



Estimation of Excess Properties of Binary Liquids Systems of Tetrahydrofurfuryl Alcohol with Short-Chain Carboxylic Acids at 298.15 K

HUSAM S. KHALAF^{*a}, ZAINAB A.H. AL-DULAIMY^b, AHMED M. ABBAS^b and MAIDA H. SALEEM^b

Department of Chemistry, College of Education for Pure Science/Ibn Al-Haitham, University of Baghdad, Baghdad, Iraq

*Corresponding author: E-mail: husamkhalaf250@gmail.com

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At the temperature 298.15 K, some physical properties such as: refractive indices (n_D), viscosities (η) and densities (ρ) were studied in four liquid-liquid mixtures: carboxylic acids (HCOOH , CH_3COOH , $\text{CH}_3\text{CH}_2\text{COOH}$ and $\text{CH}_3\text{CH}_2\text{CH}_2\text{COOH}$) with tetrahydrofurfuryl alcohol (THFA) with the identified configuration set. These empirical data were utilized to estimate the excess molar volumes (V_m^E), refractive index perversions (ΔR), viscosity deviations (η^E) and excess molar Gibbs free energy (ΔG^{*E}). Values of V_m^E , η^E , ΔG^{*E} and ΔR were plotted *versus* mole fraction of tetrahydrofurfuryl alcohol. In all cases, the values of V_m^E , η^E , ΔG^{*E} and ΔR that obtained in this study were found to be negative at 298.15 K. The excess parameters were applied in the Redlich-Kister equation by utilizing multi-parameter coefficients that concluded binary coefficients and in respect to the standard deviation. The difference of these characteristics with the formation of binary liquid systems indicates the absence of bipolar bond, variation in the shape and size of component molecules, hydrogen bonding and dipolar interaction among unlike molecules.

Keywords: Binary liquid systems, Refractive index, Excess properties, Carboxylic acids, Tetrahydrofurfuryl alcohol.

INTRODUCTION

The changes in the thermodynamic properties to the mixtures and the degree of it aberration from idealization were studied to be an excellent quantitative and qualitative method for obtaining knowledge in terms of molecular forces and structure between molecules into binary mixtures. This led to the theory and experiential understanding of extra thermodynamic characteristics of binary liquid systems [1,2]. Thus, the data of some physical characteristic related to liquids and binary mixtures such as refractive index, viscosity and application of density in the theory of solutions and molecular dynamics. This conclusion is necessary for the data version obtained from electrochemical and kinetic studies [3]. The current conclusion is the continuation of previous research on the properties of thermodynamics of liquid-liquid mixtures [4-8]. Fluids were selected in the present study on the basis of their importance in the industry.

Tetrahydrofurfuryl alcohol is a biodegradable solvent having utilities in many agricultural and industrial applications. Carboxylic acids are significant chemicals utilized in a diversity of

industrial implementation like the process of separation, cleaning agents, food, beverage, polyester resins and in different chemical industries [9,10]. The objective of the current investigation is to resolve the perturbation of self-association disorder in formic acid, acetic acid, propanoic acid, butyric acid and the breaking of dipole-dipole interaction of tetrahydrofurfuryl alcohol with the reaction among the hydroxyl group of tetrahydrofurfuryl alcohol and the carboxyl group in the formic acid, acetic acid, propanoic acid and butanoic acid *via* formation of hydrogen bonding.

In this paper, we have calculated V_m^E , η^E , ΔG^{*E} and ΔR values for liquid-liquid blends of tetrahydrofurfuryl alcohol (THFA) with carboxylic acid at 298.15 K. Changes in these parameters with a composition of mixtures are beneficial in conception the nature and scope of molecular interactions between the unlike molecules.

EXPERIMENTAL

The source and purity of chemical compounds used in this study are shown in Table-1 along with experimental measurements of viscosity, density and refractive index, as well as values

TABLE-1
AT 298.15 K, SHOWS THE SOURCE, PURITY, ρ , η AND n_D IN THE PURE LIQUIDS

Liquid	Source	Purity (%)	ρ (g cm ⁻³)		η (mpa.s)		n_D	
			Measured	Literature	Measured	Literature	Measured	Literature
THFA	Aldrich	> 99	1.04761	1.055100 [11]	4.89883		1.45322	1.45170 [11]
Formic acid	Fluka	≥ 98	1.21405	1.214050 [12]	1.52212	1.607 [17] 1.510 [14]	1.36974	1.36932 [12]
				1.213400 [13]				1.36943 [14]
				1.213800 [14]				1.36970 [14]
Acetic acid	Fluka	> 99.8	1.04391	1.043920 [12]	1.05752	1.056 [17]	1.37081	1.36980 [12]
				1.043220 [13]				1.37046 [14]
				1.044000 [14]				1.37070 [15]
Propanoic acid	Fluka	≥ 99.5	0.98809	0.988080 [12]	1.03043	1.030 [17]	1.38291	1.38430 [12]
				0.987930 [13]				1.38479 [14]
				0.990930 [14]				1.38500 [15]
Butanoic acid	Fluka	≥ 99.5	0.95277	0.987847 [16]	1.39589	1.396 [12]	1.39635	1.39580 [12]
				0.953200 [12]				1.39630 [15]
				0.952800 [18]				1.39615 [19]

obtained from other researchers [11-19]. All the chemicals and reagents were used without further purification.

Density: By using anton paar digital densimeter (model DMA 60/602) with an accuracy of $\pm 10^{-5}$ g/cm³, density measurements were made for pure components and dual mixtures. Air and double distilled water used for the calibration of the densimeter. At least three times for each composition in experiments were generally repeated and the results were treated.

Refractive index: A digital Abbe refractometer (model: BOE 32400) were specified the refractive indices of pure components and their binary liquid system. The measuring refractive indices of double distilled water and toluene were used to calibrate the refractometer at 298.15 K.

Viscosities: The viscosity was determined by using a suspended-level Ubbelohde viscometer controlled to adjust the temperature of bath with an accuracy of ± 0.01 K at 298.15 K. To give the final values, the experiments were conducted thrice.

RESULTS AND DISCUSSION

Depending on the density data, the V_m^E were measured according to the following equation:

$$V_m^E = \sum_{i=1}^N X_i M_i (\rho^{-1} - \rho_i^{-1}) \quad (1)$$

where ρ_i is the density, X_i is the mole fraction and M_i is the molar mass of the component i , ρ is the density of mixtures and N is the number of components. Fig. 1 shows the values of V_m^E in all liquid-liquid mixtures as they were negative to the full composition at $T = 298.15$ K. The negative values of V_m^E indicate a certain interaction between mixture components [20]. The observed trend of excess volume change, can be explained by two conditions with opposite effects [21,22] (i) firmness of big molecules leading to cellular gap that can be filled by smaller particles and bipolar interaction additional to hydrogen bonds among molecules, (ii) dipole-dipole and donor-acceptor interaction among molecules. However, the effective alteration in size relies on the relative strength of these two influences. The blending of tetrahydrofurfuryl alcohol with short chain carboxylic acids might encourage to the reciprocal dissociation of hydrogen-bonded structures in pure components with a next formation of new hydrogen bonds (O-H...H-O) among the

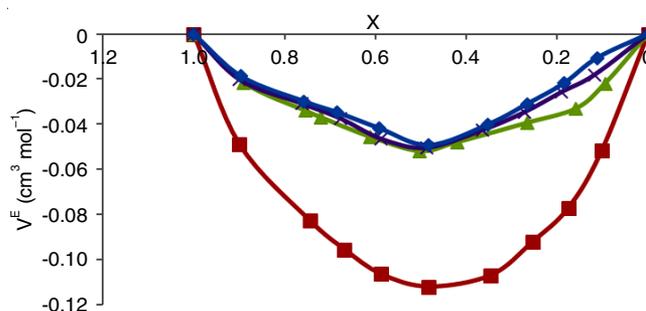


Fig. 1. Curves of V_m^E against X_1 for the binary mixtures of THFA + methanoic acid (■), ethanoic acid (▲), propanoic acid (×) and butyric acid (◆) at 298.15 K

hydroxyl group belonging to tetrahydrofurfuryl alcohol and the hydroxyl group of carboxylic acids in liquid-liquid mixtures. An increase in the negative values of V_m^E for tetrahydrofurfuryl alcohol + formic acid indicated the strong fixed interaction in the hydrogen bonding between tetrahydrofurfuryl alcohol and formic acid (Table-2). However, V_m^E values of tetrahydrofurfuryl alcohol + butyric acid mixtures decreases in the negative values indicate that the molecular bond through hydrogen bonding among the various molecules becomes weak [23] (Table-2). This probably because of an increase in the size of alkyl group in butyric acid and also influenced by the steric factor.

The increase in the number of methyl groups in butyric acid is the path of tetrahydrofurfuryl alcohol. Thus, the molecules of butyric acid are increasingly impeded by the variable mechanism [24] resulting in lower negative values for V_m^E .

The η^E values are calculated for two composite mixtures as follows:

$$\eta^E = \eta - \sum_{i=1}^N X_i \eta_i \quad (2)$$

where η_i is the absolute viscosity of pure component i and η is the absolute viscosity of mixture. In Fig. 2, we observed a representation of values η^E graphically as a function of mole fraction at 298.15 K. In Fig. 2, we also observed that the aberration in viscosity for whole binary liquid systems is negative through the entire structure, suggesting that the viscosity of pure component is less than the viscosity of the mixture and hence the flow of the mixtures is high [25].

TABLE-2
SHOWS ρ , η , n_D , V_m^E , η^E , ΔG^{*E} AND ΔR OF THFA AND CARBOXYLIC ACIDS AT 298.15 K

X_1	ρ (g cm ⁻³)	V_m^E (cm ³ mol ⁻¹)	n_D	ΔR (cm ³ mol ⁻¹)	η (map.s)	η^E (mpa.s)	ΔG^{*E} (kJ mol ⁻¹)
Tetrahydrofurfuryl alcohol + formic acid							
0.00000	1.21405	0.00000	1.36974	0.00000	1.52212	0.0000	0.00000
0.1022	1.17775	-0.05179	1.37199	-2.59787	1.53682	-0.10985	-0.14238
0.1754	1.15705	-0.07719	1.38959	-3.40653	1.58854	-0.16231	-0.20955
0.2542	1.13829	-0.09221	1.39999	-4.01737	1.69229	-0.19115	-0.23589
0.3463	1.12012	-0.10708	1.40898	-4.35613	1.85768	-0.20556	-0.24329
0.4832	1.02835	-0.11211	1.41237	-4.52492	2.16613	-0.21197	-0.25637
0.5883	1.08483	-0.10650	1.42235	-4.00049	2.48823	-0.19208	-0.24088
0.6684	1.07581	-0.09558	1.42862	-3.45918	2.82340	-0.16345	-0.21947
0.7438	1.0683	-0.08293	1.43282	-2.89703	3.15301	-0.14117	-0.17222
0.8997	1.05509	-0.04889	1.44889	-1.08126	4.05849	-0.07095	-0.09784
1.0000	1.04761	0.00000	1.45322	0.00000	4.89883	0.00000	0.00000
Tetrahydrofurfuryl alcohol + acetic acid							
0.00000	1.04391	0.00000	1.37082	0.00000	1.05752	0.00000	0.00000
0.0952	1.04475	-0.02204	1.37222	-1.10620	1.12753	-0.08185	-0.16861
0.1588	1.04530	-0.03306	1.37899	-1.50684	1.17838	-0.13524	-0.28346
0.2662	1.04584	-0.039404	1.38121	-2.27614	1.34078	-0.17078	-0.35078
0.4209	1.04627	-0.04805	1.38599	-2.92837	1.66897	-0.18899	-0.38301
0.5022	1.04662	-0.05195	1.39109	-3.00471	1.88306	-0.19108	-0.38698
0.6118	1.04689	-0.045621	1.40085	-2.82156	2.29229	-0.16431	-0.32932
0.7209	1.04710	-0.03695	1.41673	-2.18463	2.78509	-0.13684	-0.27542
0.7532	1.04741	-0.03413	1.41899	-2.07440	2.95650	-0.12663	-0.25558
0.8889	1.04760	-0.02169	1.43798	-1.02083	3.85258	-0.06993	-0.14359
1.00000	1.04761	0.00000	1.45322	0.00000	4.89883	0.00000	0.00000
Tetrahydrofurfuryl alcohol + propionic acid							
0.00000	0.98809	0.00000	1.38291	0.00000	1.03043	0.0000	0.00000
0.1185	0.99718	-0.01822	1.38312	-0.68962	1.15428	-0.07125	-0.16707
0.1909	1.00239	-0.02581	1.38873	-0.83188	1.23638	-0.11541	-0.27214
0.2721	1.00799	-0.03473	1.39224	-1.06574	1.35805	-0.14824	-0.34962
0.3647	1.01405	-0.04269	1.39699	-1.24698	1.56124	-0.15308	-0.35922
0.4878	1.02162	-0.05054	1.40122	-1.50206	1.88502	-0.15653	-0.36668
0.5869	1.02725	-0.04665	1.40932	-1.42619	2.21932	-0.14776	-0.34593
0.6758	1.03200	-0.03765	1.41572	-1.34970	2.62403	-0.11885	-0.27649
0.7618	1.03642	-0.03092	1.42219	-1.22053	3.09352	-0.08833	-0.20416
0.8988	1.04309	-0.02037	1.43569	-0.77497	4.03284	-0.03675	-0.08384
1.00000	1.04761	0.00000	1.45322	0.00000	4.89883	0.0000	0.00000
Tetrahydrofurfuryl alcohol + butyric acid							
0.00000	0.95277	0.00000	1.39635	0.00000	1.39589	0.00000	0.00000
0.1125	0.96406	-0.01066	1.39685	-0.32546	1.52041	-0.05581	-0.13794
0.1846	0.97127	-0.02181	1.39772	-0.50332	1.59876	-0.09606	-0.23769
0.2664	0.97935	-0.03109	1.39811	-0.73184	1.72773	-0.12118	-0.29970
0.3537	0.98789	-0.04031	1.40032	-0.88153	1.91311	-0.12886	-0.31862
0.4829	1.00034	-0.04941	1.40372	-1.08897	2.24575	-0.13076	-0.32325
0.5922	1.01058	-0.04187	1.41105	-1.03091	2.60189	-0.12078	-0.29855
0.6843	1.01911	-0.03487	1.41781	-0.94643	2.98849	-0.09789	-0.24187
0.7572	1.02581	-0.02999	1.42425	-0.82075	3.34327	-0.07721	-0.19078
0.8965	1.03845	-0.01878	1.43725	-0.53626	4.18129	-0.02843	-0.07017
1.00000	1.04761	0.00000	1.45322	0.00000	4.89883	0.00000	0.00000

By activating the viscous influx, ΔG^{*E} was obtained by eqn 3:

$$\Delta G^{*E} = RT[\ln(\eta_m V_m) - (X_1 \ln \eta_1 V_1) - (X_2 \ln \eta_2 V_2)] \quad (3)$$

where R is the constant of gases. T is the degree of heat in units of Kelvin, X_1 , X_2 are the mole fraction of components 1 and 2, V_1 , V_2 are the molar volumes of components 1 and 2, and η_1 , η_2 and η_m are the viscosity of component 1,2 and mixture respectively, V_m values were obtained from eqn 4:

$$V_m = (X_1 V_1 + X_2 V_2) / \rho_m \quad (4)$$

Fig. 3 shows the excess Gibbs energy to activate the viscous flow of all systems. The ΔG^{*E} values are negative for all liquid-liquid mixtures. These results can be explained by the acceleration of hydrogen bonding between tetrahydrofurfuryl alcohol and carboxylic acids beyond dipole-dipole molecular interaction between them. The deviation in ΔR of eqn 5 was calculated by utilizing the molar refractivity of pure, blended components and volume fraction of the component [26].

$$\Delta R \text{ (cm}^3 \text{ mol}^{-1}\text{)} = R_m - \sum_i \phi_i R_i \quad (5)$$

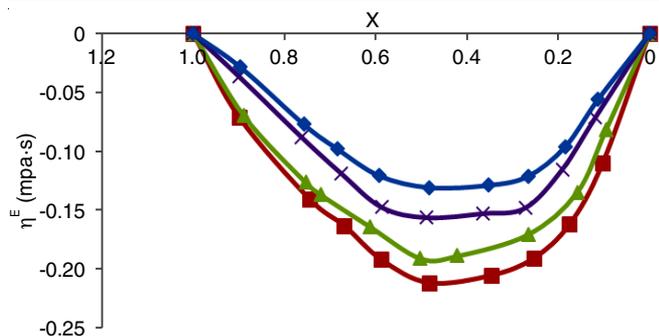


Fig. 2. Curves of η^E against X_1 for the binary mixtures of THFA + methanoic acid (■), ethanoic acid (▲), propanoic acid (×) and butyric acid (◆) at 298.15 K

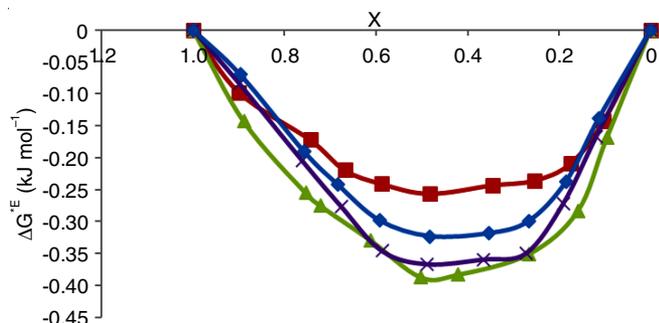


Fig. 3. Curves of ΔG^{*E} against X_1 for the binary mixtures of THFA + methanoic acid (■), ethanoic acid (▲), propanoic acid (×) and butyric acid (◆) at 298.15 K

$$R_m = \left(\frac{n_D^2 - 1}{n_{D,i}^2 + 2} \right) \left(\frac{\sum_i X_i M_i}{\rho_m} \right) \quad (6)$$

$$R_i = \left(\frac{n_D^2 - 1}{n_{D,i}^2 + 2} \right) \left(\frac{M_i}{\rho_i} \right) \quad (7)$$

$$\phi_i = \frac{X_i V_i}{\sum_i X_i V_i} \quad (8)$$

where R_i , R_m , ϕ_i , n_D , $n_{D,i}$ and V_i symbolize the molar refractivity of pure component i and blend, volume fraction of component i in blend, refractive index of blend of pure component i and

molar volume of pure component i , respectively. As shown in Fig. 4, the experimental values ΔR showed a negative deviation from the ideal behavior.

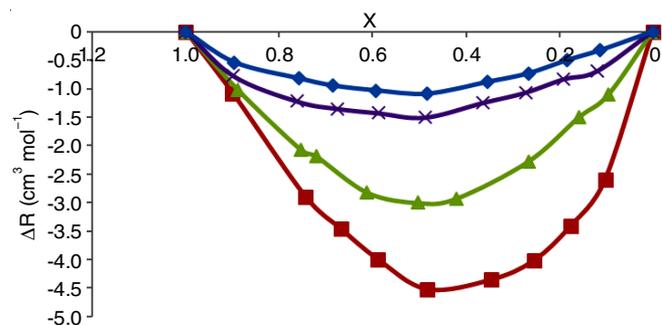


Fig. 4. Curves of ΔR against X_1 for the binary mixtures of THFA + methanoic acid (■), ethanoic acid (▲), propanoic acid (×) and butyric acid (◆) at 298.15 K

There was an increase in negativity with decreasing in the molecular weight of components in mixtures. This increase in negativity can be attributed mainly to the association between the O atom in tetrahydrofurfuryl alcohol (non-associated) and with (-OH) group from C1 to C4 in carboxylic acids by the genesis of hydrogen bonds, and the interactions between unlike molecules were stronger than interactions between like molecules [27-29].

The values of V_m^E , η^E , ΔG^{*E} and ΔR were correlated with the Redlich-Kister polynomial to correlate binary parameters and estimate standard deviation [30].

$$V_m^E, \eta^E, \Delta G^{*E}, \Delta R = X_1 X_2 \sum_{i=1}^n A_i (X_1 - X_2)^{i-1} \quad (9)$$

where n refers to a number of parameters, A_i fitted Redlich-Kister parameter where X is the mole fraction. The corresponding standard deviation σ from eqn 9 is calculated as follows:

$$\sigma_{st} = \left(\frac{\sum_i^N (X_i^{calc.} - X_i^{exp.})^2}{(N - n)} \right)^{1/2} \quad (10)$$

where $X_i^{exp.}$ and $X_i^{calc.}$ are the experimental and calculated values of the component i , respectively, N is the numeral of empirical points and n is the numeral of fitted parameter (Table-3).

TABLE-3
PARAMETERS THE STANDARD DEVIATION OF eqn. 9 AND 10 FOR THFA + THE SMALL CARBOXYLIC ACIDS AT 298.15 K

System		A_0	A_1	A_2	σ
Tetrahydrofurfuryl alcohol+ formic acid	V_m^E	-0.439427	0.046753	-0.144996	0.01225
	η^E	-0.820908	0.273198	-0.249268	0.01149
	ΔG^{*E}	-1.00108	0.291767	-0.468927	0.01524
	ΔR	-17.4882	7.122081	-3.46811	0.05709
Tetrahydrofurfuryl alcohol+ acetic acid	V_m^E	-0.194771	0.028191	-0.044610	0.01110
	η^E	-0.745219	0.210639	-0.168612	0.00902
	ΔG^{*E}	-1.50424	0.461761	-0.411901	0.01471
	ΔR	-11.9254	0.817647	1.716198	0.06515
Tetrahydrofurfuryl alcohol+ propionic acid	V_m^E	-0.188737	-0.005527	0.029580	0.01355
	η^E	-0.633031	0.216575	0.098713	0.01166
	ΔG^{*E}	-1.48386	0.528901	0.245812	0.01831
	ΔR	-5.65647	-1.22896	-2.10417	0.05975
Tetrahydrofurfuryl alcohol+ butyric acid	V_m^E	-0.181996	-0.012945	0.071805	0.01092
	η^E	-0.530470	0.184153	0.089533	0.01498
	ΔG^{*E}	-1.31142	0.456505	0.221553	0.02361
	ΔR	-4.16470	-1.01684	0.042513	0.04796

Conclusion

This paper presents experimental data on ρ , η and n_D of tetrahydrofurfuryl alcohol with short-chain carboxylic acids on a full scale of mole fraction at 298.15 K. The values of pure ingredients generally correspond to the literature that can be obtained. The experimental data values were utilized to calculate V_m^E , viscosity deviations, ΔG^{*E} and refractive index deviations, and correlated using the Redlich-Kister multiplex equation. The negative values of these parameters for the mixtures of carboxylic acids C1-C4 are due to the strong bond of hydrogen between molecules.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

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