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Excess Molar Volumes and Viscosities for Binary Liquid Mixtures of Methyl *tert*-Butyl Ether and of *tert*-Amyl Methyl Ether with Methanol, 1-Propanol, and 1-Pentanol at 298.15 K

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Excess molar volumes V_m^E and viscosities η have been measured as a function of composition for binary liquid mixtures of methyl *tert*-butyl ether and of *tert*-amyl methyl ether with methanol, 1-propanol, and 1-pentanol at 298.15 K and atmospheric pressure. The excess volumes V_m^E are negative for all the mixtures over the entire range of composition. From the experimental results, the deviation in the viscosity ($\Delta \ln \eta$) has been calculated. These properties are fitted to the Redlich−Kister equation to estimate the binary coefficients. The results for V_m^E and $\Delta \ln \eta$ are discussed on the basis of molecular interactions between the components of the mixtures.

Introduction

This paper is part of our systematic program of research concerning the experimental study of several thermodynamic and transport properties of binary liquid mixtures containing branched ethers (Pal and Singh, 1997; Pal et al., 1997) used as oxygenating agents in gasoline technology.

In this communication we report on the measured values of excess molar volumes (V_m^E) and viscosities (η) for binary mixtures of methyl *tert*-butyl ether (MTBE) and of *tert*-amyl methyl ether (TAME) with methanol, 1-propanol, and 1-pentanol over the whole mole fraction range at 298.15 K and atmospheric pressure. The thermodynamic properties of mixtures of branched ethers with either alkanes, dialkyl carbonates, di-*n*-alkyl ethers, or tributylamine (Zhu et al., 1994; Tong et al., 1996; Francesconi and Comelli, 1997; Liao et al., 1997; Latcher and Domanska, 1994) have been studied. A few other studies involving excess enthalpies of binary mixtures of methyl *tert*-butyl ether with methanol (Tussel-Langer et al., 1991) and with propan-1-ol or propan-2-ol (Nagata and Tamura, 1995) have also been performed. We are not aware of any volumetric and viscometic data in the literature for the systems presented in this study. The aim of this work is to provide a set of values for the characterization of the molecular interactions of these mixtures and to examine the effect of the enlargement of the CH_2 unit in the branched ether.

Experimental Section

Materials. Methanol (S.R.L., Bombay, GCmin 99.8 mol %), 1-propanol (S.R.L., Bombay, GCmin 99.5 mol %), and 1-pentanol (Acros, USA, 99 mol %) were the same as those used in our earlier studies (Pal and Kumar, 1999; Pal and Sharma, 1998). The MTBE was obtained from Acros Chemical Co. The purities stated by the manufacturers were at least 99 mol %. The TAME, with stated purity exceeding 97 mol %, was obtained from Aldrich Chemical Co. Both of these chemicals were used without further purification. All liquids were stored in dark bottles over 0.4 nm molecular sieves to reduce water content. Before the measurements, all liquids were partially degassed under vacuum. The purities of the liquids were checked by comparing the densities and viscosities at (298.15 ± 0.01) K with their corresponding literature values in Table 1. The densities were measured with a bicapillary pycnometer that gave an accuracy of 5 parts in 10^5.

Table 1. Comparison of Experimental Densities ρ and Viscosities η of Pure Liquids with Literature Values at 298.15 K

	ρ/g cm^{-3}		η/mPa s	
liquid	exptl	lit.	exptl	lit.
tert-butyl methyl ether	0.7359	0.73531[a]	0.340	
		0.73584[b]		
		0.7353[c]		
tert-amyl methyl ether	0.7663	0.76626[b]	0.438	
		0.7656[c]		
methanol	0.7864	0.78637[d]	0.550	0.5513[d]
		0.7866[e]		0.538[e]
		0.7866[f]		0.550[f]
		0.78664[g]		0.5531[g]
1-propanol	0.7994	0.79960[d]	1.951	1.9430[d]
		0.7998[e]		1.927[e]
		0.7996[f]		1.943[f]
		0.79975[g]		1.9430[g]
1-pentanol	0.8111	0.81080[d]	3.511	3.5128[d]
		0.8110[e]		3.421[e]
		0.8107[f]		3.510[f]
		0.8115[g]		3.5128[g]

[a] Francesconi et al., 1997. [b] Tong et al., 1996. [c] TRC Tables, 1988a,b. [d] Riddick et al., 1986. [e] Aminabhavi et al., 1998. [f] Sastry et al., 1998. [g] TRC Tables, 1998.

Apparatus and Procedure. The excess molar volumes, which are reproducible to ±0.003 cm^3 mol^{-1}, were measured by means of a continuous dilution dilatometer in a fashion similar to that described by Dickinson et al. (1975). The details of its calibration, experimental setup, and measuring procedure have been described previously (Pal

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and Singh, 1994). Each run covered just over half of the mole fraction range so as to give an overlap between two runs.

The mole fraction of each mixture was obtained with an accuracy of 1×10^{-4} from the measured apparent masses of the components. All apparent masses were corrected for buoyancy. All molar quantities are based on the relative atomic mass table of 1986 issued by IUPAC (1986).

The kinematic viscosites in both the pure liquids and their mixtures were measured at 298.15 K and atmospheric pressure, by means of a suspended Ubbelohde viscometer, which was calibrated with doubly distilled water and benzene. The detailed procedure was described earlier (Pal and Singh, 1996). Care was taken to reduce evaporation during the measurements. An average of four or five sets of readings for flow times were taken for each pure liquid and liquid mixture. The value of the absolute viscosity (η) was obtained by multiplying the measured values of kinematic viscosity (ν) by the density. The sensivity of the viscometer corresponded to a precision in the viscosity of 1×10^{-4} mPa s. The reproducibility of the viscosity estimates was found to be within ± 0.003 mPa s. The time of efflux was measured with a digital stopwatch capable of measuring time to within ±0.01 s. All the measurements were carried out in a thermostatically controlled well-stirred water bath with temperature controlled to within ±0.01 K.

Results and Discussion

Table 2 summarizes the experimental values of the excess molar volumes for binary mixtures at number of mole fractions at 298.15 K and atmospheric pressure.

From the measured values of excess molar volumes, the densities of liquid mixtures were calculated from

$$\rho = (x_1 M_1 + x_2 M_2)/(V_m^E + x_1 V_1 + x_2 V_2) \qquad (1)$$

where M_1 and M_2 represent molar masses.

The viscosity deviations from a mole fraction average can be calculated from the following relationship (Aucejo et al., 1996; Ramadevi et al., 1996)

$$\Delta \ln \eta = \ln \eta - [x_1 \ln \eta_1 + x_2 \ln \eta_2] \qquad (2)$$

where x_1 and x_2 are the mole fractions, η is the dynamic viscosity of the mixtures, and η_1 and η_2 are the viscosities of components 1 and 2.

The derived densities and viscosities for the systems at 298.15 K are given in Table 3. The estimated accuracy of ρ is about 1×10^{-1} kg m^{-3}. The values of V_m^E and $\Delta \ln \eta$ for each mixture have been fitted to the Redlich−Kister type equation (Redlich−Kister, 1948)

$$Y = x_1 x_2 \sum_{i=1}^{k} A_i (x_1 - x_2)^{i-1} \qquad (3)$$

where A_i are the polynomial coefficients, which were obtained by fitting the equation to the experimental result with a least-squares regression method. The correlated results are given in Table 4, in which the tabulated standard deviation was defined as

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

where n is the number of measurements and k is the number of estimated parameters. Y refers to V_m^E/cm^3 mol^{-1} or Δ ln[η/mPa s]. For the mixtures $\sigma(V_m^E)$ <0.003 cm^3 mol^{-1}, in accord with the precision attainable with the dilatometer used.

The variation of the excess molar volume, and the viscosity varying with the whole mole fraction of ether for the investigated binary mixtures at 298.15 K are presented in Figures 1−4. Figures 1 and 2 show that the excess molar volumes are negative for all the mixtures over the whole mole fraction range. However, the excess molar volumes are less negative for the *tert*-amyl methyl ether−alkanol systems. It means that volume contraction occurs less by the addition of a CH$_2$ unit at the middle of the MTBE molecule. Similar variations in V_m^E occur in mixtures of branched alkoxy ethanols with water (Davis and Chacon,

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}
		tert-Butyl Methyl Ether (1) + Methanol (2)			
0.0120	−0.035	0.1938	−0.404	0.5974	−0.612
0.0360	−0.102	0.2018	−0.416	0.6227	−0.605
0.0443	−0.122	0.2687	−0.490	0.6799	−0.583
0.0549	−0.149	0.3106	−0.529	0.6980	−0.573
0.0861	−0.219	0.3700	−0.568	0.8064	−0.465
0.1018	−0.254	0.4185	−0.596	0.9001	−0.294
0.1101	−0.271	0.4758	−0.614	0.9467	−0.175
0.1497	−0.345	0.5380	−0.618	0.9898	−0.040
		tert-Butyl Methyl Ether (1) + 1-Propanol (2)			
0.0274	−0.061	0.3130	−0.537	0.5385	−0.636
0.0609	−0.135	0.3298	−0.552	0.5894	−0.635
0.0808	−0.180	0.3321	−0.554	0.6451	−0.614
0.1047	−0.230	0.3734	−0.590	0.6839	−0.589
0.1468	−0.310	0.3766	−0.588	0.7751	−0.512
0.1884	−0.381	0.4061	−0.611	0.8344	−0.439
0.2218	−0.433	0.4213	−0.619	0.9119	−0.289
0.2655	−0.488	0.4514	−0.625	0.9567	−0.163
0.3029	−0.531	0.4977	−0.638	0.9935	−0.027
		tert-Butyl Methyl Ether (1) + 1-Pentanol (2)			
0.0123	−0.031	0.3886	−0.694	0.5860	−0.761
0.0482	−0.123	0.3955	−0.705	0.6223	−0.749
0.0915	−0.230	0.4222	−0.722	0.6579	−0.730
0.1470	−0.349	0.4436	−0.734	0.7091	−0.699
0.1994	−0.448	0.4780	−0.755	0.7771	−0.626
0.2563	−0.544	0.4958	−0.760	0.8655	−0.470
0.3151	−0.626	0.5202	−0.762	0.9125	−0.340
0.3508	−0.662	0.5507	−0.770	0.9659	−0.160
0.3666	−0.676	0.5562	−0.762		
		tert-Amyl Methyl Ether (1) + Methanol (2)			
0.0243	−0.046	0.1784	−0.282	0.4572	−0.433
0.0345	−0.067	0.2171	−0.324	0.5365	−0.430
0.0481	−0.091	0.2473	−0.351	0.6210	−0.404
0.0652	−0.120	0.2735	−0.373	0.7210	−0.337
0.0904	−0.162	0.3017	−0.390	0.8131	−0.251
0.1139	−0.196	0.3344	−0.406	0.8946	−0.153
0.1392	−0.232	0.3937	−0.426	0.9418	−0.090
		tert-Amyl Methyl Ether (1) + 1-Propanol (2)			
0.0329	−0.066	0.3500	−0.433	0.6328	−0.446
0.0601	−0.112	0.3872	−0.452	0.7145	−0.400
0.1119	−0.199	0.4292	−0.462	0.7897	−0.334
0.1544	−0.262	0.4763	−0.470	0.8298	−0.284
0.1875	−0.295	0.5075	−0.472	0.9017	−0.184
0.2631	−0.374	0.5380	−0.470	0.9511	−0.096
0.3029	−0.404	0.5618	−0.468		
		tert-Amyl Methyl Ether (1) +1-Pentanol (2)			
0.0375	−0.071	0.3370	−0.437	0.6220	−0.494
0.0644	−0.126	0.3485	−0.447	0.6814	−0.468
0.0897	−0.162	0.3911	−0.471	0.7569	−0.410
0.1241	−0.218	0.4336	−0.492	0.8430	−0.308
0.1721	−0.278	0.4772	−0.501	0.9126	−0.190
0.2311	−0.347	0.5190	−0.508	0.9565	−0.102
0.2943	−0.413	0.5714	−0.509		

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
tert-Butyl Methyl Ether (1) + Methanol (2)											
0.0048	0.7860	0.549	0.1987	0.7707	0.496	0.4299	0.7577	0.433	0.7768	0.7441	0.370
0.0234	0.7843	0.547	0.2377	0.7682	0.483	0.4810	0.7553	0.420	0.8313	0.7421	0.363
0.0483	0.7822	0.544	0.2656	0.7665	0.475	0.5141	0.7539	0.413	0.8980	0.7398	0.356
0.0818	0.7794	0.535	0.3162	0.7635	0.462	0.5796	0.7512	0.399	0.9429	0.7381	0.346
0.1136	0.7769	0.526	0.3736	0.7604	0.446	0.6163	0.7498	0.394			
0.1601	0.7734	0.510				0.7002	0.7468	0.382			
tert-Butyl Methyl Ether (1) + 1-Propanol (2)											
0.0111	0.7986	1.902	0.2501	0.7816	0.998	0.4004	0.7717	0.729	0.7585	0.7502	0.427
0.0293	0.7972	1.804	0.3008	0.7782	0.889	0.4451	0.7688	0.671	0.8246	0.7464	0.397
0.0591	0.7950	1.651	0.3240	0.7767	0.850	0.4880	0.7661	0.624	0.8813	0.7432	0.378
0.1133	0.7911	1.416	0.3581	0.7744	0.792	0.5535	0.7621	0.562	0.9491	0.7391	0.356
0.1374	0.7894	1.327	0.3641	0.7740	0.779	0.6157	0.7584	0.512	0.9860	0.7368	0.344
0.2015	0.7849	1.123				0.6770	0.7548	0.471			
tert-Butyl Methyl Ether (1) + 1-Pentanol (2)											
0.0301	0.8091	3.179	0.3286	0.7892	1.346	0.5239	0.7750	0.843	0.8339	0.7507	0.440
0.0484	0.8080	3.008	0.3767	0.7858	1.174	0.5817	0.7707	0.725	0.8897	0.7460	0.398
0.1133	0.8037	2.461	0.4229	0.7825	1.052	0.6305	0.7670	0.653	0.9343	0.7420	0.370
0.1663	0.8002	2.091	0.4688	0.7791	0.939	0.6824	0.7629	0.587	0.9822	0.7376	0.349
0.2109	0.7973	1.840	0.5137	0.7758	0.843	0.7282	0.7593	0.538			
0.2824	0.7924	1.560				0.7788	0.7553	0.487			
tert-Amyl Methyl Ether (1) + Methanol (2)											
0.0050	0.7863	0.550	0.1987	0.7814	0.546	0.4252	0.7764	0.508	0.6944	0.7713	0.468
0.0339	0.7855	0.554	0.2009	0.7814	0.546	0.4676	0.7755	0.500	0.7437	0.7704	0.462
0.0488	0.7851	0.556	0.2338	0.7806	0.541	0.5142	0.7746	0.491	0.7936	0.7696	0.457
0.0714	0.7845	0.557	0.2861	0.7794	0.532	0.5503	0.7739	0.486	0.8642	0.7685	0.450
0.1129	0.7835	0.555	0.3409	0.7782	0.524	0.6236	0.7726	0.477	0.9029	0.7678	0.446
0.1508	0.7826	0.552	0.3790	0.7774	0.516	0.6574	0.7719	0.473			
tert-Amyl Methyl Ether (1) + 1-Propanol (2)											
0.0142	0.7989	1.889	0.2075	0.7918	1.180	0.4452	0.7836	0.767	0.7588	0.7737	0.530
0.0405	0.7979	1.759	0.2876	0.7890	1.002	0.4890	0.7821	0.720	0.8347	0.7714	0.496
0.0642	0.7970	1.647	0.2890	0.7889	0.999	0.5363	0.7806	0.677	0.9093	0.7691	0.455
0.0889	0.7961	1.548	0.3381	0.7872	0.912	0.5867	0.7789	0.633	0.9470	0.7679	0.454
0.1148	0.7952	1.461	0.3924	0.7853	0.833	0.6450	0.7772	0.594			
0.1520	0.7938	1.330				0.6933	0.7757	0.564			
tert-Amyl Methyl Ether (1) + 1-Pentanol (2)											
0.0219	0.8102	3.286	0.2110	0.8023	1.930	0.4353	0.7926	1.125	0.7266	0.7795	0.652
0.0388	0.8095	3.133	0.2721	0.7997	1.651	0.4908	0.7901	1.003	0.8027	0.7760	0.579
0.0752	0.8081	2.891	0.3347	0.7970	1.417	0.5534	0.7874	0.886	0.8732	0.7726	0.521
0.1143	0.8064	2.511	0.3464	0.7965	1.374	0.6141	0.7846	0.789	0.9484	0.7689	0.469
0.1631	0.8043	2.192	0.3850	0.7948	1.260	0.6759	0.7818	0.710	0.9936	0.7666	0.441

Table 4. Smoothing Coefficients A_i and Standard Deviations σ ($Y(x)$) of Eq 3 for the Binary Mixtures at 298.15 K

$Y(x)$	A_1	A_2	A_3	A_4	A_5	σ ($Y(x)$)
tert-Butyl Methyl Ether (1) + Methanol (2)						
V_m^E/cm^3 mol^{-1}	−2.460	−0.292	−0.880	−0.037		0.002
$\Delta \ln[\eta$/mPa s]	−0.158	0.010	0.221	−0.201		0.002
tert-Butyl Methyl Ether (1) + 1-Propanol (2)						
V_m^E/cm^3 mol^{-1}	−2.541	−0.217	−0.645	−0.696		0.003
$\Delta \ln[\eta$/mPa s]	−1.141	0.198	−0.184	−0.100	0.452	0.003
tert-Butyl Methyl Ether (1) + 1-Pentanol (2)						
V_m^E/cm^3 mol^{-1}	−3.033	−0.499	−0.474	−0.667	−0.382	0.003
$\Delta \ln[\eta$/m Pas]	−0.898	0.026	−0.086	−0.113		0.004
tert-Amyl Methyl Ether (1) + Methanol (2)						
V_m^E/cm^3 mol^{-1}	−1.746	0.215	−0.094	−0.018		0.002
$\Delta \ln[\eta$/mPa s]	0.0314	−0.194	0.214	−0.079		0.002
tert-Amyl Methyl Ether (1) + 1-Propanol (2)						
V_m^E/cm^3 mol^{-1}	−1.892	−0.043	−0.237	0.005		0.002
$\Delta \ln[\eta$/mPa s]	−1.053	0.289	−0.033	−0.248		0.006
tert-Amyl Methyl Ether (1) + 1-Pentanol (2)						
V_m^E/cm^3 mol^{-1}	−2.027	−0.271	−0.257	0.049		0.003
$\Delta \ln[\eta$/mPa s]	−0.930	0.139	0.129	−0.164		0.006

1991). For each ether, V_m^E decreases with increasing chain length of the alcohol. This behavior may be compared with the V_m^E results for mixtures of the branched ether 2-isopropoxyethanol with 1-alcohols (Pal and Sharma, 1998):

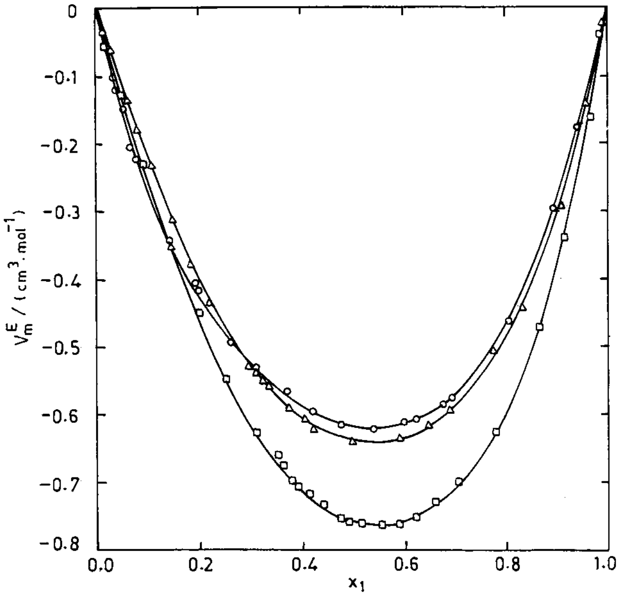

Figure 1. Excess molar volume (V_m^E) for *tert*-butyl methyl ether (1) + methanol (2) (○), + 1-propanol (2) (△), + 1-pentanol (2) (□), at 298.15 K. Full curves represent the smoothing of eq 3 with the coefficients of Table 4.

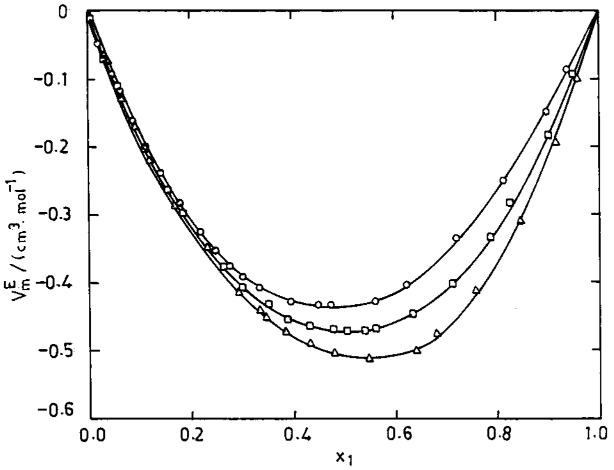

Figure 2. Excess molar volume (V_m^E) for *tert*-amyl methyl ether (1) + methanol (2) (○), + 1-propanol (2) (△), + 1-pentanol (2) (□), at 298.15 K. Full curves represent the smoothing of eq 3 with the coefficients of Table 4.

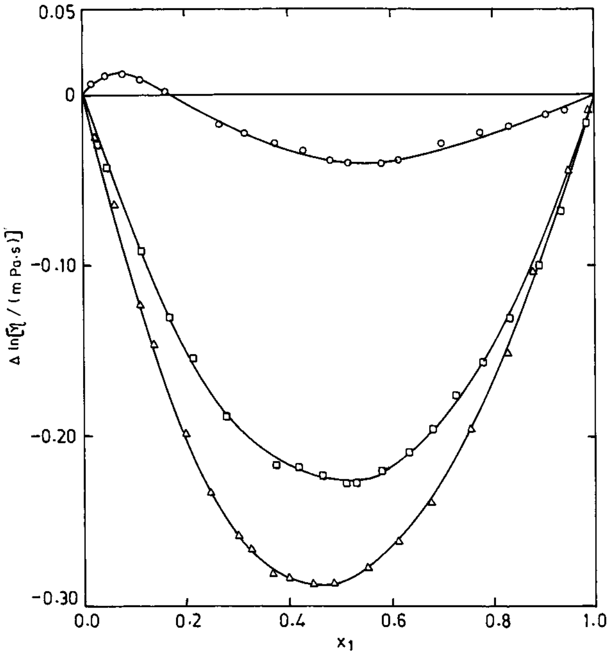

Figure 3. Viscosity deviations (Δ ln η) for *tert*-butyl methyl ether (1) + methanol (2) (○), + 1-propanol (2) (△), + 1-pentanol (2) (□), at 298.15 K. Full curves represent the smoothing of eq 3 with the coefficients of Table 4.

V_m^E increases as the alkyl chain length of the 1-alcohol increases. The magnitude of V_m^E is the result of a contribution from different effects: (i) specific interactions due to formation of multimers of alkanol and ether molecules; (ii) possible breaking of the hydrogen-bonded alkanol structure; and (iii) geometrical fitting of the ether into the remaining alkanol structure. Figures 3 and 4 illustrate that the viscosity deviations are negative for all the investigated systems and change sign for mixtures of methanol with MTBE and TAME. The absolute values of the deviations increase with increasing chain length of 1-alcohols. It can be observed that viscosity decreases to a greater extent on the initial addition of ether to alkanols for all mixtures. This type of behavior, however, does not explain the viscosity deviation in the present mixtures. The behavior of Δ ln η suggests that the strength of the specific interaction is not the only factor, but the molecular sizes and shapes of the components are equally important factors.

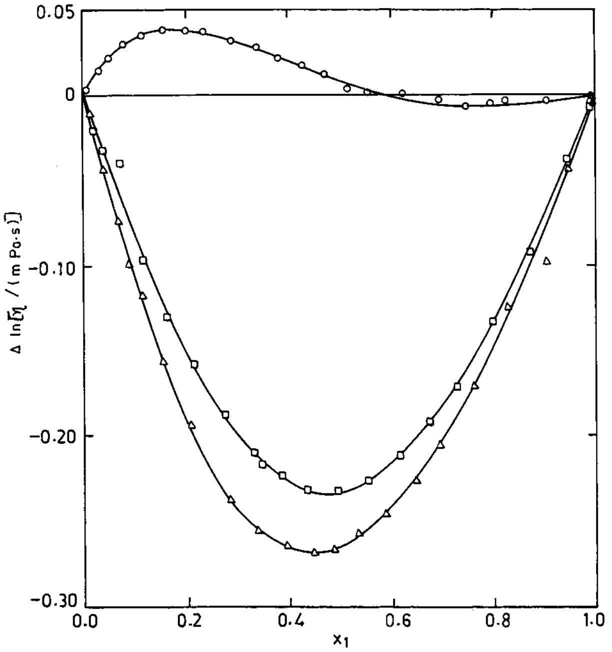

Figure 4. Viscosity deviations (Δ ln η) for *tert*-amyl methyl ether (1) + methanol (2) (○), + 1-propanol (2) (△), + 1-pentanol (2) (□) at 298.15 K. Full curves represent the smoothing of eq 3 with the coefficients of Table 4.

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