Articles

Densities and Viscosities of Binary Mixtures of Polyethylene Glycol 350 Monomethyl Ether with *n*-Butanol and *n*-Pentanol and Tetraethylene Glycol Dimethyl Ethers with *n*-Propanol, *n*-Butanol, and *n*-Pentanol from 278.15 K to 318.15 K

Marta E. Ferreyra de Ruiz Holgado, Cecilia R. de Schaefer, and Eleuterio L. Arancibia*

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

The density and viscosity of binary mixtures of polyethylene glycol 350 monomethyl ether with 1-butanol and 1-pentanol and tetraethylene glycol dimethyl ether with 1-propanol, 1-butanol, and 1-pentanol were measured at five different temperatures (278.15, 288.15, 298.15, 308.15, and 318.15 K) and at atmospheric pressure. The measurements were carried out over the whole range of composition. Viscosity values were used in the determination of the viscosity deviation, $\Delta\eta$, which was fitted to the Redlich−Kister equation and was compared with the predicted values obtained by the viscosity regular term.

Introduction

The physical and thermodynamic properties of polyethylene glycol ether plus polar organic molecules mixtures are interesting because they have been suggested to be used instead of saline solutions in refrigeration technology. Mixtures containing methanol + polyethylene glycol 250 dimethyl ether have been studied by Esteve[1] from this point of view, and their physical and thermodynamic properties (V^E, H^E) have been measured at 303.15 K. Ruiz Holgado et al.[2] also report physical properties (density and viscosity) of mixtures containing polyethylene glycol 350 monomethyl ether (PEGMME) with 1-propanol at different temperatures and at low and high range of PEGMME concentration. They also predict the behavior of those mixtures over all the range of concentration, starting from dilute solutions data.

In this study, the density and viscosity of binary mixtures of 1-propanol, 1-butanol, and 1-pentanol in tetraethylene glycol dimethyl ether and of 1-butanol and 1-pentanol in polyethylene glycol 350 monomethyl ether were measured over the whole concentration range at five temperatures, from 278.15 to 318.15 K. The present paper analyzes the viscosimetric behavior of the mentioned mixtures and compares the experimental viscosity values with those obtained by using the viscosity regular term defined by Nakagawa,[3] which were calculated from dynamic viscosity values of dilute solutions of polyether in alcohol taken from Ruiz Holgado's reference.[4]

Experimental Section

Materials. 1-Propanol, 1-butanol, and 1-pentanol (Merck pro-analysis) were kept over molecular sieves (4 Å), and their purities were verified by GC. Polyethylene glycol monomethyl ether (molecular weight, 350 average; PEGMME; Sigma) and tetraethylene glycol dimethyl ether (TEGDME; Aldrich >99%) were used without further purification and were also kept over molecular sieves. Experimental values of densities, ρ, and viscosities, η, for the pure liquids are compared with the published results at 298.15 K in Table 1.

Table 1. Comparison of Experimental Densities, ρ, and Viscosities, η, of Pure Liquids with Literature Values at 298.15 K

	ρ/g·cm^{-3}		η/mPa·s	
liquid	exptl	lit.	exptl	lit.
tetraethyleneglycol dimethyl ether (Aldrich >99)	1.0059 1.00653[b]	1.0063[a]	3.313	3.394[a]
polyethylene glycol 350 monomethyl ether (Sigma)	1.0850		28.43	
1-propanol	0.7994	0.7996[c]	1.977	1.943[c]
(Merck p.a.)		0.7995[d]		1.953[d]
		0.7994[e]		1.898[e]
1-butanol	0.8060	0.80575[c]	2.623	2.571[c]
(Merck p.a.)		0.8057[d]		2.593[d]
		0.8056[e]		2.524[e]
1-pentanol	0.8116	0.81080[c]	3.505	3.5128[c]
(Merck p.a.)		0.8109[d]		3.497[d]

[a] Pal et al., 1999.[5] [b] Carmona et al., 1999.[6] [c] Riddick et al., 1986.[7] [d] Shan et al., 1999.[8] [e] Rauf et al., 1983.[9]

Equipment. Kinematic viscosity measurements of pure liquids and their mixtures were made with an automatic Schott Gerate AVS 400 viscosity-measuring system equipped with series of Ubbelhode viscosimeters. In all cases, the experiments were generally performed at least in five replicates for each composition at each temperature and the results were averaged. A thermostated bath constant to 0.02 K was used, and the temperatures were read from calibrated thermometers. The experimental uncertainty in the kinematic viscosity was approximately equal to $\pm 3 \times 10^{-9}$ m^2·s^{-1}.

* To whom correspondence should be addressed. Fax: 54-0381-4363004. E-mail: earancibia@herrera.unt.edu.ar.

10.1021/je010182s CCC: $22.00
Published on Web 01/18/2002

Table 2. Experimental Densities, ρ, Dynamic Viscosities, η, and Viscosity Deviations, $\Delta\eta$, for 1-Propanol (1) + TEGDME (2)

x_2	ρ	η	$\Delta\eta$	x_2	ρ	η	$\Delta\eta$
	g·cm^{-3}	mPa·s	mPa·s		g·cm^{-3}	mPa·s	mPa·s
	T = 278.15 K				T = 288.15 K		
0.0000	0.8157	3.315	0.000	0.0000	0.8073	2.517	0.000
0.0946	0.8661	3.200	−0.304	0.0946	0.8594	2.490	−0.160
0.1998	0.9071	3.223	−0.504	0.1998	0.8985	2.482	−0.323
0.3000	0.9300	3.478	−0.474	0.3000	0.9199	2.684	−0.278
0.4069	0.9577	3.818	−0.389	0.4069	0.9483	2.936	−0.202
0.4908	0.9765	4.175	−0.245	0.4908	0.9653	3.149	−0.135
0.6025	0.9935	4.752	0.034	0.6025	0.9811	3.506	0.017
0.7005	1.0027	5.079	0.083	0.7005	0.9914	3.743	0.063
0.7971	1.0109	5.372	0.084	0.7971	0.9980	3.947	0.070
0.9256	1.0210	5.760	0.059	0.9256	1.0106	4.194	0.037
1.0000	1.0248	5.955	0.000	1.0000	1.0166	4.328	0.000
	T = 298.15 K				T = 308.15 K		
0.0000	0.7994	1.977	0.000	0.0000	0.7920	1.593	0.000
0.0946	0.8494	1.958	−0.118	0.0946	0.8410	1.598	−0.078
0.1998	0.8897	1.982	−0.210	0.1998	0.8813	1.629	−0.145
0.3000	0.9114	2.121	−0.187	0.3000	0.9030	1.741	−0.132
0.4069	0.9394	2.304	−0.135	0.4069	0.9308	1.873	−0.111
0.4908	0.9570	2.479	−0.068	0.4908	0.9482	2.001	−0.075
0.6025	0.9721	2.741	0.043	0.6025	0.9631	2.209	0.005
0.7005	0.9830	2.922	0.083	0.7005	0.9739	2.334	0.011
0.7971	0.9920	3.079	0.096	0.7971	0.9829	2.453	0.005
0.9256	1.0016	3.238	0.050	0.9256	0.9921	2.619	−0.005
1.0000	1.0059	3.313	0.000	1.0000	0.9960	2.731	0.000
	T = 318.15 K				T = 318.15 K		
0.0000	0.7850	1.295	0.000	0.6025	0.9534	1.821	0.032
0.0946	0.8324	1.308	−0.052	0.7005	0.9638	1.921	0.035
0.1998	0.8719	1.375	−0.065	0.7971	0.9730	2.018	0.031
0.3000	0.8935	1.456	−0.063	0.9256	0.9829	2.135	0.005
0.4069	0.9211	1.559	−0.051	1.0000	0.9875	2.217	0.000
0.4908	0.9382	1.666	−0.019				

Table 3. Experimental Densities, ρ, Dynamic Viscosities, η, and Viscosity Deviations, $\Delta\eta$, for 1-Butanol (1) + TEGDME (2)

x_2	ρ	η	$\Delta\eta$	x_2	ρ	η	$\Delta\eta$
	g·cm^{-3}	mPa·s	mPa·s		g·cm^{-3}	mPa·s	mPa·s
	T = 278.15 K				T = 288.15 K		
0.0000	0.8210	4.594	0.000	0.0000	0.8133	3.452	0.000
0.0955	0.8633	3.831	−0.878	0.0955	0.8551	2.896	−0.631
0.1736	0.8901	3.713	−1.093	0.1736	0.8818	2.895	−0.695
0.2991	0.9268	3.720	−1.245	0.2991	0.9170	2.882	−0.811
0.3995	0.9502	3.848	−1.247	0.3995	0.9405	2.938	−0.840
0.5021	0.9684	4.227	−1.006	0.5021	0.9586	3.146	−0.721
0.6109	0.9832	4.583	−0.800	0.6109	0.9734	3.409	−0.555
0.6798	0.9930	4.880	−0.601	0.6798	0.9834	3.610	−0.416
0.7906	1.0054	5.411	−0.229	0.7906	0.9961	3.958	−0.170
0.9101	1.0150	5.735	−0.082	0.9101	1.0062	4.181	−0.060
1.0000	1.0248	5.955	0.000	1.0000	1.0166	4.328	0.000
	T = 298.15 K				T = 308.15 K		
0.0000	0.8060	2.623	0.000	0.0000	0.7985	2.049	0.000
0.0955	0.8472	2.304	−0.378	0.0955	0.8387	1.867	−0.239
0.1736	0.8735	2.300	−0.432	0.1736	0.8651	1.853	−0.302
0.2991	0.9050	2.279	−0.534	0.2991	0.8959	1.922	−0.314
0.3995	0.9302	2.351	−0.529	0.3995	0.9208	1.942	−0.360
0.5021	0.9486	2.500	−0.449	0.5021	0.9391	2.043	−0.328
0.6109	0.9638	2.686	−0.339	0.6109	0.9543	2.169	−0.278
0.6798	0.9745	2.833	−0.241	0.6798	0.9652	2.268	−0.229
0.7906	0.9869	3.066	−0.089	0.7906	0.9783	2.459	−0.120
0.9101	0.9966	3.218	−0.026	0.9101	0.9889	2.605	−0.065
1.0000	1.0059	3.313	0.000	1.0000	0.9996	2.741	0.000
	T = 318.15 K				T = 318.15 K		
0.0000	0.7902	1.625	0.000	0.6109	0.9456	1.769	−0.195
0.0955	0.8307	1.476	−0.198	0.6798	0.9558	1.855	−0.152
0.1736	0.8569	1.482	−0.233	0.7906	0.9681	2.008	−0.069
0.2991	0.8891	1.533	−0.250	0.9101	0.9778	2.120	−0.036
0.3995	0.9131	1.566	−0.273	1.0000	0.9875	2.217	0.000
0.5021	0.9310	1.659	−0.240				

Table 4. Experimental Densities, ρ, Dynamic Viscosities, η, and Viscosity Deviation, $\Delta\eta$, for 1-Pentanol (1) + TEGDME (2)

x_2	ρ	η	$\Delta\eta$	x_2	ρ	η	$\Delta\eta$
	g·cm^{-3}	mPa·s	mPa·s		g·cm^{-3}	mPa·s	mPa·s
	T = 278.15 K				T = 288.15 K		
0.0000	0.8261	6.546	0.000	0.0000	0.8189	4.720	0.000
0.1018	0.8595	4.970	−1.514	0.1018	0.8520	3.658	−1.021
0.1500	0.8748	4.622	−1.832	0.1500	0.8669	3.444	−1.215
0.2036	0.8932	4.344	−2.077	0.2036	0.8854	3.259	−1.378
0.3306	0.9255	4.130	−2.214	0.3306	0.9169	3.060	−1.527
0.5011	0.9604	4.619	−1.623	0.5011	0.9511	3.419	−1.100
0.5994	0.9794	4.870	−1.315	0.5994	0.9702	3.592	−0.889
0.7097	0.9938	5.134	−0.987	0.7097	0.9849	3.783	−0.655
0.7984	1.0073	5.336	−0.734	0.7984	0.9980	4.008	−0.397
0.9308	1.0197	5.738	−0.256	0.9308	1.0094	4.285	−0.069
1.0000	1.0248	5.955	0.000	1.0000	1.0166	4.328	0.000
	T = 298.15 K				T = 308.15 K		
0.0000	0.8116	3.504	0.000	0.0000	0.8039	2.657	0.000
0.1018	0.8444	2.801	−0.683	0.1018	0.8367	2.169	−0.496
0.1500	0.8591	2.648	−0.826	0.1500	0.8511	2.078	−0.591
0.2036	0.8775	2.529	−0.935	0.2036	0.8696	2.007	−0.667
0.3306	0.9085	2.368	−1.071	0.3306	0.8998	1.886	−0.799
0.5011	0.9425	2.675	−0.732	0.5011	0.9338	2.143	−0.556
0.5994	0.9610	2.785	−0.603	0.5994	0.9526	2.233	−0.474
0.7097	0.9745	2.923	−0.444	0.7097	0.9671	2.364	−0.353
0.7984	0.9884	3.104	−0.246	0.7984	0.9804	2.486	−0.238
0.9308	1.0008	3.307	−0.019	0.9308	0.9919	2.648	−0.087
1.0000	1.0059	3.313	0.000	1.0000	0.9996	2.741	0.000
	T = 318.15 K				T = 318.15 K		
0.0000	0.7964	2.046	0.000	0.5994	0.9450	1.828	−0.318
0.1018	0.8287	1.700	−0.363	0.7097	0.9593	1.931	−0.235
0.1500	0.8436	1.635	−0.436	0.7984	0.9715	2.026	−0.155
0.2036	0.8615	1.596	−0.483	0.9308	0.9826	2.143	−0.062
0.3306	0.8913	1.507	−0.594	1.0000	0.9875	2.217	0.000
0.5011	0.9277	1.764	−0.365				

Dynamic viscosity was calculated with the following equation:

$$\eta = \frac{\nu}{\rho} = k(t_m - f) \qquad (1)$$

where k is the viscosimeter constant, f is the Hagenbach correction factor (both provided by the manufacturer), t_m is time, ν is the kinematic viscosity, and ρ is the density.

Densities were determined with an AP digital densimeter, model DMA 45. Calibration was carried out with air and doubly distilled water. The estimated uncertainty in the density measurement was approximately equal to $\pm 2 \times 10^{-4}$ g·cm^{-3}.

All weighings were performed on a Metler H20T balance, and the estimated uncertainty in mole fraction was $\pm 1.4 \times 10^{-4}$.

Results and Discussion

The experimental densities, dynamic viscosities, and deviations of the viscosity for five studied systems are given in Tables 2−6.

The excess volumes were calculated but not reported because they were obtained with poor density values. They are negative and close to zero at all the temperatures, for TEGDME + alcohol systems. For PEGMME + alcohol systems, they are negative and become more negative with increasing length of the alcohol hydrocarbon chain.

Experimental values of the dynamic viscosity of PEGMME + alcohol mixtures increase when the glycol derivative concentration increases. But, in the systems TEGDME + alcohol, the behavior is different; at first, the experimental dynamic viscosity values decrease to a minimum up to a given mole fraction, and then, they increase as glycol

Table 5. Experimental Densities, ρ, Dynamic Viscosities, η, and Viscosity Deviations, $\Delta\eta$, for 1-Butanol (1) + PEGMME (2)

x_2	ρ	η	$\Delta\eta$	x_2	ρ	η	$\Delta\eta$
	g·cm^{-3}	mPa·s	mPa·s		g·cm^{-3}	mPa·s	mPa·s
	T = 278.15 K				T = 288.15 K		
0.0000	0.8210	4.59	0.00	0.0000	0.8133	3.45	0.00
0.1089	0.9176	8.58	2.38	0.1089	0.9099	6.13	1.56
0.1500	0.9362	11.27	4.33	0.1500	0.9283	7.59	2.51
0.1940	0.9640	13.18	5.35	0.1940	0.9560	9.07	3.37
0.2500	0.9880	17.23	8.11	0.2500	0.9801	10.93	4.35
0.3255	1.0104	22.17	10.95	0.3255	1.0024	14.51	6.51
0.4500	1.0512	31.20	15.41	0.4500	1.0433	18.96	7.93
0.5462	1.0666	38.10	17.54	0.5462	1.0588	23.53	9.39
0.6000	1.0738	42.80	18.97	0.6000	1.0659	25.96	9.71
0.7722	1.0884	54.37	16.15	0.7722	1.0807	34.42	9.08
0.8595	1.0936	62.24	13.68	0.8595	1.0849	39.20	7.44
1.0000	1.1026	71.40	0.00	1.0000	1.0940	45.64	0.00
	T = 298.15 K				T = 308.15 K		
0.0000	0.8060	2.62	0.00	0.0000	0.7985	2.05	0.00
0.1089	0.9017	4.58	1.18	0.1089	0.8937	3.51	0.90
0.1500	0.9201	5.57	1.82	0.1500	0.9122	4.21	1.34
0.1940	0.9475	6.56	2.39	0.1940	0.9394	4.93	1.76
0.2500	0.9716	7.84	3.08	0.2500	0.9634	5.83	2.24
0.3255	0.9938	10.07	4.37	0.3255	0.9856	7.40	3.13
0.4500	1.0344	12.99	5.33	0.4500	1.0259	9.46	3.82
0.5462	1.0500	15.79	6.15	0.5462	1.0414	11.15	4.16
0.6000	1.0569	17.25	6.29	0.6000	1.0483	12.26	4.36
0.7722	1.0717	22.23	5.71	0.7722	1.0628	15.26	3.63
0.8595	1.0757	24.94	4.60	0.8595	1.0672	17.82	3.66
1.0000	1.0850	28.43	0.00	1.0000	1.0764	19.42	0.00
	T = 318.15 K				T = 318.15 K		
0.0000	0.7902	1.62	0.00	0.4500	1.0159	6.91	2.64
0.1089	0.8851	2.77	0.72	0.5462	1.0305	8.19	2.94
0.1500	0.9122	3.29	1.05	0.6000	1.0382	8.85	2.96
0.1940	0.9306	3.81	1.35	0.7722	1.0539	10.93	2.41
0.2500	0.9540	4.42	1.64	0.8595	1.0580	12.12	1.85
0.3255	0.9760	5.59	2.32	1.0000	1.0683	13.88	0.00

Table 6. Experimental Densities, ρ, Dynamic Viscosities, η, and Viscosity Deviations, $\Delta\eta$, for 1-Pentanol (1) + PEGMME (2)

x_2	ρ	η	$\Delta\eta$	x_2	ρ	η	$\Delta\eta$
	g·cm^{-3}	mPa·s	mPa·s		g·cm^{-3}	mPa·s	mPa·s
	T = 278.15 K				T = 288.15 K		
0.0000	0.8261	6.55	0.00	0.0000	0.8189	4.72	0.00
0.0953	0.9104	7.94	−0.28	0.0953	0.9028	5.69	−0.16
0.1905	0.9745	11.94	1.62	0.1905	0.9675	8.34	1.07
0.2613	1.0124	15.68	3.46	0.2613	1.0033	11.01	2.47
0.4104	1.0565	23.94	6.49	0.4104	1.0481	14.85	2.87
0.4997	1.0701	29.72	8.11	0.4997	1.0614	18.60	3.94
0.6274	1.0771	39.16	9.85	0.6274	1.0684	24.14	4.54
0.6977	1.0809	44.22	9.54	0.6977	1.0721	27.57	4.58
0.7989	1.0836	52.56	8.40	0.7989	1.0749	32.82	3.90
0.9041	1.0922	62.41	5.63	0.9041	1.0837	38.91	2.20
1.0000	1.1026	71.40	0.00	1.0000	1.0940	45.64	0.00
	T = 298.15 K				T = 308.15 K		
0.0000	0.8116	3.50	0.00	0.0000	0.8039	2.66	0.00
0.0953	0.8947	4.27	−0.00	0.0953	0.8872	3.17	−0.04
0.1905	0.9585	6.03	0.81	0.1905	0.9510	4.55	0.67
0.2613	0.9949	7.90	1.84	0.2613	0.9869	5.85	1.38
0.4104	1.0394	11.42	3.15	0.4104	1.0310	7.54	1.53
0.4997	1.0528	13.06	3.09	0.4997	1.0443	9.08	1.90
0.6274	1.0600	15.98	2.94	0.6274	1.0512	11.29	2.04
0.6977	1.0638	18.38	3.28	0.6977	1.0549	12.61	1.96
0.7989	1.0666	21.37	2.70	0.7989	1.0576	14.60	1.58
0.9041	1.0751	24.92	1.66	0.9041	1.0664	16.79	0.74
1.0000	1.0850	28.43	0.00	1.0000	1.0764	19.42	0.00
	T = 318.15 K				T = 318.15 K		
0.0000	0.7964	2.05	0.00	0.6274	1.0494	8.34	1.54
0.0953	0.8829	2.47	0.01	0.6977	1.0529	9.27	1.49
0.1905	0.9488	3.52	0.57	0.7989	1.0549	10.62	1.18
0.2613	0.9784	4.51	1.13	0.9041	1.0629	12.05	0.49
0.4104	1.0273	5.72	1.23	1.0000	1.0683	13.88	0.00
0.4997	1.0417	6.81	1.48				

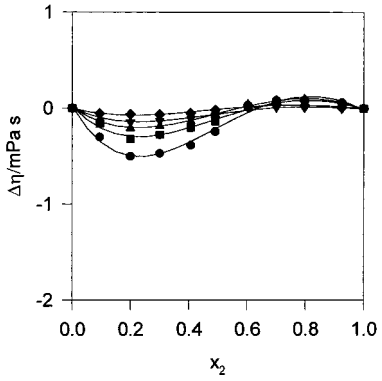

Figure 1. Viscosity deviation for the 1-propanol (1) + TEGDME (2) system at various temperatures: ●, 5 °C; ■, 15 °C; ▲, 25 °C; ▼, 35 °C; ◆, 45 °C; solid lines, Redlich−Kister correlations.

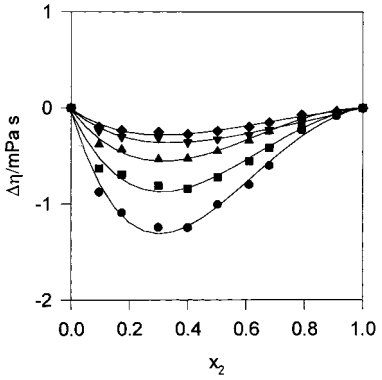

Figure 2. Viscosity deviation for the 1-butanol (1) + TEGDME (2) system at various temperatures: ●, 5 °C; ■, 15 °C; ▲, 25 °C; ▼, 35 °C; ◆, 45 °C; solid lines, Redlich−Kister correlations.

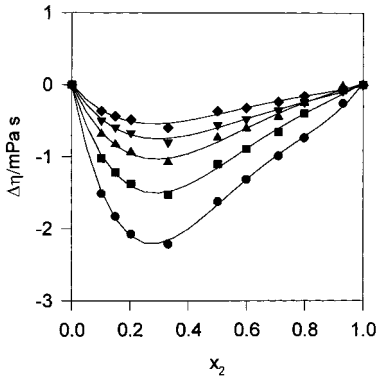

Figure 3. Viscosity deviation for the 1-pentanol (1) + TEGDME (2) system at various temperatures: ●, 5 °C; ■, 15 °C; ▲, 25 °C; ▼, 35 °C; ◆, 45 °C; solid lines, Redlich−Kister correlations.

derivative concentration increases. For the TEGDME + 1-propanol system, the minimum reaches a mole fraction close to 0.2 at 278.15 K and 288.15 K. For the TEGDME + 1-butanol system, the minimum reaches higher mole fraction values and it includes all the concentration range when the mixture contains 1-pentanol at all temperatures.

The deviation of the viscosity from the mole fraction average, $\Delta\eta$, is given by

$$\Delta\eta = \eta - (x_1\eta_1 + x_2\eta_2) \qquad (2)$$

where η, η_1, η_2, x_1, and x_2 are the dynamic viscosity of the mixture, the dynamic viscosities of the pure components, and the molar fractions, respectively. Figures 1−3, for TEGDME + alcohols systems, and Figures 4 and 5, for

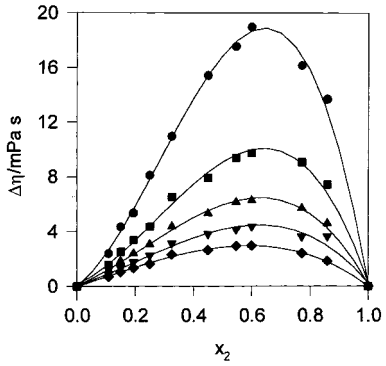

Figure 4. Viscosity deviation for the 1-butanol (1) + PEGMME (2) system at various temperatures: ●, 5 °C; ■, 15 °C; ▲, 25 °C; ▼, 35 °C; ◆, 45 °C; solid lines, Redlich−Kister correlations.

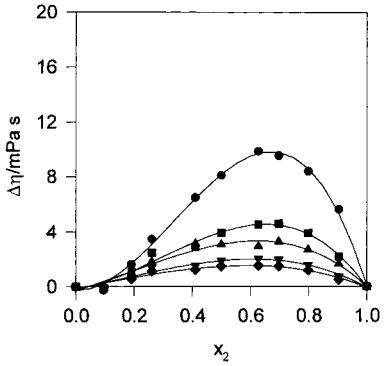

Figure 5. Viscosity deviation for the 1-pentanol (1) + PEGMME (2) system at various temperatures: ●, 5 °C; ■, 15 °C; ▲, 25 °C; ▼, 35 °C; ◆, 45 °C; solid lines, Redlich−Kister correlations.

PEGMME + alcohols systems, show the dependence of the deviation of the viscosity on the composition and temperature.

The viscosity deviation represents deviations from a rectilinear dependence of viscosity on mole fraction. The values of $\Delta\eta$ shown in Figure 1, for the systems containing TEGDME + 1-propanol, were sigmoids with negative values in the alcohol-rich region at all the temperatures. For TEGDME + 1-butanol and 1-pentanol, the values of $\Delta\eta$ were all negative throughout the whole concentration range at all the temperatures, and in the PEGMME + alcohols systems, the values of $\Delta\eta$ were all positive throughout the whole concentration range at all the temperatures.

The viscosity deviations were represented mathematically by the Redlich−Kister equation.

$$\Delta\eta/\mathrm{mPa\cdot s} = x_2(1 - x_2)\sum_{i=0}^{n} a_i(2x_2 - 1)^i \qquad (3)$$

where a_i are the fitting coefficients and x_2 is the mole fraction of glycol derivatives. The coefficients and the standard deviation σ are presented in Table 7.

Following Nakagawa,[3] who has applied a theoretical development of Matsubayashi and Nakahara,[10] the dynamic viscosity of a nonelectrolyte binary mixture can be given by

$$\eta = x_2\eta_2 + (1 - x_2)\eta_1 + x_2(1 - x_2)\eta_{12} \qquad (4)$$

where η_{12} is the viscosity regular term.

A methodology to calculate it was developed by Nakagawa[3] and applied by Ruiz Holgado[2] in mixtures containing glycol methyl derivatives with 1-propanol.

Table 7. Redlich−Kister Equation Fitting Coefficients of the Viscosity Deviation, $\Delta\eta$/mPa·s, for Different Systems at Various Temperatures

T/K	a_0	a_1	a_2	a_3	a_4	a_5	σ
TEGDME + 1-Propanol							
278.15	−0.798	3.841	−1.075	−1.940			0.03
288.15	−0.432	2.334	−0.642	−1.149			0.02
298.15	−0.214	1.809	−0.272	−0.746			0.01
308.15	−0.237	1.067	−0.434	−0.820			0.01
318.15	−0.040	0.711	−0.241	−0.488			0.01
TEGDME + 1-Butanol							
278.15	−4.120	4.011	−1.265	2.396			0.06
288.15	−2.824	2.351	−0.722	2.237			0.06
298.15	−1.779	1.742	−0.262	0.940			0.03
308.15	−1.277	0.588	−0.404	1.036			0.02
318.15	−0.936	0.619	−0.310	0.795			0.02
TEGDME + 1-Pentanol							
278.15	−6.6780	6.986	−5.747	0.663			0.04
288.15	−4.6150	4.739	−2.686	1.521			0.05
298.15	−3.1710	3.393	−1.549	0.940			0.05
308.15	−2.3870	2.474	−1.469	−0.253			0.04
318.15	−1.6450	2.035	−1.228	−0.521			0.04
PEGMME + 1-Butanol							
278.15	67.77	52.76	−19.81	−50.44	52.70	114.5	0.3
288.15	35.38	22.46	6.725	9.648	3.922	9.065	0.2
298.15	23.38	12.82	2.984	4.896	3.139	6.269	0.1
308.15	16.59	7.028	−9.881	−8.530	32.38	41.99	0.1
318.15	11.44	3.963	−0.415	0.7285	1.254	2.803	0.08
PEGMME + 1-Pentanol							
278.15	32.90	34.90	0.799	−23.61	−6.533	53.71	0.1
288.15	15.02	15.07	16.89	−20.37	−34.41	36.57	0.2
298.15	12.63	−1.545	2.669	44.36	−12.38	-36.83	0.1
308.15	7.305	4.220	7.905	−6.649	−20.01	13.94	0.1
318.15	5.685	2.330	6.600	−2.617	−16.82	6.930	0.08

Table 8. Regular Viscosity Terms, η_{12}, of Different Systems at Various Temperatures

	PEGMME			TEGDME		
T/K	PR	BU	PE	PR[a]	BU	PE
278.15	−10.9	−15.3	−26.8	−33.9	−27.7	−28.4
288.15	−7.1	−10.4	−17.8	−16.7	−12.8	−15.6
298.15	−5.7	−7.5	−12.6	−5.4	−3.4	−7.0
308.15	−4.4	−5.7	−8.8	0.0	0.0	−2.9
318.15	−3.6	−4.3	−6.5	2.3	1.8	−1.2

[a] Values taken from Ruiz Holgado et al.[2]

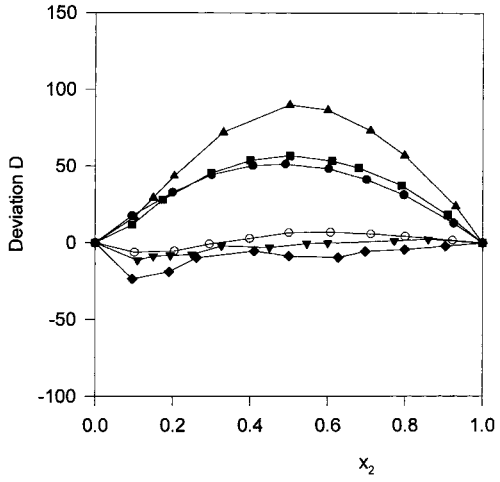

Figure 6. Deviation, *D*, of dynamic viscosity predicted with respect to experimental values for different systems at 298.15 K: ●, TEGDME + 1-propanol; ■, TEGDME + 1-butanol; ▲, TEGDME + 1-pentanol; ▼, PEGMME + 1-butanol; ◆, PEGMME + 1-pentanol; ○, PEGMME + 1-propanol; calculated values are from ref 2.

In the present paper, the viscosity regular term, η_{12}, was calculated from Jones Dole *B* coefficient values, which were

obtained from a plot of $((\eta/\eta_1 - 1)/c)$ versus c, where c is the solution molarity in moles per cubic centimeter, using dynamic viscosity data from dilute solutions of TEGDME in 1-propanol, 1-butanol, and 1-pentanol and of PEGMME in 1-butanol and 1-pentanol, taken from the literature.[11]

The viscosity regular term values calculated for the studied systems are shown in Table 8. They have allowed the prediction of the dynamic viscosity values of TEGDME and PEGMME in *n*-alcohols, over the whole concentration range in binary mixtures.

The deviation of the values calculated with the viscosity regular term with respect to the experimental ones has been defined by the following equation:

$$D = 100\left(\frac{\eta_{\text{exp}} - \eta_{\text{cal}}}{\eta_{\text{exp}}}\right) \tag{5}$$

where η is the dynamic viscosity and the subindices exp and cal mean experimental and calculated, respectively.

The deviation values (D) versus the mole fraction of the glycol derivative for all the systems were plotted in Figure 6. The smaller deviations correspond to those systems containing PEGMME, and the greater ones, to those with TEGDME, with values close to 75% in the intermediate concentration area.

Literature Cited

Esteve, X.; Boer, D.; Patil, K. R.; Chaudhari, S. K.; Coronas, A. Densities, Viscosities, and Enthalpies of Mixing of the Binary System Methanol + Polyethylene Glycol 250 Dimethyl Ether at 303.15 K. *J. Chem. Eng. Data* **1994**, *39*, 767−769.

Ruiz Holgado, M. E. de; Schaefer, C. R. de; Arancibia, E. L. Viscosity Study of 1-Propanol with Polyethylene Glycol 350 Monomethyl Ether Systems at Different Temperatures. *J. Mol. Liquids* **1999**, *79*, 257−267.

Nakagawa, T. Is Viscosity B Coefficient Characteristic for Solute−Solvent Interaction? *J. Mol. Liquids* **1995**, *63*, 303−316.

Ruiz Holgado, M. E. de; Schaefer, C. R. de; Arancibia, E. L. Viscosimetric Study of the Behaviour of Monomethyl Ether and Dimethyl Ether of Polyethylene Glycol in n-Alcohols. *J. Mol. Liquids*, in press.

Pal, A.; Dass, G.; Kumar, A. Excess Molar Volumes, Viscosities, and Refractive Indices of Tetraethylene Glycol Dimethyl Ether + Dimethyl Carbonate, + Diethyl Carbonate, and + Propylene Carbonate at 298.15 K. *J. Chem. Eng. Data* **1999**, *44*, 2−5.

Carmona, F. J.; Arroyo, F. J.; Darcia de la Fuente, I.; Gonzáles, J. A.; Cobos, J. C. Excess Molar Volumes of Methanol or Ethanol + n-Polyethers at 298.15 K. *Can. J. Chem.* **1999**, *77*, 1608−1616.

Riddick, A. R.; Bunger, W. B.; Sakano, T. K. *Organic Solvents. Techniques of Chemistry*, 4th ed.; Wiley-Interscience: New York, 1986.

Shan, Z.; Asfour, A.-F. A. Viscosities and Densities of Nine Binary 1-Alkanol Systems at 293.15 K and 298.15 K. *J. Chem. Eng. Data* **1999**, *44*, 118−123.

Rauf, M. A.; Stewart, G. H.; Farhataziz, V. Viscosities and Densities of Binary Mixtures of 1-Alkanols from 15 to 55 °C. *J. Chem. Eng. Data* **1983**, *28*, 324−328.

Matsubayashi, N.; Nakahara, M. Dynamics in Regular Solutions: Concentration and Viscosity Dependence Orientational Correlation Time of a Benzene Molecule. *J. Chem. Phys.* **1991**, *94*, 653−661.

Ruiz Holgado, M. E. de; Schaefer, C. R. de; Arancibia, E. L. Thermodynamic Functions of Activation for Viscous Flow for Solutions of Polyethylene Glycol Methyl Derivatives with n-Alcohols at Different Temperatures. *Fluid Phase Equilib.* **2000**, *169*, 237−247.

Received for review June 21, 2001. Accepted October 12, 2001. This work was supported by a CIUNT research grant. E.L.A. is member of CONICET.

JE010182S