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Densities and Viscosities of Triethylene Glycol Monomethyl Ether +Water Solutions in the Temperature Interval 25 °C−80 °C

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The density and viscosity of aqueous solutins of triethylene glycol monomethyl ether have been measured at five temperatures in the range 25 °C−80 °C throughout the whole concentration range. Excess volume and viscosity deviation values were calculated and correlated as a function of composition. The results are compared with other data when these were available. The derived excess molar volumes, viscosity deviations, apparent molar volumes, and partial molar volumes at infinite dilution and the Grunberg−Nissan interaction energy constants are reported. The values obtained can be used to understand the nature and extent of the patterns of molecular aggregation that exist in binary (water + organic) mixtures and their sensitivity to variations in compositions, temperature, and molecular structure. Knowledge of the effect of temperature and composition on changes in the liquid density and viscosity is required in the modeling of mass transfer, heat transfer, and fluid flow.

Introduction

Previously published data (Henni and Mather, 1995a−c) revealed that triethylene glycol monomethyl ether (TEGMME) absorbs more CO_2, less methane, but more ethane than solvents currently used in the gas industry. In a recent survey of the most suitable polar solvents for CO_2 removal from gas streams (Henni and Mather, 1997), TEGMME also was found to have the highest boiling point. In a solubility study Henni and Mather (1995a) chose TEGMME among other suitable solvents mainly because of its lower cost compared to solvents of the same chemical family like tetraethylene glycol dimethyl ether (TEGDME), a solvent recommended by Zawacki et al. (1981).

The present work was undertaken to measure the viscosity and density of this promising solvent in its pure form and in aqueous solution. As TEGMME can be used with chemical solvents (alkanolamine) in aqueous solutions for acid gas removal, transport properties measurement provides data necessary to model the rate of reactions and mass transfer rates in absorption and desorption operations.

Measurements of the density and viscosity of the aqueous TEGMME were performed at various temperatures (25, 35, 40, 50, 60, and 80 °C) in order to cover the range of temperatures found in absorbers and regenerators. The results were represented by mole fraction polynomials, and calculated values of the excess volumes and the deviation of the viscosity were correlated by a Redlich−Kister type equation. The variations of the apparent molar volumes with the mole fraction and the second derivative of the partial molar volumes at infinite dilution with respect to temperature were calculated. The Grunberg−Nissan constants (d_{12}) were derived. The results obtained were used as a basis for deducing a “picture” of the solute−water interactions over the range of composition and temperature for the system TEGMME + water.

Experimental Section

Triethylene glycol monomethyl ether (>97% pure) was purchased from Fluka and used without further purification. Water used was deionized and distilled in glass.

Densities of the aqueous solutions were measured with a platinium−rhodium vibrating densimeter (Sodev Inc., model 03 Dpr, Rock Forest, Québec, Canada) with a 10^{-6} $g{\cdot}cm^{-3}$ resolution. The densimeter was calibrated with dry air and distilled water. The reproducibility of the measurements was about 3×10^{-5} $g{\cdot}cm^{-3}$. The temperature was controlled by means of a digital controller (Cole Parmer, model H-01158−65, Anjou, Québec, Canada) in a well-stirred water bath (model H-01158-10) to ±0.004 °C and was known to within ±0.01 °C as measured with a digital thermometer (Cole Parmer, model H-08502-12).

The density was determined by the two-parameter equation

$$\rho = A + B\tau^2 \tag{1}$$

where ρ is the liquid density and τ is the period of oscillation. The values of *A* and *B* were determined at each temperature using deionized water (Kell, 1975), and an air equation for the determination of the density of dry air was taken from the annual book of ASTM standards (ASTM, 1986).

Viscosities were determined with two different viscometers to cover with precision the range of temperature from 25 °C to 80 °C. For temperatures up to 50 °C, an Ubbelohde suspended level viscometer No. 10 (J. Toonen Glass,

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Table 1. Densities of Water (1) + TEGMME (2) at Various Temperatures

	ρ/g·cm^3						
x_2	25 °C	35 °C	40 °C	50 °C	60 °C	70 °C	80 °C
0.000	0.997 04	0.994 03	0.992 21	0.988 04	0.983 12	0.977 77	0.971 79
0.0110	1.008 95	1.003 04	1.001 03	0.997 17	0.991 13	0.985 09	0.976 97
0.0268	1.021 49	1.012 52	1.010 12	1.004 53	0.999 39	0.992 76	0.982 12
0.0451	1.032 70	1.021 85	1.019 81	1.014 24	1.007 42	1.000 15	0.992 39
0.0686	1.043 25		1.029 57	1.022 16	1.014 44	1.006 72	0.998 94
0.0995	1.051 14		1.036 01	1.028 47	1.020 38	1.011 72	1.003 24
0.1420	1.056 08		1.040 75	1.033 55	1.024 24	1.016 02	1.006 34
0.2050	1.057 22		1.042 64	1.034 86	1.025 19	1.016 04	1.007 43
0.3055	1.053 36		1.040 36	1.033 42	1.023 36	1.014 07	1.005 18
0.4215	1.051 09		1.038 57	1.030 52	1.022 58	1.017 23	1.005 21
0.4991	1.051 99		1.035 75	1.028 65	1.020 38	1.014 36	1.004 00
0.609	1.051 02		1.034 40	1.026 80	1.018 34	1.012 92	1.002 41
0.7626	1.049 14		1.031 85	1.023 90	1.015 89	1.011 01	1.000 93
0.8318	1.047 60		1.031 09	1.023 80	1.015 68	1.009 99	1.000 08
0.9000	1.047 30		1.030 90	1.022 30	1.014 12	1.009 32	0.999 42
1.0000	1.045 24	1.032 62	1.029 08	1.021 27	1.013 27	1.008 51	0.997 96

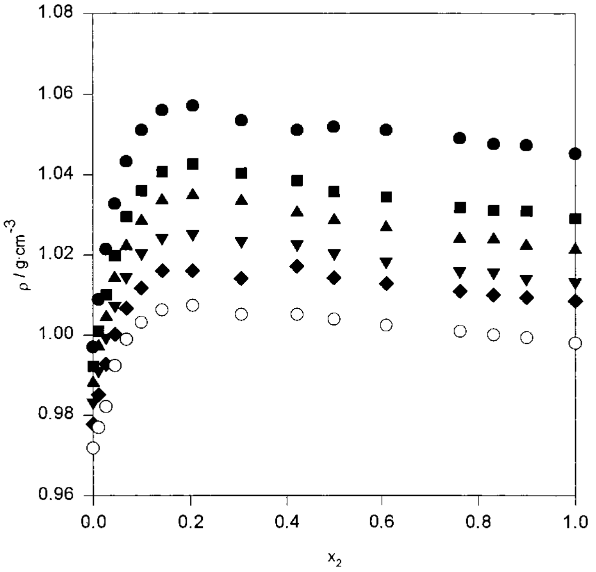

Figure 1. Densities of TEGMME (2) + water (1) system at various temperatures: (●) 25 °C; (■) 40 °C; (▲) 50 °C; (▼) 60 °C; (◆) 70 °C; (○) 80 °C.

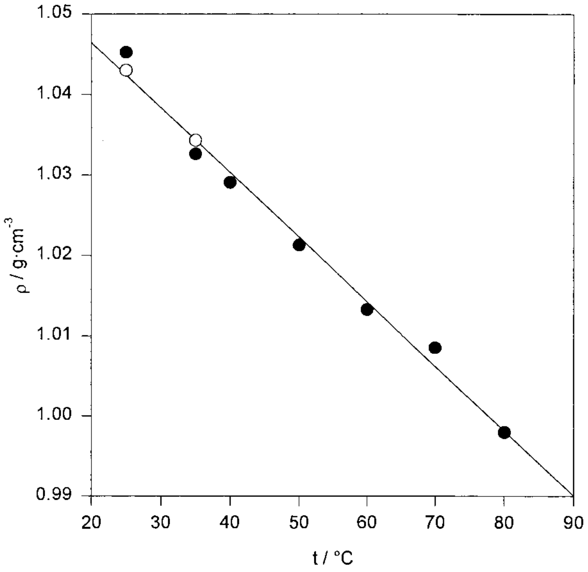

Figure 2. Density of pure TEGMME at various temperatures: (●) this work; (○) Pal and Singh (1995, 1996); (—) linear regression.

Edmonton, Canada) was used. For higher temperatures, a Cannon-Ubbelohde viscometer (Cole-Parmer, E-98934-11) was utilized. The efflux time was measured with a hand-held digital stopwatch capable of measuring time to within ±0.01s. Experiments were repeated a minimum of four times at each temperature for all compositions. The equation for kinematic viscosity, according to Poiseuille's law, is

$$\nu = k_1 t - k_2 / t \tag{2}$$

where t is the efflux time and k_1 and k_2 are the viscometer constants. The second term representing the correction due to the kinetic energy was found to be negligible. The value of the absolute viscosity (η) was obtained by multiplying the measured kinematic viscosity (ν) with the measured density.

Calibration constants for the two viscometers were checked using high-purity ethylene glycol (EG), diethylene glycol (DEG), and water. A review of published values of the kinematic and dynamic viscosities of EG and DEG, with new experimental data, was published by Lee and Teja (1990). Values of the viscosity of water were taken from Stokes and Mills (1965). The values of the viscosities were reproducible to ±0.003 mPa·s.

Results and Discussion

Experimentally measured densities of the binary aqueous solutions of TEGMME at (25, 35, 40, 50, 60, 70, and 80 °C) throughout the whole concentration range are listed in Table 1. The values obtained are shown in Figure 1. The values of pure TEGMME at 25 °C and 35 °C are in poor agreement (<0.2%) with the values obtained by Pal and Singh (1995, 1996) using a doubled-armed pycnometer, i.e., 1.0430 and 1.0343 g·cm^{-3}, respectively. Some values of the density of the mixture at different compositions were measured at 35 °C in order to (indirectly) compare the data obtained with the values of excess volumes published by Pal and Singh (1996). The effect of the temperature on the density of pure TEGMME is depicted in Figure 2.

All data for the density of pure TEGMME (g·cm^{-3}) were correlated as a polynomial equation in temperature (°C) in the range from 25 °C to 80 °C.

$$\rho_{\mathrm{TEGMME}} = 1.022\,17 + 3.043\,38 \times 10^{-3} t - 1.338\,46 \times 10^{-4} t^2 + 1.942\,75 \times 10^{-6} t^3 - 9.903\,126 \times 10^{-9} t^4 \tag{3}$$

The standard deviation was 0.001 g·cm^{-3}. The density measurements of the aqueous solution were correlated in a polynomial form ($\rho = \sum_0^n a_k x^k$); the coefficients and the standard deviation (s) are presented in Table 2.

Table 2. Coefficients of the Polynomial ρ (g·cm^{-3}) = $\sum_0^n a_k x^k$ and the Standard Deviation for Water (1) + TEGMME (2) at Various Temperatures

t/°C	a_0 a_6	a_1 a_7	a_2 s/g·cm^{-3}	a_3	a_4	a_5
25	0.997 721	1.045 21	−69.826 4	23.047 8	−4209.61	433.393
	−23.590 9	5.284 96	0.000 546			
40	0.992 626	0.780 235	−4.532 27	12.636 4	−18.498 1	13.612 8
	−3.962 67		0.002 41			
50	0.988 902	0.706 242	−4.011 26	10.925 3	−15.657 7	11.304 37
	−3.234 69		0.000 51			
60	0.9.839 62	0.665 453	−3.973 09	11.352 27	−16.918 89	12.590 2
	−3.686 85		0.000 917			
70	0.979 936	0.513 458 6	−2.457 67	5.164 59	−4.945 87	1.754 73
			0.003 02			
80	0.972 291	0.497 263 5	−2.381 029	4.932 48	−46.393 1	16.168 1
			0.002 41			

Table 3. Redlich−Kister Equation Fitting Coefficients of the Excess Volumes (V_m^E) for Water (1) + TEGMME (2) at Various Temperatures

t/°C	a_0 a_7	a_1 s/g·cm^{-3}	a_2	a_3	a_4	a_5	a_6
25	−3.9599	3.6453	−9.9257	−13.139	27.9838	51.719	−31.8056
	−47.786	0.0142					
40	−3.8355	3.0640	−2.6083	−0.99239	1.47731	13.8122	−5.4638
	−17.713	0.0162					
50	−3.8541	2.3576	1.1930	9.01386	−18.291	−25.5726	19.7805
	23.329	0.0286					
60	−3.6782	2.8891	1.1687	3.1322	−17.762	−12.571	19.458
	15.507	0.0255					
70	−3.323	3.6271	−4.7124	−8.2214	8.7403	29.237	−8.7582
	−24.756	0.0249					
80	−3.4803	6.3028	−4.6443	−35.402	11.5142	99.0442	−14.627
	−80.415	0.0787					

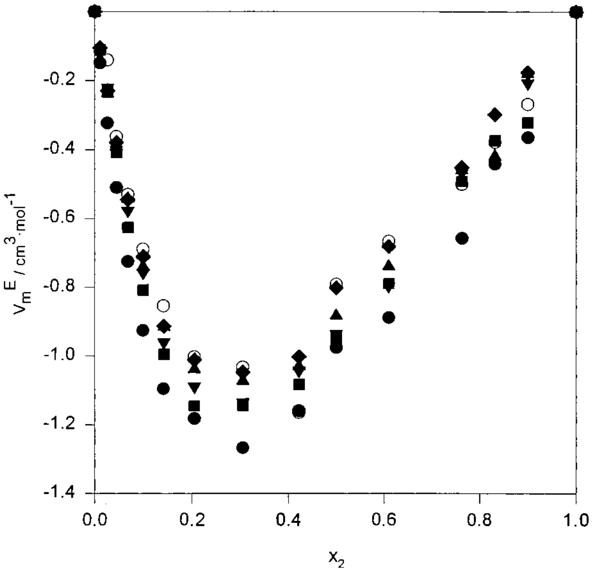

Figure 3. Excess molar volumes of TEGMME (2) + water (1) system at various temperatures: (●) 25 °C; (■) 40 °C; (▲) 50 °C; (▼) 60 °C; (◆) 70 °C; (○) 80 °C.

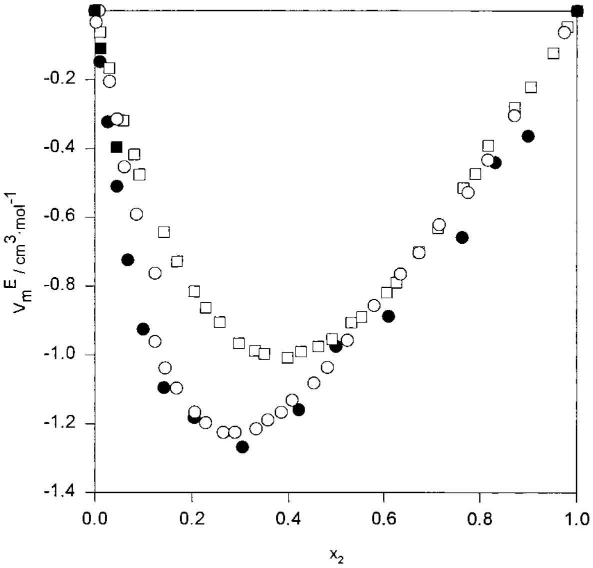

Figure 4. Comparison of the excess molar volumes of TEGMME (2) + water (1) system at 25 °C and 35 °C: (●) 25 °C, this work; (○) 25 °C, Pal and Singh, 1995; (■) 35 °C, this work; (□) 35 °C, Pal and Singh, 1996.

Experimental density values of the binary mixtures were used to calculate the excess molar volume, V_m^E, as

$$V_m^E = V_m - V_1^\circ x_1 - V_2^\circ x_2 \tag{4}$$

where V_m^E is the molar volume of the mixture and V_1° and V_2° are those of pure water and pure TEGMME, respectively; x_1 and x_2 are the mole fractions of the pure components.

Figure 3 displays the dependence of V_m^E on the composition and temperature. The excess molar volume values are recorded in Table 3. The excess volumes were fitted to the Redlich and Kister (1948) relation

$$V_m^E = x_1 x_2 \sum_{j=0}^{n} a_j (x_2 - x_1)^j \tag{5}$$

where a_j's are the adjustable parameters. The values of the parameters and the standard deviations s

$$s\,(V_m^E) = [(V_{m,exp}^E - V_{m,calc}^E)^2/(n - p)]^{1/2} \tag{6}$$

where n is the total number of experimental points and p

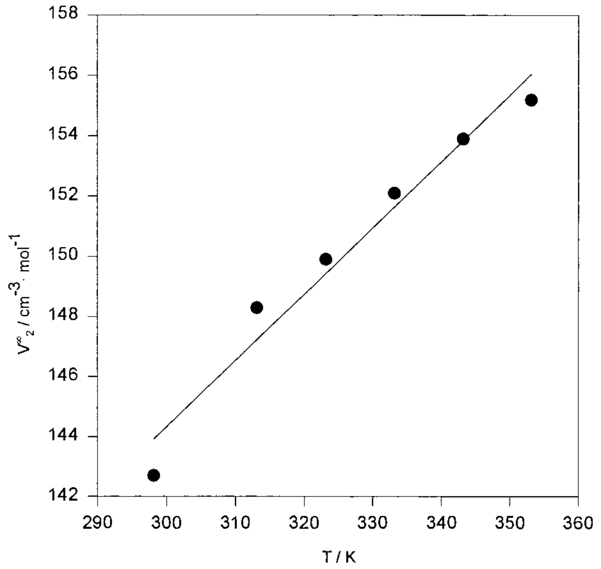

Figure 5. Partial molar volumes of TEGMME at infinite dilution in water at various temperatures: (●) 25 °C; (—) linear regression.

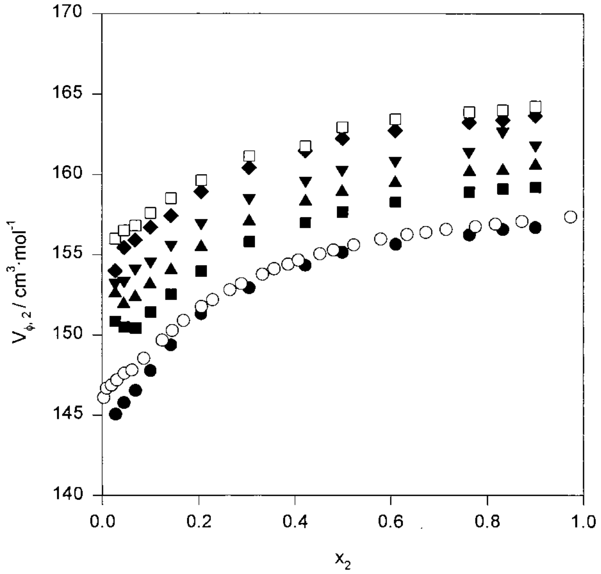

Figure 6. Apparent molar volumes of TEGMME in water at various temperatures: (○) 25 °C, Pal and Singh, 1995; (●) 25 °C, this work; (■) 40 °C; (▲) 50 °C; (▼) 60 °C; (◆) 70 °C; (□) 80 °C.

Table 4. Partial Molar Volumes of TEGMME at Infinite Dilution in Water (V^∞_2) and of Water at Infinite Dilution in TEGMME (V^∞_1)

t/°C	V^∞_2/ cm^{3}mol^{-1}	V^∞_1/ cm^{3}mol^{-1}	t/°C	V^∞_2/ cm^{3}mol^{-1}	V^∞_1/ cm^{3}mol^{-1}
25	142.8	17.9	60	151.4	18.2
40	148.2	18.0	70	152.9	18.3
50	149.1	18.1	80	155.2	18.4

is the number of parameters in the regression, are reported in Table 3. Figure 4 compares the values of V^E_m calculated in this study with those measured by Pal and Singh (1995, 1996) using a continuous-dilution dilatometer. There is reasonable agreement for 25 °C. At 35 °C, V^E_m values calculated in this work are in agreement with Pal and Singh's values at low TEGMME concentrations. At other concentrations their values are less negative than what could be predicted from our calculated values at 25 and 40 °C.

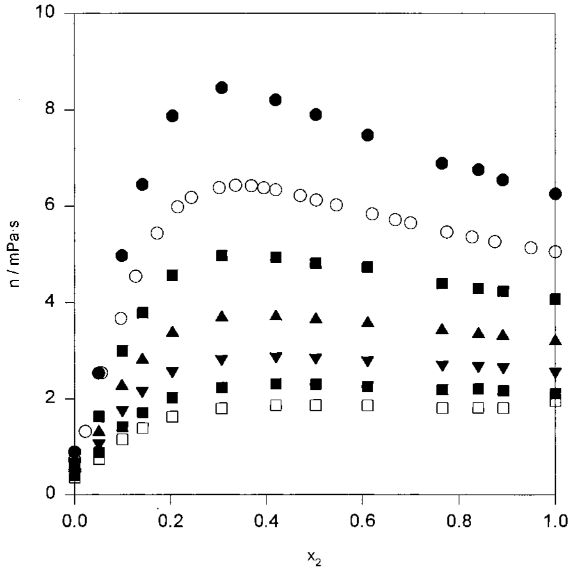

Figure 7. Viscosities of TEGMME (2) + water (1) system at various temperatures: (●) 25 °C, this work; (○) 35 °C, Pal and Singh, 1995; (■) 40 °C; (▲) 50 °C; (▼) 60 °C; (◆) 70 °C; (□) 80 °C.

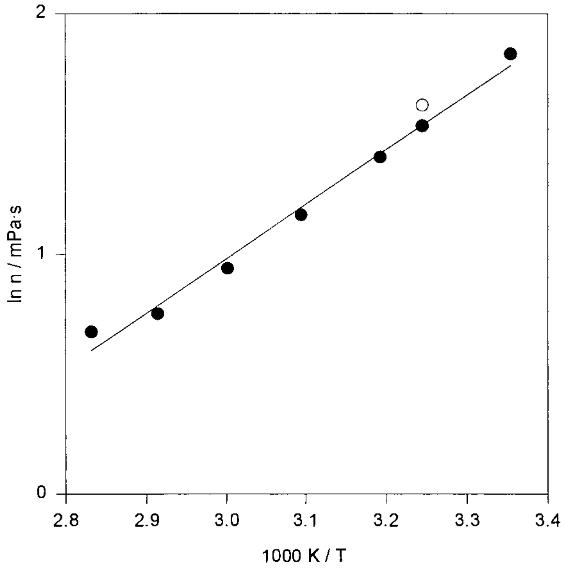

Figure 8. Dynamic viscosity of pure TEGMME at various temperatures: (●) 25 °C, this work; (○) 25 °C, Pal and Singh, 1996; (—) linear regression.

Table 5. Experimental Viscosities for TEGMME (2) + Water (1) Mixtures at Various Temperatures

	η /mPa·s					
x_2	25 °C	40 °C	50 °C	60 °C	70 °C	80 °C
0.000	0.8903	0.6531	0.547	0.466	0.4049	0.3554
0.0497	2.531	1.632	1.300	1.072	0.883	0.745
0.0989	4.964	2.994	2.266	1.764	1.417	1.152
0.1415	6.443	3.785	2.812	2.167	1.710	1.388
0.2035	7.869	4.556	3.375	2.576	2.022	1.631
0.3069	8.456	4.963	3.689	2.826	2.230	1.799
0.4204	7.901	4.933	3.709	2.881	2.307	1.862
0.5037	7.472	4.812	3.647	2.853	2.300	1.866
0.6109	6.897	4.728	3.566	2.805	2.255	1.858
0.7651	6.754	4.394	3.428	2.720	2.189	1.811
0.8400	6.754	4.295	3.352	2.688	2.210	1.816
0.8917	6.594	4.236	3.307	2.662	2.217	1.811
1.0000	6.253	4.074	3.202	2.618	2.121	1.968

All excess volumes were negative, as is common for other completely miscible (water + polar organic) solvents. These

Table 6. Coefficients of the Polynomial $\ln \eta$ (mPa·S) $= \ln \eta_0 + \sum_1^n a_k x^k$ and the Standard Deviation for the Binary Solutions of TEGMME (2) + Water (1) at Various Temperatures

	a_0	a_1	a_2	a_3		
t/°C	a_6	a_7	a_8	s/mPa·s	a_4	a_5
25	25.75	−87.40	−134.91	1920.55	−6435.71	11182.0
	−10952.0	5733.70	−1249.80	0.0360		
40	22.07	−63.74	−248.94	2395.5	−7892.10	14031.0
	−14238.0	7757.0	−1761.07	0.0362		
50	21.34	−72.328	−130.20	1818.5	−6350.88	11543.7
	−11816.0	6447.90	−1460.20	0.0286		
60	21.30	−97.514	144.51	460.16	−2567.6	5264.50
	−5666.30	3171.0	−728.46	0.0002		
70	20.26	−99.58	229.69	−119.07	−577.70	1372.80
	1312.10	−583.10	−95.82	0.0003		
80	19.394	−101.49	287.27	−417.25	150.2797	434.24
	−692.19	412.25	−90.79	0.00013		

Table 7. Redlich−Kister Equation Fitting Coefficients for the Viscosity Deviations ($\Delta\eta$/MPa·S) and the Standard Deviation for Water (1) +TEGMME (2) at Various Temperatures

t/°C	a_0	a_1	a_2	a_3	a_4	a_5	s/mPa·s
25	16.908	−22.1230	29.1170	5.3049	−0.9174		0.225
40	9.6902	−11.1990	14.3250	0.7023	−14.111		0.292
50	5.3280	−4.4756	5.6209	−7.5873	−3.0047	10.1490	0.046
60	5.1893	−4.5307	6.6728	−6.3424	−5.2644	7.3824	0.041
70	4.1067	−3.5969	3.5201	−4.6038	−1.1874	−6.1776	0.003
80	4.0846	−4.2469	3.9838	0.0230	−2.2034		0.005

V_m^E values became less negative with increasing temperatures, as is also common.

Figures 1 and 3 show a sharp change in the density and V_m^E in the water-rich region. The excess molar volume reached its minimum value at around 30 mol % of TEGMME, i.e., in the water-rich region. This minimum shifts to higher mole fractions of TEGMME as the temperature increases. Pal and Singh (1996) concluded that the contraction in the volume is due to the ability of the −OH group to form hydrogen bonds with water molecules and the formation of the hydrogen bonds is enhanced by the presence of the ether oxygen (Buckley and Brochu, 1972). The second interpretation given was that such marked change in V_m^E might also be due to the accommodation of the nonaqueous molecules occupying in the structured water lattice a void space.

It is known that the number of cross-associated H-bonds decreases with increasing temperature, which leads to positive contribution to V_m^E. As a consequence V_m^E values became more negative with decrease in temperature. In comparing the excess volumes, Pal and Singh (1996) concluded that the order of the strength of the hydrogen bonding is higher in TEGMME than in diethylene glycol monomethyl ether (DEGMME) followed by ethylene glycol monomethyl ether (EGMME). The large change noticed in the density as a function of composition and temperature must be taken into account in heat transfer, mass transport, and fluid flow models.

Hepler (1969) and Neal and Goring (1970) recognized the usefulness of thermal expansion data for revealing correlations between molecular structure of solutes and their effects on water structure. They suggested using the sign of the second derivative of the partial molar volume at infinite dilution of the solute with respect to the temperature ($d^2V_2^\infty/dT^2$) to classify solutes as “structure makers” or “ structure breakers”. A positive sign corresponds to structure-making while a negative sign corresponds to structure-breaking solute.

The availability of excess volume data for the present system [TEGMME (2) + water (1)] at six different temperatures provided an opportunity to further investigate the proposed criterion. Both the partial molar volume of water at infinite dilution (V_1^∞) in TEGMME and the partial molar volume of TEGMME at infinite dilution (V_2^∞) in water were obtained using the method proposed by Maham et al. (1995). The apparent molar volume of water ($V_{\phi,1}$) and the apparent molar volume of TEGMME in water ($V_{\phi,2}$) were first calculated as

$$V_{\phi,1} = V_1^\circ + [V_m^E/(1 - x_2)] \tag{7}$$

and

$$V_{\phi,2} = V_2^\circ + (V_m^E/x_2) \tag{8}$$

where V_1° and V_2° are the molar volumes of pure water and TEGMME, respectively. An analytical extrapolation of $V_{\phi,1}$ to $x_2 = 1$ led to V_1^∞, and a similar extrapolation to $x_2 = 0$ led to V_2^∞. Partial molar volumes at infinite dilution are listed in Table 4. As shown in Figure 5, V_2^∞ values varied linearly with temperature. The second derivative with respect to the temperature was found to be equal to zero. According to the criterion described above, the solute (TEGMME) would be considered as a having no net effect on the structure of water. All values of the molar volumes at infinite dilution (V_2^∞) were smaller than the corresponding molar values of pure TEGMME (V_2°). The change in the excess volume is then explained by the TEGMME molecule fitting (partially) into the open or empty space in liquid water. This picture does not invoke either the structure-making or the breaking properties and is consistent with the observation that ($d^2V_2^\infty/dT^2$) values are approximately zero.

Values of the apparent molar volumes of TEGMME in water ($V_{\phi,2}$) are shown in Figure 6. The values are in close agreement with the values calculated by Pal and Singh (1995) except at mole fractions less than 0.2, keeping in mind that the experimental data were obtained using a different equipment. Having fewer values of V_m^E for x_2 near unity, accuracies of V_1^∞ may be lower. The values obtained showed a linear variation with respect to the temperature. It is still clear that dV_1^∞/dT values were positive. All values of V_1^∞ were smaller than the corresponding molar volumes of pure water (V_1°) at the same

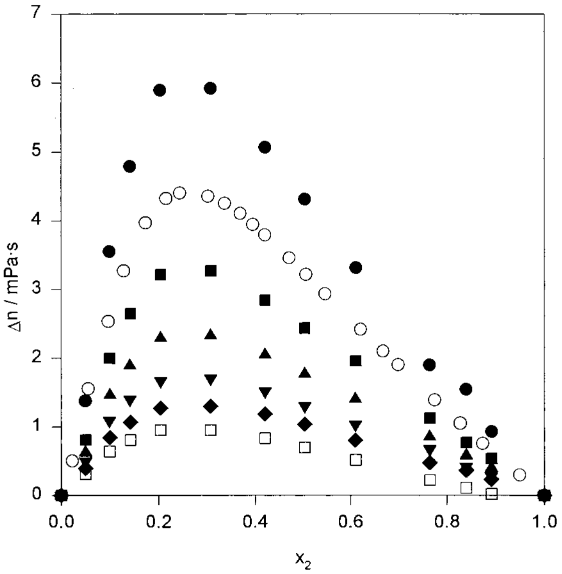

Figure 9. Viscosity deviations for TEGMME (2) + water (1) system at various temperatures: (●) 25 °C, this work; (○) 35 °C, calcd Pal and Singh, 1996; (■) 40 °C; (▲) 50 °C; (▼) 60 °C; (◆) 70 °C; (□) 80 °C.

temperature. As mentioned by Maham et al. (1995), this is consistent with the idea that the molar volume of pure water is the sum of the actual molecular volumes plus the "empty" volume that arises from the hydrogen-bonded open structure of water.

Experimentally measured viscosities of the binary aqueous solutions of TEGMME at (25, 40, 50, 60, 70, and 80 °C) are listed in Table 5 and shown in Figure 7. Only few viscosity measurements were made for comparison with values published by Pal and Singh (1996) at 35 °C. Figure 7 shows a sharp increase in the viscosity of the mixture after the addition of TEGMME. The effect of the composition on viscosity decreases with increase in temperature. The largest variation occurs at 25 °C. The viscosity at 30 mol % TEGMME was 9.5 times larger than that of pure water and 1.35 times that of pure TEGMME. The effect of the temperature on the viscosity of pure TEGMME is shown in Figure 8. The viscosities of the solutions at each temperature were fitted by the following polynomial

$$\ln \eta = \ln \eta_0 + \sum_{j=0}^{n} a_k\, x^k \tag{9}$$

where η is the viscosity of the binary solution (mPa·s), η_0 is the viscosity of pure water, and x is the mole fraction of TEGMME. The values of polynomial coefficients a_k are listed in Table 6.

Experimental viscosity values of the binary mixtures were used to calculate the viscosity deviation defined by

$$\Delta\eta = \eta - \eta_1 x_1 - \eta_2 x_2 \tag{10}$$

where η is the viscosity of the mixture and η_1 and η_2 are those of pure water and pure TEGMME, respectively; x_1 and x_2 are the mole fractions of the pure components. The viscosity deviation represents deviations from a rectilinear dependence of viscosity on mole fraction. Figure 9 displays the dependence of $\Delta\eta$ on the composition and temperature. Values of $\Delta\eta$ were all positive throughout the whole concentration range for all temperatures.

The calculated values of $\Delta\eta$ were fitted to the Redlich and Kister (1948) relation

$$\Delta\eta = x_1 x_2 \sum_{j=o}^{n} a_j\,(x_2 - x_1)^j \tag{11}$$

where a_j's are the adjustable parameters and n is the total number of experimental points. The values of the parameters and the standard deviations are reported in Table 7.

According to Grunberg and Nissan (1949), the value of the binary mixture of components can be described in terms of the viscosities of the pure components and an interaction term as

$$\ln \eta = x_1 \ln \eta_1 + x_2 \ln \eta_2 + x_1 x_2 d_{12} \tag{12}$$

where d_{12} is regarded as a measure of the strength of interactions between the mixing species. Values of d_{12} were positive for all temperatures and compositions. The values obtained varied from 20.07 to 2.66 at 25 °C and from 13.89 to 1.10 at 80 °C. Positive values of d_{12} indicate a negative deviation from Raoult's law (Grunberg and Nissan, 1949) and support the notion of the formation of intermolecular complexes between the unlike molecules through hydrogen bonding (Pal and Singh, 1996).

Conclusions

This paper reports experimental data for the density and viscosity of the system TEGMME + water over a range of temperature from 25 °C to 80 °C. The criterion proposed by Hepler (1969) suggests that the addition of TEGMME to water has no effect on its structure. The calculated Grunberg−Nissan constants were found to be all positive. More experimental work is needed to analyze the interactions between alkyl poly(ethylene glycol) ethers and water in order to further test the criterion proposed by Hepler (1969) and others.

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