

Excess Molar Volumes and Viscosities of Binary Liquid Mixtures of Ethylene Glycol Dimethyl Ether + Ethylene Glycol Monomethyl, + Diethylene Glycol Monomethyl, and + Triethylene Glycol Monomethyl Ethers at 298.15 and 308.15 K

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Excess molar volumes V_m^E and viscosities η have been measured as a function of mole fraction for binary liquid mixtures of ethylene glycol dimethyl ether (1,2-dimethoxyethane), $\text{CH}_3\text{OC}_2\text{H}_4\text{OCH}_3$, with diethylene glycol monomethyl ether (2-(2-methoxyethoxy)ethanol), $\text{CH}_3(\text{OC}_2\text{H}_4)_2\text{OH}$, and triethylene glycol monomethyl ether (2-[2-(2-methoxyethoxy)ethoxy]ethanol), $\text{CH}_3(\text{OC}_2\text{H}_4)_3\text{OH}$, at 298.15 and 308.15 K and at atmospheric pressure. Measurements of viscosity at 298.15 and 308.15 K and excess molar volume at 308.15 K have also been made for the mixtures ethylene glycol dimethyl ether + ethylene glycol monomethyl ether (2-methoxyethanol), $\text{CH}_3\text{OC}_2\text{H}_4\text{OH}$, over the whole composition range. Density ρ of the mixtures have been calculated from the results. All of the systems show negative deviation from a mole fraction average in viscosity. The excess molar volumes for all of the mixtures are negative over the entire composition range.

Introduction

As a part of our studies on thermodynamic, transport, and acoustic properties of mixtures containing polyethers, we have recently reported excess molar volumes, speeds of sound, and viscosities of binary mixtures of polyethers either with an alkoxyethanol (Pal et al., 1999) or with alkanol (Pal and Sharma, 1998a,b). These investigations were concerned with the sensitivity to the alkyl chain length as well as the polar headgroup of the composition dependence of a variety of thermodynamic properties and functions. In continuation of our studies on binary mixtures containing polyethers, we have measured excess molar volumes V_m^E and viscosity η for binary mixtures of ethylene glycol dimethyl ether ($\text{CH}_3\text{OC}_2\text{H}_4\text{OCH}_3$), with diethylene glycol monomethyl ether ($\text{CH}_3(\text{OC}_2\text{H}_4)_2\text{OH}$) and triethylene glycol monomethyl ether, ($\text{CH}_3(\text{OC}_2\text{H}_4)_3\text{OH}$), over the whole composition range at 298.15 and 308.15 K and at atmospheric pressure. We have also measured the viscosity of ethylene glycol dimethyl ether ($\text{CH}_3\text{OC}_2\text{H}_4\text{OCH}_3$) with ethylene glycol monomethyl ether ($\text{CH}_3\text{OC}_2\text{H}_4\text{OH}$) at 298.15 and 308.15 K and excess molar volume for the same mixture at 308.15 K. The excess molar volumes at 298.15 K for ethylene glycol dimethyl ether with ethylene glycol monomethyl ether has been reported in our earlier paper (Pal et al., 1999). A survey of literature reveals that a few other studies involving excess molar volumes and excess heat capacities of polyethers + esters (Tovar et al., 1997), enthalpies of alkanol + polyethers (Cobos et al., 1984, 1990; Villamañan et al., 1982), and excess molar volumes and enthalpies of propan-1-ol + polyether (Mohren and Heintz, 1997) have also been studied. Therefore there is no systematic study on the behavior of ethylene glycol dimethyl ether with alkoxyethanols.

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Table 1. Properties of the Pure Liquids at 298.15 K and Atmospheric Pressure

liquid	$\rho/(\text{g}\cdot\text{cm}^{-3})$		$\eta/(\text{mPa}\cdot\text{s})$	
	exptl	lit.	exptl	lit.
$\text{CH}_3\text{OC}_2\text{H}_4\text{OCH}_3$	0.8626	0.86262 ^a	0.417	0.424 ^b
$\text{CH}_3\text{OC}_2\text{H}_4\text{OH}$	0.9602	0.96024 ^c	1.532	1.5414 ^d
$\text{CH}_3(\text{OC}_2\text{H}_4)_2\text{OH}$	1.0164	1.0167 ^c	3.565	3.480 ^c
$\text{CH}_3(\text{OC}_2\text{H}_4)_3\text{OH}$	1.0430	1.04310 ^e	6.586	

^a Douhéret et al. (1993). ^b Das et al. (1994). ^c Riddick et al. (1986). ^d Muhuri and Hazra (1995). ^e Douhéret et al. (1995).

Experimental Section

Materials. Ethylene glycol monomethyl ether (E. Merck; GC minimum, 99.5%), and ethylene glycol dimethyl ether (Lancaster, England; 99%) were dried over ferrous sulfate (A.R., BDH) and then fractionally distilled under reduced nitrogen gas pressure (Riddick et al., 1986; Perrin et al., 1980). Diethylene glycol monomethyl ether (Fluka; purum, GC > 98%) and triethylene glycol monomethyl ether (Fluka; purum, GC > 97%) were used without further purification. Prior to measurements, all liquids were stored in contact with 0.4 nm molecular sieves to reduce water contents and were partially degassed under vacuum. The purities of the liquids were checked by measuring their densities and viscosities at (298.15 ± 0.01) K. These are compared with the literature values in Table 1. The densities were measured with a bicapillary pycnometer which gave an accuracy of 5 parts in 10^5 . The composition of each mixture was obtained with an accuracy of 1×10^{-4} from the measured apparent masses of the components. All the masses were corrected for buoyancy. All molar quantities are based on the relative atomic mass table of 1986 issued by IUPAC (1986).

Apparatus and Procedure. The measurements of excess molar volumes were carried out by a continuous dilution dilatometer in a fashion similar to that described

Table 2. Excess Molar Volume (V_m^E) for the Mixtures of [$\text{CH}_3\text{OC}_2\text{H}_4\text{OCH}_3$ (1) + $\text{CH}_3(\text{OC}_2\text{H}_4)_n\text{OH}$ (2) ($n = 1, 2,$ and 3)] at 298.15 and 308.15 K and Atmospheric Pressure

x_2	$V_m^E/(\text{cm}^3\cdot\text{mol}^{-1})$	x_2	$V_m^E/(\text{cm}^3\cdot\text{mol}^{-1})$	x_2	$V_m^E/(\text{cm}^3\cdot\text{mol}^{-1})$
$\text{CH}_3\text{OC}_2\text{H}_4\text{OCH}_3$ (1) + $\text{CH}_3\text{OC}_2\text{H}_4\text{OH}$ (2)					
$T = 308.15$ K					
0.0107	-0.014	0.3505	-0.228	0.7750	-0.182
0.0260	-0.030	0.4052	-0.234	0.8290	-0.154
0.0591	-0.064	0.4666	-0.243	0.8625	-0.131
0.0888	-0.090	0.5206	-0.246	0.9157	-0.091
0.1199	-0.117	0.5554	-0.244	0.9631	-0.041
0.1661	-0.147	0.6011	-0.238	0.9827	-0.020
0.2110	-0.169	0.6557	-0.228		
0.2778	-0.200	0.7334	-0.201		
$\text{CH}_3\text{OC}_2\text{H}_4\text{OCH}_3$ (1) + $\text{CH}_3(\text{OC}_2\text{H}_4)_2\text{OH}$ (2)					
$T = 298.15$ K					
0.0258	-0.046	0.3769	-0.383	0.8136	-0.208
0.0423	-0.077	0.4572	-0.387	0.8645	-0.160
0.0629	-0.111	0.5197	-0.382	0.9173	-0.102
0.0873	-0.146	0.5590	-0.371	0.9452	-0.065
0.1383	-0.210	0.6029	-0.352	0.9814	-0.024
0.1868	-0.265	0.6349	-0.338		
0.2594	-0.331	0.6915	-0.305		
0.3313	-0.367	0.7541	-0.262		
$T = 308.15$ K					
0.0100	-0.020	0.3104	-0.390	0.7172	-0.330
0.0263	-0.053	0.3587	-0.410	0.7966	-0.258
0.0417	-0.082	0.4061	-0.421	0.8611	-0.193
0.0696	-0.132	0.4491	-0.426	0.8930	-0.153
0.0969	-0.175	0.4817	-0.424	0.9331	-0.101
0.1260	-0.218	0.4971	-0.423	0.9660	-0.055
0.1573	-0.258	0.5431	-0.416	0.9903	-0.016
0.2128	-0.320	0.6074	-0.397		
0.2681	-0.364	0.6648	-0.363		
$\text{CH}_3\text{OC}_2\text{H}_4\text{OCH}_3$ (1) + $\text{CH}_3(\text{OC}_2\text{H}_4)_3\text{OH}$ (2)					
$T = 298.15$ K					
0.0129	-0.038	0.3006	-0.475	0.6278	-0.440
0.0294	-0.080	0.3352	-0.491	0.6805	-0.402
0.0576	-0.150	0.3677	-0.502	0.7214	-0.367
0.0793	-0.198	0.4027	-0.510	0.7688	-0.319
0.1092	-0.255	0.4268	-0.513	0.8476	-0.226
0.1507	-0.326	0.4702	-0.509	0.8929	-0.168
0.1943	-0.384	0.5071	-0.500	0.9213	-0.126
0.2316	-0.422	0.5530	-0.486	0.9550	-0.075
0.2673	-0.454	0.5864	-0.467		
$T = 308.15$ K					
0.0129	-0.042	0.3002	-0.535	0.6952	-0.428
0.0346	-0.110	0.3680	-0.560	0.7300	-0.396
0.0480	-0.147	0.4088	-0.564	0.7934	-0.326
0.0800	-0.229	0.4496	-0.565	0.8435	-0.258
0.1060	-0.286	0.4920	-0.556	0.8861	-0.200
0.1338	-0.340	0.5483	-0.534	0.9171	-0.151
0.1682	-0.402	0.5925	-0.511	0.9473	-0.103
0.2227	-0.471	0.6594	-0.461	0.9709	-0.058

by Dickinson et al. (1975). Details of its calibration, experimental setup, and measuring procedure have been described previously (Pal and Singh, 1994). The mercury meniscus, as well as the reference mark, were measured with the help of a cathetometer that could read to within 0.01 mm. The results for excess molar volumes are reproducible to $\pm 0.003 \text{ cm}^3\cdot\text{mol}^{-1}$. Each run covered just over half of the mole fraction range, giving an overlap between two runs.

The kinematic viscosities of the pure liquids and the binary mixtures were measured at 298.15 and 308.15 K and at atmospheric pressure, by using an Ubbelohde suspended-level viscometer, and the detailed procedure was described in our earlier paper (Pal and Singh, 1997). The measured values of kinematic viscosities, ν , were converted to dynamic viscosities, η , after multiplication by the

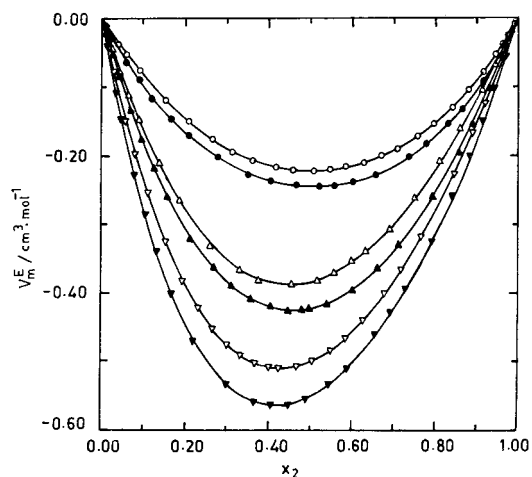


Figure 1. Excess molar volume, V_m^E , for $\text{CH}_3\text{OC}_2\text{H}_4\text{OCH}_3$ (1) + $\text{CH}_3\text{OC}_2\text{H}_4\text{OH}$ (2), (\circ , 298.15 K (Pal et al., 1999); \bullet , 308.15 K), + $\text{CH}_3(\text{OC}_2\text{H}_4)_2\text{OH}$ (2), (Δ , 298.15 K; \blacktriangle , 308.15 K), + $\text{CH}_3(\text{OC}_2\text{H}_4)_3\text{OH}$ (2), (∇ , 298.15 K; \blacktriangledown , 308.15 K). Solid curves were calculated from eq 3 using coefficient A_j of Table 4.

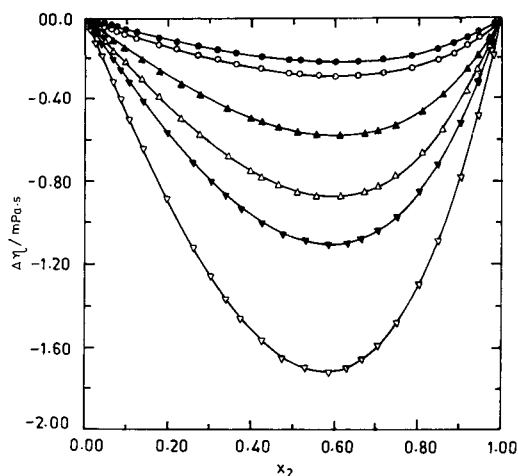


Figure 2. Deviation in viscosity, $\Delta\eta$, for $\text{CH}_3\text{OC}_2\text{H}_4\text{OCH}_3$ (1) + $\text{CH}_3\text{OC}_2\text{H}_4\text{OH}$ (2), (\circ , 298.15 K; \bullet , 308.15 K), + $\text{CH}_3(\text{OC}_2\text{H}_4)_2\text{OH}$ (2), (Δ , 298.15 K; \blacktriangle , 308.15 K), + $\text{CH}_3(\text{OC}_2\text{H}_4)_3\text{OH}$ (2), (∇ , 298.15 K; \blacktriangledown , 308.15 K). Solid curves were calculated from eq 3 using coefficient A_j of Table 4.

density. The values of η thus obtained are reproducible to $\pm 0.003 \text{ mPa}\cdot\text{s}$. All of the measurements were carried out in a well-stirred water bath whose temperature was controlled to $\pm 0.01 \text{ K}$. The densities of the mixtures were obtained from the densities of the pure components and measurements on V_m^E for their binary mixtures according to the equation

$$\rho = (x_1 M_1 + x_2 M_2) / (V_m^E + x_1 v_1^0 + x_2 v_2^0) \quad (1)$$

where x_1 and x_2 are the mole fractions and M_1 , M_2 and v_1^0 and v_2^0 represent molar masses and molar volumes of components 1 and 2, respectively.

Results and Discussion

The experimental values of the excess molar volumes and the viscosities of the different binary mixtures at 298.15 and 308.15 K are listed as a function of mole fraction in Tables 2 and 3. The plots of excess molar volumes against the mole fraction of alkoxyethanol are given in Figure 1.

The experimental values of dynamic viscosities η of the mixtures have been used to calculate the viscosity devia-

Table 3. Kinematic Viscosity, ν , and Densities, ρ , for the Mixtures of [CH₃OC₂H₄OCH₃ (1) + CH₃(OC₂H₄)_{*n*}OH (2) (*n* = 1, 2, and 3)] at 298.15 and 308.15 K and at Atmospheric Pressure

x_2	$\nu/(\text{mm}^2\cdot\text{s}^{-1})$	$\rho/(\text{g}\cdot\text{cm}^{-3})$	$\nu/(\text{mm}^2\cdot\text{s}^{-1})$	$\rho/(\text{g}\cdot\text{cm}^{-3})$	x_2	$\nu/(\text{mm}^2\cdot\text{s}^{-1})$	$\rho/(\text{g}\cdot\text{cm}^{-3})$	$\nu/(\text{mm}^2\cdot\text{s}^{-1})$	$\rho/(\text{g}\cdot\text{cm}^{-3})$	
CH ₃ OC ₂ H ₄ OCH ₃ (1) + CH ₃ OC ₂ H ₄ OH(2)										
T = 298.15 K			T = 308.15 K			T = 298.15 K			T = 308.15 K	
0.0000	0.483	0.8626	0.438	0.8507	0.4860	0.758	0.9055	0.661	0.8950	
0.0157	0.490	0.8639	0.442	0.8520	0.5142	0.782	0.9083	0.684	0.8978	
0.0290	0.495	0.8650	0.448	0.8532	0.5596	0.826	0.9127	0.717	0.9024	
0.0479	0.503	0.8665	0.454	0.8548	0.5992	0.865	0.9167	0.750	0.9065	
0.0624	0.507	0.8678	0.459	0.8561	0.6512	0.926	0.9219	0.797	0.9120	
0.0782	0.512	0.8691	0.465	0.8574	0.7115	1.006	0.9282	0.859	0.9185	
0.0978	0.519	0.8707	0.470	0.8591	0.7803	1.110	0.9355	0.939	0.9261	
0.1227	0.530	0.8728	0.478	0.8613	0.8427	1.220	0.9424	1.025	0.9331	
0.1763	0.554	0.8774	0.499	0.8660	0.8812	1.297	0.9467	1.083	0.9376	
0.2178	0.574	0.8810	0.515	0.8697	0.9158	1.374	0.9506	1.143	0.9416	
0.2703	0.605	0.8856	0.540	0.8745	0.9439	1.441	0.9538	1.195	0.9449	
0.3145	0.628	0.8896	0.560	0.8786	0.9802	1.540	0.9579	1.273	0.9491	
0.3599	0.659	0.8937	0.585	0.8828	1.0000	1.595	0.9602	1.316	0.9515	
0.4210	0.704	0.8994	0.619	0.8887						
CH ₃ OC ₂ H ₄ OCH ₃ (1) + CH ₃ (OC ₂ H ₄) ₂ OH(2)										
T = 298.15 K			T = 308.15 K			T = 298.15 K			T = 308.15 K	
0.0000	0.483	0.8626	0.438	0.8507	0.4618	1.115	0.9417	0.942	0.9313	
0.0142	0.496	0.8653	0.449	0.8534	0.5088	1.230	0.9489	1.023	0.9387	
0.0270	0.507	0.8677	0.458	0.8559	0.5545	1.351	0.9557	1.108	0.9457	
0.0472	0.522	0.8715	0.471	0.8597	0.6107	1.521	0.9640	1.225	0.9541	
0.0686	0.541	0.8754	0.486	0.8638	0.6691	1.719	0.9724	1.364	0.9627	
0.0957	0.567	0.8804	0.509	0.8688	0.7008	1.840	0.9768	1.443	0.9673	
0.1352	0.607	0.8875	0.543	0.8761	0.7422	2.010	0.9826	1.558	0.9732	
0.1767	0.653	0.8948	0.580	0.8835	0.8077	2.319	0.9915	1.763	0.9823	
0.2290	0.717	0.9038	0.632	0.8927	0.8557	2.568	0.9979	1.939	0.9888	
0.2748	0.779	0.9116	0.681	0.9006	0.9154	2.921	1.0057	2.188	0.9967	
0.3395	0.877	0.9223	0.760	0.9115	0.9426	3.100	1.0092	2.313	1.0003	
0.3948	0.977	0.9312	0.840	0.9206	0.9712	3.297	1.0128	2.455	1.0040	
0.4212	1.031	0.9353	0.878	0.9249	1.0000	3.507	1.0164	2.607	1.0076	
CH ₃ OC ₂ H ₄ OCH ₃ (1) + CH ₃ (OC ₂ H ₄) ₃ OH(2)										
T = 298.15 K			T = 308.15 K			T = 298.15 K			T = 308.15 K	
0.0000	0.483	0.8626	0.438	0.8507	0.4740	1.738	0.9703	1.392	0.9603	
0.0120	0.500	0.8661	0.451	0.8543	0.5274	2.007	0.9794	1.575	0.9695	
0.0262	0.519	0.8702	0.468	0.8585	0.5840	2.325	0.9885	1.792	0.9788	
0.0398	0.539	0.8741	0.484	0.8624	0.6280	2.601	0.9953	1.975	0.9857	
0.0672	0.581	0.8816	0.522	0.8702	0.6621	2.835	1.0003	2.135	0.9909	
0.0854	0.609	0.8865	0.544	0.8751	0.7011	3.128	1.0059	2.333	0.9966	
0.1068	0.643	0.8922	0.574	0.8809	0.7458	3.488	1.0121	2.574	1.0029	
0.1402	0.703	0.9007	0.624	0.8895	0.7997	3.972	1.0193	2.898	1.0102	
0.1983	0.822	0.9147	0.719	0.9038	0.8450	4.421	1.0250	3.201	1.0160	
0.2612	0.972	0.9288	0.841	0.9182	0.8981	5.003	1.0315	3.596	1.0226	
0.3019	1.088	0.9375	0.927	0.9270	0.9421	5.506	1.0366	3.958	1.0278	
0.3383	1.201	0.9449	1.011	0.9345	0.9792	6.024	1.0407	4.290	1.0320	
0.3754	1.331	0.9522	1.105	0.9419	1.0000	6.314	1.0430	4.488	1.0343	
0.4243	1.521	0.9614	1.239	0.9513						

tions from a linear dependence in the mole fraction average from the following relationship

$$\Delta\eta = \eta - \sum_{i=1}^2 x_i \eta_i \quad (2)$$

where η_i refers to the dynamic viscosities of the pure component i for which the mole fraction in the mixture is x_i . The values of $\Delta\eta$ for the various mixtures have been plotted against the mole fraction of alkoxyethanol in Figure 2.

The composition dependence of the excess molar volume and viscosity deviation is correlated by the Redlich–Kister polynomial

$$F(x) = x_1 x_2 \sum_{j=0}^n A_j (x_2 - x_1)^j \quad (3)$$

where A_j is the polynomial coefficient and n is the polynomial degree. The values of the coefficient A_j were evaluated by the method of least squares with all points weighted

equally and are summarized in Table 4, along with the standard deviation.

Excess volume versus composition plots in Figure 1 show that V_m^E is negative over the whole range of composition at both temperatures in the three mixtures. The negative values of V_m^E suggested the specific intermolecular interaction occurring in the mixing process. This is important with higher alkoxyethanol. Excess volumes become more negative with the rise in temperature. For the same value of x_2 , the magnitude of the V_m^E at both temperatures decreases with each addition of a $-\text{OC}_2\text{H}_4$ group in the molecule of ethylene glycol dimethyl ether. The behavior is similar to that of V_m^E of n -alkoxyethanol with 2-methoxyethanol or ethanol (Cobos et al., 1989; Pal and Singh, 1996a,b; Pal and Kumar, 1997, 1998) and ethylacetate with mono- and polyethers (Tovar et al., 1997). Figure 2 shows that the deviations in viscosity $\Delta\eta$ are negative for all systems over the entire range of composition at both temperatures. Viscosities of all ethylene glycol dimethyl ether–alkoxyethanols binary mixtures decrease with an increase in temperature. The values of $\Delta\eta$ at equimolar concentrations of ethylene glycol dimethyl ether and

Table 4. Smoothing Coefficients A_j and Standard Deviation $\sigma[F(x)]$ of Equation 3 for the Binary Mixtures

system	$F(x)$	A_0	A_1	A_2	A_3	$\sigma[F(x)]$
$T = 298.15 \text{ K}$						
CH ₃ OC ₂ H ₄ OCH ₃ (1) + CH ₃ OC ₂ H ₄ OH(2)	$\Delta\eta/(\text{mPa}\cdot\text{s})$	-1.1103	-0.4545	-0.2166	-0.1285	0.001
	$V_m^E/(\text{cm}^3\cdot\text{mol}^{-1})$	-1.5409	0.3072	-0.0624		0.002
CH ₃ (OC ₂ H ₄) ₂ OH(2)	$\Delta\eta/(\text{mPa}\cdot\text{s})$	-3.3899	-1.2510	-0.2690	-0.0533	0.001
	$V_m^E/(\text{cm}^3\cdot\text{mol}^{-1})$	-2.0056	0.5452	-0.2935		0.002
CH ₃ (OC ₂ H ₄) ₃ OH(2)	$\Delta\eta/(\text{mPa}\cdot\text{s})$	-6.7367	-2.0204	-0.3600	-0.2321	0.001
$T = 308.15 \text{ K}$						
CH ₃ OC ₂ H ₄ OCH ₃ (1) + CH ₃ OC ₂ H ₄ OH(2)	$V_m^E/(\text{cm}^3\cdot\text{mol}^{-1})$	-0.9774	-0.0233	-0.2157		0.002
	$\Delta\eta/(\text{mPa}\cdot\text{s})$	-0.8380	-0.3439	-0.1821	-0.1116	0.001
CH ₃ (OC ₂ H ₄) ₂ OH(2)	$V_m^E/(\text{cm}^3\cdot\text{mol}^{-1})$	-1.6962	0.2509	-0.1759		0.001
	$\Delta\eta/(\text{mPa}\cdot\text{s})$	-2.2257	-0.8874	-0.3973	-0.1260	0.001
CH ₃ (OC ₂ H ₄) ₃ OH(2)	$V_m^E/(\text{cm}^3\cdot\text{mol}^{-1})$	-2.2101	0.6579	-0.4752		0.002
	$\Delta\eta/(\text{mPa}\cdot\text{s})$	-4.3231	-1.4455	-0.4098	-0.0982	0.001

alkoxyethanols follow the order ethylene glycol monomethyl ether > diethylene glycol monomethyl ether > triethylene glycol monomethyl ether. This sequence, highly related to the strengthening of the heteroassociation of molecules with the increase of a -OC₂H₄ group in ethylene glycol monomethyl ether, is also observed for the excess molar volumes V_m^E of these mixtures.

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