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THE REDUCED REDLICH-KISTER EQUATIONS FOR CORRELATING EXCESS PROPERTIES OF 1,2-DIMETHOXYETHANE + WATER BINARY MIXTURES AT TEMPERATURES FROM 303.15 K TO 323.15 K.

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ABSTRACT

Excess molar volumes, viscosity deviations and excess molar Gibbs energy of activation of viscous flow in 1,2-dimethoxyethane + water binary mixtures at (303.15, 308.15, 313.15, 318.15 and 323.15) K. were calculated from experimental density and viscosity data presented in previous work. Here these experimental values were used to test the applicability of the correlative reduced Redlich-Kister equation and the Herráez and Belda equations as well as their corresponding relative properties. Their correlation ability at different temperatures, and the use of appropriate numbers of parameters, is discussed for the case of limited experimental data. The relative functions are important to reduce the effect of temperature and, consequently, to reveal the effects of different factors and types of interactions. Values of limiting excess partial molar thermodynamic properties at infinite dilution were deduced from different methods.

Keywords: Binary liquid mixture, viscosity, Reduced Redlich-Kister equation, Molecular interaction, 1,2 dimethoxyethane.

INTRODUCTION

Physical and chemical properties of fluid mixtures are important for understanding their thermodynamic behavior. One of the most important considerations is that these properties may provide information about molecular interactions. This paper is a continuation of our previous works and those in literature that include the study of the binary liquid mixture of 1,2-dimethoxyethane (DMOE) + water (W)[1-14]. The measured density and viscosity values were used to calculate the excess properties and then fitted to the Redlich-Kister equation. The DMOE + water solvent mixture is mostly attractive, and a great deal of works has been well explored by many researchers[15-26] with different detailed experimental studies, to understand the non-bonding interactions such as hydrogen bonding, electrostatic, hydrophobic hydration and hydrophobic interaction that play an important role in the stabilization of native conformations of biological macromolecules. Also, dynamic viscosity and specific density deviations are fitted to the Herráez and the reduced Redlich-Kister (R-K) equations[3-8], through the Legendre polynomials, and are interpreted in terms of molecular interactions and structural effects. Note that similar study were made in previous papers for isobutyric acid + water mixtures both near to, and far away from, the critical temperatures and for 1,4-dioxane + water ones for different temperatures[27-33]. In addition, the reduced R-K function relative to the molar volume and the Gibbs free energy which are equivalent to the apparent molar property is more

sensitive than the corresponding excess property to interaction and gives more information[27-34]. We have noted that, at infinite dilution of one component in the other one, the Herráez exponential polynomials' values converge to the universal exponents (0.5 or 1.0) independent of temperature which is justified[27-29,32-34] and considered as an interaction's indicator characterizing existing or predominant solute-solute or solute-solvent interaction at infinite dilution.

EXPERIMENTAL

General procedure

Redlich-Kister Equations

Excess Properties Results:

The excess molar volumes V^E, viscosity deviation $\Delta\eta$ and excess molar Gibbs free energy ΔG^{*E} were correlated with the mole fraction x_1 of DMOE using the Redlich-Kister[1] expression as described below which was fitted with least-squares polynomials at three different temperatures (303.15, 308.15, 313.15, 318.15 and 323.15) K. Note that the measured density and viscosity values of DMOE and W binary mixtures were used to calculate the excess molar volumes, viscosity deviations and excess molar Gibbs free energy of this system and then fitted to the Redlich-Kister equation[27-32]:

$$Y^E = x_1(1- x_1).\sum_{p=0}^{p=n} A_{p,T}(2x_1 - 1)^p \quad (1)$$

Where Y^E denotes V^E, $\Delta\eta$ and ΔG^{*E}, and n is the optimal number of parameters (n = 5). The coefficients $A_{p,T}$ in Eq.-1 are regressed by employing the least-square fit method.

The corresponding standard deviation $\sigma(Y)$ was calculated by Eq.-2:

$$\sigma(Y) = \sqrt{\sum_{i=1}^{i=N} \frac{\left(Y_{i,\exp} - Y_{i,calc}\right)^2}{N-m}} \quad (2)$$

Where N and m are the numbers of data points and of adjustable parameters, respectively.

In the same context, Desnoyers et al.[31] showed that an examination of the trends of the dependence of V^E, $\Delta\eta$ and ΔG^{*E} on x_1 suggested that many of these systems are similar, and the differences in interactions are mostly significant in solutions very rich in component 2. From the treatment of excess thermodynamic quantities for liquid mixtures proposed by Desnoyers et al.[31], we can conclude that the excess quantity (V^E, $\Delta\eta$ and ΔG^{*E}) gives an overall view of the origin of the non-ideality in the mixtures but still can be quite misleading, especially for systems that show strong interactions at high dilution. Desnoyers et al.[31] suggested that, in agreement with the original statements of Redlich and Kister[30], it is better to use the ratios $V^E / x_1(1 - x_1)$, $\Delta\eta / x_1(1 - x_1)$ or $\Delta G^{*E} / x_1(1 - x_1)$ for this purpose.

RESULTS AND DISCUSSION

The Reduced Redlich-Kister Equation

The precedent ratios represent the experimental reduced Redlich-Kister excess properties and denoted $Q_{Y,\exp,T}(x_1)$ which are expressed by Eq.-3,

$$Q_{Y,\exp,T}(x_1) = Y^E/(x_1(1- x_1)) \quad (3)$$

Where Y^E denotes V^E, $\Delta\eta$ and ΔG^{*E}. Also, the excess thermodynamic quantities have the advantage of illustrating the sign and magnitude of their non-ideality, but the reduced Redlich and Kister function $Y^E / x_1(1 - x_1)$ gives a much better handle on the origin of the non-ideality. The experimental reduced Redlich-Kister excess values (Eq.-3) applied for the molar volume $Q_{V,T}(x_1)$, the viscosity $Q_{\eta,T}(x_1)$ and the molar Gibbs free energy $Q_{\Delta G^*,T}(x_1)$ for DMOE (1) + W (2) mixtures against the mole fraction x_1 in DMOE at temperatures 303.15, 308.15, 313.15, 318.15 and 323.15 K are presented in Table-1.

We note that the reduced Redlich-Kister excess property is more sensitive than the direct excess property, V^E, $\Delta\eta$ or ΔG^{*E}, to interactions that occurs at low concentrations[30, 31]. We note that Redlich and Kister treated the data with a power series, putting all the weight on data near 0.5 mole fraction. As Desnoyers et *al*. have shown[31], this is not always the best approach for mixtures having specific interactions such as association at low concentration. Therefore, elimination of the factor $[x_1(1 - x_1)]$ (Eq.-1) in the reduced Redlich-Kister excess function $Q_{Y,\exp,T}(x_1)$ (Eq.-3) removes this effect and gives a specific reduced function $Q_{Y,\exp,T}(x_1)$ characterizing the viscosity or other property and also gives evidence to the existence of important interactions.For the system studied in this work, the experimental reduced excess molar volume, viscosity and excess molar Gibbs free energy $Q_{Y,\exp,T}(x_1)$, Eq.-3, *versus* the mole fraction x_1 of DMOE is plotted in Figures- 1,2 and 3.

Table-1: Experimental reduced Redlich-Kister excess values (Eq.-3) relative to the molar volume $Q_{V,T}(x_1)$ /10^{-6} $m^3 \cdot mol^{-1}$, the viscosity $Q_{\eta,T}(x_1)$ / mPa·s and the molar Gibbs free energy $Q_{\Delta G^*,T}(x_1)$ kJ·mol^{-1} for DMOE (1) + W (2) mixtures against the mole fraction x_1 in DMOE at the temperatures: (303.15, 308.15, 313.15, 318.15 and 323.15) K.

x_1	$Q_{V,T}(x_1)$	$Q_{\square,T}(x_1)$	$Q_{\square G^*,T}(x_1)$	x_1	$Q_{V,T}(x_1)$	$Q_{\square,T}(x_1)$	$Q_{\square G^*,T}(x_1)$
303.15 K							
0.0000	-11.610	14.181	-	0.3598	-7.4190	2.0283	9.7872
0.0471	-11.628	12.680	36.413	0.4455	-6.5270	1.1586	7.4311
0.0951	-11.562	11.336	29.397	0.5389	-5.9586	0.64309	5.6948
0.1421	-11.156	9.1135	23.850	0.6952	-5.1353	0.23266	3.8821
0.1877	-9.9078	6.7050	19.268	0.8030	-4.4646	0.09103	3.0507
0.2322	-9.4534	4.9304	15.893	0.9001	-4.1013	0.00445	2.4010
0.2770	-8.6190	3.5512	13.185	1.0000	-3.9280	-	-
308.15 K							
0.0000	-11.661	11.889	-	0.3598	-7.4048	1.8950	9.7167
0.0471	-11.636	10.530	35.041	0.4455	-6.5250	1.0157	7.4096
0.0951	-11.579	9.3799	28.430	0.5389	-5.9900	0.57709	5.7351
0.1421	-11.106	7.5697	23.168	0.6952	-5.1845	0.22181	3.9851
0.1877	-9.8502	5.5900	18.765	0.8030	-4.4993	0.09862	3.1887
0.2322	-9.4574	4.2203	15.678	0.9001	-4.1833	0.01779	2.5559
0.2770	-8.5980	3.0389	13.001	1.0000	-3.9955	-	-
313.15 K							
0.0000	-11.712	9.9699	-	0.3598	-7.4216	1.5459	9.6628
0.0471	-11.703	8.8611	33.884	0.4455	-6.5805	0.90273	7.4100
0.0951	-11.526	7.9367	27.741	0.5389	-6.0867	0.52115	5.7738
0.1421	-11.097	6.3474	22.538	0.6952	-5.2397	0.20718	4.0565
0.1877	-9.8289	4.7295	18.351	0.8030	-4.5391	0.08850	3.2260
0.2322	-9.4294	3.5859	15.363	0.9001	-4.2944	0.01779	2.6134
0.2770	-8.6311	2.6175	12.821	1.0000	-4.0630	-	-
318.15 K							
0.0000	-11.763	8.5617	-	0.3598	-7.4338	1.3619	9.6110
0.0471	-11.577	7.5488	32.893	0.4455	-6.6006	0.80517	7.4141
0.0951	-11.487	6.6782	26.869	0.5389	-6.1380	0.47045	5.8090
0.1421	-11.062	5.3647	21.936	0.6952	-5.3050	0.19774	4.1546
0.1877	-9.7974	4.0461	17.990	0.8030	-4.4554	0.09166	3.3454
0.2322	-9.4299	3.0435	15.001	0.9001	-4.2927	0.03336	2.7897
0.2770	-8.6221	2.2724	12.662	1.0000	-4.1305	-	-
323.15 K							
0.0000	-11.804	7.4282	-	0.3598	-7.5015	1.2012	9.5420
0.0471	-11.760	6.5105	32.031	0.4455	-6.6797	0.72178	7.4181
0.0951	-11.471	5.6870	26.132	0.5389	-6.0465	0.42900	5.8611

0.1421	-11.044	4.5871	21.428	0.6952	-5.3767	0.18075	4.1949
0.1877	-10.037	3.5083	17.695	0.8030	-4.5075	0.08471	3.3945
0.2322	-9.4423	2.6278	14.742	0.9001	-4.2593	0.02335	2.7913
0.2770	-8.6458	1.9723	12.477	1.0000	-4.1980	-	-

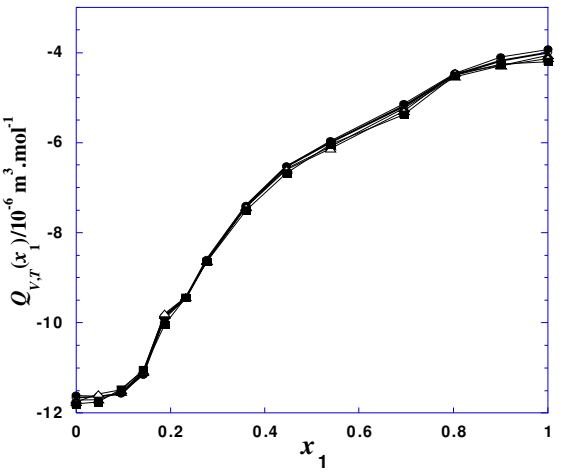

Fig.-1: Experimental reduced Redlich-Kister excess properties $Q_{V,T}(x_1) = V^E / x_1(1 - x_1)$ of excess molar volume for DMOE (1) + W (2) mixtures against mole fraction x_1 in DMOE at the temperatures, (●): 303.15 K; (○): 308.15 K; (▲): 313.15 K; (Δ): 318.15 K; (■): 323.15 K.

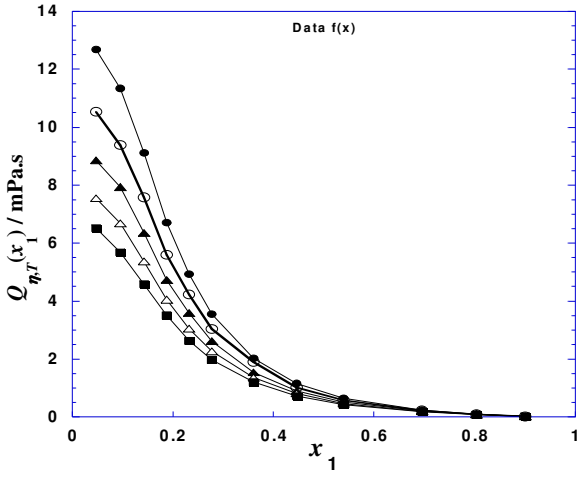

Fig.-2: Experimental reduced Redlich-Kister excess properties $Q_{\eta,T}(x_1) = \Delta\eta/x_1(1 - x_1)$ / mPa·s of viscosity deviation for DMOE (1) + W (2) mixtures against mole fraction x_1 in DMOE at the temperatures, (●): 303.15 K; (○): 308.15 K; (▲): 313.15 K; (Δ): 318.15 K; (■): 323.15 K.

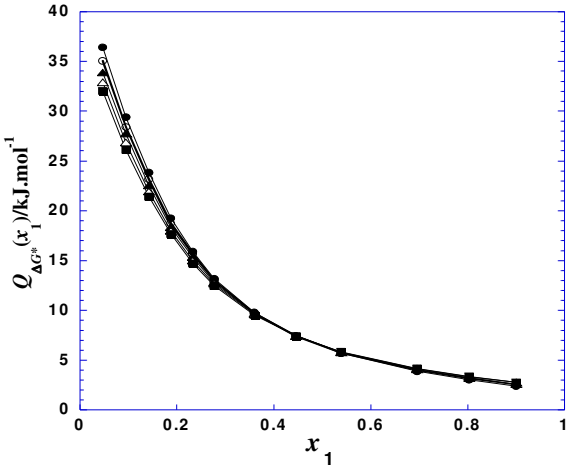

Fig.-3: Experimental reduced Redlich-Kister excess properties $Q_{\Delta G^*,T}(x_1) = \Delta G^{*E} / x_1(1 - x_1)$ / kJ·mol^{-1} of excess molar Gibbs free energy for DMOE (1) + W (2) mixtures against mole fraction x_1 in DMOE at the temperatures, (●): 303.15 K; (○): 308.15 K; (▲): 313.15 K; (Δ): 318.15 K; (■): 323.15 K.

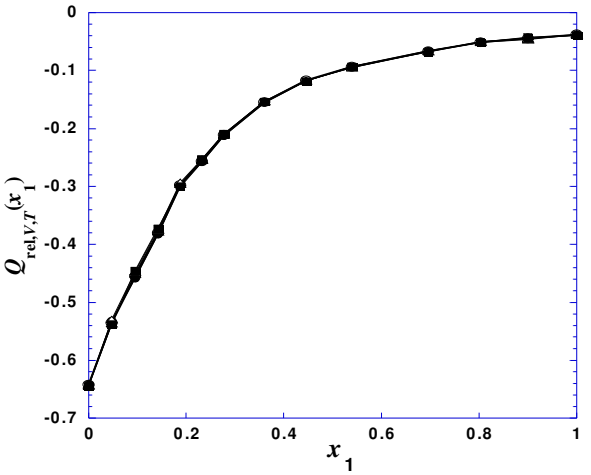

Fig.-4: Experimental relative reduced Redlich-Kister excess properties $Q_{rel,V,T}(x_1)$ for the ratio = $Q_{V,T}(x_1)/V$ of excess molar volume for DMOE (1) + W (2) mixtures against mole fraction x_1 in DMOE at the temperatures, (●): 303.15 K; (○): 308.15 K; (▲): 313.15 K; (Δ): 318.15 K; (■): 323.15 K.

We suppose that the changes in curvature exhibited in these Figures-1 and 2 are due to hydrogen bonding between unlike molecules of the mixture leading to strong intermolecular correlation. The less decrease in

$Q_{V,\exp,T}(x_1)$ with increase in temperature (Fig.-1) is explained by considering the differences in the molar volumes of the two liquids at different temperatures. In case of molar volume and viscosity properties it is observed that the shape of the present curves (Figs.-1 and 2) is almost similar to the excess molar volume and viscosity deviation versus mole fraction curves, reported earlier[1] with a slight shift in their minima and maxima values, respectively, but for isentropic compressibility the present curve (Fig.-3) shows a monotonous increase with mole fraction of DMOE (x_1) without any minimum which was shown in earlier curve [1]. For the present system the reduced Redlich-Kister isentropic compressibility $Q_{\kappa s,T}(x_1)$ values are negative over the entire composition range at all the three temperatures investigated. The existence of these negative values means that the mixture is less compressible than the corresponding ideal mixture suggesting a predominant hydrogen bond interaction between DMOE and water. The strictly monotonous increase of the reduced isentropic compressibility values with the mole fraction of DMOE can be also explained by the geometrical effects allowing for the accommodation of molecules of different size into the cavities of molecular network. The observed trend indicates that interstitial accommodation of water in DMOE molecular network is much greater than the reverse case and the disappearance of the minima involves the formation of some stable structure between the two unlike molecules[1,15-26].

Although DMOE+W mixtures are nonelectrolyte solutions, we can adopt an expansion equivalent to the Debye-Hückel and Jones-Dole expressions[35-39] for very dilute nonelectrolyte solution where the first parameter a_1 (of Eq. 4) is related to solute-solute interaction while the second parameter a_2 is related to solute-solvent interaction. Generally, the experimental reduced Redlich-Kister excess viscosity (Eq.-3) $Q_{\eta,\exp,T}(x_1)$ increases exponentially and diverges at infinite dilution in several binary mixtures (as $x_1 \rightarrow 0$) [2,27-32,40-42], according to the following equation:

$$\eta = \eta_2(1+a_1x_1^{1/2} +a_2x_1 +a_3x_1^{3/2} + \cdots) \quad (4)$$

We note that, at higher temperatures, there will be a competition between molecular interactions and thermal disordered movement of molecules. On the other hand, the absence of a divergence of the experimental reduced Redlich-Kister viscosity deviation (Fig.-2) at infinite dilution of water in DMOE ($x_1 \rightarrow 1$) gives evidence for a negligible a_1 parameter. In fact, for nonelectrolyte solutions, the absence of solute-solute interactions leads us to consider that the a_1 value is always zero. We can also state that the large separation of curves near $x_1 \approx 0$ and their closeness near $x_1 \approx 1$ suggest that the solute-solvent parameter a_2 (Eq. 4), which is equivalent to B parameter of the Jones-Dole equation[35-39], is much larger for DMOE in W than for the reverse case, and we can justify the existing hydrophobic hydration even in infinite dilution[1,15-26].

In addition, values of reduced Redlich-Kister functions $Q_{Y,T}(x_i)$ (Eqs.-1 and 3) at infinite dilution represent values of the equivalent of the partial excess physical magnitudes at infinite dilution ($x_1 \rightarrow 0^+$) [43], which can be also calculated from the adjustable parameters using Eqs.-5 and 6:

$$Q_{\eta,T}(x_1=0) = A_{0,T} - A_{1,T} + \ldots + (-1)^p \cdot A_{p,T+\ldots+} (-1)^n \cdot A_{n,T} \quad .$$
$$= (\eta_1 - \eta_2) \cdot (1 + (\partial \ln(\eta - \eta_2)/\partial x_1)_{T,x1=0}) \quad (5)$$

and

$$Q_{\eta,T}(x_1=1) = A_{0,T} + A_{1,T} + \ldots + A_{p,T+\ldots+} A_{n,T} \quad .$$
$$= (\eta_1 - \eta_2) \cdot (1 - (\partial \ln(\eta - \eta_2)/\partial x_1)_{T,x1=1}) \quad (6)$$

As discussed in literature[44-47], the variation of $Q_{V,\exp,T} = V^E/x_1(1-x_1)$ (Eq.-3) with composition was used in every case to test the quality of the data; this function is extremely sensitive to experimental errors, particularly in the dilute ranges. In addition, its values at infinite dilution represent the values of the partial excess molar volume at infinite dilution, $\bar{V}_i^{E,\infty}$ [48], which can be also calculated from the adjustable parameters using the Eqs.-7 and 8 for Redlich-Kister expression:

$$Q_{V,T}(x_1=0) = A_{0,T} - A_{1,T} + \ldots + (-1)^p \cdot A_{p,T+\ldots+} (-1)^n \cdot A_{n,T} \quad .$$
$$= \bar{V}_1^{E,\infty} = \bar{V}_1^{\infty} - V_1 \quad (7)$$

and

$$Q_{V,T}(x_1=1) = A_{0,T} + A_{1,T} + \ldots + A_{p,T+\ldots+} A_{n,T} \quad .$$

$= \bar{V}_2^{E,\infty} = \bar{V}_2^{\infty} - V_2$ (8)

In Eqs. 7 and 8, V_i is the molar volume of pure component '*i*'. Molar volume of the mixture (V) and partial molar volumes of DMOE ($\bar{V}_1$) and water ($\bar{V}_2$) for DMOE (1) + W (2) mixtures against the mole fraction x_1 in DMOE at the temperatures 303.15, 308.15, 313.15, 318.15 and 323.15 K are reported in Table-2. The values of $\bar{V}_i^{E,\infty}$ calculated by using Eqs.-7 and 8 are listed in Table-3.

As discussed in literature[3,31,41,49,50], it can also readily be shown that, for many properties, such as enthalpies, heat capacities, volumes, compressibilities and expansibilities, this quantity ($Q_{Y,T}(x_1)$) is directly related to the apparent molar quantities of both components:

$$Q_{Y,T}(x_1) = Y^E/x_1(1-x_1) = \frac{(Y_{2,\varphi} - Y_2^*)}{x_1} = \frac{Y_{1,\varphi} - Y_1^*}{x_2} \quad (9)$$

This function is therefore equivalent to an apparent molar quantity over the whole mole fraction range, and its extrapolation to $x_1 = 0$ and $x_1 = 1$ (parameters sum $Q_{Y,T}(x_1=0)$ and $Q_{Y,T}(x_1=1)$ of Eqs. 7 and 8) will give two excess partial molar quantities $\bar{Y}_i^{E,\infty}$ (where i = 1 or 2). These two limiting parameters are of fundamental importance since they are, measures of the solute-solvent interactions of both components.

It is therefore important to determine precisely the Redlich-Kister interaction parameters, and especially those corresponding to $x_1 = 0$ and $x_1 = 1$. Desnoyers et al.[31] suggested that one of the simplest ways of determining these parameters was from a plot of $Y^E/x_1(1-x_1)$ (or dY^E/dx_1 for free energies). Note that the Redlich-Kister excess quantity (Eq.-1) cannot appropriately represent the dependence of viscosity with composition.

We can add that these parameters can be obtained from three other manners:

(i) A local extrapolation by fitting partially the curves $Q_{V,T}(x_1)$ separately in each rich-region of one component (1) or (2). The obtained values are truer than those given from Eqs.-7 and 8.

(ii) The fit of the molar volume $V(x_1)$ against the molar fraction (x_1) gives directly the partial molar volume $\bar{V}_i$ of the component 'i' by the following equation:

$$\bar{V}_i = V(x_i) - x_j \cdot \left(\frac{\partial V(x_j)}{\partial x_j} \right)_{T,P} \quad (10)$$

Table-2: Values of molar volume of the mixture $V/10^{-6}$ $m^3 \cdot mol^{-1}$ and partial molar volumes of DMOE ($\bar{V}_1$) $/10^{-6}$ $m^3 \cdot mol^{-1}$ and water ($\bar{V}_2$) $/10^{-6}$ $m^3 \cdot mol^{-1}$ for DMOE (1) + W (2) mixtures against the mole fraction x_1 in DMOE at the temperatures: (303.15, 308.15, 313.15, 318.15 and 323.15) K.

x_1	V	$\bar{V}_1$	$\bar{V}_2$	x_1	V	$\bar{V}_1$	$\bar{V}_2$
303.15 K							
0.0000	18.093	90.567	18.093	0.3598	47.719	103.82	16.189
0.0471	21.678	94.273	18.090	0.4455	55.279	104.40	15.816
0.0951	25.372	97.093	17.835	0.5389	63.545	104.69	15.453
0.1421	29.109	99.180	17.502	0.6952	77.550	105.00	14.951
0.1877	32.929	100.80	17.247	0.8030	87.320	105.16	14.622
0.2322	36.630	101.89	16.893	0.9001	96.114	105.19	14.358
0.2770	40.491	102.77	16.629	1.0000	105.18	105.18	14.446
308.15 K							
0.0000	18.122	91.401	18.122	0.3598	47.990	104.50	16.230
0.0471	21.733	95.034	18.110	0.4455	55.604	105.08	15.857
0.0951	25.471	97.822	17.867	0.5389	63.924	105.36	15.491
0.1421	29.238	99.882	17.537	0.6952	78.030	105.66	15.000
0.1877	33.092	101.49	17.287	0.8030	87.876	105.84	14.668
0.2322	36.812	102.57	16.926	0.9001	96.720	105.86	14.330
0.2770	40.708	103.45	16.668	1.0000	105.88	105.88	14.271
313.15 K							
0.0000	18.155	92.285	18.155	0.3598	48.264	105.17	16.280

0.0471	21.795	95.820	18.136	0.4455	55.927	105.76	15.893
0.0951	25.573	98.552	17.904	0.5389	64.300	106.05	15.508
0.1421	29.369	100.57	17.575	0.6952	78.524	106.37	15.018
0.1877	33.256	102.16	17.333	0.8030	88.449	106.55	14.680
0.2322	37.008	103.24	16.979	0.9001	97.344	106.56	14.297
0.2770	40.923	104.11	16.713	1.0000	106.59	106.59	14.175
318.15 K							
0.0000	18.192	92.955	18.192	0.3598	48.536	105.83	16.333
0.0471	21.869	96.536	18.178	0.4455	56.253	106.43	15.936
0.0951	25.676	99.276	17.941	0.5389	64.680	106.76	15.497
0.1421	29.504	101.29	17.613	0.6952	79.007	107.15	14.807
0.1877	33.421	102.86	17.375	0.8030	89.030	107.37	14.293
0.2322	37.198	103.92	17.021	0.9001	97.988	107.34	13.745
0.2770	41.144	104.78	16.763	1.0000	107.29	107.29	13.574
323.15 K							
0.0000	(-11.804)	18.232	93.471	18.232	48.805	106.54	16.360
0.0471	-11.760	21.922	97.138	18.204	56.577	107.11	15.973
0.0951	-11.471	25.783	99.964	17.987	65.109	107.47	15.602
0.1421	-11.044	29.643	102.01	17.656	79.504	107.80	14.964
0.1877	-10.037	33.553	103.56	17.375	89.608	108.01	14.609
0.2322	-9.4423	37.395	104.66	17.053	98.664	108.02	14.378
0.2770	-8.6458	41.368	105.51	16.793	108.01	108.01	14.736

When we proceed to operations of limits of Eq.-10 at infinite dilution ($x_1 \rightarrow 0^+$ or $x_1 \rightarrow 1^-$), we can easily obtain the values of the excess partial molar volume at infinite dilution of the component '*i*' in the other one ($\bar{V}_i^{E,\infty}$) :

$$\bar{V}_1^{E,\infty} = \bar{V}_1^{\infty} - V_1 = -(V_1 - V_2) + \left(\frac{\partial V(x_1)}{\partial x_1}\right)_{T,P,x_1=0} \quad (11)$$

and,

$$\bar{V}_2^{E,\infty} = V_2^{\infty} - V_2 = (V_1 - V_2) - \left(\frac{\partial V(x_1)}{\partial x_1}\right)_{T,P,x_1=1} \quad (12)$$

(iii) We note that in recent work, Belda[41] proposes a new empirical correlation equation for four properties (density, viscosity, surface tension, and refractive index) (Eq.-13) which introduces a correcting factor to linearity as a homographic function acting upon the molar fraction of one component of the binary mixture (x_1):

$$Y_B(x_1) = Y_2 + (Y_1 - Y_2)\cdot x_1 \cdot \frac{1+m_1\cdot(1-x_1)}{1+m_2\cdot(1-x_1)} \quad (13)$$

Where $Y_B(x_1)$ is the mixture property and m_1 and m_2 are the two introduced empirical adjustable parameters.

Belda coefficients m_i for (Eq.-13) and corresponding errors of molar volume (V) for DMOE (1) + water (2) mixtures at the temperatures 298.15, 308.15 and 318.15 K are given in Table-4.

Table-3: Comparison of calculated values-with three methods-of the limiting excess partial molar volume (10^{-6} $m^3\cdot mol^{-1}$) at infinite dilution relative to DMOE (1) and of water (2) in their binary mixture at the temperatures: (303.15, 308.15, 313.15, 318.15 and 323.15) K.

$\bar{V}_i^{E,\infty}$ / 10^{-6} $m^3\cdot mol^{-1}$	*T*	Redlich-Kister	Derivation	Belda
	K	Eqs. 7 and 8	Eqs. 11 and 12	Eqs. 14 and 15

$\bar{V}_1^{E,\infty}$	303.15	-14.613	-14.740	-12.408
	308.15	-14.479	-14.280	-12.006
	313.15	-14.305	-13.918	-13.644
	318.15	-14.335	-14.051	-13.251
	323.15	-14.539	-13.975	-12.958
$\bar{V}_2^{E,\infty}$	303.15	-3.647	-3.746	-3.2.65
	308.15	-3.851	-3.712	-4.596
	313.15	-3.980	-3.720	-4.636
	318.15	-4.618	-3.722	-4.356
	323.15	-3.496	-3.715	-3.987

Table-4: Belda coefficients m_i for (Eq. 13) and corresponding errors, standard deviation $\sigma(Y)$ (Eq. 2) of molar volume (V) for DMOE(1)+water(2) mixtures at the temperatures:(303.15, 308.15, 313.15, 318.15 and 323.15) K.

T / K	m_1		m_2		σ_B (Y)
	value	Error	value	Error	
303.15	-0.4811	0.02487	-0.4555	0.02620	0.0476
308.15	-0.5011	0.02792	-0.4375	0.02894	0.0494
313.15	-0.5291	0.02879	-0.4695	0.02981	0.0487
318.15	-0.5573	0.02765	-0.4878	0.02768	0.0473
323.15	-0.5732	0.02841	-0.4911	0.02942	0.0489

In previous papers[49,50], we have discussed the validity of the Belda equation for some physicochemical properties in the 1,4-dioxane-water and isobutyric acid-water mixtures and we have concluded that this equation can give better results than the Redlich-Kister one with the same number of free parameters. We have also given physical significance[3] for their corresponding parameters (m_1 and m_2). In fact, when we proceed to some operations of limits and derivations on the Belda equation by inserting Eq.-13 in Eqs.-11 and 12, we can easily obtain the values of partial excess property at infinite dilution of the component "i" in the other one ($\bar{Y}_i^{E,\infty}$) through the following equations:

$$\bar{V}_1^{E,\infty} = \bar{V}_1^{\infty} - V_1 = \frac{m_1 - m_2}{1 + m_2} \cdot (V_1 - V_2) \quad (14)$$

and,

$$\bar{V}_2^{E,\infty} = \bar{V}_2^{\infty} - V_2 = (m_1 - m_2) \cdot (V_1 - V_2) \quad (15)$$

Comparison of calculated values-with three methods-of the limiting excess partial molar volume (10^{-6} $m^3 \cdot mol^{-1}$) at infinite dilution relative to DMOE (1) and of water (2) in their binary mixture at the temperatures 303.15, 308.15, 313.15, 318.15 and 323.15 K are given in Table-3.

The Relative Reduced Redlich-Kister Equation

Nevertheless, like the relative deviation, we can consider the relative reduced Redlich-Kister function, $Q_{Y,\mathrm{rel},T}(x_1) = Q_{Y,T}(x_1)/Y(x_1)$. The plot of $Q_{Y,\mathrm{rel},T}(x_1)$ versus mole fraction x_1 (Figures- 4,5 and 6) shows that the curves draw together and $Q_{Y,\mathrm{rel},T}(x_1)$ curves seem to depend very slightly on temperature. We conclude that the nearness in first portion and then the larger separation of curves in Fig.-2 at the two limits of infinite dilution do not depend only on the value of the solute-solvent parameter a_2 but are also affected by temperature. We remark that the differences between their Arrhenius activation energy of viscosity Ea can play an important role in this effect. Note that the relative reduced Redlich-Kister function $Q_{Y,\mathrm{rel},T}(x_1)$ is also a good tool, like the reduced Redlich-Kister function $Q_{Y,T}(x_1)$, for interpreting different types of interactions.

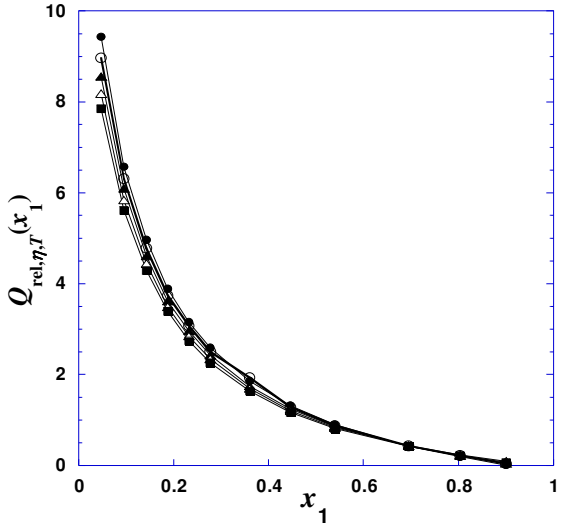

Fig.-5: Experimental relative reduced Redlich-Kister excess properties $Q_{rel,\eta,T}(x_1)$ for the ratio = $Q_{\eta,T}(x_1)/\eta$ of viscosity deviation for DMOE (1) + W (2) mixtures against mole fraction x_1 in DMOE at the temperatures, (●): 303.15 K; (○): 308.15 K; (▲): 313.15 K; (Δ): 318.15 K; (■): 323.15 K.

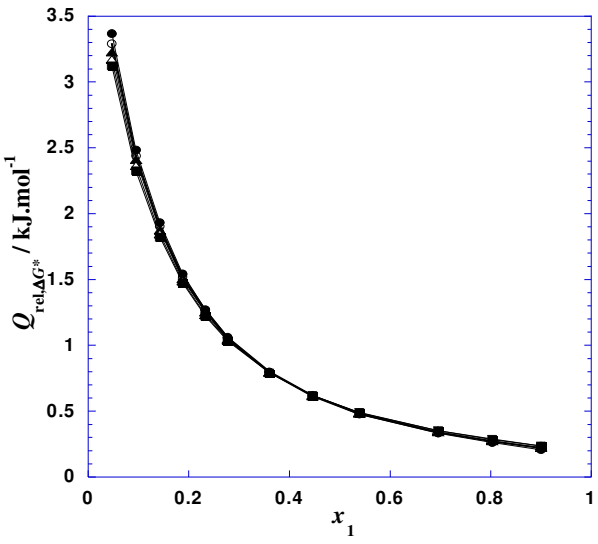

Fig.-6: Experimental relative reduced Redlich-Kister excess properties $Q_{rel,\kappa_S,T}(x_1)$ for the ratio = $Q_{\Delta G^*,T}(x_1)$ $/\Delta G^{*E}$ of the molar Gibbs free energy for DMOE (1) + W (2) mixtures against x_1 in DMOE at the temperatures, (●): 303.15 K; (○): 308.15 K; (▲): 313.15 K; (Δ): 318.15 K; (■): 323.15 K.

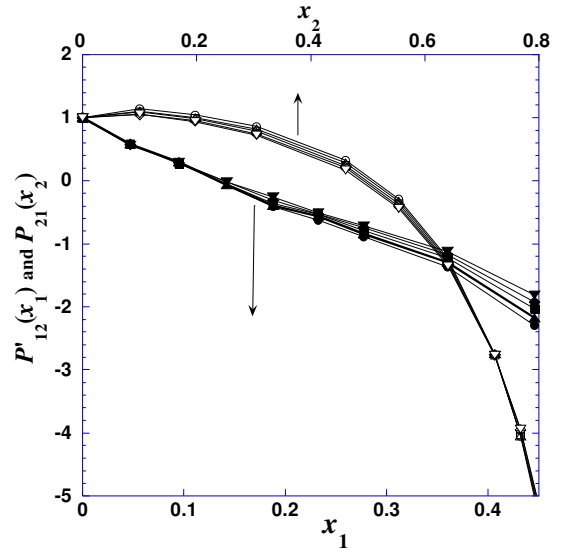

Fig.-7: Variation of the experimental Herráez exponents $P'_{12,exp}(x_1)$ (Eq. 18) and $P_{21,exp}(x_2)$ (Eq. 21) for DMOE (1) + W (2) mixtures against mole fraction x_1 in DMOE at the temperatures, $P'_{12,exp}(x_1)$: (●): 303.15 K; (▲):308.15 K; (■):313.15 K; (♦):318.15 K and (▼):323.15 K, and $P_{21,exp}(x_2)$:(○): 303.15 K; (Δ):308.15 K; (□) : 313.15 K; (◊): 318.15 K and (▽):323.15 K.

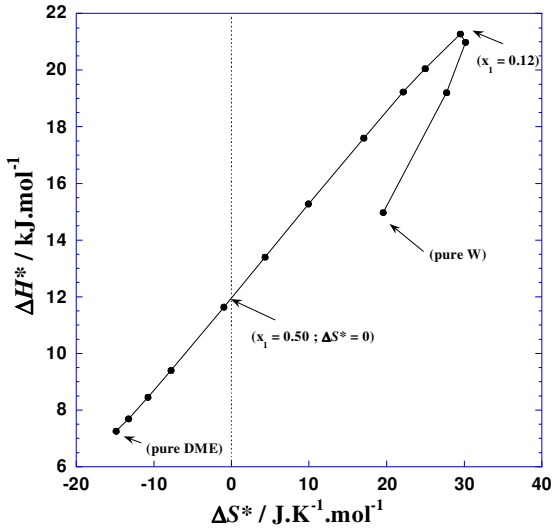

Fig.-8: Correlation between the enthalpy of activation of viscous flow ΔH^* / (kJ·mol^{-1}) and the entropy of activation of viscous flow ΔS^* / (J·mol^{-1}·K^{-1}) for DMOE (1) + W (2) mixtures against mole fraction x_1 in DMOE at the temperatures, in the temperature range (303.15 to 323.15 K).

In addition, Herráez et *al*.[33] propose a new empirical correlation equation (Eqs.-16 and 19) which introduces a correcting polynomial (Eqs.-17 and 20) as an exponential-acting upon the molar fraction of one of the mixture's components.

We note that, the viscosity deviations calculated with this model generally yield satisfactory results for many studied mixtures showing monotonous variation in viscosity values with molar fraction, but the model records inferior performance when distribution exhibits a maximum or minimum[33,34]. The following expression was proposed:

$$\eta(x_1) = \eta_2 + (\eta_1 - \eta_2)\cdot x_1^{P12,T(x1)} \quad (16)$$

Where $P_{12,T}(x_1)$ is a power polynomial with order (n) and (n+1) adjustable parameters $B_{p,T}$:

$$P_{12,T}(x_1) = \sum_{p=0}^{p=n} B_{p,T} \cdot x_1^{\,p} \quad (17)$$

Hence, the Herráez $P_{n,T}(x_1)$ polynomials of Eq.-17 can be inspected experimentally and graphically (Fig.-7) using Eq.-18.

$$P_{12,\exp,T}(x_1) = \frac{\ln\left(\dfrac{\eta_{\exp,T}(x_1) - \eta_2}{\eta_1 - \eta_2}\right)}{\ln x_1} \quad (18)$$

Where η_1 and η_2 are the dynamic viscosity of pure components (1) and (2), respectively and $\eta_{\exp,T}(x_1)$ the dynamic viscosity of mixture at molar fraction x_1 and temperature T for $[x_1 = 0,1]$.

On the other face of the model and when it is mathematically possible we can write:

$$\eta(x_2) = \eta_1 + (\eta_2 - \eta_1)\cdot x_2^{P21,T(x2)} \quad (19)$$

where $P_{21,T}(x_2)$ is a power polynomial with order (n) and (n+1) adjustable parameters $B'_{p,T}$:

$$P_{21,T}(x_2) = \sum_{p=0}^{p=n} B'_{p,T} \cdot x_2^{\,p} \quad (20)$$

Hence, the Herráez $P_{n,T}(x_2)$ polynomials of Eq.-25 can be inspected experimentally and graphically (Fig.-7) using Eq.-21.

$$P_{21,\exp,T}(x_2) = \frac{\ln\left(\dfrac{\eta_{\exp T}(x_2) - \eta_1}{\eta_2 - \eta_1}\right)}{\ln x_2} \quad (21)$$

Where η_1 and η_2 are the dynamic viscosity of pure components (1) and (2), respectively and $\eta_{\exp,T}(x_2)$ the dynamic viscosity of mixture at molar fraction x_2 and temperature T for $[x_2 = 0,1]$.

However, at infinite dilution ($x_i \rightarrow 0^+$), the $P_{\exp,T}(x_i)$ values converge to a single point ($P_{12,\exp,T}(x_1=0) = 1$ and $P_{21,\exp,T}(x_2=0) = 1.0$) independent of temperature (Fig.-7) showing a fixed value of $B_{0,T}$ or $B'_{0,T}$ constants corresponding to the first monomial of $P_{ij,T}(x_i)$ in Eqs.-17 and 20. Note that the same remark is observed in our previous works[3,27-29,32] investigating viscosity in isobutyric acid + water and 1,4-dioxane + water mixtures. We have concluded that B_0 is considered as a universal exponent characterizing the type of preponderant interaction at infinite dilution.

In the case of diluted solution of W (2) in DMOE (1), Eq.-4 can be rewritten as:

$$\eta = \eta_1(1+b_1x_2^{1/2} + b_2x_2 + \cdots) \quad (22)$$

Hence, in the basic of the extended conformal solution (ECS) theory[51,52], the $Q_{\exp,T}(x_1=1)$ is the regular viscosity term and denoted η_{21}[35,36,51,52]. Therefore, we can deduce the b_2 coefficient for a nonelectrolyte binary solution as given by Nakagawa[37] as:

$$b_2 = [(\eta_2 - \eta_1) + \eta_{21}]/\eta_1 \quad (23)$$

Where η_{21} is the famous reduced Redlich-Kister function $Q_{exp,\eta,T}(x_1=1)$; the subscripts (1) and (2) denote (DMOE) and W, respectively. The b_2 coefficient may be divided in the two parts: the first B_{id} which is based on the contribution of ideal mixture (term in parenthesis in Eq.-23), and the second B_n coefficient is based on the net interaction between the solute (W) and the solvent (DMOE)[36,37].
Considering the Eqs.-16 to 18 and the limiting expansion of Eq.-4 we can write the Herráez polynomial $P_{n,T}(x_1)$ in a limiting expansion at low concentrations of DMOE in water:

$$P_{12,\exp,T}(x_1) = 1 + \frac{\ln[\frac{\eta_2}{\eta_1-\eta_2}\cdot b_1]+o(x_1)}{\ln x_1} \tag{24}$$

We can easily justify the limit value of $P_{n,T}(x_1)$ at infinite dilution in water as: ($\lim_{x_1\to 0^+} P_{n,T}(x_1) = 1$) and we can conclude that the first monomial of $P_{n,T}(x_1)$ is a fixed value independent of temperature and equal to 1 for the dynamic viscosity η (i.e. $B_{n,0,T} = 1$) of this mixture-type. We add that the 1-value of the universal exponent B_0 leads us to affirm the absence of the $(a_1 x_1^{1/2})$-term in Eq.-4 which justifies the changes in slope observed at low mole fractions (x_1) in Fig.-2. In fact values of the experimental reduced Redlich-Kister viscosity deviation $Q_{\eta,T}(x_1)$ (Eq.-3) must diverge when ($x_1 \to 0^+$)) due to divergence of the derivative function in Eq.-5.
Nevertheless, we can add that in the case of nonelectrolyte solution when the Falkenhagen parameter[39] (solute-solute interaction) is zero or vanish small ($b_1 = 0$) and considering the Eqs.-4 and 22, we can rewrite the Eq.-21 in a new limiting asymptotic expansion at high dilution solution of W (2) in DMOE (1) or the reverse. Thus, we obtain then:

$$P_{21,\exp,T}(x_2) = 1 + \frac{\ln[\frac{\eta_1}{\eta_2-\eta_1}\cdot b_2]+o(x_2)}{\ln x_2} \tag{25}$$

from which we can conclude that: ($\lim_{xi\to 0^+} P_{\exp T}(x_i) = 1$) and at very high dilution, only the interaction solute-solvent is dominant. Note that the universal exponent B_0 of Herráez polynomial tends to the exponent of the existent term of Eq.-4.

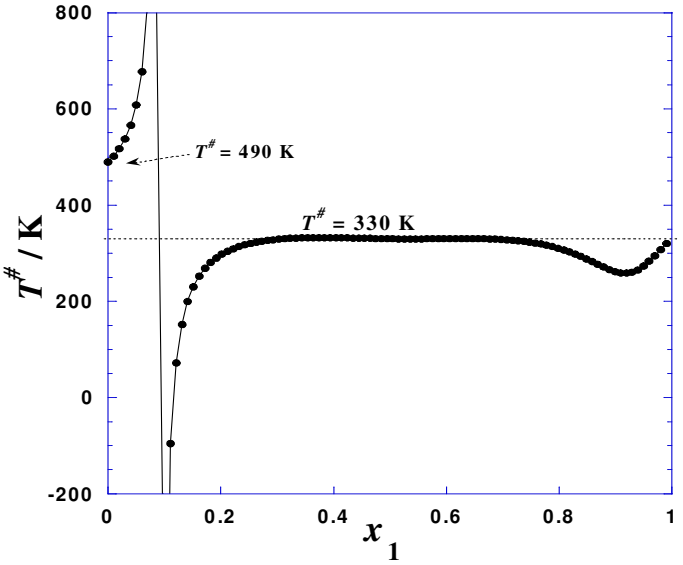

Fig.-9: Variation of the activation temperature related to the activation of viscous flow $T^{\#}$ / (K) for DMOE (1) + water (2) mixtures against mole fraction x_1 in DMOE in the temperature range (303.15 to 323.15 K).

Correlation between Arrhenius parameters

Investigation of the Gibbs free energy expression ($\Delta G^* = \Delta H^* - T\,\Delta S^*$) against the molar fraction of DMOE at different temperatures leads to estimate values of the enthalpy of activation of viscous flow ΔH^* / (kJ·mol^{-1}) and the entropy of activation of viscous flow ΔS^* / (J·mol^{-1}·K^{-1}) for DMOE (1) + W (2) mixtures against mole fraction x_1 in DMOE at the temperatures, in the temperature range (303.15 to 323.15 K)[1]. We observe two distinct behaviors clearly shown when the correlation between disorder and order is schematized through the enthalpy of activation of viscous flow ΔH^* against the entropy of activation of viscous flow ΔS^* in Fig.-8. Generally, the enthalpy of activation of viscous flow ΔH^* increases against and the entropy of activation of viscous flow ΔS^* which is also observed for some other studied binary mixtures such as water with 1.4-dioxane or N,N-dimethylacetamide (DMA)[2,6]. We note that the composition (x_1 = 0.12 ≈ 1/8) corresponds approximately to the formation of the complex r cluster (1DMOE:7W). In fact, considering the Gibbs free energy expression ($\Delta G^* = \Delta H^* - T\,\Delta S^*$) and the partial derivatives functions of Maxwell equations, we can consider the following equation at constant pressure.

$$[(\partial \Delta H^*/\partial \Delta S^*)_p = T^{\#}(x_1)] \tag{26}$$

Where $T^{\#}$ can be considered as the activation temperature related to the activation of viscous flow. Fig.-9 clearly shows the two domains separated approximately by the composition ($x_1 \approx 0.12$). In the very dilute region of DMOE in water, this temperature takes high values while in the rest of composition domain; it remains practically constant around 330 K which is near to the boiling temperatures values of the two pure components DMOE and W. We add that similar phenomenon is observed by similarity in some binary mixtures when we correlate the two viscosity Arrhenius parameters (E_a and $\ln(As)$)[2,7,53-66].

CONCLUSION

Based on experimental data of densities, viscosities and isentropic compressibilities for DMOE + water binary mixture at three different temperatures (303.15, 308.15, 313.15, 318.15 and 323.15) reported earlier, the corresponding reduced Redlich-Kister excess functions calculated from these three properties have been presented here along with a comparison of the direct excess property of earlier investigation. In addition, the relative reduced Redlich-Kister equation has been introduced to reduce the temperature effect and it also can be a good tool, like the reduced Redlich-Kister function for interpreting different types of interactions. In the present work, Arrhenius parameters of viscosity for DMOEA+W mixtures are determined at (303.15, 308.15, 313.15, 318.15 and 323.15) K. The pre-exponential entropic factor equivalent to the viscosity at infinite temperature is closely related to that of the same system in vapor-phase. Also, the Arrhenius activation energy is almost equal to the enthalpy of activation of viscous flow and it is much correlated to the vaporization enthalpy of liquid mixture. Correlation between the two Arrhenius parameters for DMOE+W mixtures can give evidence of the existence of distinct composition regions with different behaviors. Therefore, there will be a significant degree of H-bonding leading to strong correlation between the molecules; moreover, the difference in the size of the molecules can also play an important role in this respect.

Limiting excess partial molar volume at infinite dilution relative to DMOE and water in their binary mixture at the temperatures: (303.15, 308.15, 313.15, 318.15 and 323.15) K were deduced through different techniques. Among these techniques, the empirical Belda equation was successfully used in the present system. It was found that the reduced Redlich-Kister function relative to the Arrhenius activation energy which is equivalent to the apparent molar property is more sensitive than the excess property to interactions and gives more information. The Redlich–Kister polynomial is more sensitive to the number of experimental points whereas the Herráez model offers a good smoothed interpolation without any oscillation between data points, even in the case with few experimental measurements. It is observed that the viscosity deviations calculated with the Herráez model generally yield satisfactory results for many studied mixtures showing monotonous variation in viscosity values with molar fraction. In the case of some

nonelectrolyte mixtures, the net absence of solute-solute interactions leads to a fixed value of the Herráez constant of 1.0 or more.

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