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Viscosity in Water + Ethylene Glycol Dimethyl, +Diethylene Glycol Dimethyl, +Triethylene Glycol Dimethyl, and +Tetraethylene Glycol Dimethyl Ethers at 298.15 K

Amalendu Pal* and Yoginder P. Singh

Department of Chemistry, Kurukshetra University, Kurukshetra 136 119, India

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The viscosity in binary liquid mixtures of water with ethylene glycol dimethyl ether (1,2-dimethoxyethane), $CH_3OCH_2CH_2OCH_3$, diethylene glycol dimethyl ether (bis(2-methoxyethyl) ether), $CH_3OCH_2CH_2OCH_2CH_2OCH_3$, triethylene glycol dimethyl ether (1,2-bis(2-methoxyethoxy)ethane), $CH_3OCH_2(CH_2OCH_2)_2CH_2OCH_3$, and tetraethylene glycol dimethyl ether (2,5,8,11,14-pentaoxapentadecane), $CH_3OCH_2(CH_2OCH_2)_3CH_2OCH_3$, have been determined at 298.15 K over the whole range of mixture compositions. The viscosity data have been analyzed by using the viscosity equations of McAllister and Heric. The values of the quantity $\Delta\eta$, which refer to the deviations of the experimental values of the dynamic viscosities of the mixtures from the mole fraction mixture law values, have been found to be positive for all the mixtures. The deviations in viscosity data $\Delta\eta$ have been fitted to the Redlich−Kister polynomial relation to estimate the binary coefficients. Also, the values of the parameter d_{12} have been calculated from the equation $\ln \eta = x_1 \ln \eta_1 + x_2 \ln \eta_2 + x_1x_2d_{12}$, where η_1 and η_2 refer to the dynamic viscosities of the two pure liquids 1 and 2 whose mole fractions in the mixture are x_1 and x_2, respectively. The values of d_{12} indicate the existence of a specific interaction between dissimilar molecules.

Introduction

Our recent research has been concerned with the measurement, analysis, and interpretation of the thermodynamic properties of the aqueous mixtures of nonionic amphiphiles, mainly the poly(ethylene glycol) monoalkyl ethers, which have the generic formula $C_mH_{2m+1}(OC_2H_4)_nOH$ (Douhéret et al., 1989; Pal and Singh, 1994, 1995). We devoted earlier papers to the study of the changes arising from the variation of the alkyl chain length for species with a common polar head group for which different measurements like ultrasonic speeds, dielectric constants, densities, and viscosities of the aqueous mixtures have been made and reported in the literature (Pal and Singh, 1994; Douhéret et al., 1990; Douhéret and Pal, 1988; Pal and Singh, 1995, 1996). The present work deals with the study of the changes of the polar head group, by the addition of an OC_2H_4 unit, for species with a common alkyl chain. We report here the viscosities (η) for binary mixtures of $\{H_2O\ (1) + CH_3(OCH_2CH_2)_nOCH_3\ (2)\}$ for $n = 1−4$, over the whole mole fraction range at 298.15 K. There have been physical and thermodynamic investigations for these systems, but imprecision of measurements or sparseness of the viscosity results for either pure liquids or the binary systems seemed to provoke a more detailed investigation of the mole fraction dependence of the viscosities (Wallace and Mathews, 1963; Wallace et al., 1968; Das et al., 1994).

Experimental Section

Material. The water used was first deionized by means of ion-exchange resins and then distilled in glass. Its conductivity was always below 1.0×10^{-6} S cm^{-1}. Diethylene glycol dimethyl ether (Spectrochem, Bombay) and 1,2-dimethoxyethane (Ubichem Ltd.) were dried over ferrous sulfate (A.R., BDH) and then fractionally distilled two times before use under reduced nitrogen gas pressure (Riddick et al., 1986; Perrin et al., 1980). Triethylene glycol dimethyl ether (E. Merck, F.R.G., zur Synthese) and tetraethylene glycol dimethyl ether (E. Merck, F.R.G., zur Synthese) were dried over 4A molecular sieves (Fluka, AG).

Table 1. Properties of Pure Liquids at 298.15 K

	ρ/kg m^{-3}		η/mPa s	
liquid	exptl	lit.	exptl	lit.
ethylene glycol dimethyl ether	862.6	862.62[a] 862.1[b] 861.32[c] 860.5[d]	0.420	0.424[c] 0.432[d]
diethylene glycol dimethyl ether	939.1	939.24[e] 939.9[f] 939.4[g] 938.4[b]	1.003	0.991[f] 0.981[g] 0.989[b]
triethylene glycol dimethyl ether	981.1	981.71[e] 979.50[h]	2.009	1.960[h]
tetraethylene glycol dimethyl ether	1006.3	1006.27[e] 1004.7[i]	3.294	3.295[i]

[a] Douhéret et al. (1993). [b] Riddick et al. (1986). [c] Das et al. (1994). [d] Wallace and Mathews (1963). [e] Dethlefsen and Hvidt (1985). [f] Aminabhavi and Gopalakrishna (1995). [g] de Ruiz Holgado et al. (1994). [h] Wallace et al. (1968). [i] McGee et al. (1983).

Estimated purities determined by gas chromatographic analysis were better than 99.5 mol % for all the liquid samples. Karl Fischer titration gave water contents of <0.01 mass %. The densities and viscosities of these liquids are compared with the literature values in Table 1. All samples were kept tightly sealed to minimize, as far as possible, the absorption of atmospheric moisture and CO_2. Before the measurements, all liquids were partially degassed under vacuum. The composition of each mixture was obtained with an accuracy of 1×10^{-4} from the measured masses of the components. All the mass measurements were performed on an electronic balance (Dhona 200 D, India) accurate to 0.01 mg. Corrections were made for buoyancy.

Measurements. Densities (ρ) of pure liquids were measured using a double-armed pycnometer having a capacity of 15 cm^3. The pycnometer was calibrated at 298.15 K with doubly distilled water. An average of

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* To whom correspondence should be addressed.

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Table 2. Experimental Viscosities, η, Viscosity Deviations, $\Delta\eta$, and Grunberg–Nissan Interaction Parameters, d_{12}, for Water (1) + Glycol Ether (2) Solutions at 298.15 K

x_2	η/mPa s	$\Delta\eta$/mPa s	d_{12}	x_2	η/mPa s	$\Delta\eta$/mPa s	d_{12}	x_2	η/mPa s	$\Delta\eta$/mPa s	d_{12}
				H_2O (1) + $CH_3O(CH_2)_2OCH_3$ (2)							
0.0070	1.018	0.131	20.11	0.2002	2.001	1.205	6.00	0.5617	0.707	0.081	0.78
0.0184	1.189	0.307	16.80	0.2402	1.782	1.005	4.79	0.6130	0.638	0.036	0.54
0.0306	1.376	0.500	15.45	0.2886	1.550	0.795	3.76	0.6418	0.605	0.017	0.42
0.0542	1.712	0.847	13.55	0.3482	1.256	0.529	2.67	0.6575	0.586	0.005	0.34
0.0775	1.968	1.114	11.91	0.3836	1.128	0.418	2.22	0.7264	0.536	−0.013	0.20
0.1063	2.146	1.306	10.10	0.4226	1.005	0.313	1.80	0.8227	0.491	−0.012	0.17
0.1301	2.252	1.423	9.06	0.4689	0.886	0.216	1.40	0.8646	0.471	−0.013	0.11
0.1654	2.164	1.352	7.34	0.5203	0.780	0.134	1.04	0.9633	0.433	−0.004	0.08
				H_2O (1) + $CH_3\{O(CH_2)_2\}_3CH_3$ (2)							
0.0091	1.139	0.248	27.21	0.2597	2.661	1.741	5.53	0.6590	1.272	0.307	1.24
0.0188	1.403	0.511	24.52	0.3028	2.389	1.465	4.51	0.7106	1.205	0.235	1.06
0.0295	1.691	0.797	22.28	0.3461	2.153	1.224	3.72	0.7695	1.145	0.168	0.90
0.0510	2.246	1.400	19.45	0.3843	1.982	1.048	3.19	0.8171	1.103	0.221	0.78
0.0854	2.993	2.093	15.39	0.4327	1.791	0.852	2.64	0.8712	1.066	0.078	0.68
0.1194	3.328	2.424	12.41	0.4651	1.691	0.748	2.36	0.9453	1.024	0.027	0.52
0.1604	3.232	2.324	9.43	0.5196	1.536	0.587	1.94				
0.2099	2.993	2.079	7.16	0.6080	1.356	0.397	1.46				
				H_2O (1) + $CH_3\{O(CH_2)_2\}_3OCH_3$ (2)							
0.0057	1.058	0.161	29.66	0.1215	4.581	3.555	14.42	0.4857	2.706	1.272	2.87
0.0124	1.278	0.374	28.69	0.1423	4.678	3.629	12.65	0.5930	2.598	1.044	2.11
0.0176	1.444	0.534	27.13	0.1607	4.625	3.555	11.25	0.6382	2.311	0.707	1.88
0.0247	1.737	0.819	26.91	0.1907	4.512	3.408	9.51	0.7020	2.269	0.593	1.74
0.0326	2.063	1.136	25.80	0.2288	4.179	3.033	7.71	0.7475	2.220	0.493	1.62
0.0455	2.613	1.672	23.94	0.2745	3.189	2.622	6.19	0.7813	2.166	0.402	1.48
0.0673	3.462	2.496	20.76	0.3074	3.588	2.354	5.37	0.8141	2.121	0.320	1.36
0.8262	4.032	3.045	18.29	0.3653	3.236	1.937	4.28	0.8937	2.068	0.178	1.22
0.1017	4.344	3.340	16.44	0.4097	3.035	1.686	3.69	0.9665	2.035	0.063	1.19
				H_2O (1) + $CH_3\{O(CH_2)_2\}_4OCH_3$ (2)							
0.0053	1.082	0.178	35.72	0.1238	6.387	5.165	16.67	0.5962	3.839	1.350	2.83
0.0132	1.424	0.498	34.73	0.1510	6.493	5.198	13.96	0.6747	3.662	0.962	2.42
0.0263	2.173	1.212	33.50	0.1780	6.376	5.008	11.86	0.7566	3.524	0.605	2.09
0.383	2.931	1.938	31.00	0.2201	6.024	4.543	9.46	0.8127	3.436	0.366	1.88
0.0507	3.724	2.698	28.35	0.2593	5.661	4.075	7.86	0.8330	3.402	0.278	1.79
0.0640	4.511	3.449	25.69	0.3289	5.077	3.305	5.94	0.9193	3.352	−0.004	1.64
0.0658	4.591	3.524	25.28	0.3723	4.782	2.893	5.11	0.5943	3.326	−0.123	1.56
0.0770	5.162	4.065	23.31	0.4051	4.585	2.608	4.60	0.9742	3.306	−0.197	1.43
0.0865	5.540	4.418	21.70	0.4349	4.431	2.374	4.21				
0.1017	6.014	4.851	19.45	0.5545	3.959	1.582	3.10				

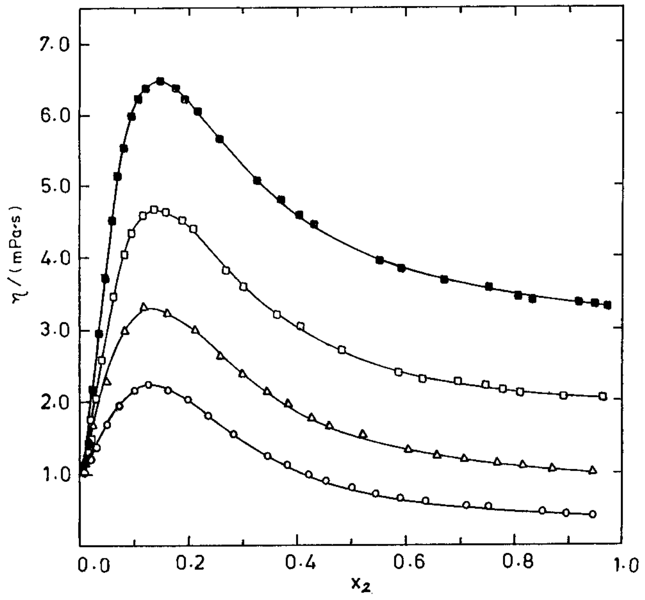

Figure 1. Viscosities (η) for $\{x_1H_2O + x_2CH_3(OCH_2CH_2)_nOCH_3\}$ at 298.15 K: (○) $n = 1$; (△) $n = 2$; (□) $n = 3$; (■) $n = 4$.

triplicate measurements was taken into account, and these are reproducible to ±0.3 kg m^{-3}. The density ρ of the mixture at any composition and excess volume derived from the density values were reported at 298.15 K by Dethlefsen and Hvidt (1985) and Douhéret et al. (1993).

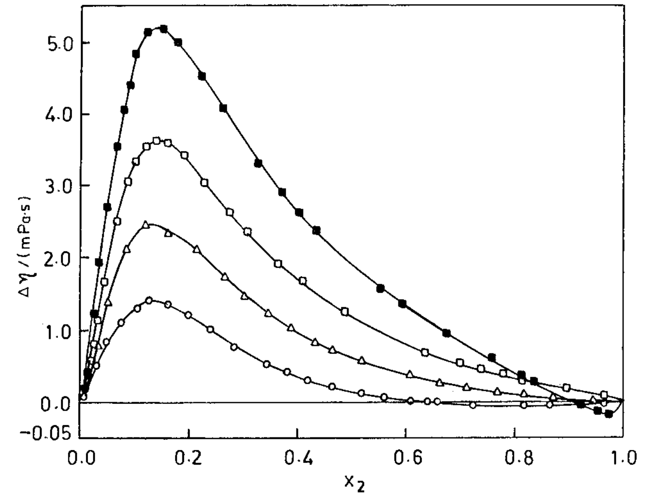

Figure 2. Viscosity deviations ($\Delta\eta$) for $\{x_1H_2O + x_2CH_3(OCH_2CH_2)_nOCH_3\}$ at 298.15 K: (○) $n = 1$; (△) $n = 2$; (□) $n = 3$; (■) $n = 4$. The solid curves have been calculated from eq 2.

The measurements of the viscosity of pure liquids and the binary systems were made with a modified suspended level Ubbelohde viscometer. The viscometer was calibrated, and two constants, A and B, of the viscometer in the relation $\eta = \rho(At - B/t)$ were obtained by measuring the flow time t with high-purity benzene, toluene, and water at the working temperature. The average of four or five sets of flow times for each fluid was taken for the purpose of the calculation of viscosity. The flow time was

Table 3. Smoothing Coefficients A_i and Standard Deviations σ for Eq 2 for Binary Mixtures at 298.15 K

function	A_0	A_1	A_2	A_3	A_4	A_5	σ
			H_2O (1) + $CH_3O(CH_2)_2OCH_3$ (2)				
$\Delta\eta$/mPa s	0.5570	−2.4732	7.3107	−14.1205	4.1239	7.0200	0.044
			H_2O (1) + $CH_3\{O(CH_2)_2\}_2OCH_3$ (2)				
$\Delta\eta$/mPa s	2.4744	−4.1721	10.1196	−22.3062	8.0289	9.9848	0.089
			H_2O (1) + $CH_3\{O(CH_2)_2\}_3OCH_3$ (2)				
$\Delta\eta$/mPa s	4.6915	−4.3772	16.8216	−45.5133	6.7964	32.2351	0.167
			H_2O (1) + $CH_3\{O(CH_2)_2\}_4OCH_3$ (2)				
$\Delta\eta$/mPa s	7.2355	−9.1001	24.1476	−52.4700	5.9565	28.6456	0.245

measured with an accurate stopwatch having a precision of ±0.01 s. Viscosities are reproducible to ±0.003 mPa s. The performance of the viscometer was assessed by measuring and comparing the viscosities of the pure components with the values reported in the literature (Riddick et al., 1986; Timmerman, 1950, 1962). A thermostatically controlled, well-stirred water bath whose temperature was controlled to ±0.01 K was used for all the measurements. The density and viscosity of water at 298.15 K were taken from the literature (Riddick et al., 1986; Kell, 1967).

Results and Discussion

The experimental values of viscosity at various mole fractions of organic solvents at 298.15 K are reported in Table 2. Our results for the water + ethylene glycol dimethyl ether system agree in the amphiphile-rich region with the data reported at 298.15 K by Das et al. (1994) and Wallace and Mathews (1963), although their results are rather sparse at mole fraction $x_2 < 0.35$. The results for water + triethylene glycol dimethyl and +tetraethylene glycol dimethyl ethers reported by Wallace et al. (1968) and McGee et al. (1983) at 298.15 K using an Ostwald viscometer are in agreement with our results over the whole mole fraction range.

The experimental values of η for the various mixtures have been used to calculate the values of the viscosity deviations from a mole fraction average, $\Delta\eta$, for the mixtures from the relation

$$\Delta\eta = \eta - (x_1\eta_1 + x_2\eta_2) \tag{1}$$

where η is the dynamic viscosity of the binary mixture and x_1 and x_2 are the mole fractions and η_1 and η_2 the dynamic viscosities of components 1 and 2, respectively. The variation of η and $\Delta\eta$ with mole fraction of organic solvents at 298.15 K are shown in Figures 1 and 2. The $\Delta\eta$ values over the whole mole fraction range were fitted to the Redlich−Kister (1948) polynomial equation

$$\Delta\eta = x_1x_2\sum_{i=0}^{m} A_i(x_2 - x_1)^i \tag{2}$$

The coefficients A_i of eq 2 were calculated by the method of least squares. The values obtained for the coefficients A_i along with the standard deviation σ are reported in Table 3.

According to Grunberg and Nissan (1949), the value of η for a binary mixture of components 1 and 2 is given by

$$\ln \eta = x_1 \ln \eta_1 + x_2 \ln \eta_2 + x_1x_2d_{12} \tag{3}$$

where d_{12} is a constant proportional to the interaction energy and is regarded as a measure of the strength of interactions between the mixing species. The values of d_{12}, as estimated from eq 3 are included in the last column of Table 2.

Table 4. Values of Adjustable Parameters and Standard Deviations for Viscosity Models at 298.15 K

	mPa s			
equation	*a*	*b*	*c*	σ
	H_2O (1) + $CH_3O(CH_2)_2OCH_3$ (2)			
Heric	2.257	−5.472	7.061	0.089
McAllister	0.160	13.390		0.327
	H_2O (1) + $CH_3\{O(CH_2)_2\}_2OCH_3$ (2)			
Heric	3.411	−6.859	9.304	0.171
McAllister	0.321	32.219		0.642
	H_2O (1) + $CH_3\{O(CH_2)_2\}_3OCH_3$ (2)			
Heric	4.245	−8.148	12.415	0.208
McAllister	0.377	138.923		1.221
	H_2O (1) + $CH_3\{O(CH_2)_2\}_4OCH_3$ (2)			
Heric	4.929	−10.547	14.805	0.289
McAllister	0.321	493.648		2.218

Assuming a three-body interaction model, McAllister (1960) proposed the following relation for the viscosity of mixtures

$$\ln \lambda = x_1^3 \ln \lambda_1 + 3x_1^2x_2 \ln a + 3x_1x_2^2 \ln b + x_2^3 \ln \nu_2 - \ln[x_1 + x_2M_2/M_1] + 3x_1^2x_2 \ln[(2 + M_2/M_1)/3] + 3x_1x_2^2 \ln[(1 + 2M_2/M_1)/3] + x_2^3 \ln(M_2/M_1) \tag{4}$$

where λ ($=\eta/\rho$) refers to the kinematic viscosity of the mixture and λ_1 and λ_2 are the kinematic viscosities of the pure components 1 and 2, respectively; *a* and *b* are adjustable parameters which are characteristic of the system.

Heric (1966) proposed the following relation for correlating the kinematic viscosity of the binary liquid mixture

$$\ln \lambda = x_1 \ln(\lambda_1M_1) + x_2 \ln(\lambda_2M_2) - \ln(x_1M_1 + x_2M_2) + x_1x_2[a + b(x_1 - x_2) + C(x_1 - x_2)^2] \tag{5}$$

in which M_1 and M_2 are molecular weights of components 1 and 2. In the above equations (4) and (5), the coefficients *a*, *b*, and *c* have been calculated using the least-squares procedure. The estimated parameters of the viscosity equations and the standard errors, σ, between the calculated and experimental values are given in Table 4. As can be clearly seen, Heric's formula fits the experimental results better as compared to the McAllister one as the σ values for the latter equation are larger in all the systems.

The values of $\Delta\eta$ presented in Figure 2 are positive for all the mixtures except at higher x_2, while the excess molar volumes for the same systems, derived from densities (Dethlefsen and Hvidt, 1985; Douhéret et al. 1993) are negative over the entire compositive range at 298.15 K. There is a very obvious increase in the magnitudes of η and $\Delta\eta$ with each addition of a $-OC_2H_4-$ group in the molecule of ethylene glycol dimethyl ether. It is also remarkable that the variations of V_m^E with each addition of a $-OC_2H_4-$ group in this homologous series are not as clear as found in the literatures. Negative excess molar volumes and rather high values of η and $\Delta\eta$ must be attributed mainly to disruption of (i) intramolecular hydrogen bonds and intramolecular dipolar interactions in amphiphile and (ii) the hydrogen bonds present in the self-associated water molecules. The presence of the etheric oxygen enhances the ability of the −OH group of the glycol to form hydrogen bonds with water molecules. The order of strength of the hydrogen bonding owing to increased molecular weight and polar head group size is as follows: $(n = 4) > (n = 3) > (n = 2) > (n = 1)$, while in the same sequence, the maximum in viscosity is shifted to lower values of x_2. That is, the maximum in viscosity in the range $0.12 > x_2 > 0.16$ increases as the ethers become more complex. The sharp changes of these properties with amphiphile content in water-rich mixtures would imply more order and hence more structure or clustering in an aqueous environment. This leads to substantial increases in the viscosities of the mixtures. Further, the molar volumes of water and glycols at 298.15 K are quite large, which might allow the glycol molecules to occupy the cavities in a more structured water lattice, thereby giving rise to a marked change in their η and $\Delta\eta$ values at the water-rich region. This change continues until a concentration is reached at which all cavities are filled. Also the Grunberg−Nissan parameter d_{12} for the four systems at 298.15 K show always positive, which supports the formation of intermolecular complexes between the unlike molecules through hydrogen bonding.

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