



Volumetric and viscosimetric properties of the binary mixture of triethylene glycol monomethyl ether + water at $T = (293.15, 303.15, 313.15, 323.15, 333.15)$ K under atmospheric pressure

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ABSTRACT

This paper reports new measurements of densities and viscosities for the binary liquid mixture of triethylene glycol monomethyl ether $\text{CH}_3\text{O}(\text{CH}_2)_2\text{O}(\text{CH}_2)_2\text{O}(\text{CH}_2)_2\text{OH}$ + water as a function of composition at $T = (293.15, 303.15, 313.15, 323.15, 333.15)$ K under atmospheric pressure. Densities were determined using a capillary pycnometer. Viscosities were measured with Ubbelohde capillary viscometer. From the experimental data, the excess molar volumes V^E , and viscosity deviations $\delta\eta$, and the excess energies of activation for viscous flow ΔG^{*E} were calculated. These data have been correlated by the Redlich–Kister type equations to obtain their coefficients and standard deviations. The results suggest that molecular interaction between triethylene glycol monomethyl ether and water is stronger than that of diethylene glycol monomethyl ether and water.

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1. Introduction

Alkoxyethanols, with the combination of ether, alcohol and hydrocarbon chain in one molecule, provide versatile characteristics with both polar and non-polar properties. This kind of short-chain glycol ethers is theoretical significance to study molecular interactions and arrangements through the mixing deviation from ideality [1]. They can also be used for the research of the hydration of polyether (PEO). As is summarized by Fenn et al. [2], PEO is important in many industrial, environmental, and biological applications because of its specific hydrophilic properties. The hydration of PEO ascribes to terminal hydroxyl groups as well as the ether oxygens along the polymer backbone. To explicate the role played by the polyether backbone, the authors had studied tetraethylene glycol dimethyl ether/water systems by using ultrafast infrared (IR) polarization selective pump-probe spectroscopy of water's hydroxyl stretch.

This paper is a continuation of our systematic program on the thermodynamic study of the binary mixtures of glycol ethers with water. Glycol ethers are widely used industrial solvents with the highest potential for gas sweetening [3]. They can be used as scrubbing liquids in the cleaning of exhaust air and gas streams from industrial production plants because of their favorable properties such as low vapor pressure, low toxicity, low viscosity, high

chemical stability, and low melting temperature [4]. Binary mixtures of glycol ethers and water have been used in the absorption of carbonyl sulfide (COS) from synthesis gas for reasons of either protecting catalyst activity in subsequent operations or preventing corrosion and air pollution. The key advantage of using the binary mixtures of glycol ethers and water as absorption liquids of COS is that both solubility and hydrolysis rate are enhanced. Triethylene glycol monomethyl ether is a physical solvent which has a strong affinity for CO_2 . It could also be used in a combination with a chemical solvent (amine), with or without water in mixed solvents [5,6]. Process design using these systems requires accurate thermophysical property data.

In addition, glycol ethers are nonionic amphiphile molecules, very effective as surfactants with a large number of applications [7]. They can be used as polar additives in anionic polymerization and automotive brake fluid. Short-chain polyethylene glycol monoalkylethers are used in various biotechnical and biomedical applications, constituting a simple model of biological systems [8].

2. Experimental

2.1. Materials

Triethylene glycol monomethyl ether (TEGMME, CAS 112-35-6) was purchased from Fluka, and its mass fraction purity was 98.5%. After purified by vacuum distillation, the purity of TEGMEE was $\text{GC} > 99\%$. Prior to measurements, it was dried over 0.4 nm molecular sieves and partially degassed under vacuum. Doubly distilled water with its conductivity lower than $10^{-7} \Omega^{-1} \cdot \text{cm}^{-1}$ was used.

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Table 1

Comparison of measured densities (ρ), viscosities (η) of TEGMME with literature values at $T = (298.15, 308.15, 313.15, 323.15, 333.15)$ K.

T/K	$\rho/\text{g cm}^{-3}$		$\eta/\text{mPa s}$	
	This work	Lit	This work	Lit
298.15	1.0432	1.04524 ^a 1.0414 ^b 1.04304 ^d 1.0430 ^e 1.04310 ^f 1.04491 ^g	6.256	6.253 ^a 6.318 ^b 6.240 ^d 6.586 ^e
308.15	1.0339	1.03262 ^a 1.0328 ^b 1.0343 ^e 1.03611 ^g	4.627	4.719 ^b 5.055 ^c 4.642 ^e
313.15	1.0296	1.02908 ^a	4.083	4.074 ^a
323.15	1.0209	1.02127 ^a	3.235	3.202 ^a
333.15	1.0124	1.01327 ^a	2.607	2.618 ^a

^a [9] A. Henni, P. Tontiwachwuthikul, A. Chakma.

^b [6] S. Aznarez, M.M.E.F. de Ruiz Holgado, E.L. Arancibia.

^c [10] A. Pal, Y.P. Singh.

^d [11] A. Henni, Y. Maham, P. Tontiwachwuthikul, A. Chakma, A.E. Mather.

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^f [13] G. Douhéret, P. Lajoie, M.I. Davis, J.L. Ratliff, J. Ulloa, H. Høiland, I.J. Fjellanger.

^g [14] T. Singh, A. Kumar.

2.2. Apparatus and procedure

The densities of the pure liquids and the mixtures were measured with a 10 cm³ capillary pycnometer. Degassed pure water was used as calibrating substances. A thermostatically controlled, well-stirred water bath whose temperature was controlled to ± 0.01 K was used for all the density measurements and the following measurements of viscosity. Binary mixtures were prepared by mass, using an electronic analytical balance (HANGPING FA2104, Shanghai, China) with a precision of ± 0.0001 g. The uncertainty in mole fraction was estimated to be ± 0.0001 . The uncertainty of the density measurements was estimated to be ± 0.1 %.

The viscosities were determined with a capillary viscometer of Ubbelohde type which was checked by measurement of the viscosity of pure water. The flow-time measurements were made using an accurate stopwatch with a precision of ± 0.01 s. The average of five or six sets of flow times for each fluid was taken for the purpose of the calculation of viscosity. The flow times were reproducible to ± 0.06 s.

Table 2

Densities ρ and viscosities η for the mixture of TEGMME (1) + water (2) at $T = (293.15, 303.15, 313.15, 323.15, 333.15)$ K.

x_1	$\rho/\text{g cm}^{-3}$					$\eta/\text{mPa s}$				
	T/K = 293.15	303.15	313.15	323.15	333.15	293.15	303.15	313.15	323.15	333.15
0.0000	0.9982	0.9957	0.9922	0.9881	0.9832	1.005	0.8007	0.656	0.5494	0.4688
0.01046	1.0098	1.0062	1.0016	0.9967	0.9909	1.629	1.205	0.9579	0.7879	0.6504
0.02067	1.0181	1.0133	1.0083	1.0025	0.9964	2.223	1.581	1.246	0.9959	0.7873
0.03394	1.0281	1.0221	1.0155	1.0089	1.0025	2.973	2.123	1.622	1.288	0.9540
0.04952	1.0371	1.0299	1.0227	1.0159	1.0086	3.758	2.666	1.998	1.538	1.163
0.06922	1.0458	1.0384	1.0303	1.0221	1.0144	4.796	3.335	2.457	1.873	1.455
0.1011	1.0533	1.0451	1.0371	1.0288	1.0205	6.215	4.247	3.084	2.291	1.831
0.1416	1.0584	1.0501	1.0418	1.0334	1.0249	7.762	5.413	3.793	2.834	2.165
0.2058	1.0602	1.0521	1.0433	1.0347	1.0264	9.057	6.345	4.572	3.418	2.615
0.3024	1.0588	1.0502	1.0414	1.0326	1.0241	9.855	6.821	4.927	3.668	2.801
0.3835	1.0573	1.0485	1.0393	1.0306	1.0221	9.886	6.881	4.962	3.712	2.875
0.4966	1.0549	1.0461	1.0371	1.0282	1.0197	9.399	6.537	4.796	3.668	2.844
0.5979	1.0533	1.0443	1.0352	1.0265	1.0179	8.987	6.272	4.662	3.576	2.824
0.6775	1.0521	1.0431	1.0341	1.0252	1.0167	8.642	6.122	4.536	3.533	2.782
0.7996	1.0505	1.0415	1.0324	1.0236	1.0151	8.207	5.821	4.363	3.408	2.698
0.8982	1.0491	1.0402	1.0311	1.0223	1.0138	7.791	5.581	4.202	3.296	2.637
1.0000	1.0476	1.0387	1.0296	1.0209	1.0124	7.458	5.375	4.083	3.235	2.607

Table 3

Parameters in Eqs. (1) and (2) for density and viscosity data of TEGMME (1) + water (2).

x_1	a_0	$a_1 \cdot 10^4$	$\sigma/\text{g cm}^{-3}$	$\eta_0 \cdot 10^4$	$E_a/\text{kJ mol}^{-1}$	$\sigma/\text{mPa s}$
0.01046	1.1492	-4.73	0.0007	6.5	19.06	0.030
0.02067	1.1775	-5.42	0.0005	3.5	21.30	0.044
0.03394	1.2171	-6.44	0.0002	2.7	22.70	0.041
0.04952	1.2452	-7.10	0.0001	2.2	23.78	0.032
0.06922	1.2779	-7.91	0.0002	1.8	24.76	0.057
0.1011	1.2934	-8.19	0.0001	1.5	25.88	0.092
0.1416	1.3038	-8.37	0.00008	1.5	26.49	0.059
0.2058	1.3095	-8.50	0.0002	2.5	25.59	0.051
0.3024	1.3139	-8.70	0.0001	2.3	25.97	0.073
0.3835	1.3161	-8.83	0.00002	2.6	25.66	0.082
0.4966	1.3137	-8.83	0.00002	3.5	24.82	0.11
0.5979	1.3129	-8.86	0.00002	4.4	24.17	0.13
0.6775	1.3120	-8.87	0.00002	5.4	23.58	0.11
0.7996	1.3104	-8.87	0.00002	6.1	23.14	0.11
0.8982	1.3084	-8.85	0.00002	7.3	22.58	0.10
1.0000	1.3060	-8.82	0.0002	9.2	21.92	0.11

The uncertainty of the viscosity measurements was estimated to be ± 0.2 %.

3. Results and discussion

A comparison of our measurements of density and viscosity with the data in the literature was shown in Table 1. The viscosity of pure TEGMME at 298.15 and 308.15 K was also measured for comparison. A reasonable agreement was found between our experimental values and those of the literatures.

The experimental results of the densities and viscosities from (293.15 to 333.15) K at various mole fractions are listed in Table 2.

In order to show the temperature dependence of density and viscosity, the measured densities are presented as functions of temperature by the linear relation, Eq. (1), while viscosities are presented by Arrhenius-like equation, Eq. (2) [15].

$$\rho = a_0 + a_1(T/K) \quad (1)$$

$$\eta = \eta_0 e^{\frac{E_a}{RT}} \quad (2)$$

where a_0 , a_1 , η_0 , and E_a are the undetermined parameters. The parameters in Eqs. (1) and (2) for density and viscosity data of mixture

Table 4Excess molar volumes V^E , and viscosity deviations $\delta\eta$ for the mixture of TEGMME (1) + water (2) at $T = (293.15, 303.15, 313.15, 323.15, 333.15)$ K.

x_1	$V^E/\text{cm}^3 \text{ mol}^{-1}$					$\delta\eta/\text{mPa s}$				
	$T/\text{K} = 293.15$	303.15	313.15	323.15	333.15	293.15	303.15	313.15	323.15	333.15
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.000	0.000	0.000	0.000	0.000
0.01046	-0.1439	-0.1335	-0.1220	-0.1149	-0.1041	0.5565	0.3565	0.2661	0.2104	0.1592
0.02067	-0.2517	-0.2260	-0.2144	-0.1955	-0.1840	1.085	0.6858	0.5192	0.3910	0.2743
0.03394	-0.4063	-0.3644	-0.3274	-0.2983	-0.2865	1.749	1.167	0.8497	0.6475	0.4126
0.04952	-0.5649	-0.5043	-0.4615	-0.4351	-0.4084	2.433	1.639	1.172	0.8556	0.5883
0.06922	-0.7459	-0.6893	-0.6324	-0.5776	-0.5467	3.344	2.218	1.564	1.138	0.8382
0.1011	-0.9343	-0.8665	-0.8231	-0.7731	-0.7320	4.558	2.984	2.082	1.470	1.146
0.1416	-1.107	-1.047	-1.006	-0.9612	-0.9197	5.843	3.965	2.652	1.904	1.393
0.2058	-1.221	-1.184	-1.137	-1.093	-1.067	6.724	4.603	3.211	2.316	1.706
0.3024	-1.221	-1.178	-1.144	-1.099	-1.070	6.899	4.637	3.235	2.306	1.686
0.3835	-1.173	-1.128	-1.077	-1.043	-1.019	6.406	4.326	2.992	2.133	1.586
0.4966	-1.026	-0.9935	-0.9677	-0.9243	-0.9061	5.189	3.465	2.438	1.785	1.313
0.5979	-0.8861	-0.8448	-0.8185	-0.798	-0.7745	4.124	2.736	1.957	1.421	1.077
0.6775	-0.7518	-0.7164	-0.7069	-0.6693	-0.6589	3.265	2.222	1.558	1.164	0.8646
0.7996	-0.5256	-0.4989	-0.4871	-0.4655	-0.4599	2.042	1.363	0.9668	0.7112	0.5195
0.8982	-0.2902	-0.2833	-0.2779	-0.2597	-0.2572	0.9899	0.6717	0.4679	0.3344	0.2477
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.000	0.000	0.000	0.000	0.000

of TEGMME with water were obtained by fitting the equations to the experimental values with a least-squares method, listed in Table 3.

Excess molar volumes V^E and viscosity deviations $\delta\eta$ were calculated from the experimental results according to the following equations, respectively

$$V^E = \frac{x_1 M_1 + x_2 M_2}{\rho_M} - \frac{x_1 M_1}{\rho_1} - \frac{x_2 M_2}{\rho_2} \quad (3)$$

$$\delta\eta = \eta_M - (x_1 \eta_1 + x_2 \eta_2) \quad (4)$$

where x_1 and x_2 are the mole fractions; M_1 and M_2 are molar masses; ρ_1 and ρ_2 are the densities; and η_1 and η_2 are the viscosities of pure components 1 and 2, respectively. The subscript M represents mixture properties. The result of V^E and $\delta\eta$ given in Table 4.

On the basis of the theory of absolute reaction rates, the excess Gibbs free energy of activation of viscous flow (ΔG^{*E}) was calculated by using the following equation: [16]

$$\Delta G^{*E} = RT[\ln(\eta V) - x_1 \ln(\eta_1 V_1) - x_2 \ln(\eta_2 V_2)] \quad (5)$$

where R is the universal constant of gases; T is the absolute temperature; V , V_1 and V_2 are the molar volumes of the binary mixtures and pure component 1 and 2, respectively.

The values of V^E , $\delta\eta$, and ΔG^{*E} were correlated by a Redlich–Kister type polynomial: [17]

$$Y = x_1 x_2 \sum_{k=0}^m A_k (x_1 - x_2)^k \quad (6)$$

where $Y = V^E$, $\delta\eta$, or ΔG^{*E} , and the coefficients of A_k are parameters that were obtained by fitting the equations to the experimental values with a least-squares method, which are given in Table 5.

The correlated results for densities, viscosities, excess mole volumes, viscosity deviations and excess Gibbs free energy of activation of viscous flow were also given in Tables 3 and 5, in which the tabulated standard deviation σ was defined as

$$\sigma = \left[\frac{\sum (Y_{\text{exp}} - Y_{\text{cal}})^2}{n-p} \right]^{1/2} \quad (7)$$

where Y refers to ρ , η , V^E , $\delta\eta$, or ΔG^{*E} ; n is the number of data points; and p is the number of coefficients. The subscripts exp and cal represent the experimental value and the calculated value, respectively.

Fig. 1 shows that the excess molar volumes are negative over the entire range of composition with a minimum around mole fraction 0.26 for TEGMME at all temperatures. The negative V^E indicate that there is a volume contraction on mixing. Fig. 2 shows that the viscosity deviations are positive over the entire range of composition with a maximum around mole fraction of 0.28 for TEGMME at all

Table 5

Coefficients of the Redlich–Kister equation and standard deviation for excess molar volume, viscosity deviation, and excess free energies of activation for TEGMME (1) + water (2).

T/K	Property	A_0	A_1	A_2	A_3	A_4	A_5	σ
293.15	$V^E/\text{cm}^3 \text{ mol}^{-1}$	-4.0384	2.5322	-4.0752	3.1262			0.013
	$\delta\eta/\text{mPa s}$	20.5822	-21.1956	22.1726	-15.0311	-10.0787	15.3718	0.051
	$\Delta G^{*E}/\text{kJ mol}^{-1}$	17.0510	-17.5577	6.5699	17.0516	25.6664	-52.7755	0.15
303.15	$V^E/\text{cm}^3 \text{ mol}^{-1}$	-3.9027	2.6843	-3.7155	2.2221			0.011
	$\delta\eta/\text{mPa s}$	13.7372	-14.2897	16.4675	-11.3951	-9.1138	12.6215	0.046
	$\Delta G^{*E}/\text{kJ mol}^{-1}$	16.7372	-16.7802	7.8698	11.9281	21.9545	-43.5863	0.12
313.15	$V^E/\text{cm}^3 \text{ mol}^{-1}$	-3.7981	2.6125	-3.4508	1.8012			0.013
	$\delta\eta/\text{mPa s}$	9.6912	-10.0087	10.5926	-5.3549	-5.4246	4.8791	0.024
	$\Delta G^{*E}/\text{kJ mol}^{-1}$	16.5536	-16.4954	7.4529	12.8955	20.8836	-43.5275	0.13
323.15	$V^E/\text{cm}^3 \text{ mol}^{-1}$	-3.6723	2.6191	-3.1208	104063			0.015
	$\delta\eta/\text{mPa s}$	7.0330	-6.8836	7.4285	-3.8064	-3.7409	2.4750	0.031
	$\Delta G^{*E}/\text{kJ mol}^{-1}$	16.3615	-15.9424	7.4393	12.3340	19.4631	-42.0450	0.13
333.15	$V^E/\text{cm}^3 \text{ mol}^{-1}$	-3.5983	2.6037	-2.9414	1.0271			0.016
	$\delta\eta/\text{mPa s}$	5.2218	-4.6151	5.3017	-5.8360	-2.5214	5.5823	0.020
	$\Delta G^{*E}/\text{kJ mol}^{-1}$	16.1295	-14.5781	7.9889	3.6887	17.1557	-28.9468	0.095

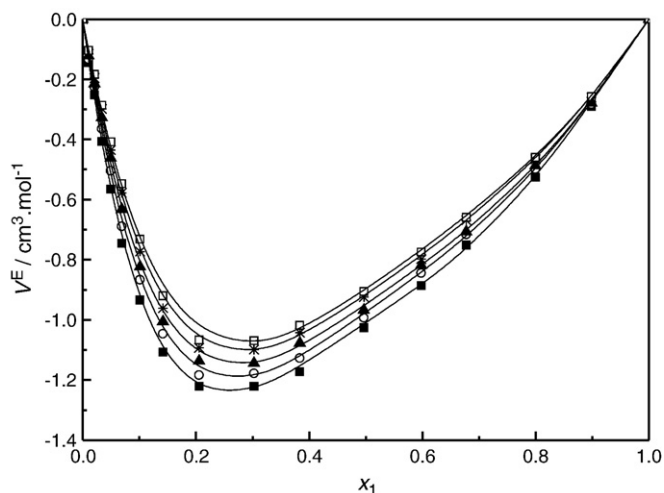


Fig. 1. Plot of excess molar volume V^E against mole fraction x of TEGMME (1) + water (2). At $T=293.15$ K (■); 303.15 K (○); 313.15 K (▲); 323.15 K (*); 333.15 K (□). The symbols represent experimental values, and the solid curves represent the values calculated from Eq. (6).

temperatures. Both V^E and $\delta\eta$ indicate that the interaction between TEGMME and water molecular is strong. It can also be observed that this kind of interaction is affected by temperature and composition. The interaction becomes stronger with decreasing temperature. The effect on viscosity of temperature is sharper than that on density.

The excess Gibbs free energy of activation of viscous flow, ΔG^{*E} , is positive over the whole mole fraction range for the binary mixtures at different temperatures, Fig. 3. The sign of the values of ΔG^{*E} can be considered as a reliable criterion for detecting or excluding the presence of interaction between unlike molecules. The positive ΔG^{*E} values are also indicative of the strong molecular interaction between TEGMME and water.

As is suggested by other authors [8], the molecular interactions in the aqueous solution of alkoxyethanols are complex due to the presence of the O and OH groups in the same molecule, which allows self-association via intermolecular hydrogen bonds. The conformational behavior around the C—C bond and C—O bonds in oxyethylene chain in water suggest the presence of several types of hydrogen bonds between the molecule of amphiphile and water. ^1H NMR spectra of aqueous solutions of some n-alkoxyethanols (C_1E_m , $m=1, 2, 3$) over the whole

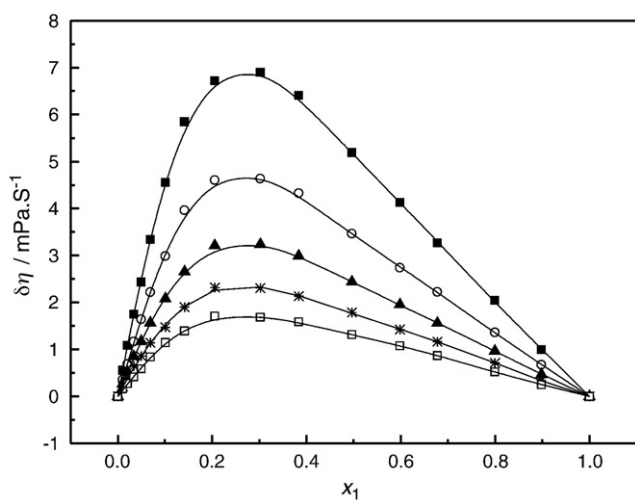


Fig. 2. Plot of viscosity deviation $\delta\eta$ against mole fraction x of TEGMME (1) + water (2). At $T=293.15$ K (■); 303.15 K (○); 313.15 K (▲); 323.15 K (*); 333.15 K (□). The symbols represent experimental values, and the solid curves represent the values calculated from Eq. (6).

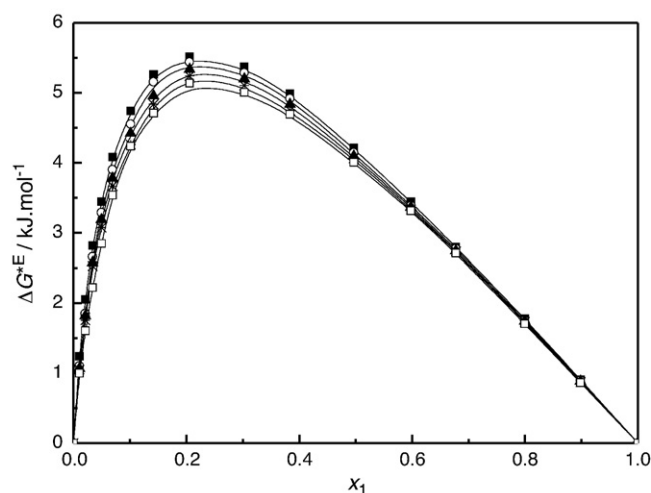


Fig. 3. Plot of the excess free energies of activation for viscous flow ΔG^{*E} against mole fraction x of TEGMME (1) + water (2). At $T=293.15$ K (■); 303.15 K (○); 313.15 K (▲); 323.15 K (*); 333.15 K (□).

composition range at 298.15 K had been used to studied the behavior of binary liquid mixtures. Spectroscopic results confirm the strong interactions between components involving hydrogen bonds [18]. Katsumoto et al. [19] had investigated the molecular interaction occurring in the system 2-butoxyethanol in water by infrared (IR) spectroscopy combined with quantum chemical calculations. They found that the blue shift of the ν_{CH} bands due to the butoxy group of C_4E_1 originates mainly from the formation of H-bonds between the ether oxygen atom and water molecules.

The properties of the TEGMME + water can be compared with that of diethylene glycol monomethyl ether (DEGMME) + water measured by us previously [20]. The negative values of the excess molar volumes and the positive values of the viscosity deviations are larger than that of DEGMME, which suggest that the interactions between TEGMME and water through hydrogen bonding is stronger than that of DEGMME and water. The only difference between TEGMME and DEGMME molecule lies in the former has three oxyethylene units while the latter has two. Thus it can be concluded that the molecular interaction of glycol ether with water is enhanced by increasing the oxyethylene units within the same hydrocarbon chain.

In our previous study, densities and viscosities for aqueous solutions of diethylene glycol monomethyl ether (DEGMME), diethylene glycol monoethyl ether (DEGMEE), diethylene glycol monobutyl ether

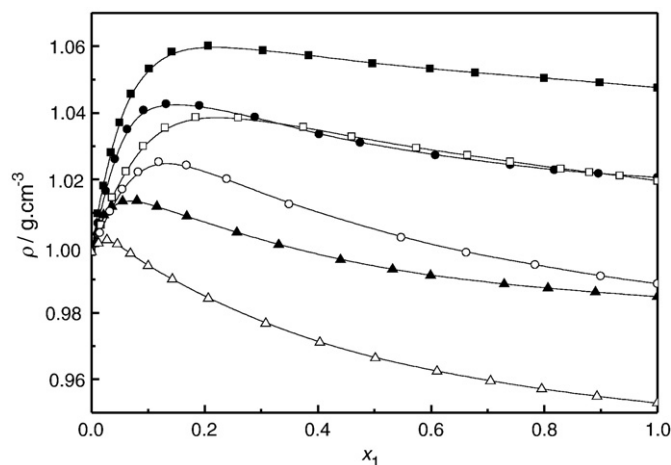


Fig. 4. Plot of density ρ against mole fraction x of alkoxyethanol for alkoxyethanol (1) + water (2) at $T=273.15$ K. TEGMME (■); TEGMEE (●); TEGMBE (▲); DEGMME (□); DEGMEE (○); DEGMBE (Δ).

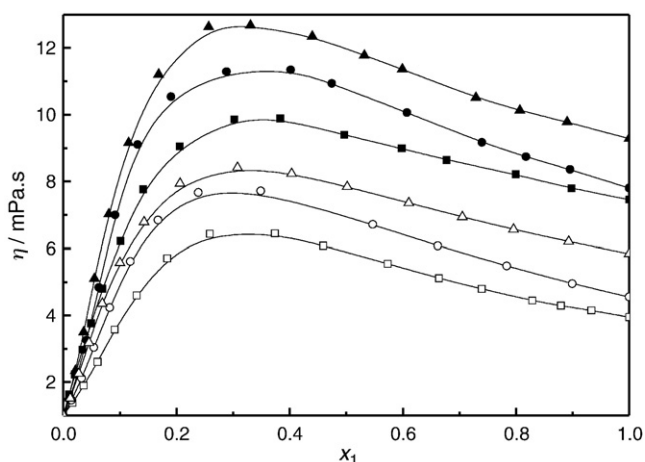


Fig. 5. Plot of viscosity η against mole fraction x of alkoxyethanol (1) + water (2) at $T = 273.15$ K. TEGMME (■); TEGMEE (●); TEGMBE (▲); DEGMME (□); DEGMEE (○); DEGMBE (△).

(DEGMBE), triethylene glycol monoethyl ether (TEGMEE) and triethylene glycol monobutyl ether (TEGMBE) had been measured over the entire composition at different temperatures. Figs. 4 and 5 give the comparison of densities and viscosities for aqueous solution of these alkoxyethanols at 293.15 K.

Fig. 4 shows clearly that the densities of pure alkoxyethanols and their aqueous solutions are affected by the molecular structure. The density of pure alkoxyethanol decreases as alkyl chain length increasing with the common oxyethylene units. For alkoxyethanols with the same alkyl chain, the density increases with increasing the number of oxyethylene units. The density value for aqueous solutions of alkoxyethanols increases sharply in water-rich region up to a maximum with increasing the mole fraction. The density maximum shifts to lower

alkoxyethanol mole fraction as alkyl chain length increasing with the common oxyethylene units. Fig. 5 gives some information about the effect of molecular structure on the viscosities of pure alkoxyethanols and their aqueous solutions. The viscosity of pure alkoxyethanol increases as alkyl chain length increasing with the common oxyethylene units. The viscosity of pure alkoxyethanol also increases with increasing the number of oxyethylene units with the same alkyl chain. The viscosity value for aqueous solutions of alkoxyethanols increases sharply up to a maximum with increasing the mole fraction. Therefore these macro phenomena suggest that the interaction forces between alkoxyethanol molecule and water molecule are complicated and affected by molecular structure.

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