Excess Molar Volumes and Viscosities of Mixtures Containing Some Polyethers + Acetonitrile at 298.15 K

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Excess molar volumes V_m^E and viscosities η for ethylene glycol dimethyl ether (monoglyme), $CH_3OC_2H_4OCH_3$, diethylene glycol dimethyl ether (diglyme), $CH_3(OC_2H_4)_2OCH_3$, triethylene glycol dimethyl ether (triglyme), $CH_3(OC_2H_4)_3OCH_3$, tetraethylene glycol dimethyl ether (tetraglyme), and $CH_3(OC_2H_4)_4OCH_3$ + acetonitrile (CH_3CN) have been measured as a function of composition at 298.15 K and atmospheric pressure. The measurements of excess volume have been carried out with a continuous dilution dilatometer. The excess volumes are all negative over the entire range of composition and decrease with increasing number of oxygen atoms in the glyme molecule. The results are discussed in terms of dipole−dipole interactions between glymes and acetonitrile and their magnitudes increasing with the dipole character of the glyme molecules.

Introduction

As a continuation of our earlier studies (Pal and Sharma,-1998a,b; Pal et al., 1999; Pal and Kumar, 1999) on the thermodynamic properties of nonaqueous binary mixtures of strongly polar liquids, we report in this paper new experimental excess molar volumes V_m^E and viscosities η of binary mixtures of acetonitrile (CH_3CN) with ethylene glycol dimethyl ether ($CH_3OC_2H_4OCH_3$), diethylene glycol dimethyl ether ($CH_3(OC_2H_4)_2OCH_3$), triethylene glycol dimethyl ether ($CH_3(OC_2H_4)_3OCH_3$), and tetraethylene glycol dimethyl ether ($CH_3(OC_2H_4)_4OCH_3$) over the whole mole fraction range at 298.15 K and atmospheric pressure in order to gain a better understanding of the intermolecular interactions between the component molecules. Interactions between monoether + alkane (Berti et al., 1989), polyether + alkane (Peleteire et al., 1994;Tovar et al., 1997b), (monoether or polyether) + ester (Tovar et al., 1997a), polyether + alkanol (Cobos et al., 1990;Villamanan et al., 1982; Mohren and Heintz, 1997), and (mono and branched ether) or cyclic ether + acetonitrile (Letcher and Domanska, 1994a,b; Francesconi and Comelli, 1988; De Schaefer et al., 1990), have been studied extensively while the interaction between (polyether + acetonitrile) is not well-known. The aim of this work is to provide results for the characterization of the influence of the size, shape, and polarity of the ether on the thermodynamic properties of mixtures when we pass from a molecule with one -O- functional group to a molecule with several -O- functional groups.

Experimental Section

Materials. Acetonitrile (SRL, Bombay, HPLC grade, GC minimum, 99.8 mol %) was purified by fractional distillation as described elsewhere (Riddick et al., 1986). The samples diethylene glycol dimethyl ether, Merck Schuchardt product with stated purity >99 mol %, and ethylene glycol dimethyl ether, Acros product with stated purity 99 mol %, 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). Triethylene glycol dimethyl ether (Merck Schuchardt, GC > 98 mol %) and tetraethylene glycol dimethyl ether (Merck Schuchardt, GC > 98 mol %) were used without further purification. All liquids were stored in dark bottles to prevent contamination from air and carefully dried over 0.4 nm molecular sieves to reduce the water content. The purities of the liquid were checked by measuring and comparing the densities at 298.15 K and atmospheric pressure with their corresponding literature values, as shown in Table 1. The densities were measured with a bicapillary pycnometer that gives an accuracy of five parts in 10^5 at (298.15 ± 0.01) K. Before the measurements, all liquids were partially degassed under vacuum. The com-

Table 1. Properties of Pure Liquids at 298.15 K

	ρ/g cm^3		η/mPa s	
liquid	exptl	lit.	exptl	lit.
acetonitrile	0.7769	0.7766[a] 0.7765[b]	0.341	0.341[c]
ethylene glycol dimethyl ether	0.8626	0.86262[d] 0.8621[e] 0.86132[f] 0.8605[g] 0.86124[h]	0.417	0.424[f] 0.432[g]
diethylene glycol dimethyl ether	0.9389	0.9399[b] 0.9384[e] 0.93873[h] 0.93924[i] 0.9394[j]	0.985	0.991[b] 0.989[e] 0.981[j]
triethylene glycol dimethyl ether	0.9811	0.98067[h] 0.98171[i] 0.97950[k] 0.98001[l]	2.007	1.960[k]
tetraethylene glycol dimethyl ether	1.0063	1.00627[i] 1.00662[l] 1.0047[m]	3.295	3.295[m]

[a] Riddick and Bunger (1970). [b] Aminabhavi and Gopalakrishna (1995). [c] Gill and Bakshi (1989). [d] Douhéret et al. (1993). [e] Riddick et al. (1986). [f] Das et al. (1994). [g] Walllace and Mathews (1963). [h] Tover et al. (1997a). [i] Dethlefsen and Hvidt (1985). [j] de Ruiz Holgado et al. (1994). [k] Wallace et al. (1968). [l] Treszczanowicz et al. (1990). [m] McGee et al. (1983).

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Table 2. Excess Molar Volume V_m^E for the Binary Mixtures at 298.15 K

x_1	V_m^E/cm^3 mol^{-1}	x_1	V_m^E/cm^3 mol^{-1}	x_1	V_m^E/cm^3 mol^{-1}	x_1	V_m^E/cm^3 mol^{-1}	x_1	V_m^E/cm^3 mol^{-1}	x_1	V_m^E/cm^3 mol^{-1}
Ethylene Glycol Dimethyl Ether (1) + Acetonitrile (2)											
0.0476	−0.087	0.2346	−0.398	0.4140	−0.552	0.5063	−0.576	0.6401	−0.557	0.7591	−0.449
0.0787	−0.161	0.2830	−0.454	0.4175	−0.555	0.5409	−0.578	0.6708	−0.536	0.7868	−0.419
0.1232	−0.238	0.3235	−0.491	0.4485	−0.565	0.5713	−0.576	0.6973	−0.516	0.8430	−0.331
0.1691	−0.310	0.3562	−0.516	0.4696	−0.572	0.5952	−0.572	0.7151	−0.498	0.8699	−0.283
0.2077	−0.363	0.3911	−0.540	0.4996	−0.574	0.6178	−0.564	0.7391	−0.471	0.9323	−0.158
Diethylene Glycol Dimethyl Ether (1) + Acetonitrile (2)											
0.0239	−0.095	0.2052	−0.574	0.3486	−0.743	0.4084	−0.772	0.5554	−0.758	0.7639	−0.540
0.0558	−0.211	0.2613	−0.654	0.3522	−0.751	0.4228	−0.778	0.5843	−0.736	0.8008	−0.471
0.0933	−0.322	0.2825	−0.686	0.3751	−0.761	0.4772	−0.779	0.6245	−0.710	0.8322	−0.411
0.1243	−0.403	0.3070	−0.712	0.3794	−0.766	0.5048	−0.777	0.6645	−0.669	0.9235	−0.215
0.1514	−0.469	0.3187	−0.726	0.3912	−0.771	0.5269	−0.770	0.6985	−0.624	0.9470	−0.157
0.1784	−0.524	0.3343	−0.734								
Triethylene Glycol Dimethyl Ether (1) + Acetonitrile (2)											
0.0209	−0.114	0.0828	−0.407	0.1893	−0.727	0.4173	−0.964	0.6136	−0.832	0.7556	−0.610
0.0369	−0.196	0.0944	−0.447	0.2030	−0.757	0.4482	−0.962	0.6491	−0.793	0.8099	−0.498
0.0429	−0.227	0.1081	−0.496	0.2461	−0.841	0.4594	−0.958	0.6920	−0.720	0.8762	−0.350
0.0501	−0.264	0.1346	−0.590	0.2792	−0.885	0.5199	−0.928	0.6921	−0.723	0.9181	−0.245
0.0594	−0.306	0.1510	−0.632	0.3552	−0.950	0.5446	−0.904	0.7260	−0.665	0.9767	−0.081
0.0720	−0.359	0.1659	−0.671	0.4013	−0.968	0.5771	−0.872				
Tetraethylene Glycol Dimethyl Ether (1) + Acetonitrile (2)											
0.0066	−0.053	0.1102	−0.676	0.2421	−1.021	0.3326	−1.118	0.4379	−1.116	0.6884	−0.805
0.0330	−0.260	0.1279	−0.740	0.2545	−1.037	0.3461	−1.124	0.4619	−1.106	0.7471	−0.681
0.0481	−0.362	0.1602	−0.846	0.2733	−1.064	0.3548	−1.125	0.4868	−1.089	0.8035	−0.543
0.0655	−0.457	0.1996	−0.939	0.2931	−1.088	0.3853	−1.128	0.5088	−1.068	0.8882	−0.337
0.0839	−0.556	0.2163	−0.972	0.3116	−1.101	0.4131	−1.125	0.5889	−0.967	0.9516	−0.166
0.0980	−0.624	0.2268	−0.990								

position of each mixture was obtained with an accuracy of 1×10^{-4} from the measured apparent masses of the components. All masses were corrected for buoyancy. All molar quantities were based upon the IUPAC (1986) table of atomic weights.

Apparatus and Procedure. Excess molar volumes, which are accurate to ±0.003 cm^3 mol^{-1}, were measured in a continuous dilution dilatometer in a fashion similar to that described by Dickinson et al. (1975). Details of its calibration and operational procedures have been described elsewhere (Pal and Singh, 1994; Pal et al., 1994). The dilatometer was clamped vertically into the water bath. The mercury meniscus, as well as the reference mark, were measured with a cathetometer having a precision of ±0.001 cm. Each run covered just over half of the mole fraction range, giving an overlap between two runs.

The measurements of kinematic viscosity in both the pure liquids and their mixtures were performed with a modified suspended level Ubbelohde viscometer. The viscometer was calibrated with doubly distilled water and benzene. The details of the apparatus and procedure have been described previously (Pal and Singh, 1996; Pal and Singh, 1997). The arithmetic mean of four or five sets of flow times for each pure liquid or liquid mixture was taken for the purpose of the calculation of viscosity. The flow-time measurements were performed using an accurate stopwatch with a precision of ±0.01 s. The overall uncertainty of the viscosity measurements is dependent on the equilibrium stability of the viscometer, the time of flow, and the change of concentration, which are of the order of 1×10^{-2}, 1×10^{-2}, and 1×10^{-4}, respectively; viscosity values are accurate to within the range ±0.003 mPa s. All measurements were carried out in a well-stirred water bath with temperature controlled to within ±0.01 K.

Results and Discussion

Excess molar volumes V_m^E are listed in Table 2, and viscosities η are listed in Table 3. The plots of the excess molar volume against the mole fraction of the glycol ether are given in Figure 1. The V_m^E values are used to compute

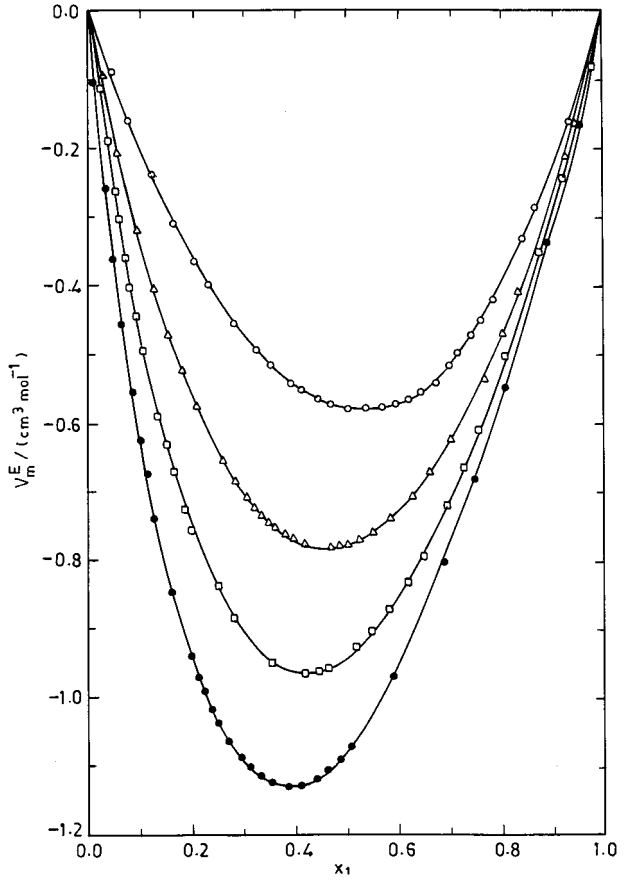

Figure 1. Excess molar volume V_m^E for ethylene glycol dimethyl ether (1) (○), diethylene glycol dimethyl ether (1) (△), triethylene glycol dimethyl ether (1) (□), and tetraethylene glycol dimethyl ether (1) (●) + acetonitrile (2) at 298.15 K. The solid curves have been drawn from eq 3.

Table 3. Densities ρ and Viscosities η for the Binary Mixtures at 298.15 K

x_1	ρ/g cm^{-3}	η/mPa s	x_1	ρ/g cm^{-3}	η/mPa s	x_1	ρ/g cm^{-3}	η/mPa s	x_1	ρ/g cm^{-3}	η/mPa s
Ethylene Glycol Dimethyl Ether (1) + Acetonitrile (2)											
0.0062	0.7781	0.342	0.1843	0.8078	0.373	0.3528	0.8272	0.396	0.5792	0.8447	0.414
0.0339	0.7835	0.348	0.2035	0.8104	0.377	0.3672	0.8286	0.397	0.6374	0.8481	0.416
0.0506	0.7866	0.351	0.2366	0.8146	0.380	0.3901	0.8307	0.400	0.6911	0.8510	0.416
0.0637	0.7889	0.352	0.2722	0.8188	0.386	0.4107	0.8325	0.401	0.7816	0.8551	0.418
0.0854	0.7927	0.357	0.2923	0.8210	0.387	0.4383	0.8347	0.404	0.8356	0.8573	0.418
0.1070	0.7963	0.360	0.3080	0.8227	0.390	0.4905	0.8388	0.408	0.8598	0.8582	0.418
0.1367	0.8009	0.366	0.3167	0.8236	0.392	0.5391	0.8422	0.411	0.8879	0.8591	0.413
0.1610	0.8045	0.377									
Diethylene Glycol Dimethyl Ether (1) + Acetonitrile (2)											
0.0037	0.7786	0.344	0.1437	0.8321	0.480	0.3505	0.8801	0.663	0.6100	0.9144	0.836
0.0121	0.7826	0.352	0.1763	0.8415	0.507	0.3800	0.8851	0.682	0.6524	0.9183	0.856
0.0230	0.7876	0.362	0.2139	0.8514	0.538	0.4214	0.8916	0.714	0.7402	0.9253	0.900
0.0444	0.7968	0.381	0.2405	0.8578	0.563	0.4482	0.8955	0.732	0.7995	0.9293	0.923
0.0620	0.8039	0.397	0.2538	0.8608	0.565	0.5026	0.9026	0.771	0.8671	0.9332	0.948
0.0827	0.8117	0.416	0.2671	0.8638	0.592	0.5263	0.9055	0.785	0.9455	0.9368	0.974
0.1207	0.8248	0.452	0.3139	0.8733	0.629						
Triethylene Glycol Dimethyl Ether (1) + Acetonitrile (2)											
0.0024	0.7786	0.345	0.1477	0.8579	0.619	0.3885	0.9244	1.137	0.6661	0.9616	1.616
0.0088	0.7833	0.355	0.1809	0.8705	0.689	0.4353	0.9327	1.235	0.7499	0.9685	1.720
0.0210	0.7918	0.376	0.2182	0.8829	0.768	0.4731	0.9387	1.305	0.7727	0.9701	1.749
0.0301	0.7979	0.392	0.2528	0.8932	0.843	0.5015	0.9428	1.349	0.7887	0.9712	1.769
0.0575	0.8146	0.442	0.2809	0.9007	0.908	0.5698	0.9515	1.467	0.8757	0.9762	1.860
0.0925	0.8333	0.509	0.2958	0.9045	0.938	0.6518	0.9602	1.580	0.9459	0.9793	1.938
0.1237	0.8479	0.569	0.3483	0.9164	1.052						
Tetraethylene Glycol Dimethyl Ether (1) + Acetonitrile (2)											
0.0021	0.7790	0.346	0.0590	0.8270	0.505	0.2677	0.9222	1.283	0.6128	0.9832	2.511
0.0056	0.7824	0.355	0.0874	0.8459	0.596	0.2904	0.9285	1.376	0.6695	0.9887	2.665
0.0107	0.7873	0.369	0.1165	0.8626	0.695	0.3252	0.9373	1.514	0.7087	0.9919	2.765
0.0144	0.7908	0.378	0.1388	0.8739	0.776	0.3733	0.9480	1.699	0.7633	0.9962	2.885
0.0189	0.7949	0.390	0.1615	0.8843	0.860	0.4510	0.9622	1.993	0.8021	0.9985	2.965
0.0240	0.7994	0.404	0.1908	0.8964	0.974	0.5325	0.9740	2.268	0.9071	1.0037	3.153
0.0364	0.8099	0.437	0.2188	0.9066	1.085	0.5725	0.9789	2.393	0.9588	1.0054	3.227

Table 4. Smoothing Coefficients A_i and Standard Deviations σ for Binary Mixtures at 298.15 K

function	A_1	A_2	A_3	A_4	A_5	σ
Ethylene Glycol Dimethyl Ether (1) + Acetonitrile (2)						
V_m^E/cm^3 mol^{-1}	−2.3129	−0.2647	−0.2107	0.0905	0.2875	0.003
$\Delta\eta$/mPa s	0.1180	−0.0121	−0.0383	−0.0260		0.001
Diethylene Glycol Dimethyl Ether (1) + Acetonitrile (2)						
V_m^E/cm^3 mol^{-1}	−3.1129	0.4175	−0.2545	0.0140	−0.3202	0.003
$\Delta\eta$/mPa s	0.4217	0.0131	−0.0814	0.0986		0.002
Triethylene Glycol Dimethyl Ether (1) + Acetonitrile (2)						
V_m^E/cm^3 mol^{-1}	−3.7600	1.1812	−0.4914	0.0670	−0.3726	0.003
$\Delta\eta$/mPa s	0.7128	0.2950	−0.5882	−0.1061		0.003
Tetraethylene Glycol Dimethyl Ether (1) + Acetonitrile (2)						
V_m^E/cm^3 mol^{-1}	−4.3168	1.7559	−0.5522	0.7393	−1.3427	0.004
$\Delta\eta$/mPa s	1.3700	0.8540	−0.9047	0.1677		0.003

the density ρ of mixtures from

$$\rho = (x_1M_1 + x_2M_2)/(V_m^E + x_1v_1^\circ + x_2v_2^\circ) \quad (1)$$

where x_1 and x_2 are the mole fractions, M_1 and M_2 are the molar masses, and v_1° and v_2° are the molar volumes of glycol ether (1) and acetonitrile (2), respectively. The deviation in the viscosity from a mole fraction average is given by

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

where η, η_1, and η_2 are the viscosities of the mixture and components 1 and 2, respectively. The variations of $\Delta\eta$ with the mole fraction at 298.15 K are given in Figure 2.

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

$$Y = x_1x_2\sum_{i=1}^{n}A_i(x_1 - x_2)^{i-1} \quad (3)$$

where A_i are the polynomial coefficients, which were evaluated from the least-squares method. The values of the parameters along with the standard deviations are given in Table 4. Values of σ were obtained from the equation

$$\sigma = [\sum(Y_{exp} - Y_{cal})^2/(k - n)]^{1/2} \quad (4)$$

where k is the number of experimental points. Y refers to V_m^E/cm^3 mol^{-1} or $\Delta\eta$/mPa s.

For each of the mixtures studied, the V_m^E values are negative over the whole mole fraction range and show a minimum in the sequence monoglyme < diglyme < triglyme < tetraglyme, and in the same sequence the minimum is shifted to lower values of x_1. Thus, the presence of diethylene glycol units [CH_2CH_2O] in the glyme chain lowers the excess volume. The negative values of V_m^E suggested a strong chemical or specific interaction between the components which is maximum in the case of tetraglyme−acetonitrile. Further, interactions between glymes and the acetonitrile cannot imply hydrogen bonding; the

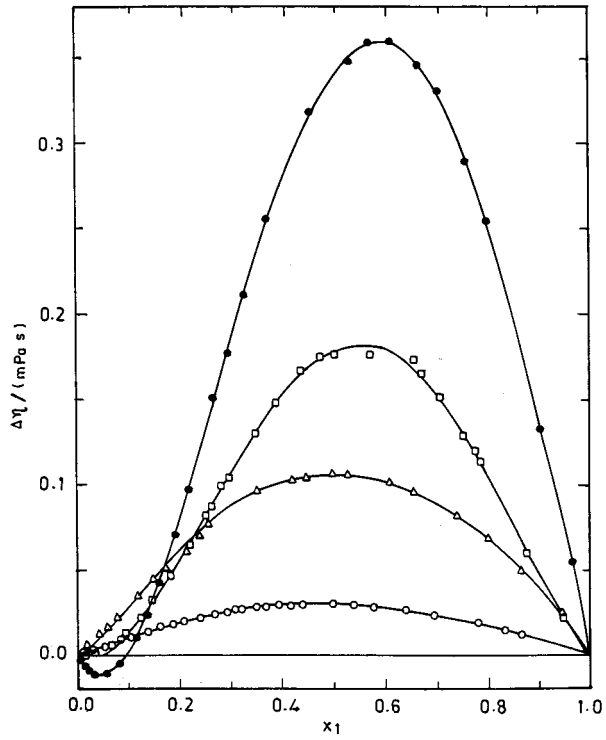

Figure 2. Viscosity deviations $\Delta\eta$ for ethylene glycol dimethyl ether (1) (○), diethylene glycol dimethyl ether (1) (△), triethylene glycol dimethyl ether (1) (□), and tetraethylene glycol dimethyl ether (1) (●) + acetonitrile (2) at 298.15 K. The solid curves have been drawn from eq 3.

differences in molecular volumes and free volumes (Prolongo et al., 1984) and the relatively larger values of the molecular dipoles of both acetonitrile and glymes may be responsible for the decrease in the magnitude of V_m^E.

The deviations in viscosity $\Delta\eta$ presented in Figure 2 are positive for all mixtures except tetraglyme−acetonitrile mixtures at lower x_1. There is a very obvious increase in the magnitude of η and $\Delta\eta$ with each addition of a OC_2H_4 group in the molecule of ethylene glycol dimethyl ether. Negative excess molar volumes and rather higher values of η and $\Delta\eta$ may be attributed to (i) the existence of dispersion and dipolar forces between unlike molecules and (ii) the differences in size and shape of unlike molecules. The absolute values of $\Delta\eta$ for the mixtures fall in the same order as the V_m^E values but with the opposite variation.

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