Revue Roumaine de Chimie, **2006**, *51*(10), 1011–1018

EXPERIMENTAL AND CALCULATED PROPERTIES IN BINARY AND TERNARY SYSTEMS WITH WATER, 1,4-DIOXANE AND DIETHYLENE GLYCOL AT 298.15 K

Marinela CATRINCIUC,[a] Olga IULIAN,[a*] Ligia Maria OMOTA[a] and Irina NIȚĂ[b]

[a] Department of Applied Physical Chemistry and Electrochemistry, "Politehnica" University of Bucharest, 313 Splaiul Independenței, Bucharest 060042, Roumania
[b] Department of Technology and Chemical Engineering, "Ovidius" University of Constanta, 124 Mamaia Blvd., 900527, Roumania

Received February 16, 2006

The kinematic viscosities and the densities of three binary liquid mixtures: water+1,4-dioxane, water+diethylene glycol, and 1,4-dioxane+diethylene glycol, as well as of the corresponding ternary mixture water+1,4-dioxane+diethylene glycol were measured at 298.15K over the whole range of the mixture compositions. In order to see how some models in the literature work for these mixtures, the experimental data for binary and ternary mixtures were correlated by means of McAllister, Soliman, Heric, Redlich–Kister, and NRTL/*v*2 models. It could be seen that the calculated viscosities agree well with the experimental measurements. The excess properties of binary and ternary systems above mentioned were calculated and fitted by means of the Redlich–Kister's equations.

INTRODUCTION

The viscosity and density of non-electrolyte liquid mixtures are two of the most important physico-chemical properties, that are important not just as transport and volumetric properties, but also because they allow some considerations about the internal organization of the liquid phase.[1–3] This study is interesting both in order to try making a prediction of the mixture's viscosity from the properties of the pure components, and to have a better understanding of the liquid mixture behavior. For this study, the ternary system water+1,4-dioxane+diethylene glycol (DEG) was chosen, as well as the corresponding binary ones: water+1,4-dioxane, water+DEG, and 1,4-dioxane+DEG. These systems have a large industrial use; the solvent of the type water+organic component is frequently used as a reaction medium. DEG is an important intermediate for polyester resins, polyurethane, and plastics. A literature study, by our knowledge, shows that the ternary system has not been studied yet and for the binary ones, information regarding viscometric and volumetric properties is limited,[4] with the exception of the system water+1,4-dioxane.[5–7]

This paper reports the results concerning the density and viscosity of the binary and ternary systems containing water, 1,4-dioxane and DEG at 298.15K. Both the binary and ternary data were correlated by means of five models in the literature: McAllister,[8] Soliman,[9] Heric,[10] Redlich–Kister,[11] and with our model, NRTL/*v*2.[6] The excess properties for the binary and ternary mixtures were correlated by the Redlich–Kister's equations.[11]

EXPERIMENTAL

The kinematic viscosities and the densities of the pure liquids and of their mixtures were experimentally measured at 298.15K for the entire range of the composition. The kinematic viscosity (v) was measured using an Ubbelohde viscometer, and the density (ρ) by conventional pycnometric method. The viscometer was calibrated using triple-distilled water and pure solvents. The time of fall always exceeded 60 seconds; the accuracy of the measurement of the time was 0.1 seconds. Consequently, the errors of the measured values of the viscosity may be estimated as less as 1%. All measurements were done in a thermostat maintained at $T \pm 0.05$K. The used substances were purified by distillation. Analytical-reagent-grade 1,4-dioxane from Merck (Germany) was distilled at 374.15K

* Corresponding author: o_iulian@chim.upb.ro

to collect the middle fractions; the water was distilled twice and DEG, from Merck, was distilled at 473.15K in vacuum (around 3.87KPa). The purity of the materials was checked by means of gas chromatographic analysis. The comparison of the values of the density and dynamic viscosity of the used materials with values from the literature is presented in Tab.1. The mixtures were prepared volumetrically. The accuracy of the mole fraction was estimated at 0.002. All mixtures were completely miscible over the whole composition range.

Table 1
Comparison of the experimental densities and dynamic viscosities of pure liquids with the literature values at 298.15K

Component	ρ, g·cm^{-3}		η, mPa·s	
	Exp.	Lit.	Exp.	Lit.
Water	0.9973	0.9973[5] 0.9972[12]	0.9166	0.891[5] 0.9471[12]
1,4-Dioxane	1.0280	1.0286[5] 1.0280[13]	1.1902	1.172[5] 1.181[16]
DEG	1.1136	1.1135[5] 1.11351[14]	28.1790	26.812[5] 30.012[14] 30.0[15]

RESULTS AND DISCUSSION

Binary systems

The experimental data, densities (ρ) and kinematic viscosities (ν), measured for all three binary mixtures are presented in Tab. 2.

Table 2
Densities and kinematic viscosities for the water (1)+DEG (2), water (1)+1,4-dioxane (2), and 1,4-dioxane (1)+DEG (2) systems at 298.15K

x_1	ρ, g·cm^{-3}	ν, mm^2·s^{-1}	x_1	ρ, g·cm^{-3}	ν, mm^2·s^{-1}
Water (1)+DEG (2)					
0.0000	1.1136	25.3044	0.8084	1.0787	6.0685
0.0762	1.1129	24.2580	0.8448	1.0701	4.6112
0.1833	1.1120	22.4509	0.8900	1.0574	3.1950
0.2744	1.1113	20.8498	0.9315	1.0401	2.0661
0.3829	1.1096	18.7185	0.9551	1.0277	1.5930
0.4722	1.1076	16.8486	0.9795	1.0125	1.1899
0.5828	1.1031	13.9060	1.0000	0.9973	0.9191
0.7040	1.0940	9.8828			
Water (1)+1,4-Dioxane (2)					
0.0000	1.0280	1.1578	0.7530	1.0368	1.9667
0.1007	1.0294	1.1789	0.8197	1.0343	1.8590
0.1995	1.0308	1.2484	0.8710	1.0298	1.7446
0.3422	1.0328	1.4483	0.9130	1.0234	1.5239
0.4256	1.0340	1.5960	0.9470	1.0158	1.3248
0.5379	1.0357	1.7823	0.9760	1.0068	1.0881
0.6640	1.0372	1.9677	1.0000	0.9973	0.9191
1,4-Dioxane (1)+DEG (2)					
0.0000	1.1136	25.3044	0.6252	1.0634	3.2312
0.1106	1.1056	17.2011	0.7211	1.0547	2.5173
0.2175	1.0972	12.2066	0.8167	1.0460	2.0104
0.3223	1.0887	8.6008	0.9109	1.0364	1.5415
0.4254	1.0810	6.0688	1.0000	1.0280	1.1578
0.5269	1.0715	4.3518			

The experimental data were correlated with McAllister, Soliman (a modified form of McAllister equation), Heric, Redlich–Kister, and NRTL/*v*2 models, represented by eqs. (1)-(5), respectively. The obtained results for each of the three binary systems are presented in Tab. 3. Also, for a better understanding of the behavior of these systems, the excess thermodynamic functions (excess molar volume and excess viscosity) were calculated and correlated with the Redlich–Kister equation and graphically represented, as shown in Fig. 1(a) and (b).

$$\ln \nu = x_1^3 \ln \nu_1 + 3x_1^2 x_2 \ln \nu_{12} + 3x_1 x_2^2 \ln \nu_{21} + x_2^3 \ln \nu_2 - \ln\left(x_1 M_1 + x_2 M_2\right) + \\ + x_1^3 \ln M_1 + 3x_1^2 x_2 \ln M_{12} + 3x_1 x_2^2 \ln M_{21} + x_2^3 \ln M_2 \tag{1}$$

$$M_{12} = \left(2M_1 + M_2\right)/3 \qquad M_{21} = \left(M_1 + 2M_2\right)/3$$

$$\ln \nu = x_1^3 \ln \nu_1 + x_2^3 \ln \nu_2 + 3x_1^2 x_2 \ln A_{12} + 3x_1 x_2^2 \ln A_{21} + \frac{x_1 x_2 B_{12}\left(x_1 + x_2\right)^3}{\left(\frac{M_1}{M_2}\right)^2 x_1 + x_2} \tag{2}$$

$$\ln \nu = x_1 \ln \nu_1 + x_2 \ln \nu_2 + x_1 \ln M_1 + x_2 \ln M_2 - \ln\left(x_1 M_1 + x_2 M_2\right) + \delta_{12} \tag{3}$$

$$\delta_{12} = x_1 x_2 \left[\alpha_{12} + \alpha'_{12}\left(x_1 - x_2\right)\right]$$

$$\ln \nu = x_1 \ln\left(\nu_1 M_1\right) + x_2 \ln\left(\nu_2 M_2\right) - \ln\left(x_1 M_1 + x_2 M_2\right) + \\ + x_1 x_2 A_o + x_1 x_2\left(x_1 - x_2\right) A_1 + x_1 x_2\left(x_1 - x_2\right)^2 A_2 + x_1 x_2\left(x_1 - x_2\right)^3 A_3 \tag{4}$$

$$\ln \nu = x_1 \ln\left(\nu_1 M_1\right) + x_2 \ln\left(\nu_2 M_2\right) - \ln\left(x_1 M_1 + x_2 M_2\right) + \\ + x_1 x_2 \left(\frac{\tau_{21}\theta_{21}}{x_1 + x_2\theta_{21}} + \frac{\tau_{12}\theta_{12}}{x_2 + x_1\theta_{12}}\right) \tag{5}$$

$$\theta_{12} = \exp \tau_{12} \qquad \theta_{21} = \exp \tau_{21} \qquad g_{12} - g_{22} = \tau_{12} RT \qquad g_{21} - g_{11} = \tau_{21} RT$$

x_i is the mole fraction of component i in the mixture; ν is the kinematic viscosity of the binary mixture; M_i, ν_i are the molar mass and kinematic viscosities, respectively, of the pure components; ν_{ij} are McAllister's parameter; A_{ij} , B_{ij} , Soliman's parameters; α_{ij} , α'_{ij} , Heric's parameters; A_i , Redlich's parameters and g_{ij}-g_{ii} are parameters in NRTL/*v*2 model.

The excess molar volumes (V^E) and the excess viscosity (or viscosity deviations, $\Delta\nu$) were computed through the equations(6)-(7) and fitted with Redlich-Kister equation (8):

$$V^E = \sum_{i=1}^{N} X_i M_i \left(\rho^{-1} - \rho_i^{-1}\right) \tag{6}$$

$$\Delta\nu = \nu - \sum_{i=1}^{N} \nu_i x_i \tag{7}$$

$$Y^E = x_i x_j \sum_{k=0}^{p} A_k \left(x_j - x_i\right)^k \tag{8}$$

where ρ and ν are the density and kinematic viscosity, respectively, of the of the binary or ternary mixture; M_i ,ρ_i , ν_i are the molar mass, density and kinematic viscosities of the pure components, respectively, Y^E is the excess property and N represents the number of components in the mixture. For all correlative and predictive calculations for binary and ternary systems, the standard deviations were computed using the following equation:

$$\sigma = \left[\frac{\sum\left(Y_{\exp} - Y_{calc}\right)^2}{N_{\exp} - N_{par}}\right]^{0.5} \tag{9}$$

where Y_{exp} is the experimental property or the experimental excess property, Y_{calc}, the calculated property or the calculated excess property, A_k, Redlich's parameters, N_{exp}, the number of experimental data and N_{par} is the number of parameters model.

Generally, the binary systems are well represented by the utilized models, exception being the water+DEG system, which gives higher deviations. At the same time, it could be noticed that McAllister and Heric models give the same deviations, meaning that it could be used either one of them for the correlation of these binary systems.

The excess functions represent a measure of the non-ideality of the liquid mixture that is due to a various factors as structural, geometric factors, or to the interactions in mixtures. In Fig. 1(a) all the values for the excess molar volumes are negative. From Fig. 1(b) it could be noticed that each system has a different behavior regarding the deviation in viscosity: the water+1,4-dioxane system presents positive viscosity deviation, the water+DEG system, positive and negative and 1,4-dioxane+DEG system, the most negative viscosity deviation. Generally, the first two systems have a normal behavior: negative V^E and an opposite sign for the viscosity deviation, but the third system, the system 1,4-dioxane+DEG, presents a particularity, having the both excess functions negative.

This complex behavior of the different systems is the result of the structure and interactions between the components upon mixing. Water is a polar protic solvent (dipole moment μ=1.85D) with small size, which is able to support extensive hydrogen-bonding networks.[9] The diethylene glycol itself, as well as glycol mixed with water, can also associate by hydrogen bonding; it has a high dipole moment (μ=2.31D). Dioxane (μ=0.4D) is a nonpolar aprotic solvent, but is able to associate by hydrogen bonding, due to its etheric oxygens.

Consequently, the positive Δv values indicate specific interactions between water and 1,4-dioxane molecules, according to the literature,[5] and between water and DEG molecules for the most concentrations. The significantly large negative values of Δv indicate much weaker interactions between DEG and 1,4-dioxane molecules than in the DEG+water system, and the difference in size and shape of unlike molecules, 1,4-dioxane and DEG. The trends of curves and the values of V^E and Δv at 298.15 K for the binary systems water+1,4-dioxane and water+DEG, correspond well with those in literature.[5]

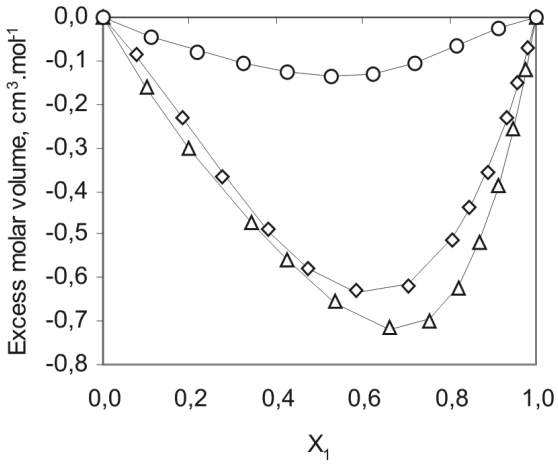
Table 3
Parameters and standard deviations of McAllister, Soliman, Heric, Redlich–Kister, and NRTL/*v*2 models for the binary systems at 298.15K

Model	Model Parameters				$\sigma^2 \cdot 10^2$
Water+1,4-Dioxane					
McAllister	ν_{12}	ν_{21}			1.17
	8.4521	0.7142			
Soliman	A_{12}	A_{21}	B_{12}		0.08
	2.9577	0.8774	0.4012		
Heric	α_{12}	α'_{12}			1.17
	3.3886	3.9442			
Redlich–Kister	A_o	A_1	A_2	A_3	0.05
	3.1690	2.8425	1.8877	1.9900	
NRTL/ν2	$g_{12}-g_{22}$ (cal·mol^{-1})	$g_{21}-g_{11}$ (cal·mol^{-1})			0.30
	-474.9141	4744.3783			
Water+DEG					
McAllister	ν_{12}	ν_{21}			57.19
	54.4942	16.0521			
Soliman	A_{12}	A_{21}	B_{12}		11.73
	17.2554	14.7516	0.6564		
Heric	α_{12}	α'_{12}			57.19
	6.4056	3.6543			
Redlich–Kister	A_o	A_1	A_2	A_3	0.26
	6.2372	4.3274	2.5381	0.6957	
NRTL/ν2	$g_{12}-g_{22}$ (cal·mol^{-1})	$g_{21}-g_{11}$ (cal·mol^{-1})			92.73
	2106.186	6257.229			

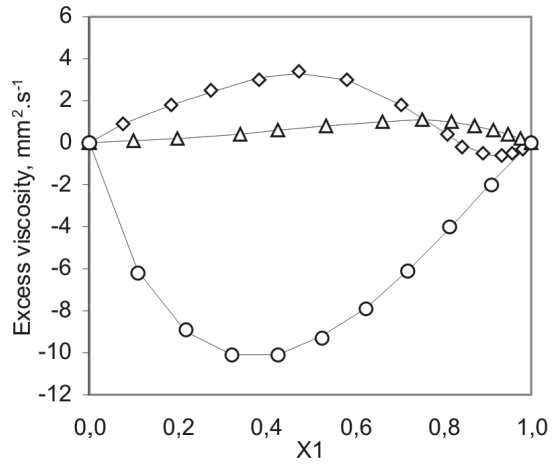
Table 3 (continues)

Table 3 (continued)

1,4-Dioxane+DEG					
McAllister	ν_{12}	ν_{21}			0.80
	2.7131	8.0201			
Soliman	A_{12}	A_{21}	B_{12}		0.90
	1.0842	4.1683	1.9839		
Heric	α_{12}	α'_{12}			0.80
	-0.4347	-0.0833			
Redlich–Kister	A_o	A_1	A_2	A_3	0.03
	-0.5155	-0.3297	0.7031	1.2739	
NRTL/ν 2	$g_{12}-g_{22}$ (cal·mol^{-1})	$g_{21}-g_{11}$ (cal·mol^{-1})			0.80
	-1222.0217	-148.3377			



(a)



(b)

Fig. 1 – (a) Excess molar volume versus composition; (b) Excess viscosity versus composition; (◊) water (1)+DEG (2); (Δ) water (1)+1,4-dioxane (2); (o) 1,4-dioxane (1)+DEG (2); (—)Redlich–Kister's equation

Ternary system

Experimental densities and dynamic viscosities, measured at 298.15K, are listed in Tab. 4. The experimental data for the ternary viscosity were correlated with the same five proposed models: McAllister Soliman, Heric, Redlich–Kister, and NRTL/*v*2 given by eqs. (10)-(14), respectively, for ternary systems. The respective capacities of correlation with various models were tested. The correlation results are gathered in Tab. 5: the values of the models parameters and of standard deviations. The standard deviation was computed using the equation (9).

$$\begin{aligned}\ln\nu = {} & x_1^3\ln\left(\nu_1 M_1\right)+x_2^3\ln\left(\nu_2 M_2\right)+x_3^3\ln\left(\nu_3 M_3\right)+\\ & +3x_1^2x_2\ln\left(\nu_{12}M_{12}\right)+3x_1x_2^2\ln\left(\nu_{21}M_{21}\right)+3x_1^2x_3\ln\left(\nu_{13}M_{13}\right)+\\ & +3x_1x_3^2\ln\left(\nu_{31}M_{31}\right)+3x_2^2x_3\ln\left(\nu_{23}M_{23}\right)+3x_2x_3^2\ln\left(\nu_{32}M_{32}\right)-\\ & -\ln\left(x_1M_1+x_2M_2+x_3M_3\right)+x_1x_2x_3\ln\left(\nu_{123}M_{123}\right)\end{aligned} \tag{10}$$

$$M_{12}=\left(2M_1+M_2\right)/3 \quad M_{21}=\left(M_1+2M_2\right)/3 \quad M_{13}=\left(2M_1+M_3\right)/3,\ M_{31}=\left(M_1+2M_3\right)/3$$

$$M_{23}=\left(2M_2+M_3\right)/3 \quad M_{32}=\left(M_2+2M_3\right)/3 \quad M_{123}=\left(M_1+M_2+M_3\right)/3$$

$$\begin{aligned}\ln\nu = {} & x_1^3\ln\nu_1+x_2^3\ln\nu_2+x_3^3\ln\nu_3+3x_1^2x_2\ln A_{12}+3x_1^2x_3\ln A_{13}+3x_2^2x_3\ln A_{23}+\\ & +\frac{x_1x_2B_{12}\left(x_1+x_2\right)^3}{\left(\frac{M_1}{M_2}\right)^2x_1+x_2}+\frac{x_1x_3B_{13}\left(x_1+x_3\right)^3}{\left(\frac{M_1}{M_3}\right)^2x_1+x_3}+\frac{x_2x_3B_{23}\left(x_2+x_3\right)^3}{\left(\frac{M_2}{M_3}\right)^2x_2+x_3}+6x_1x_2x_3\ln C_{123}\end{aligned} \tag{11}$$

$$\begin{aligned}\ln\nu = {} & x_1\ln\nu_1+x_2\ln\nu_2+x_3\ln\nu_3+x_1\ln M_1+x_2\ln M_2+x_3\ln M_3-\\ & -\ln\left(x_1M_1+x_2M_2+x_3M_3\right)+\delta_{123}\end{aligned} \tag{12}$$

$$\begin{aligned}\delta_{123} = {} & x_1x_2\left[\beta_{12}+\beta'_{12}\left(x_1-x_2\right)\right]+x_1x_3\left[\beta_{13}+\beta'_{13}\left(x_1-x_3\right)\right]+\\ & +x_2x_3\left[\beta_{23}+\beta'_{23}\left(x_2-x_3\right)\right]+x_1x_2x_3\alpha_{123}\end{aligned}$$

$$\begin{aligned}\ln\nu = {} & x_1\ln\left(\nu_1M_1\right)+x_2\ln\left(\nu_2M_2\right)+x_3\ln\left(\nu_3M_3\right)-\ln\left(x_1M_1+x_2M_2+x_3M_3\right)+\\ & +x_1x_2\left[A_o+A_1\left(x_1-x_2\right)+A_2\left(x_1-x_2\right)^2+A_3\left(x_1-x_2\right)^3\right]+\\ & +x_2x_3\left[B_o+B_1\left(x_2-x_3\right)+B_2\left(x_2-x_3\right)^2+B_3\left(x_2-x_3\right)^3\right]+\\ & +x_3x_1\left[C_o+C_1\left(x_3-x_1\right)+C_2\left(x_3-x_1\right)^2+C_3\left(x_3-x_1\right)^3\right]+x_1x_2x_3\gamma\end{aligned} \tag{13}$$

$$\begin{aligned}\ln\nu = {} & x_1\ln\left(\nu_1M_1\right)+x_2\ln\left(\nu_2M_2\right)+x_3\ln\left(\nu_3M_3\right)-\\ & -\ln\left(x_1M_1+x_2M_2+x_3M_3\right)+x_1x_2\left(\frac{\tau_{21}\theta_{21}}{x_1+x_2\theta_{21}}+\frac{\tau_{12}\theta_{12}}{x_2+x_1\theta_{12}}\right)+\\ & +x_1x_3\left(\frac{\tau_{31}\theta_{31}}{x_1+x_3\theta_{31}}+\frac{\tau_{13}\theta_{13}}{x_3+x_1\theta_{13}}\right)+x_2x_3\left(\frac{\tau_{32}\theta_{32}}{x_2+x_3\theta_{32}}+\frac{\tau_{23}\theta_{23}}{x_3+x_2\theta_{23}}\right)\end{aligned} \tag{14}$$

ν_{ij} are McAllister's parameters; A_{ij}, Bij, C_{ijk}, Soliman's parameters; $\beta_{ij,}$, β'_{ij}, α_{ijk}, Heric's parameters; A_i , B_i, C_i, and γ, Redlich's parameters and g_{ij}-$g_{ii,}$, parameters in NRTL/*v*2 model.

The comparison of the equations used for fitting the ternary experimental data for viscosity shows that Soliman's model leads to the best results. This equation is more complex than that the others. The correlation capability of the equation NRTL/*v*2 could be improved by the addition of a ternary parameter.

Table 4
Densities and viscosities for the water (1)+1,4-dioxane (2)+DEG (3) system at 298.15K

x_1	x_2	ρ, g·cm^{-3}	ν, mm^2·s^{-1}	x_1	x_2	ρ, g·cm^{-3}	ν, mm^2·s^{-1}
0.1574	0.4213	1.0768	5.0858	0.6993	0.2100	1.0589	3.1128
0.3031	0.3521	1.0761	5.1297	0.8498	0.1044	1.0471	2.3642
0.4879	0.2576	1.0757	5.0968	0.1446	0.2557	1.0930	9.5229
0.6981	0.1511	1.0707	4.2984	0.2960	0.2106	1.0921	9.1836
0.8495	0.0751	1.0546	2.7904	0.5008	0.1496	1.0899	8.2395
0.1525	0.7157	1.0458	1.8034	0.7006	0.0895	1.0833	5.9745
0.3009	0.5891	1.0469	1.9860	0.8501	0.0450	1.0628	3.3183
0.5004	0.4178	1.0500	2.3643	0.1465	0.1315	1.1043	14.9810
0.7000	0.2503	1.0497	2.4780	0.2993	0.1048	1.1025	14.0588
0.8499	0.1249	1.0414	2.0928	0.5010	0.0744	1.0997	11.5179
0.1546	0.5914	1.0586	2.7335	0.6015	0.2454	1.0666	3.7963
0.2987	0.4889	1.0612	3.0090	0.8492	0.0206	1.0685	3.8074
0.5008	0.3484	1.0608	3.1876				

Table 5
Parameters and standard deviations for McAllister, Soliman, Heric, Redlich–Kister, and NRTL/*v*2 models for the water (1)+1,4-dioxane (2)+DEG (3) ternary system at 298.15K

Model	Model Parameters							$\sigma^2\cdot10^2$
McAllister	ν_{12}	ν_{21}	ν_{13}	ν_{31}	ν_{23}	ν_{32}	ν_{123}	59.97
	17.2284	1.3752	106.1931	24.5309	16.3180	30.8535	99.4240	
Soliman	A_{12}	A_{21}	A_{13}	A_{31}	A_{23}			0.42
	2.3547	0.9236	20.3490	16.9305	1.5796			
	A_{32}	B_{12}	B_{13}	B_{23}	C_{123}			
	7.9077	0.3946	0.3941	0.9518	5.5026			
Heric	β_{12}	β'_{12}	β_{13}	β'_{13}	β_{23}	β'_{23}	α_{123}	7.69
	3.3149	4.2959	6.4844	3.7482	1.0143	-0.5952	-11.7442	
Redlich–Kister	A_o	A_1	A_2	A_3	B_o	B_1	B_2	0.28
	2.8740	1.8345	2.1765	2.9661	-0.1185	-0.6981	0.4873	
	B_3	C_o	C_1	C_2	C_3	γ		
	-3.8870	6.1306	-3.3031	0.5851	-3.7917	-5.1607		
NRTL/ν2	$g_{12}-g_{22}$ (cal·mol^{-1})	$g_{21}-g_{11}$ (cal·mol^{-1})	$g_{13}-g_{33}$ (cal·mol^{-1})	$g_{31}-g_{11}$ (cal·mol^{-1})	$g_{23}-g_{33}$ (cal·mol^{-1})	$g_{32}-g_{22}$ (cal·mol^{-1})		14.90
	-1026.46	1283.89	2485.21	6067.41	-3600.52	-2517.71		

The excess properties of ternary system were calculated with eqs. (6) and (7) and correlated with Redlich–Kister type equation. The isolines at 298.15K at constant values of V^E and Δv were drawn in Fig. 2 and Fig. 3, respectively. The values are negative for all excess volumes and for majority of viscosity deviations.

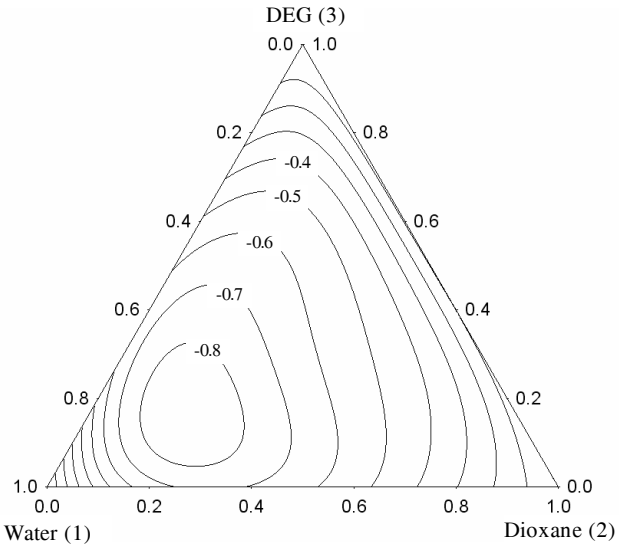


Fig. 2 – Isolines at constant V^E for the ternary system water (1)+1,4-dioxane (2)+DEG (3) at 298.15K

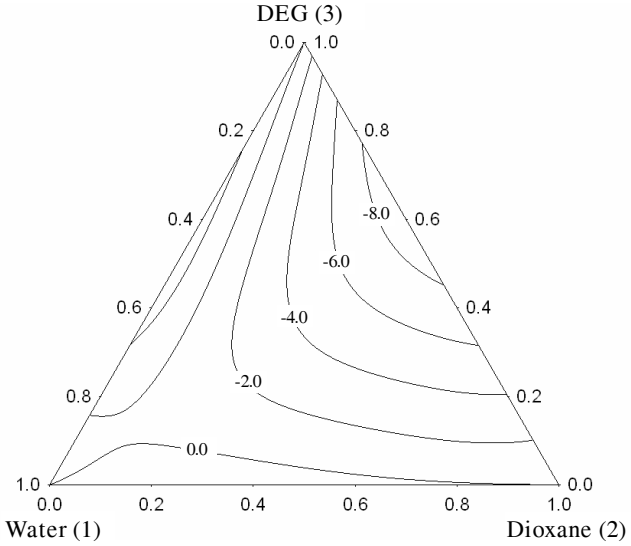


Fig. 3 – Isolines at constant Δv for the ternary system water (1)+1,4-dioxane (2)+DEG (3) at 298.15K

CONCLUSIONS

Densities and viscosities of binary and ternary systems with water, 1,4-dioxane, and DEG were experimentally measured at 298.15K and correlated by means of McAllister, Soliman, Heric, Redlich–Kister and NRTL/*v*2 models. Generally, the binary systems are well represented by the utilized models. For the ternary system, Soliman's model leads to the best results. Excess functions have been calculated and fitted to the Redlich-Kister equation. For binary and ternary systems, the excess molar volumes are negative and the deviations in viscosity are positive and negative. The systems present important deviations from ideality, as a consequence of their structures and interactions.

REFERENCES

1. B. E. Poling, J. M. Prausnitz and J. P. O'Connell**,** "The Properties of Gases and Liquids", McGraw-Hill, New York, 2001**,** p.9.77-9.90
2. S. Glasstone, K. J. Leidler and H. Eyring, "The Theory of Rate Processes", McGraw-Hill, New York, 1941
3. W. D. Monnery, A. K. Mehrota and W. Y. Svrcek, *Can. J. Chem. Eng*., **1991**, *69*, 1213–1219
4. E. A. Muller and P. Rasmussen, *J. Chem. Eng. Data*, **1991**, *36*, 214–217
5. T. Aminabhavi and B. Gopalakrishna, *J. Chem. Eng. Data*, **1995**, *40*, 856–861
6. O. Iulian, G. Jinescu, M. Iliuta and L. M. Hamplea, *Hung. J. Ind. Chem*., **1994**, *22*, 95–100
7. O. Landauer, C. Mateescu, O. Iulian and D. Geana, *Rev. Roum. Chim*., **1987**, *32*, 1129–1139
8. R. A. McAllister, *AIChE J*., **1960**, *6*, 427–431
9. K. Soliman and E. Marschall, *J. Chem. Eng. Data*, **1990**, *35*, 375–381
10. E. L. Heric, *J. Chem. Eng. Data*, **1966**, *11*, 66–70
11. O. Redlich and A. T. Kister, *Ind. Eng. Chem*., **1948**, *40*, 345–348
12. N. G. Tsierkezos and I. E. Molinou, *J. Chem. Eng. Data*, **1998**, *43*, 989–993
13. S. K. Suri and H. Naorem, *J. Chem. Eng. Data*, **1987**, *32*, 462–467
14. N. B. Sastry and M. C. Patel, *J. Chem. Eng. Data*, **2003**, *48*, 1019–1027;
15. J. A. Riddick, W. B. Bunger and T. K. Sakano, "Organic Solvents", vol.no 2, 4th ed., Wiley Interscience, New York, 1986
16. I. I. Acevedeo, G. C. Pedrosa and M. Katz, *Can. J. Chem*., **1991**, *69*, 1005–1007