

Viscosities of mixtures of 2-alkanols with tetraethyleneglycol dimethyl ether at different temperatures

S. Aznarez^a, M.M.E.F. de Ruiz Holgado^b, E.L. Arancibia^{b,*}

^a Departamento de Química, Facultad de Ingeniería, Universidad Nacional del Comahue, Buenos Aires 1400, 8300 Neuquén, Argentina

^b Departamento de Química, Facultad de Ciencias Exactas y Tecnología, Universidad Nacional de Tucumán, Av. Independencia 1800, 4000 San Miguel de Tucumán, Argentina

Received 29 April 2005; accepted 10 September 2005

Abstract

Kinematic viscosities and densities were measured over the entire range of composition and at atmospheric pressure for 2-propanol-tetraethyleneglycol dimethyl ether, 2-butanol-tetraethylene glycol dimethyl ether, and 2-pentanol-tetraethylene glycol dimethyl ether from 288.15 to 318.15 K employing pure species and solutions covering the whole range expressed by the condition $0 \leq x \leq 1$. Starting from the experimental data, the dynamic viscosity, deviation of dynamic viscosity, and the excess Gibbs energies of activation for viscous flow have been calculated. The results were fitted by the Redlich–Kister equation. Free-volume type correlation with one parameter is compared with an extension of the generalized corresponding states principle for these systems.

© 2005 Elsevier B.V. All rights reserved.

Keywords: Viscosities; Densities; Tetraethyleneglycol dimethyl ether; 2-Alkanol

1. Introduction

The industrial interest in improved non-pollutant fuels by oxygenated additive and their possible use as new working fluid pairs are responsible for an increase in experimental and theoretical studies on the thermophysical properties of ether and alkanol mixtures [1]. These mixtures represent a permanent challenge to the predictive model of thermophysical properties because of the complex behavior of self-association in alcohol molecules and cross-association between alcohol and ether molecules. In polyethers where there are various ether groups in the same molecules, the problem is most severe because of the proximity effect. There exist in the literature several works on methanol with different polyethers [2,3] and few cases of other alkanol molecules [4,5].

As part of our systematic studies on the thermophysical properties of binary liquid mixtures of polyether with alkanols, we reported the experimental results of density and viscosity at atmospheric pressure and at different temperatures for tetra-

ethyleneglycol dimethyl ether (TEGDME) and 2-propanol (2-PR), TEGDME and 2-butanol (2-BU), and TEGDME and 2-pentanol (2-PE) binary mixtures.

2. Experimental section

2-Propanol (2-PR) and 2-butanol (2-BU), Merck pro-analysis, and 2-pentanol (PE), Fluka pro-analysis, were carefully dried with molecular sieves type 4 Å. The purity of the 2-alkanols were verified by GC. Tetraethylene glycol dimethyl ether (TEGDME), Sigma >99% was used without further purification and was also kept over molecular sieves. The experimental and literature values of physical properties of pure liquids are shown in Table 1.

Kinematic viscosity (ν /cSt) ($1 \text{ cSt} = 10^{-6} \text{ m}^2 \text{ s}^{-1}$) measurement of pure liquids and their mixtures was performed using a Schott–Geräte, model AVS 400, viscosity measuring system, equipped with a series of Ubbelohde suspended-level viscosimeters covering the ($0.3 \leq \nu \leq 300$) cSt range. All the experiments were performed at least in five replicates for each composition and at each temperature and the results were averaged. The temperatures were read from a calibrated thermometer. The temperature was kept constant at $\pm 0.02 \text{ K}$.

* Corresponding author.

E-mail address: earancibia@herrera.unt.edu.ar (E.L. Arancibia).

Table 1

Comparison of density, ρ , and viscosity, η , with literature data for pure components at 298.15 K

Components	$\rho/(\text{g cm}^{-3})$		$\eta/(\text{mPa s})$	
	Exp.	Lit.	Exp.	Lit.
2-Propanol	0.7809	0.7809 ^a 0.78126 ^b 0.7809 ^c	2.089 2.0436 ^b 2.045 ^c	1.936 ^a
2-Butanol	0.8023	0.80241 ^b 0.80240 ^d	3.150	2.998 ^b
2-Pentanol	0.8047	0.8054 ^b	3.381	3.510 ^{b,*}
TEGDME	1.0059	1.0063 ^c 1.00653 ^f	3.313	3.394 ^c

^a Ref. [6].

^b Ref. [7].

^c Ref. [8].

^d Ref. [9].

^e Ref. [10].

^f Ref. [11].

* Interpolated value.

The error in the kinematic viscosity data was estimated ± 0.003 cSt.

The dynamic viscosity ($\eta/\text{mPa s}$) (1 cP = 1 mPa s) was calculated with the equation $\eta = \rho k(t - \gamma)$, where t is time, k is the viscosimeter constant, γ is the Hagenbach correction factor, η is the dynamic viscosity and ρ is the density.

A digital densimeter Anton Paar, model DMA 45, was employed for the determination of the densities of the pure components and the binary mixtures. Immediately prior to each series of measurements, doubly distilled water and air were used to calibrate the densimeter. The estimated error in the density measurement was approximately equal to $\pm 2 \cdot 10^{-4}$ g cm^{-3} . Binary mixtures were prepared using a Mettler H20T balance. The error in the mole fractions was estimated in $\pm 4 \cdot 10^{-4}$.

3. Results and discussion

In Table 2 the measurement of kinematic viscosity (ν) and density (ρ) values of the two pure species TEGDME and 2-PR, and 9 mixtures, determined at four temperatures together with

Table 2

Experimental densities (ρ) and kinematic viscosities (ν) for TEGDME(2) in 2-PR(1) binary system at different temperatures

x_2	288.15		298.15		308.15		318.15	
	ν/cSt	$\rho/\text{g cm}^{-3}$	ν/cSt	$\rho/\text{g cm}^{-3}$	ν/cSt	$\rho/\text{g cm}^{-3}$	ν/cSt	$\rho/\text{g cm}^{-3}$
0.0000	3.599	0.7891	2.675	0.7809	2.025	0.7725	1.560	0.7635
0.0994	2.810	0.8442	2.190	0.8356	1.746	0.8269	1.417	0.8178
0.2026	2.729	0.8856	2.158	0.8769	1.746	0.8681	1.441	0.8589
0.2989	2.802	0.9149	2.223	0.9060	1.810	0.8971	1.505	0.8878
0.4193	3.036	0.9445	2.397	0.9356	1.939	0.9265	1.610	0.9170
0.4967	3.201	0.9577	2.508	0.9486	2.038	0.9396	1.697	0.9301
0.5969	3.430	0.9735	2.659	0.9647	2.218	0.9555	1.800	0.9462
0.6987	3.626	0.9871	2.856	0.9777	2.364	0.9685	1.906	0.9589
0.7924	3.828	0.9977	3.033	0.9882	2.487	0.9787	2.026	0.9697
0.9026	4.079	1.0074	3.160	0.9976	2.613	0.9878	2.130	0.9790
1.0000	4.257	1.0166	3.294	1.0059	2.742	0.9960	2.245	0.9875

Table 3

Experimental densities (ρ) and kinematic viscosities (ν) for TEGDME(2) in 2-BU(1) binary system at different temperatures

x_2	288.15		298.15		308.15		318.15	
	ν/cSt	$\rho/\text{g cm}^{-3}$	ν/cSt	$\rho/\text{g cm}^{-3}$	ν/cSt	$\rho/\text{g cm}^{-3}$	ν/cSt	$\rho/\text{g cm}^{-3}$
0.0000	5.789	0.8103	3.926	0.8023	2.745	0.7939	1.995	0.7851
0.0988	3.709	0.8520	2.771	0.8437	2.128	0.8351	1.656	0.8263
0.1979	3.263	0.8855	2.513	0.8769	1.989	0.8682	1.612	0.8593
0.3132	3.199	0.9167	2.497	0.9078	2.005	0.8987	1.645	0.8903
0.3974	3.234	0.9354	2.534	0.9264	2.045	0.9174	1.689	0.9088
0.4961	3.355	0.9543	2.635	0.9454	2.130	0.9362	1.762	0.9278
0.5941	3.537	0.9705	2.780	0.9613	2.236	0.9521	1.849	0.9438
0.6954	3.732	0.9848	2.915	0.9756	2.354	0.9664	1.947	0.9581
0.7959	3.913	0.9972	3.058	0.9877	2.467	0.9783	2.039	0.9701
0.8988	4.189	1.0078	3.209	0.9980	2.613	0.9886	2.160	0.9804
1.0000	4.257	1.0166	3.294	1.0059	2.742	0.9960	2.245	0.9875

the binary composition are reported. Tables 3 and 4 show the same properties for the TEGDME+2-BU and for the TEGDME+2-PE binary systems, respectively.

In the literature [12], different correlation functions of the general form $\nu = \nu(x_i)$ have been used in order to investigate the relation to model the kinematic viscosity of binary liquids as a property of pure species. The trend is non-linear and a polynomial expansion of the type:

$$\ln \nu = \sum_0^i a_i x_i^i / \sum_{i=1}^i (1 + b_i x_i^i) \quad (1)$$

has been employed to fit the isothermal experimental kinematic viscosity data. The coefficients a_i and b_i for the 2-PR(1)+TEGDME(2), 2-BU(1)+TEGDME(2), and 2-PE(1)+TEGDME(2) binary systems are listed in Table 5 together with the average absolute percent deviations (AAD%) evaluated following Baylaucq et al. [13].

$$\text{AAD}\% = \frac{100}{n} \sum_n |1 - \nu_{\text{calcd}}/\nu_{\text{exptl}}| \quad (2)$$

where “calcd” and “exptl” mean calculated and experimental values and n is the number of experimental points.

Table 4

Experimental densities (ρ) and kinematic viscosities (ν) for TEGDME(2) in 2-PE(1) binary system at different temperatures

x_2	288.15		298.15		308.15		318.15	
	ν/cSt	$\rho/\text{g cm}^{-3}$	ν/cSt	$\rho/\text{g cm}^{-3}$	ν/cSt	$\rho/\text{g cm}^{-3}$	ν/cSt	$\rho/\text{g cm}^{-3}$
0.0000	6.249	0.8128	4.202	0.8047	2.938	0.7966	2.125	0.7881
0.1008	4.098	0.8492	3.032	0.8411	2.294	0.83270	1.796	0.8242
0.1530	3.709	0.8649	2.812	0.8566	2.177	0.84820	1.751	0.8396
0.2024	3.464	0.8791	2.653	0.8708	2.089	0.8623	1.686	0.8537
0.3053	3.266	0.9055	2.546	0.8970	2.068	0.8885	1.668	0.8796
0.3916	3.337	0.9249	2.592	0.9163	2.082	0.9076	1.716	0.8987
0.5015	3.357	0.9463	2.643	0.9375	2.137	0.9287	1.773	0.9198
0.6045	3.476	0.9639	2.785	0.9549	2.249	0.9460	1.842	0.9371
0.6974	3.678	0.9781	2.850	0.9691	2.306	0.9601	1.918	0.9511
0.7934	3.807	0.9913	2.991	0.9822	2.421	0.9732	2.009	0.9640
0.8903	3.996	1.0034	3.136	0.9943	2.541	0.9852	2.113	0.9761
1.0000	4.257	1.0166	3.294	1.0059	2.742	0.9960	2.245	0.9875

Table 5
Coefficients, standard deviations and AAD% of Eq. (1) for 2-PR(1)+TEGDME(2), 2-BU(1)+TEGDME(2), and 2-PE(1)+TEGDME(2) systems

	a_1	a_2	a_3	b_1	b_2	AAAD%
<i>2-Propanol TEGDME system</i>						
288.15	1.281	2.130	15.150	5.261	6.544	0.21
	$\pm 4 \cdot 10^{-3}$	± 0.737	± 0.619	± 0.786	± 0.459	
298.15	0.984	1.431	12.070	5.306	5.817	0.30
	$\pm 4 \cdot 10^{-3}$	± 0.708	± 0.637	± 1.017	± 0.512	
308.15	0.706	7.104	11.730	18.000		1.53
	± 0.017	± 3.608	± 4.666	± 8.115		
318.15	0.444	0.583	7.492	6.196	3.342	0.32
	$\pm 3 \cdot 10^{-3}$	± 0.343	± 0.667	± 1.268	± 0.338	
<i>2-Butanol TEGDME system</i>						
288.15	1.756	2.712	13.340	5.968	5.218	0.37
	$\pm 9 \cdot 10^{-3}$	± 1.523	± 2.242	± 1.260	± 1.696	
298.15	1.369	6.103	6.193	10.260		0.87
	± 0.013	± 1.220	± 0.573	± 1.479		
308.15	1.010	1.698	6.901	6.362	2.159	0.19
	$\pm 2.4 \cdot 10^{-3}$	± 0.312	± 0.291	± 0.464	± 0.390	
318.15	0.690	1.477	6.413	7.693	1.865	0.34
	$\pm 3.6 \cdot 10^{-3}$	± 0.489	± 0.351	± 1.107	± 0.668	
<i>2-Pentanol TEGDME system</i>						
288.15	1.834	3.739	4.691	6.047		0.49
	$\pm 9 \cdot 10^{-3}$	± 0.500	± 0.165	± 0.445		
298.15	1.437	2.638	4.118	5.788		0.55
	$\pm 9 \cdot 10^{-3}$	± 0.431	± 0.179	± 0.498		
308.15	1.079	1.983	3.731	5.878		0.46
	$\pm 6 \cdot 10^{-3}$	± 0.265	± 0.148	± 0.415		
318.15	0.755	0.887	3.131	4.960		0.84
	$\pm 8 \cdot 10^{-3}$	± 0.253	± 0.233	± 0.606		

When the viscosity data are plotted against the composition at given temperature and pressure, some liquid mixtures have no maximum or minimum viscosity, some have either a maximum or a minimum viscosity, and some have both maximum and minimum viscosities. In this case the 2-BU(1)+TEGDME(2) have a minimum viscosity and a similar behavior is found in the 2-PR(1)+TEGDME(2) and 2-PE(1)+TEGDME systems. The TEGDME+1-alkanols also present a minimum viscosity [4] and, in general, alcohol+hydrocarbon mixtures present this type of behavior [8,14,15].

In Fig. 1 we have plotted the viscosity deviation ($\Delta\eta$) for the 2-propanol+TEGDME, 2-butanol+TEGDME, and 2-pentanol+TEGDME mixtures in function of mole fraction of TEGDME at different temperatures, obtained from the following equation:

$$\Delta\eta = \eta - \sum x_i \eta_i \quad (3)$$

where η refers to the viscosity of the mixture, η_i are the properties of pure species having mole fractions x_i .

On the basis of the theory of reaction rate [16], the excess Gibbs free energies of activation for viscous flow (ΔG^{*E}) were obtained from the expression

$$\Delta G^{*E}/RT = \ln \eta V - \left(\sum_{i=1}^2 x_i \ln \eta_i V_i \right) \quad (4)$$

where V , and V_i are the molar volumes of the binary mixture and pure components.

The viscosity deviation and the excess Gibbs free energies of activation for viscous flow were fitted with a Redlich–Kister equation of the type [17].

$$X^E = x_1 x_2 \sum_0^k c_k (x_2 - x_1)^k \quad (5)$$

where X^E represents $\Delta\eta$ or ΔG^{*E} .

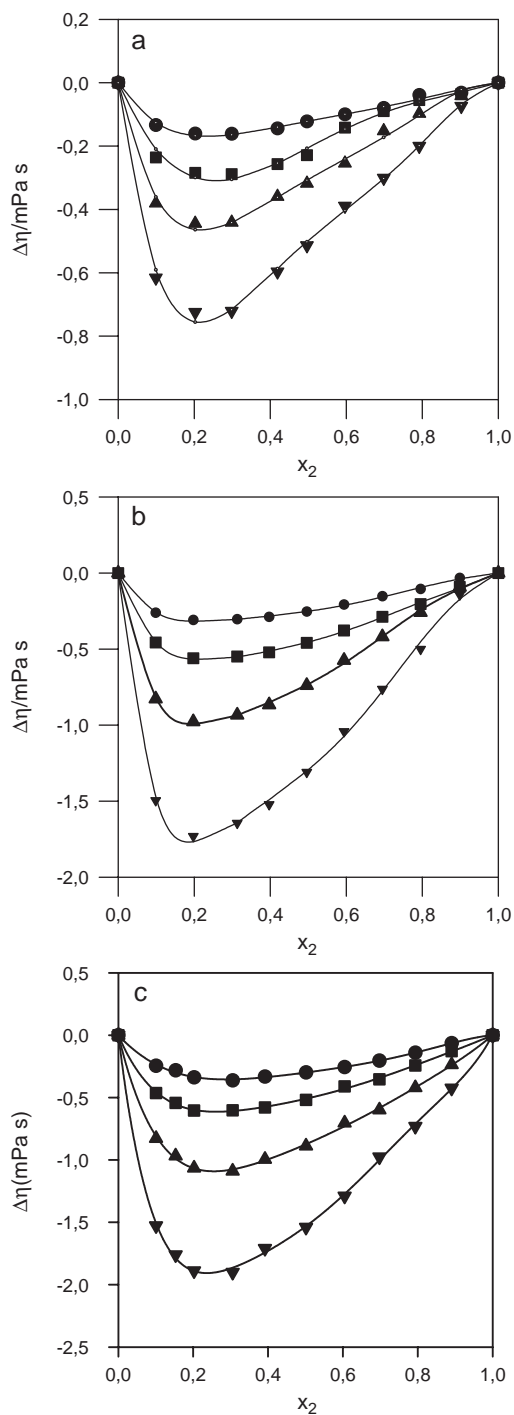


Fig. 1. (a) Viscosity deviation for the 2-propanol(1)+TEGDME(2) system at various temperatures: (▼) 288.15 K; (▲) 298.15 K; (■) 308.15 K; (●) 318.15 K; solid lines, Redlich–Kister correlations. (b) Viscosity deviation for the 2-butanol(1)+TEGDME(2) system and (c) Viscosity deviation for the 2-pentanol(1)+TEGDME(2) system at various temperatures: same notation.

The values of coefficient c_k obtained for 2-PR(1)+TEGDME(2), 2-BU(1)+TEGDME(2), and 2-PE(1)+TEGDME(2) systems are listed in Table 6 together with a standard deviation, respectively.

The viscosity deviation represents deviations from a rectilinear dependence of viscosity on mole fraction. The values of $\Delta\eta$ shown in Fig. 1(c), for the system containing 2-pentanol(1)+TEGDME(2), were asymmetrical and all negative throughout the whole concentration range at all the temperatures, with the most negative values in the alcohol rich region. A similar behaviour is observed in Fig. 1(a) and (b) for the 2-propanol+TEGDME and 2-butanol+TEGDME systems but with less negative values.

The negative values of viscosity deviations for the binary systems investigated suggest that the viscosities of associates formed between unlike molecules are relatively less than those of the pure components. Negative viscosity deviation may also occur where dispersion forces are dominant, particularly, for the systems having different molecular sizes [18,19]. We have observed such behaviour in another system studied in this laboratory for TEGDME+1-alkanols [4]. Similar to 1-alkanol+TEGDME mixtures, the values of viscosity deviation for 2-alkanol+TEGDME also increase with the chain length of 2-alkanols. However, the values of viscosity deviation for 1-alkanol mixtures are significantly higher than those mixtures involving the corresponding 2-alkanol. Thus, the values of viscosity deviation are dependent on the position of the –OH group in the alkanol molecule, indicating a different extent of molecular interactions in isomeric alkanols.

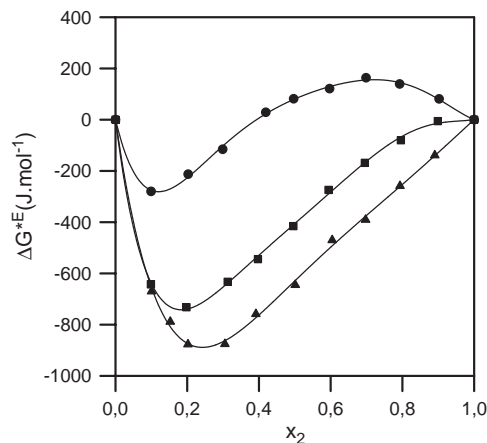


Fig. 2. Excess Gibbs energy of activation of viscous flow versus x_2 at 298.15 K: (●), 2-propanol(1)+TEGDME(2) system; (■), 2-butanol(1)+TEGDME(2) system; (▲), 2-pentanol(1)+TEGDME(2) system.

The values of ΔG^{*E} for the 2-pentanol+TEGDME, and 2-butanol+TEGDME mixtures are negative while for the 2-propanol+TEGDME mixtures, the ΔG^{*E} versus x_2 curve is sigmoid as shown in Fig. 2.

The ΔH^E values [5], for 1-alkanol+TEGDME, are positive and increase as the carbon atom number increases in the alkanol and agrees with other polyether and alkanol antecedents. The endothermicity of those mixtures shows that breaking the hydrogen bonds in the alkanol and the dipole–dipole interactions in the polyethers are energetically more

Table 6
Coefficients and standard deviations of Eq. (5) for 2-PR(1)+TEGDME(2), 2-BU(1)+TEGDME(2), and 2-PE(1)+TEGDME(2) systems

T (K)	Property	c_0	c_1	c_2	c_3	c_4	σ
<i>2-Propanol+ TEGDME system</i>							
288.15	$\Delta\eta$ (mPa s)	–2.030	2.072	–1.981	2.414	–1.280	0.011
	ΔG^E (J mol ^{–1})	9.2	1160.	–1444.	1684.	–891.	7.
298.15	$\Delta\eta$ (mPa s)	–1.270	1.356	–0.535	1.521	–1.788	0.007
	ΔG^E (J mol ^{–1})	322.	1214.	–907.	1973.	–2021.	7.
308.15	$\Delta\eta$ (mPa s)	–0.870	1.154	0.163	0.290	–1.841	0.006
	ΔG^E (J mol ^{–1})	629.	1259.	–138.	848.	–2517.	8.
318.15	$\Delta\eta$ (mPa s)	–0.507	0.458	–0.133	0.423	–0.758	0.004
	ΔG^E (J mol ^{–1})	1064.	433.	–369.	1099.	–1466.	6.
<i>2-Butanol+ TEGDME system</i>							
288.15	$\Delta\eta$ (mPa s)	–5.177	4.152	–2.955	7.686	–5.075	0.025
	ΔG^E (J mol ^{–1})	–2337.	2852.	–2225.	4079.	–2270.	12.
298.15	$\Delta\eta$ (mPa s)	–2.924	2.605	–1.412	4.717	–3.201	0.011
	ΔG^E (J mol ^{–1})	–1623.	2450.	–1620.	2959.	–2369.	8.
308.15	$\Delta\eta$ (mPa s)	–1.819	1.319	–0.956	1.811	–1.559	0.005
	ΔG^E (J mol ^{–1})	–1094.	1679.	–1425.	2173.	–1769.	6.
318.15	$\Delta\eta$ (mPa s)	–1.007	0.706	–0.454	1.281	–0.865	0.005
	ΔG^E (J mol ^{–1})	–395.	1096.	–878.	2160.	–1697.	8.
<i>2-Pentanol+ TEGDME system</i>							
288.15	$\Delta\eta$ (mPa s)	–6.1373	4.4098	–3.3518	5.0504	–5.5730	0.020
	ΔG^E (J mol ^{–1})	–3449.	3272.	–2580.	2307.	–2028.	16.
298.15	$\Delta\eta$ (mPa s)	–3.5036	2.6059	–2.5638	2.2712	–1.3844	0.010
	ΔG^E (J mol ^{–1})	–2519.	2696.	–2677.	1483.	–275.	13.
308.15	$\Delta\eta$ (mPa s)	–2.0420	1.3616	–1.2378	1.4798	–0.8575	0.009
	ΔG^E (J mol ^{–1})	–1726.	1874.	–1710.	1607.	–703.	11.
318.15	$\Delta\eta$ (mPa s)	–1.1996	0.7910	–0.8570	0.6773	0.3183	0.007
	ΔG^E (J mol ^{–1})	–1038.	1412.	–1520.	944.	577.	12.

important than the formation of hydrogen bonds between the unlike molecules.

The viscosity data of liquid mixtures are required in many chemical engineering calculations involving transfer processes. It is therefore of interest to test different predictive or correlation methods to describe such mixtures. Various different types of methods have been shown in the review of Monnery et al. [20]. We have tested two ways of analysis of the behavior of this type of mixture: one is the one-constant Teja and Rice method [21] based on the corresponding states principle and the another one is the free-volume type equation [22].

Liu et al. in 1991 [23] present an extension of the previous revised free-volume theory with temperature independent parameters for the calculation of viscosities for liquid mixtures. The model is used to calculate the viscosities of binary and ternary mixtures for diverse systems, including aqueous solutions. The equation to correlate viscosities of mixtures is

$$\ln\phi_m = \sum_i \theta_i \ln_i + \ln \frac{\bar{V}_m - 1}{\prod_i (\bar{V}_i - 1)^{\theta_i}} + \frac{x_1 x_2 G_{12}}{V_m R T} \quad (6)$$

where ϕ is the fluidity, the reciprocal of the viscosity and G_{12} is a binary interaction parameter. R is the gas constant, T is the temperature, and x is the mole fraction. \bar{V}_i , \bar{V}_m and θ_i are defined as follows

$$\theta_i = x_i V_i / V_m$$

$$\bar{V}_i = V_i / V_{0,i}$$

$$\bar{V}_m = V_m / V_{0m}$$

where V_0 is a characteristic parameter in the free-volume theory for the viscosity and V_m is the molar volume of the mixture. In this case we have applied the V_0 values calculated for pure components.

The mixing rule for V_{0m} is given by

$$V_{0m} = \sum_i \sum_j x_i x_j V_{0ij} \quad (7)$$

and the combining rule for V_{0ij} is

$$V_{0ij} = (1 - k_{ij}) \left(V_{0i}^{1/3} + V_{0j}^{1/3} \right)^3 / 8 \quad (8)$$

where k_{ij} is another binary interaction parameter.

Liu et al. [23] analyzed symmetric systems whose constituent molecules are similar in size and polarity where both the parameters G_{12} in expression (6) and k_{12} in expression (8) can be taken as zero. In this case, they obtain a predictive model and an overall absolute average deviation percent (AAD)% of 0.96% for binary systems. In the asymmetric polar+polar systems in the classification by Liu et al. [22], k_{12} is equal to zero. The results obtained for G_{12} and the AAD% values are included in Table 7.

Based on an extension of the generalized corresponding states principle treatment, Teja and Rice [21] propose a method for the treatment of the viscosity of liquid mixtures. We have

Table 7

Correlation results for the dynamic viscosity for 2-Propanol(1)+TEGDME(2), 2-Butanol(1)+TEGDME(2), and 2-Pentanol(1)+TEGDME(2) systems

	$G_{12} \pm \sigma(G_{12})$	AAD%	ψ_{12}	AAD%
<i>2-Propanol+TEGDME system</i>				
288.15	-0.178 ± 0.006	4.2	0.85	6.4
298.15	-0.133 ± 0.005	4.2	0.85	6.9
308.15	-0.102 ± 0.004	5.0	0.85	5.9
318.15	-0.090 ± 0.003	6.4	0.85	6.6
<i>2-Butanol+TEGDME system</i>				
288.15	-0.144 ± 0.004	2.5	0.80	6.5
298.15	-0.098 ± 0.003	2.3	0.80	8.1
308.15	-0.070 ± 0.002	1.8	0.85	8.9
318.15	-0.060 ± 0.002	2.8	0.85	7.3
<i>2-Pentanol+TEGDME system</i>				
288.15	-0.111 ± 0.003	1.9	-0.75	12.6
298.15	-0.069 ± 0.003	2.6	-0.75	10.5
308.15	-0.052 ± 0.003	2.5	-0.75	10.2
318.15	-0.044 ± 0.003	2.0	-0.75	11.7

defined reference fluids as the constituent pure substance TEGDME and 2-PR, TEGDME and 2-BU, and TEGDME and 2-PE and the simplified equation [24] was used.

$$\ln(\eta_m \varepsilon_m) = x_1 \ln(\eta_1 \varepsilon_1)^{(r_1)} + x_2 \ln(\eta_2 \varepsilon_2)^{(r_2)} \quad (9)$$

where ε_1 and ε_2 are functions of V_{ci} , T_{ci} , and M_i , and $\eta^{(r_1)}$ and $\eta^{(r_2)}$ are functions of the “characteristic” temperature T^* ($=TT_{ci}/T_{cm}$).

The rules suggested by the authors to compute mixture parameters are:

$$V_{cm} = \sum_i \sum_j x_i x_j V_{cij}$$

$$T_{cm} = \sum_i \sum_j x_i x_j T_{cij} V_{cij} / V_{cm}$$

$$V_{cij} = \left(V_{ci}^{1/3} + V_{cj}^{1/3} \right)^3 / 8$$

$$T_{cij} V_{cij} = \Psi_{ij} (T_{ci} T_{cj} V_{ci} V_{cj})^{1/2}$$

where Ψ_{ij} is an interaction parameter. The critical values V_{ci} and T_{ci} for TEGDME were obtained from Ref. [13].

The results obtained for 2-PR+TEGDME, 2-BU+TEGDME, and 2-PE+TEGDME systems from the Teja and Rice method together with the ψ_{12} interaction coefficient are included in Table 7 for comparison. In Table 7 we can see that the AAD% calculated with the free-volume type correlation with one interaction parameter proposed by Liu et al. works better than the Teja and Rice method for these systems.

4. Conclusions

The 2-PR(1)+TEGDME(2), 2-BU(1)+TEGDME(2), and 2-PE(1)+TEGDME(2) systems present a minimum in the plot of experimental viscosity values vs mole fraction of TEGDME. The minimum in viscosity for these mixtures could be

explained in terms of the rupture of the hydrogen bond of the alcohol in a similar way to hydrocarbon dissolution in alcohols.

The Liu et al. model based on the free-volume model works better than the Teja and Rice model based on the states corresponding principles when they are used to calculate the viscosity of these mixtures.

Acknowledgments

The present work was financed by a CIUNT research grant. E.L.A. is a member of the CONICET (Consejo Nacional de Investigaciones Científicas y Técnicas de la Republica Argentina).

References

- [1] I. Hatzioannidis, E. Voutsas, E. Lois, D.P. Tassios, *J. Chem. Eng. Data* 43 (1998) 386.
- [2] J. Harraiz, S. Shen, J. Fernandez, A. Coronas, *Fluid Phase Equilib.* 155 (1999) 327.
- [3] F.J. Carmona, F.J. Arroyo, I. Garcia de la Fuente, J.A. Gonzales, J.C. Cobos, *Can. J. Chem.* 77 (1999) 1608.
- [4] M.E. Ferreyra de Ruiz Holgado, C.R. de Schaefer, E.L. Arancibia, *J. Chem. Eng. Data* 47 (2002) 144.
- [5] M.E.F. de Ruiz Holgado, J. Fernandez, M.I. Paz Andrade, E.L. Arancibia, *Can. J. Chem.* 80 (5) (2002) 462.
- [6] T.M. Aminabhavi, B. Gopalakrishna, *J. Chem. Eng. Data* 40 (1995) 462.
- [7] J.A. Riddick, W.B. Bunger, T.K. Sakano, *Techniques of Chemistry, Organic Solvents, Physical Properties and Methods of Purification*, vol. II, Wiley, New York, 1986.
- [8] B. González, A. Domínguez, J. Tojo, *J. Chem. Thermodyn.* 35 (2003) 939.
- [9] C. Lafuente, J. Pardo, V. Rodriguez, F.M. Royo, J.S. Urieta, *J. Chem. Eng. Data* 38 (1993) 554.
- [10] A. Pal, G. Dass, A. Kumar, *J. Chem. Eng. Data* 44 (1999) 2.
- [11] F.S. Carmona, I.J. Arroyo, I. Garcia de la Fuente, J.A. Gonzales, J.C. Cobos, *Can. J. Chem.* 77 (1999) 1608.
- [12] M. Cocchi, M. Manfredini, D. Manzini, A. Marchetti, S. Sighinolfi, L. Tassi, A. Ulrici, M. Vignali, P. Zannini, *J. Mol. Liq.* 102 (2003) 309.
- [13] A. Baylaucq, M.J.P. Camuñas, C. Boned, A. Allal, J. Fernandez, *Fluid Phase Equilib.* 199 (2002) 249.
- [14] N.V. Sastry, M.K. Valand, *J. Chem. Eng. Data* 41 (1996) 1426.
- [15] C. Franjo, E. Jimenez, T.P. Iglesias, J.L. Legido, M.I. Paz Andrade, *J. Chem. Eng. Data* 40 (1995) 68.
- [16] S. Glasstone, K.J. Laidler, H. Eyring, *The Theory of Rate Process*, McGraw-Hill, New York, 1941, p. 514.
- [17] O. Redlich, A.T. Kister, *Ind. Eng. Chem.* 40 (1948) 341.
- [18] R.J. Fort, W.R. Moore, *Trans. Faraday Soc.* 62 (1966) 1112.
- [19] B. González, A. Domínguez, J. Tojo, *J. Chem. Eng. Data* 49 (2004) 1225.
- [20] W.D. Monnery, W.Y. Svrcek, A.K. Mehrotra, *Can. J. Chem.* 73 (1995) 3.
- [21] A.S. Teja, P. Rice, *Ind. Eng. Chem. Fundam.* 20 (1981) 77.
- [22] H. Liu, W. Wang, C.-H. Chang, *Ind. Eng. Chem. Res.* 30 (1991) 1617.
- [23] H. Liu, C.M. Silva, E.A. Macedo, *Fluid Phase Equilib.* 202 (2002) 89.
- [24] R.C. Reid, J.M. Prausnitz, B.E. Poling, *The Properties of Gases and Liquids*, 4th edition, McGraw Hill, N.Y., 1987, p. 479.