $X_1$ ) for THFA is shown in Fig-3.

### Excess molar Gibbs free energy

Excess molar Gibbs free energy ( $\Delta G^{*E}$ ) of activation of viscous flow was estimated by using equation (7)<sup>30</sup>:

$$\Delta G^{*E} = RT [\ln(\eta_m V_m) - (X_1 \ln \eta_1 V_1) - (X_2 \ln \eta_2 V_2)] \quad (7)$$

Where R is the universal constant for gases and T is the absolute temperature (K). The  $V_1$ ,  $V_2$  and  $V_m$  represent the molar volumes of solvent 1, solvent 2, and binary mixture solvents, respectively.  $V_m$  was obtained from equation (8)<sup>31</sup>:

$$V_m = (X_1 V_1 + X_2 V_2) / \rho_m \quad (8)$$

Table-2 lists the  $\Delta G^{*E}$  while Fig- 4 shows the relation between  $\Delta G^{*E}$  and the mole fraction ( $X_1$ ) for THFA. The Redlich-Kister<sup>32</sup> (Eq. 9) was used to calculate the excess functions of the binary systems:

$$Y = X_1 X_2 \sum_{k=0}^p A_k (X_1 - X_2)^k \quad (9)$$

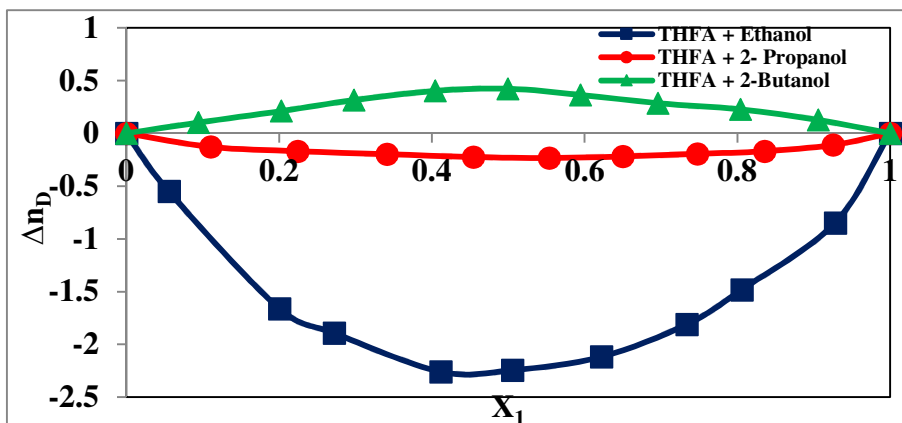


Fig.-2: Plots ( $\Delta n_D$ ) refractive index deviation against  $X_1$  For ( $X_1$ THFA +  $X_2$ - Mono alcohols) 298.15K.

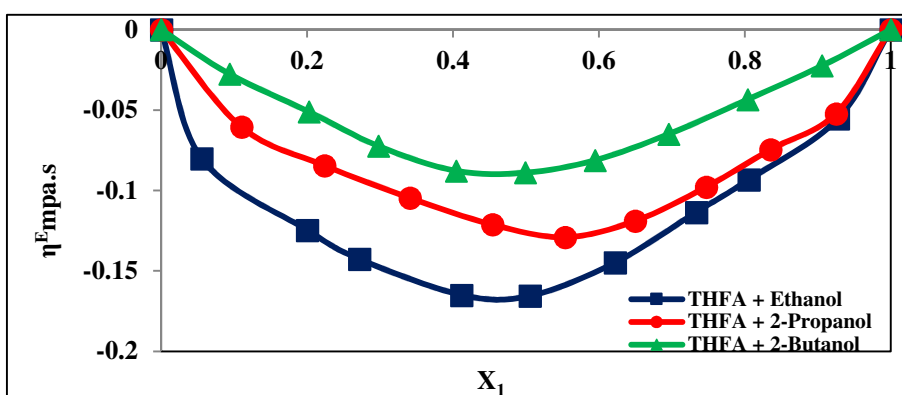


Fig.-3: Plots ( $\eta^E$ ) excess Molar viscosities against  $X_1$  for ( $X_1$ -THFA+  $X_2$ -Mono alcohols) at 298.15K.

Where  $Y$  is  $V^E$ ,  $\Delta n_D$ , and  $\eta^E$ ,  $p$  is the degree of polynomial expansion,  $X_1$  and  $X_2$  are the mole fractions of the components 1 and 2, respectively, and  $A_k$  denotes the polynomial coefficients. The standard deviation was calculated by the following equation<sup>26</sup>:

$$\sigma = \left[ \sum \frac{(X_{\text{Cal}}^E - X_{\text{exp}}^E)^2}{m-n} \right]^{0.5} \quad (10)$$

Where,  $m$  and  $n$  are the number of data points and the estimated parameters, respectively. The values of these coefficients and the standard deviation are given in Table-3. The standard deviation values agreed satisfactorily with the experimental values.

The obtained negative values of  $V_m^E$ , are believed to be attributable mainly to the association between THFA and alcohols compound through between the  $-OH$  groups in mono-alcohols and the  $-OH$  group in the THFA via the formation of hydrogen bonds. The strength of the associations arising from the interactions between the unlike molecules was stronger than the strength of the associations between like molecules<sup>33</sup>. THFA as a highly polar, an aprotic solvent has the capability of destroying the self-associated molecules of alcohols compound and orienting its molecules to interact strongly with it and forming new complexes have smaller volume. Fig.- 2 is shown refractive index deviation ( $\Delta n_D$ ) of mixtures (THFA + mono-alcohols) against mole fraction ( $X_1$ ) for THFA.

For ethanol-THFA and 2-propanol-THFA mixtures, over the range of mole fractions,  $\Delta n$  was found to have negative values. This behavior suggests that the molecular interaction between the  $-OH$  of alcohols compound and the oxygen atom of (THFA) (non-associated molecule) is greater than the molecule interaction between ethanol and 2-propanol molecules (self-associated). This leads to destroying the self-associated structure of alcohols compound and giving the highest negative values of  $\Delta n_D$ .

As the count of carbon atoms increases in the alcohol chain,  $\Delta n_D$  becomes positive for mixture of (2-butanol-THFA), such behavior is a result of lower electronic perturbation of the molecular orbital of the mixing liquids and the enhanced structure of THFA as dipolar and aprotic solvent<sup>34-35</sup>. The  $\eta^E$ , Fig.-3 and  $\Delta G^{*E}$ , Fig. -4 found to be negative along the range of mole fraction, at 298.15 K.

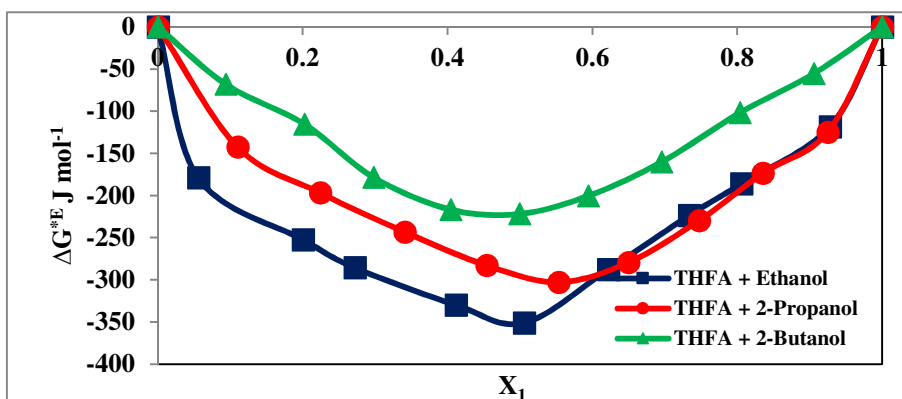


Fig.-4: Plots of ( $\Delta G^{*E}$ ) excess molar Gibbs free energy of activation of viscous flow against ( $X_1$ ) for ( $X_1$ -THFA+  $X_2$ -Mono alcohols) at 298.15.

These results may be related to the formation of hydrogen bonding between unlike molecules through. This study results suggest that the formation of the mixture leads to the structure of the alcohol to be broken since the (-OH) group is located at the end of the molecular chain which means the formation of hydrogen bonds with THFA is highly expected<sup>36</sup>.

Table- 3: Fitting Coefficient ( $A_k$ ) and Standard Deviations ( $\sigma$ ) for All Binary Mixtures at 298.15K.

Function	System	$A_0$	$A_1$	$A_2$	$\sigma$
$V^E$ ( $\text{cm}^3 \cdot \text{mol}^{-1}$ )	THFA + Ethanol	-0.82916	0.49235	-0.33529	0.01894
$V^E$ ( $\text{cm}^3 \cdot \text{mol}^{-1}$ )	THFA + 2- propanol	-9.10026	-0.07495	5.49486	0.02098
$V^E$ ( $\text{cm}^3 \cdot \text{mol}^{-1}$ )	THFA + 2- butanol	-1.52662	0.451045	-9.34118	0.09010
Function	System	$A_0$	$A_1$	$A_2$	$\sigma$
$\Delta n_D$ ( $\text{cm}^3 \cdot \text{mol}^{-1}$ )	THFA + Ethanol	-0.62798	0.17401	-0.29913	0.01692
$\Delta n_D$ ( $\text{cm}^3 \cdot \text{mol}^{-1}$ )	THFA + 2- propanol	-0.88270	-0.10255	-0.64391	0.03180
$\Delta n_D$ ( $\text{cm}^3 \cdot \text{mol}^{-1}$ )	THFA + 2- butanol	1.60546	-0.01332	-0.62052	0.03339
Function	System	$A_0$	$A_1$	$A_2$	$\sigma$
$\eta^E$ (mPa.s)	THFA + Ethanol	-8.96561	0.36658	-2.85256	0.02086
$\eta^E$ ( mPa.s)	THFA + 2- propanol	-0.48809	-0.04308	-0.14539	0.02559
$\eta^E$ ( mPa.s)	THFA + 2- butanol	-0.35173	0.04250	0.12705	0.01351
Function	System	$A_0$	$A_1$	$A_2$	$\sigma$
$\Delta G^{*E}$ J.mol <sup>-1</sup>	THFA + Ethanol	-1269.934	376.455	-639.646	0.86779
$\Delta G^{*E}$ J.mol <sup>-1</sup>	THFA + 2-propanol	-1141.320	-108.683	-350.962	1.28422
$\Delta G^{*E}$ J.mol <sup>-1</sup>	THFA + 2- butanol	-872.512	95.157	385.575	0.84470

## CONCLUSION

New values of  $\rho$ ,  $\eta$  and  $n_D$  of binary mixtures of THFA with ethanol, 2-propanol and 2-butanol at 298.15K were experimentally obtained. The  $V_m^E$ ,  $\Delta n_D$ ,  $\eta^E$  and  $\Delta G^{*E}$  were correlated using the Redlich-Kister polynomial equation. The deviations in refractive indices were negative for the systems THFA + ethanol

and THFA + 2-propanol due to the abolishing the self-associated structure of alcohols compound. On the other hand, the results indicate the positive deviation for THFA + 2-butanol mixture which may relate to the increase number of carbon atoms in alcohol chain. The values of  $V_m^E$ ,  $\eta^E$  and  $\Delta G^{*E}$  resulted in from this study, showed to be negative over the range of mole fractions at 298.15K.

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