

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

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Excess molar volumes  $V_m^E$  and viscosities  $\eta$  of the binary liquid mixtures of ethylene glycol diethyl ether (1,2-diethoxyethane),  $C_2H_5OC_2H_4OC_2H_5$ , with ethylene glycol monomethyl ether (2-methoxyethanol),  $CH_3OC_2H_4OH$ , diethylene glycol monomethyl ether (2-(2-methoxyethoxy)ethanol),  $CH_3(OC_2H_4)_2OH$ , and triethylene glycol monomethyl ether (2-[2-(2-methoxyethoxy)ethoxy]ethanol),  $CH_3(OC_2H_4)_3OH$ , have been measured at 298.15 and 308.15 K and at atmospheric pressure as a function of composition. From these results deviations of the viscosity from a mole fraction average have been calculated. Both the excess molar volumes and viscosity deviations for all the mixtures were negative over the entire composition range. Excess molar volumes and viscosity deviations have been correlated with the Redlich–Kister equation to estimate the coefficients and standard errors. The experimental and calculated quantities are used to discuss the mixing behavior of the components.

## Introduction

In our previous paper (Pal and Sharma, 1999), we studied the excess molar volumes and viscosities of binary mixtures of ethylene glycol dimethyl ether with alkoxyethanols at atmospheric pressure and at 298.15 and 308.15 K. In continuation of these investigations on the thermodynamic, transport, and acoustic properties of some mixtures of polyethers with organic solvents (Pal et al., 1999; Pal and Sharma, 1998), the present paper reports the excess molar volumes  $V_m^E$  and viscosities  $\eta$  for mixtures containing ethylene glycol diethyl ether and ethylene glycol monomethyl ether, diethylene glycol monomethyl ether, or triethylene glycol monomethyl ether over the whole composition range at 298.15 and 308.15 K. Investigations into the literature have shown that excess properties of binary mixtures containing polyethers have previously been studied by several authors (Tovar et al., 1997; Cobos et al., 1990; Mohren and Heintz, 1997). The present study was therefore undertaken in order to compare the excess molar volumes and deviations in viscosity of ethylene glycol diethyl ether + alkoxyethanols with those of our previous results for ethylene glycol dimethyl ether + alkoxyethanols (Pal and Sharma, 1999) by increasing the alkyl group size with a common polar head group. Also, an attempt has been made to interpret the results in terms of intermolecular interactions between ethylene glycol diethyl ether and alkoxyethanol.

## Experimental Section

**Materials.** Ethylene glycol diethyl ether (Lancaster, England, 98 mol %) was used without further purification. The purity of the sample was checked by density determination at 298.15 K. The density of ethylene glycol diethyl

ether at 298.15 K was found to be  $0.8362 \text{ g}\cdot\text{cm}^{-3}$ , in good agreement with literature values (Villamañán et al., 1982; Serna et al., 1997). Ethylene glycol monomethyl ether, diethylene glycol monomethyl ether, and triethylene glycol monomethyl ether were of the same quality; their purity was checked as described earlier (Pal and Sharma, 1999). Before the measurements, all liquids were stored in contact with 0.4 nm molecular sieves to reduce water content and were partially degassed at low pressure.

**Apparatus and Procedure.** Excess molar volumes were measured using a continuous dilution dilatometer in a similar fashion to that described by Dickinson et al. (1975). Calibration and operational procedures have been described previously (Pal and Singh, 1994). The measured excess molar volumes are reproducible to  $\pm 0.003 \text{ cm}^3\cdot\text{mol}^{-1}$ . All the measurements were carried out in a thermostat-controlled, well-stirred water bath, whose temperature was controlled to  $\pm 0.01 \text{ K}$ . The mole fraction of each mixture was obtained with an accuracy of  $1 \times 10^{-4}$  from the measured apparent masses of the components. All apparent masses were corrected for buoyancy. All the molar quantities were based upon the IUPAC table of atomic weights (1986). Each run covered just over half of the mole fraction range in order to give an overlap between the two runs.

The kinematic viscosities  $\nu$  of both the pure liquids and their mixtures were determined at 298.15 and 308.15 K and at atmospheric pressure with a calibrated Ubbelohde suspended level viscometer. Experimental details have been given previously (Pal and Singh, 1997). Care was taken to reduce evaporation during the measurements. The average of four or five sets of flow times was taken for the purpose of the calculation of viscosity. The flow time measurements were made with an electronic stopwatch having a precision of  $\pm 0.01 \text{ s}$ . The measured values of the kinematic viscosities were converted to dynamic viscosities  $\eta$  after multiplication by the density. The values of  $\eta$  obtained are reproducible to  $\pm 0.003 \text{ mPa}\cdot\text{s}$ . Densities of

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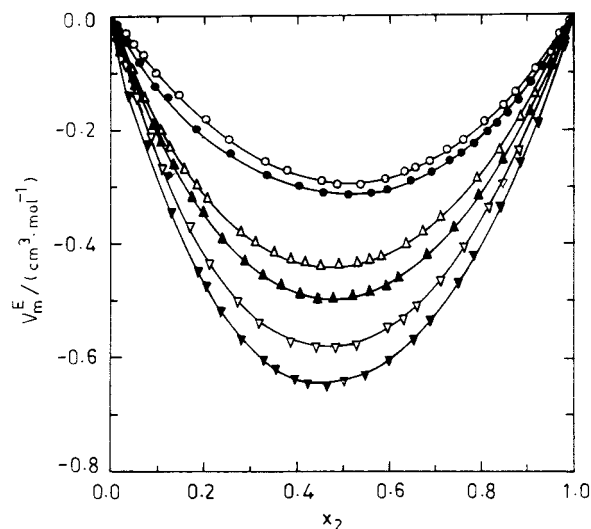
**Table 1. Excess Molar Volume  $V_m^E$  for the Mixtures of  $C_2H_5OC_2H_4OC_2H_5$  (1) +  $CH_3(OC_2H_4)_nOH$  (2) ( $n = 1, 2,$  and  $3$ ) at 298.15 K and 308.15 K and at Atmospheric Pressure**

$x_2$	$V_m^E/(\text{cm}^3\cdot\text{mol}^{-1})$	$x_2$	$V_m^E/(\text{cm}^3\cdot\text{mol}^{-1})$	$x_2$	$V_m^E/(\text{cm}^3\cdot\text{mol}^{-1})$
$C_2H_5OC_2H_4OC_2H_5$ (1) + $CH_3OC_2H_4OH$ (2)					
$T = 298.15$ K					
0.0132	-0.014	0.3761	-0.269	0.7615	-0.218
0.0302	-0.033	0.4501	-0.291	0.8080	-0.189
0.0478	-0.050	0.4847	-0.293	0.8465	-0.158
0.0653	-0.067	0.5387	-0.292	0.8750	-0.134
0.0944	-0.099	0.5907	-0.284	0.9144	-0.093
0.1441	-0.139	0.6327	-0.274	0.9456	-0.063
0.2010	-0.180	0.6584	-0.266	0.9712	-0.034
0.2531	-0.217	0.6868	-0.254	0.9921	-0.008
0.3304	-0.254	0.7267	-0.235		
$T = 308.15$ K					
0.0123	-0.015	0.4597	-0.310	0.7825	-0.222
0.0325	-0.042	0.5106	-0.312	0.8102	-0.200
0.0613	-0.079	0.5595	-0.309	0.8319	-0.186
0.0923	-0.122	0.5952	-0.304	0.8553	-0.166
0.1197	-0.141	0.6449	-0.289	0.8810	-0.143
0.1840	-0.196	0.6910	-0.274	0.9078	-0.115
0.2556	-0.239	0.7298	-0.253	0.9322	-0.087
0.3359	-0.278	0.7335	-0.249	0.9655	-0.047
0.4030	-0.298	0.7559	-0.238	0.9823	-0.025
$C_2H_5OC_2H_4OC_2H_5$ (1) + $CH_3(OC_2H_4)_2OH$ (2)					
$T = 298.15$ K					
0.0224	-0.050	0.3105	-0.395	0.6406	-0.395
0.0452	-0.092	0.3532	-0.412	0.6740	-0.375
0.0695	-0.138	0.3898	-0.424	0.7125	-0.350
0.1055	-0.195	0.4166	-0.429	0.7953	-0.280
0.1240	-0.225	0.4523	-0.435	0.8422	-0.235
0.1576	-0.265	0.4910	-0.434	0.8886	-0.174
0.1870	-0.297	0.5304	-0.430	0.9158	-0.137
0.2092	-0.320	0.5583	-0.425	0.9507	-0.084
0.2809	-0.379	0.5805	-0.417	0.9795	-0.037
$T = 308.15$ K					
0.0138	-0.035	0.2897	-0.426	0.6191	-0.457
0.0462	-0.103	0.3288	-0.452	0.6849	-0.415
0.0517	-0.114	0.3682	-0.470	0.7433	-0.369
0.0902	-0.190	0.4089	-0.486	0.7977	-0.313
0.1084	-0.219	0.4467	-0.494	0.8489	-0.248
0.1358	-0.260	0.4808	-0.493	0.9065	-0.166
0.1748	-0.315	0.5223	-0.489	0.9562	-0.084
0.1989	-0.343	0.5582	-0.480	0.9820	-0.035
0.2402	-0.389	0.5951	-0.470		
$C_2H_5OC_2H_4OC_2H_5$ (1) + $CH_3(OC_2H_4)_3OH$ (2)					
$T = 298.15$ K					
0.0153	-0.044	0.3178	-0.540	0.7094	-0.470
0.0305	-0.086	0.3843	-0.575	0.7621	-0.409
0.0533	-0.147	0.4406	-0.583	0.8182	-0.340
0.0851	-0.220	0.4826	-0.584	0.8784	-0.245
0.1097	-0.270	0.5291	-0.580	0.9323	-0.146
0.1678	-0.373	0.5943	-0.550	0.9777	-0.053
0.2128	-0.437	0.6285	-0.535		
0.2726	-0.504	0.6576	-0.512		
$T = 308.15$ K					
0.0166	-0.055	0.3270	-0.605	0.6501	-0.570
0.0362	-0.144	0.3528	-0.620	0.6861	-0.537
0.0784	-0.227	0.3939	-0.638	0.7475	-0.471
0.1289	-0.346	0.4236	-0.645	0.7822	-0.421
0.1861	-0.450	0.4644	-0.650	0.8396	-0.338
0.2025	-0.475	0.5018	-0.641	0.8842	-0.258
0.2350	-0.520	0.5484	-0.630	0.9208	-0.188
0.2788	-0.569	0.5981	-0.605	0.9780	-0.055

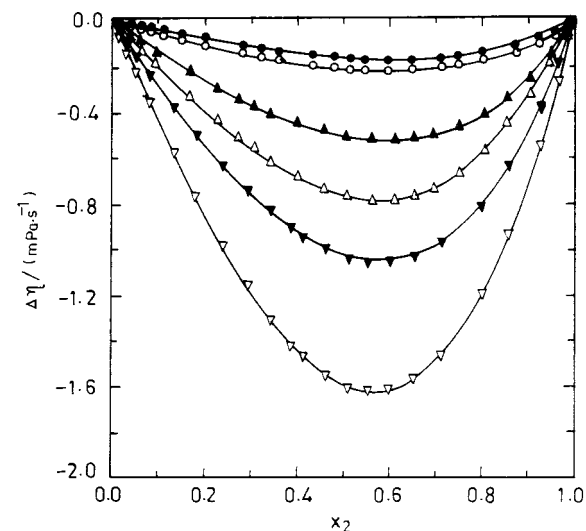
pure liquids were measured using a double-armed pycnometer with an accuracy of 5 parts in  $10^5$ .

## Results and Discussion

The experimental values of the excess molar volumes and the viscosities of the different binary mixtures at 298.15



**Figure 1.** Excess molar volume  $V_m^E$  for  $C_2H_5OC_2H_4OC_2H_5$  (1) +  $CH_3OC_2H_4OH$  (2) [(○) 298.15 K; (●) 308.15 K] or +  $CH_3(OC_2H_4)_2OH$  (2) [(△) 298.15 K; (▲) 308.15 K] or +  $CH_3(OC_2H_4)_3OH$  (2) [(▽) 298.15 K; (▼) 308.15 K]. Solid curves were calculated with eq 3 using the coefficients  $a_i$  of Table 3.



**Figure 2.** Deviation in viscosity  $\Delta\eta$  for  $C_2H_5OC_2H_4OC_2H_5$  (1) +  $CH_3OC_2H_4OH$  (2) [(○) 298.15 K; (●) 308.15 K] or +  $CH_3(OC_2H_4)_2OH$  (2) [(△) 298.15 K; (▲) 308.15 K] or +  $CH_3(OC_2H_4)_3OH$  (2) [(▽) 298.15 K; (▼) 308.15 K]. Solid curves were calculated with eq 3 using the coefficients  $a_i$  of Table 3.

and 308.15 K are reported as a function of mole fraction in Tables 1 and 2. A graphical representation of the excess molar volume results together with the smoothing curves against the mole fraction of alkoxyethanol appears in Figure 1.

From measured values of excess molar volumes, densities of liquid mixtures were computed with the equation

$$\rho = (x_1M_1 + x_2M_2)/(V_m + x_1V_1 + x_2V_2) \quad (1)$$

where  $x_1$  and  $x_2$  are the mole fractions and  $M_1$ ,  $M_2$ ,  $V_1$ , and  $V_2$  represent the molar masses and the molar volumes of components 1 and 2, respectively.

The deviation in viscosity  $\Delta\eta$  from a linear dependence on mole fraction average has been calculated from the following relation:

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

**Table 2. Dynamic Viscosity,  $\eta$  and Densities  $\rho$  for the Mixtures of [C<sub>2</sub>H<sub>5</sub>OC<sub>2</sub>H<sub>4</sub>OC<sub>2</sub>H<sub>5</sub> (1) + CH<sub>3</sub>(OC<sub>2</sub>H<sub>4</sub>)<sub>*n*</sub>OH (2) (*n* = 1, 2, and 3)] at 298.15 K and 308.15 K and at Atmospheric Pressure**

<i>x</i> <sub>2</sub>	$\frac{\rho}{\text{g}\cdot\text{cm}^{-3}}$	$\frac{\eta}{\text{mPa}\cdot\text{s}}$	$\frac{\rho}{\text{g}\cdot\text{cm}^{-3}}$	$\frac{\eta}{\text{mPa}\cdot\text{s}}$	<i>x</i> <sub>2</sub>	$\frac{\rho}{\text{g}\cdot\text{cm}^{-3}}$	$\frac{\eta}{\text{mPa}\cdot\text{s}}$	$\frac{\rho}{\text{g}\cdot\text{cm}^{-3}}$	$\frac{\eta}{\text{mPa}\cdot\text{s}}$	<i>x</i> <sub>2</sub>	$\frac{\rho}{\text{g}\cdot\text{cm}^{-3}}$	$\frac{\eta}{\text{mPa}\cdot\text{s}}$	$\frac{\rho}{\text{g}\cdot\text{cm}^{-3}}$	$\frac{\eta}{\text{mPa}\cdot\text{s}}$																																																																																																																																																																																																																																																																																																																																																									
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0.0000	0.8362	0.602	0.8276	0.522	0.0000	0.8362	0.602	0.8276	0.522	0.0000	0.8362	0.602	0.8276	0.522	0.0142	0.8373	0.608	0.8287	0.529	0.0162	0.8389	0.621	0.8303	0.538	0.0126	0.8393	0.622	0.8307	0.539	0.0354	0.8389	0.617	0.8303	0.539	0.0416	0.8431	0.646	0.8344	0.559	0.0328	0.8443	0.652	0.8356	0.561	0.0658	0.8413	0.631	0.8328	0.549	0.0613	0.8463	0.664	0.8377	0.574	0.0520	0.8490	0.688	0.8403	0.586	0.0821	0.8426	0.639	0.8341	0.553	0.0967	0.8522	0.703	0.8436	0.600	0.0807	0.8559	0.730	0.8472	0.617	0.1085	0.8448	0.647	0.8362	0.561	0.1691	0.8643	0.779	0.8556	0.666	0.1339	0.8685	0.824	0.8598	0.691	0.1503	0.8483	0.663	0.8397	0.578	0.2291	0.8745	0.850	0.8658	0.718	0.1806	0.8794	0.917	0.8706	0.765	0.2016	0.8528	0.685	0.8442	0.592	0.2741	0.8822	0.916	0.8734	0.765	0.2394	0.8928	1.047	0.8840	0.872	0.2800	0.8600	0.723	0.8513	0.620	0.3082	0.8880	0.964	0.8793	0.809	0.2918	0.9045	1.180	0.8957	0.978	0.3202	0.8639	0.742	0.8552	0.639	0.3440	0.8942	1.014	0.8855	0.849	0.3420	0.9155	1.332	0.9067	1.097	0.3604	0.8679	0.767	0.8592	0.655	0.4006	0.9041	1.115	0.8953	0.929	0.3850	0.9248	1.473	0.9160	1.198	0.4322	0.8755	0.812	0.8667	0.691	0.4593	0.9145	1.232	0.9057	1.017	0.4103	0.9301	1.588	0.9213	1.262	0.4797	0.8808	0.847	0.8720	0.718	0.5082	0.9232	1.346	0.9144	1.097	0.4604	0.9407	1.801	0.9319	1.417	0.5152	0.8849	0.875	0.8761	0.739	0.5635	0.9332	1.492	0.9244	1.203	0.5091	0.9508	2.040	0.9419	1.573	0.5455	0.8885	0.901	0.8797	0.758	0.6145	0.9426	1.650	0.9337	1.306	0.5505	0.9592	2.273	0.9504	1.725	0.5957	0.8947	0.946	0.8859	0.792	0.6552	0.9501	1.793	0.9431	1.423	0.5978	0.9687	2.561	0.9598	1.923	0.6464	0.9014	0.996	0.8926	0.832	0.6969	0.9579	1.949	0.9490	1.503	0.6522	0.9794	2.935	0.9706	2.167	0.7119	0.9105	1.066	0.9017	0.883	0.7526	0.9684	2.186	0.9595	1.654	0.7117	0.9909	3.396	0.9821	2.476	0.7472	0.9157	1.105	0.9069	0.918	0.8075	0.9788	2.441	0.9700	1.823	0.7976	1.0070	4.169	0.9982	2.989	0.8023	0.9243	1.180	0.9155	0.969	0.8567	0.9883	2.699	0.9795	1.995	0.8577	1.0180	4.791	1.0093	3.415	0.8705	0.9357	1.275	0.9270	1.054	0.9074	0.9982	2.985	0.9893	2.190	0.9256	1.0301	5.588	1.0214	3.957	0.9222	0.9450	1.366	0.9363	1.119	0.9515	1.0068	3.253	0.9980	2.386	0.9633	1.0371	6.103	1.0283	4.311	0.9592	0.9507	1.425	0.9420	1.167	0.9854	1.0135	3.470	1.0047	2.550	1.0000	1.0430	6.586	1.0343	4.642	0.9843	0.9570	1.497	0.9483	1.223	1.0000	1.0164	3.565	1.0076	2.627	1.0000	0.9602	1.532	0.9515	1.252

**Table 3. Smoothing Coefficients *a*<sub>*i*</sub> and Standard Deviations  $\sigma[F(x)]$  of Eq 3 for the Binary Mixtures**

system	<i>F</i> ( <i>x</i> )	<i>a</i> <sub>0</sub>	<i>a</i> <sub>1</sub>	<i>a</i> <sub>2</sub>	<i>a</i> <sub>3</sub>	$\sigma[F(x)]$
<i>T</i> = 298.15 K						
C <sub>2</sub> H <sub>5</sub> OC <sub>2</sub> H <sub>4</sub> OC <sub>2</sub> H <sub>5</sub> (1) + CH <sub>3</sub> OC <sub>2</sub> H <sub>4</sub> OH (2)	$V_m^E/(\text{cm}^3\cdot\text{mol}^{-1})$	-1.1684	-0.0554	-0.0004		0.002
	$\eta/(\text{mPa}\cdot\text{s})$	-0.8116	-0.2682	-0.1441	-0.2590	0.002
CH <sub>3</sub> (OC <sub>2</sub> H <sub>4</sub> ) <sub>2</sub> OH (2)	$V_m^E/(\text{cm}^3\cdot\text{mol}^{-1})$	-1.7334	0.1810	-0.3007		0.002
	$\eta/(\text{mPa}\cdot\text{s})$	-3.0305	-1.0608	0.2280	0.1981	0.002
CH <sub>3</sub> (OC <sub>2</sub> H <sub>4</sub> ) <sub>3</sub> OH (2)	$V_m^E/(\text{cm}^3\cdot\text{mol}^{-1})$	-2.3352	0.2977	-0.3325		0.002
	$\eta/(\text{mPa}\cdot\text{s})$	-6.3895	-1.7375	0.0332	-0.2863	0.003
<i>T</i> = 308.15 K						
C <sub>2</sub> H <sub>5</sub> OC <sub>2</sub> H <sub>4</sub> OC <sub>2</sub> H <sub>5</sub> (1) + CH <sub>3</sub> OC <sub>2</sub> H <sub>4</sub> OH (2)	$V_m^E/(\text{cm}^3\cdot\text{mol}^{-1})$	-1.2438	-0.0198	-0.1578		0.002
	$\eta/(\text{mPa}\cdot\text{s})$	-0.6295	-0.2323	-0.1033	-0.2404	0.003
CH <sub>3</sub> (OC <sub>2</sub> H <sub>4</sub> ) <sub>2</sub> OH (2)	$V_m^E/(\text{cm}^3\cdot\text{mol}^{-1})$	-1.9665	0.1848	-0.2288		0.002
	$\eta/(\text{mPa}\cdot\text{s})$	-1.9612	-0.7211	-0.2650	-0.2568	0.002
CH <sub>3</sub> (OC <sub>2</sub> H <sub>4</sub> ) <sub>3</sub> OH (2)	$V_m^E/(\text{cm}^3\cdot\text{mol}^{-1})$	-2.5756	0.3751	-0.4029		0.002
	$\eta/(\text{mPa}\cdot\text{s})$	-4.1498	-1.3873	-0.2430	0.0367	0.003

where  $\eta$ ,  $\eta_1$ , and  $\eta_2$  are the dynamic viscosities of the mixtures and components 1 and 2, respectively. The variation of  $\Delta\eta$  with mole fraction of alkoxyethanol for the various mixtures is represented in Figure 2.

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

$$F(x) = x_1x_2 \sum_{i=0}^{\eta} a_i(x_2 - x_1)^i \quad (3)$$

The coefficients *a*<sub>*i*</sub> of eq 3 were calculated by the method of least squares. The values obtained for the coefficients *a*<sub>*i*</sub> along with the standard deviations  $\sigma$  are reported in Table 3. For all the mixtures,  $\sigma(V_m^E) \leq 0.003 \text{ cm}^3\cdot\text{mol}^{-1}$ , in accord with the precision attainable with the dilatometer used.

For each of the mixtures studied, the  $V_m^E$  values are negative and symmetric over the entire range of composition at both temperatures. These results and those of Pal and Sharma (1999) show that  $V_m^E$  decreases (at both temperatures) as the alkyl chain end length of the ethers

increases. In Figure 1 it can also be observed how  $V_m^E$  values vary with the number of oxygen atoms in the alkoxyethanol chain. The presence of oxygen atoms in the alkoxyethanol chain lowers the excess volume, and this becomes more negative with the rise in the temperature. This behavior may be compared with the  $V_m^E$  results of the mixtures of *n*-alkoxyethanols with 2-methoxyethanol, 1-butanol, or *n*-alkanols (Pal and Singh, 1996a,b; Cobos et al., 1988, 1989; Pal and Kumar, 1997, 1999) and ethyl acetate with (mono and poly)ethers (Tovar et al., 1997). However, the behavior is opposite to that of  $V_m^E$  of 2-methoxyethanol with polyethers (Pal et al., 1999), where  $V_m^E$  increases with the number of oxygen atoms in the polyether chain. The negative values of  $V_m^E$  suggest a specific intermolecular interaction occurs in the mixing process. This is very important with higher alkoxyethanols.

Figure 2 shows that the deviation in viscosity  $\Delta\eta$  is negative for all systems over the whole mole fraction range at both temperatures. Viscosities of all these binary mixtures decrease with the increase of temperature. The absolute values  $\Delta\eta$  for mixtures of an alkoxyethanol

molecule with a OC<sub>2</sub>H<sub>4</sub> group in the middle plus a monomethyl ether fall in the following order at both temperatures: ethylene glycol monomethyl ether > diethylene glycol monomethyl ether > triethylene glycol monomethyl ether.

The general behavior of mixtures of ethers with *n*-alkoxyethanols (Pal and Sharma, 1999) confirms that the self-association of the *n*-alkoxyethanols plays an important role, but also very important is the formation of hydrogen bonds between the functional group (OH) of the *n*-alkoxyethanol and the O atoms of the ethers as well as the increase of the polarity of *n*-alkoxyethanols. The overall result is a negative  $V_m^E$  and  $\Delta\eta$ .

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