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Thermodynamic properties of *n*-alkoxyethanols + organic solvent mixtures
XVI. Excess molar volumes at 298.15 K for 2-alkoxyethanol + *n*-polyether systems

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Abstract

Excess molar volumes ($V_{\mathrm{m}}^{\mathrm{E}}$) at 298.15 K and atmospheric pressure for 2-methoxyethanol or 2-butoxyethanol + 2,5-dioxahexane, +2,5,8-trioxanonane, +3,6,9-trioxaundecane, +5,8,11-trioxapentadecane, or +2,5,8,11,14-pentaoxapentadecane, or for 2-ethoxyethanol + 2,5,8-trioxanonane, +3,6,9-trioxaundecane, or +5,8,11-pentaoxapentadecane have been obtained from densities measured with and Anton-Paar DMA 602 vibrating-tube densimeter.

They are usually small in absolute value. It is due to a compensation between positive and negative contributions to the excess molar volume. For example, free volume effects present in solutions with the pentaether are compensated by the positive contribution to $V_{\mathrm{m}}^{\mathrm{E}}$ from the rupture of dipole–dipole interactions between ether molecules.

Keywords: Experimental; Excess volume; 2-Alkoxyethanol; *n*-Polyether

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1. Introduction

The OH/O Project is a part of a general program, the so-called TOM Project [1,2], in which mixtures containing the oxygen (–O–) and/or the hydroxyl (–OH) groups are investigated in order to characterize their interactions. Particular attention is paid to intra and intermolecular effects, related to the presence of the –O– and/or –OH groups in the same or different molecules.

We have contributed largely to this project reporting experimental data at 298.15 K for molar excess enthalpies ($H_{\mathrm{m}}^{\mathrm{E}}$) molar excess volumes ($V_{\mathrm{m}}^{\mathrm{E}}$) molar excess heat capacities at constant pressure ($C_{\mathrm{p}}^{\mathrm{E}}$) and liquid–liquid equilibrium (LLE) of binary mixtures of alkoxyethanol (2-methoxyethanol, 2-ethoxyethanol, 2-butoxyethanol, 2-(2-methoxyethoxy)ethanol, 2-(2-etoxyethoxy)ethanol and 2-(2-butoxyethoxy)ethanol) with different organic solvents (see [3,4]). More recently, we have also reported vapor–liquid equilibria (VLE) for alkoxyethanol (2-methoxyethanol, 2-ethoxyethanol, 2-butoxyethanol or 2-(2-ethoxyethoxy)ethanol) + organic solvent (*n*-hexane, *n*-heptane, cyclohexane, benzene or carbon tetrachloride) systems (see [5,6]).

On the other hand, alkoxyethanol + alkane mixtures have been successfully characterized [7] in terms of the DISQUAC group contribution model [1,2]. In addition, solutions of two alkoxyethanols have been

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studied [8] in the framework of the Flory theory [9].

As a continuation of our experimental work, we present $V^{\mathrm{E}}_{\mathrm{m}}$ of systems formed by 2-methoxyethanol or 2-butoxyethanol + 2,5-dioxahexane, +2,5,8-trioxanonane, +3,6,9-trioxaundecane, +5,8,11-trioxapentadecane or +2,5,8,11,14-pentaoxapentadecane and for 2-ethoxyethanol + 2,5,8-trioxahexane, +3,6,9-trioxaundecane, or +5,8,11-trioxapentadecane, over the whole mole fraction range at 298.15 K and atmospheric pressure.

$V^{\mathrm{E}}_{\mathrm{m}}$ data for 2-methoxyethanol +2,5-dioxahexane, +2,5,8-trioxanonane, or +3,6,9-trioxaundecane have been reported previously by different authors [10,11].

2. Experimental section

2.1. Materials

Chemicals were provided by FLUKA: 2-methoxyethanol, 2-ethoxyethanol, 2,5,8-trioxanonane (puriss p.a. >99.5 mol%), 2,5-dioxahexane (purum ∼99 mol%), 3,6,9-trioxaundecane, 5,8,11-trioxapentadecane and 2,5,8,11,14-pentaoxapentadecane (purum >98 mol%). All of them were used after being carefully dried with molecular sieve (Union Carbide type 4 Å beads, from Fluka). Pure components measured densities at 298.15 K and atmospheric pressure are listed in Table 1. Generally, good agreement is found between the reported results and those of the present study.

2.2. Apparatus and procedure

Excess volumes were calculated from experimental densities of the pure liquids and their mixtures. Densities were measured at 298.15 K with and Anton-Paar DMA 602 vibrating-tube densimeter operating under static conditions. Temperature was measured by resistance measurement of an Anton-Paar thermistor (using a PM-2634 Philips multimeter) previously tested against a HP-2804 A quartz thermometer calibrated on the basis of the ITS-90 scale of temperature using the triple point of water. The densimeter was thermostated to ±0.003 K. The accuracy in $V^{\mathrm{E}}_{\mathrm{m}}$ is better than $\pm(0.01\ V^{\mathrm{E}}_{\mathrm{max}} + 0.005\ \mathrm{cm^3\,mol^{-1}})$, where $V^{\mathrm{E}}_{\mathrm{max}}$ denotes the maximum $|V^{\mathrm{E}}_{\mathrm{m}}|$ value. More details are given elsewhere [12].

Table 1
Experimental densities, ρ_{exp}, of the pure compounds at 298.15 K and atmospheric pressure, and corresponding values found in literature, ρ_{lit}

Compound	ρ_{exp} (g cm^{-3})	ρ_{lit} (g cm^{-3})
2-Methoxyethanol	0.96012	0.96024[a] 0.9602[b] 0.96019[c] 0.96018[d] 0.96002[e]
2-Ethoxyethanol	0.92728	0.92766[c] 0.92515[d] 0.92502[e]
2-Butoxyethanol	0.89586	0.89677[c] 0.89621[d] 0.89581[e]
2,5-Dioxahexane	0.8617	0.86370[a] 0.8626[b] 0.86153[f] 0.8615[g] 0.86155[h]
2,5,8-Trioxanonane	0.9391	0.9384[a] 0.9389[b] 0.93905[f] 0.9385[g]
3,6,9-Trioxaundecane	0.901938	0.902[a,i] 0.9035[b] 0.90390[f] 0.9021[g]
5,8,11-Trioxapentadecane	0.878427	0.8781[b] 0.87862[f] 0.87830[h]
2,5,8,11,14-Pentaoxapentadecane	1.006479	1.00653[f] 1.00652[h] 1.00662[j]

[a] [14].
[b] [10].
[c] [15].
[d] [16].
[e] [17].
[f] [12].
[g] [18].
[h] [19].
[i] Extrapolated value using ρ (20 °C) and dρ/dt.
[j] [20].

3. Results and discussion

The results of $V^{\mathrm{E}}_{\mathrm{m}}$ as a function of mole fraction of the alkoxyethanol, x_1, are collected in Table 2. The data were fitted by unweighted least-squares polynomial regression to the equation:

$$V_{\mathrm{m}}^{\mathrm{E}}(\mathrm{cm^3\,mol^{-1}}) = x_1(1-x_1)\sum_{i=1}^{k} A_i(2x_1-1)^{i-1} \quad (1)$$

The number of coefficients, k, used in Eq. (1) for each mixture was determined by applying a F-test [13] at the 95% confidence level. Table 3 lists the parameters A_i obtained in the regression, together with the standard deviations, σ, defined by:

$$\sigma(V_{\mathrm{m}}^{\mathrm{E}})(\mathrm{cm^3\,mol^{-1}}) = \left[\frac{\sum(V_{\mathrm{m,\,calc}}^{\mathrm{E}} - V_{\mathrm{m,\,exp}}^{\mathrm{E}})^2}{N-k}\right]^{1/2} \quad (2)$$

where N stands for the number of direct experimental values. For all the mixtures $\sigma(V_{\mathrm{m}}^{\mathrm{E}})/V_{\mathrm{max}}^{\mathrm{E}} < 0.02$ in accord with the precission attainable with the instrument used. A graphical representation of the experimental results, smoothing curves from Eq. (1), and comparison with literature results are shown in Figs. 1–3.

Good agreement is found between results from this work and those from Pal et al. [10] for 2-methoxyethanol (1) + 2,5-dioxahexane (2) ($V_{\mathrm{m}}^{\mathrm{E}}(x = 0.5) = -0.2242\,\mathrm{cm^3\,mol^{-1}}$ versus $-0.222\,\mathrm{cm^3\,mol^{-1}}$ [10]); in contrast, results from Corradini et al. [11] show a total disagreement with both of them (see Fig. 1). The good agreement with results of Pal et al. is also observed for the 2-methoxyethanol (1) + 3,6,9-trioxaundecane (2) system ($V_{\mathrm{m}}^{\mathrm{E}}(x = 0.5) = -0.1867\,\mathrm{cm^3}$ $\mathrm{mol^{-1}}$ versus $-0.185\,\mathrm{cm^3\,mol^{-1}}$ [10]), is slightly worse for 2-methoxyethanol (1) + 2,5,8-trioxanonane (2) mixture ($V_{\mathrm{m}}^{\mathrm{E}}(x = 0.5) = -0.0944\,\mathrm{cm^3\,mol^{-1}}$ versus $-0.1011\,\mathrm{cm^3\,mol^{-1}}$ [10]). In the case of 2-methoxyethanol (1) + 5,8,11-trioxapentadecane (2) system, $V_{\mathrm{m}}^{\mathrm{E}}(x)$ from Pal et al. [10] behaves similarly to our data (Fig. 2a). Nevertheless, their values are somewhat lower than those presented here ($V_{\mathrm{m}}^{\mathrm{E}}(x = 0.5) = 0.0854\,\mathrm{cm^3\,mol^{-1}}$ versus $0.066\,\mathrm{cm^3\,mol^{-1}}$ [10]).

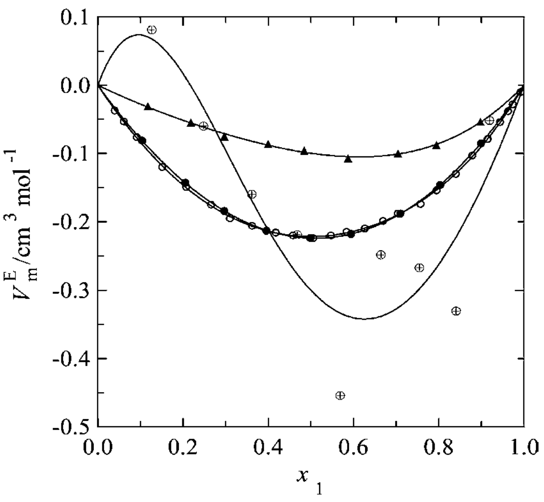

Fig. 1. Excess molar volume ($V_{\mathrm{m}}^{\mathrm{E}}$) at 298.15 K and atmospheric pressure for 2-methoxyethanol (1) + n-polyether (2) mixtures. Solid lines, results from the smoothing Eq. (1) with the coefficients listed in Table 3. Points, experimental data: +2,5-dioxahexane (this work) (●); [10] (○); [11] (⊕); +2,5,8,11,14-pentaoxapentadecane (▲).

For the investigated mixtures, $V_{\mathrm{m}}^{\mathrm{E}}$ is the result of several opposing effects. Changes in self-association (inter or intramolecular [7]) of alkoxyethanols and physical interactions between alkoxyethanol molecules, or between ether molecules lead to increased $V_{\mathrm{m}}^{\mathrm{E}}$. On the contrary, free volume effects, interstitial accommodation or interactions between unlike molecules decrease $V_{\mathrm{m}}^{\mathrm{E}}$.

In spite of the great complexity of the mixtures under study, some general trends can be remarked.

(i) Independently of the 2-alkoxyethanol, $V_{\mathrm{m}}^{\mathrm{E}}(x = 0.5)$ varies in the sequence: 2,5,8-trioxanonane > 3,6,9-trioxaundecane < 5,8,11-trioxapentadecane. The decrease of $V_{\mathrm{m}}^{\mathrm{E}}$ when passing from 2,5,8-trioxanonane to 3,6,9-trioxaundecane may be due to the larger positive contribution to $V_{\mathrm{m}}^{\mathrm{E}}$ from the rupture of the dipole–dipole interactions between oxaalkane molecules in the case of 2,5,8-trioxanonane.

Free volume effects should be large in systems with 5,8,11-trioxapentadecane as the asymmetric $V_{\mathrm{m}}^{\mathrm{E}}$ curve indicate (Fig. 2a). However, the mentioned effects are compensated by weaker interactions between unlike molecules, because the –O– atoms are more screened in 5,8,11-trioxapentadecane. In addition, more dipole–dipole interactions between alkoxyethanol molecules are disrupted. Hence, $V_{\mathrm{m}}^{\mathrm{E}}$ increases when passing from 3,6,9-trioxaundecane to 5,8,11-trioxapentadecane.

(ii) Free volume effects are also important in solutions with the penthaether. Note that $V_{\mathrm{m}}^{\mathrm{E}}$ curves are newly skewed to the component with smaller

Table 2
Excess molar volumes (V_m^E) for *n*-alkoxyethanol (1) + *n*-polyether (2) at 298.15 K and atmospheric pressure

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})
2-Methoxyethanol (1) + 2,5-dioxahexane (2)					
0.1038	−0.0810	0.3953	−0.2131	0.7101	−0.1880
0.2053	−0.1431	0.4988	−0.2238	0.8033	−0.1462
0.2966	−0.1843	0.5941	−0.2180	0.8991	−0.0852
2-Methoxyethanol (1) + 2,5,8-trioxanonane (2)					
0.0562	−0.0162	0.4040	−0.0871	0.7961	−0.0712
0.1130	−0.0311	0.4966	−0.0944	0.8809	−0.0481
0.2022	−0.0519	0.5936	−0.0948	0.9440	−0.0250
0.3038	−0.0726	0.6943	−0.0883		
2-Methoxyethanol (1) + 3,6,9-trioxaundecane (2)					
0.0706	−0.0454	0.3986	−0.1740	0.8065	−0.1341
0.1174	−0.0700	0.4954	−0.1867	0.8984	−0.0827
0.2105	−0.1140	0.6018	−0.1861	0.9477	−0.0472
0.3028	−0.1483	0.7041	−0.1691		
2-Methoxyethanol (1) + 5,8,11-trioxapentadecane (2)					
0.1184	0.0235	0.5023	0.0860	0.7029	0.0991
0.2205	0.0433	0.5068	0.0848	0.8028	0.0881
0.3154	0.0571	0.6119	0.0959	0.9051	0.0559
0.4084	0.0742				
2-Methoxyethanol (1) + 2,5,8,11,14-pentaoxapentadecane (2)					
0.1172	−0.031	0.3996	−0.086	0.7042	−0.100
0.2183	−0.055	0.4844	−0.096	0.7943	−0.087
0.2967	−0.075	0.5872	−0.107	0.8977	−0.053
2-Ethoxyethanol (1) + 2,5,8-trioxanonane (2)					
0.1004	−0.0044	0.3898	−0.0186	0.6931	−0.0221
0.1984	−0.0084	0.4892	−0.0215	0.7956	−0.0176
0.2919	−0.0137	0.5900	−0.0229	0.8977	−0.0112
2-Ethoxyethanol (1) + 3,6,9-trioxaundecane (2)					
0.0633	−0.0307	0.3956	0.1278	0.7035	0.1218
0.1103	−0.0483	0.4893	−0.1372	0.8010	−0.0944
0.1897	−0.0780	0.6046	−0.1363	0.8970	−0.0538
0.3031	−0.1100				
2-Ethoxyethanol (1) + 5,8,11-trioxapentadecane (2)					
0.1175	0.0145	0.4124	0.0404	0.7042	0.0472
0.2149	0.0252	0.5057	0.0447	0.8039	0.0389
0.3169	0.0332	0.6048	0.0475	0.9037	0.0243
2-Butoxyethanol (1) + 2,5-dioxahexane (2)					
0.1106	−0.0437	0.3931	−0.0995	0.7067	−0.0809
0.2050	−0.0725	0.4981	−0.1006	0.8039	−0.0609
0.2985	−0.0881	0.6140	−0.0941	0.9004	−0.0344
2-Butoxyethanol (1) + 2,5,8-trioxanonane (2)					
0.1081	0.0273	0.3972	0.0614	0.6393	0.0538
0.2065	0.0450	0.4973	0.0625	0.8025	0.0339
0.3025	0.0552	0.6022	0.0570	0.8967	0.0178
2-Butoxyethanol (1) + 3,6,9-trioxaundecane (2)					
0.0974	−0.0494	0.3968	−0.1431	0.7045	−0.1374
0.1964	−0.0898	0.5010	−0.1532	0.7996	−0.1094
0.2987	−0.1227	0.6010	−0.1518	0.8965	−0.0677

Table 2 (*Continued*)

x_1	V_m^E ($cm^3\,mol^{-1}$)	x_1	V_m^E ($cm^3\,mol^{-1}$)	x_1	V_m^E ($cm^3\,mol^{-1}$)
2-Butoxyethanol (1) + 5,8,11-trioxapentadecane (2)					
0.1031	−0.0278	0.4100	−0.0854	0.7046	−0.0871
0.1972	−0.0479	0.5037	−0.0938	0.8066	−0.0696
0.3109	−0.0713	0.6048	−0.0949	0.9029	−0.0416
2-Butoxyethanol (1) + 2,5,8,11,14-pentaoxapentadecane (2)					
0.1007	0.0265	0.3921	0.0762	0.6853	0.0782
0.2013	0.0518	0.4984	0.0819	0.7889	0.0633
0.3081	0.0690	0.6058	0.0802	0.8975	0.0384

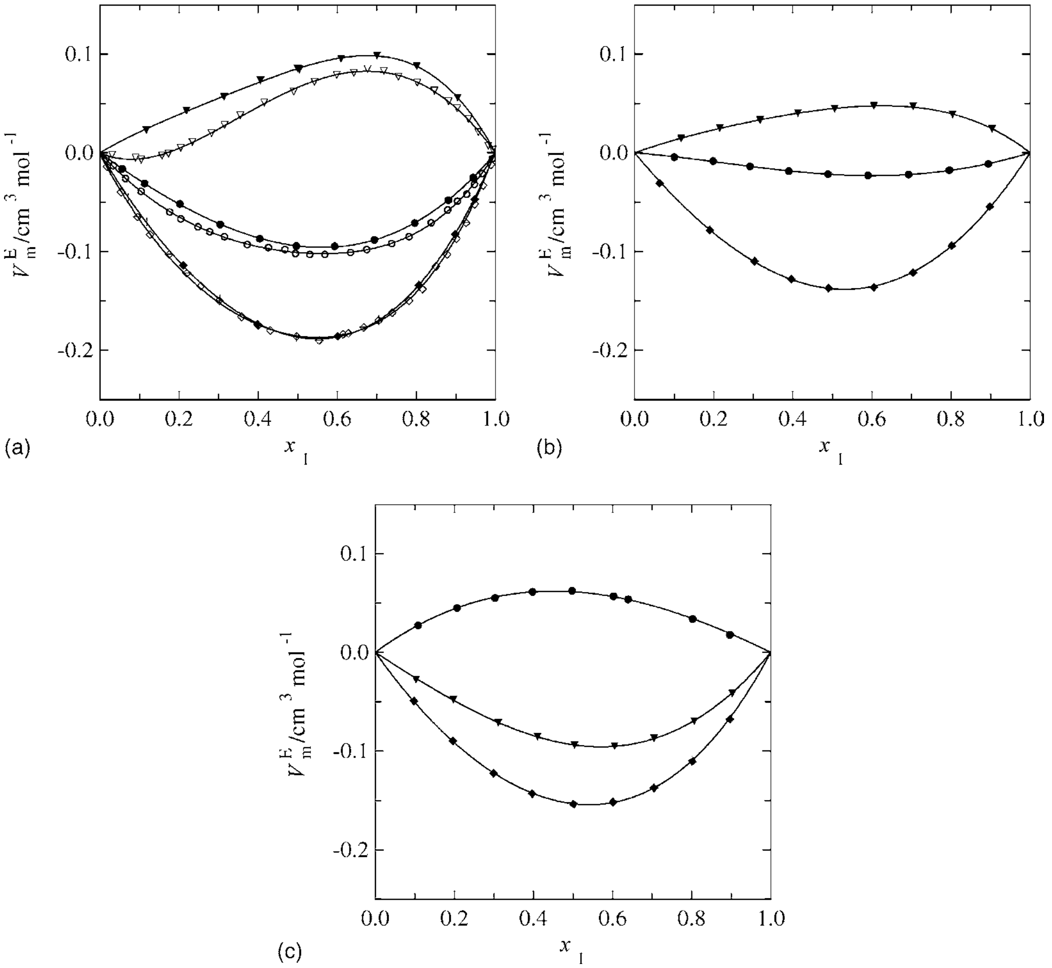

Fig. 2. Excess molar volume (V_m^E) at 298.15 K and atmospheric pressure for *n*-alkoxyethanol (1) + *n*-triether (2) mixtures. Solid lines, results from the smoothing Eq. (1) with the coefficients listed in Table 3. Points, experimental data: (a) 2-methoxyethanol + 2,5,8-trioxanonane (●); +3,6,9-trioxaundecane (◆); +5,8,11-trioxapentadecane (▼); 2,5,8-trioxanonane (○); +3,6,9-trioxaundecane (◇); +5,8,11-trioxapentadecane (▽) [10]. (b) 2-Ethoxyethanol + 2,5,8-trioxanonane (●); +3,6,9-trioxaundecane (◆); +5,8,11-trioxapentadecane (▼). (c) 2-Butoxyethanol + 2,5,8-trioxanonane (●); +3,6,9-trioxaundecane (◆); +5,8,11-trioxapentadecane (▼).

Table 3
Coefficients A_i and standard deviations, $\sigma(V_m^E)$ (Eq. (2)) for representation of the V^E at 298.15 K and atmospheric pressure by Eq. (1)

System	A_1	A_2	A_3	$\sigma(V_m^E)$ ($cm^3\ mol^{-1}$)	$V_m^E(x = 0.5)$ ($cm^3\ mol^{-1}$)
2-Methoxyethanol (1)					
+2,5-Dioxahexane (2)	−0.89666 ± 0.0008	−0.03857 ± 0.0016	−0.0120 ± 0.004	0.0004	−0.2242
+2,5,8-Trioxanonane (2)	−0.37759 ± 0.0008	−0.09501 ± 0.0016	−0.0115 ± 0.003	0.0004	−0.0944
+3,6,9-Trioxaundecane (2)	−0.7469 ± 0.0018	−0.1395 ± 0.003	−0.0707 ± 0.007	0.0007	−0.1867
+5,8,11-Trioxapentadecane (2)	0.3417 ± 0.002	0.2576 ± 0.005	0.155 ± 0.012	0.0011	0.0854
+2,5,8,11,14-Pentaoxapentadecane (2)	−0.3987 ± 0.006	−0.177 ± 0.011	−0.080 ± 0.03	0.002	−0.100
2-Ethoxyethanol (1)					
+2,5,8-Trioxanonane (2)	−0.08731 ± 0.0008	−0.0454 ± 0.0016	0.0114 ± 0.004	0.0003	−0.0218
+3,6,9-Trioxaundecane (2)	−0.5511 ± 0.002	−0.0696 ± 0.005		0.0012	−0.1378
+5,8,11-Trioxapentadecane (2)	0.1794 ± 0.0011	0.0856 ± 0.002	0.0490 ± 0.005	0.0005	0.0448
2-Butoxyethanol (1)					
+2,5-Dioxahexane (2)	−0.4043 ± 0.0018	0.0428 ± 0.003	−0.0225 ± 0.008	0.0008	−0.1011
+2,5,8-Trioxanonane (2)	0.24614 ± 0.0013	−0.0498 ± 0.003		0.0007	0.0615
+3,6,9-Trioxaundecane (2)	−0.6135 ± 0.0016	−0.0952 ± 0.003	−0.0456 ± 0.007	0.0007	−0.1534
+5,8,11-Trioxapentadecane (2)	−0.3745 ± 0.0012	−0.1126 ± 0.003		0.0007	−0.0936
+2,5,8,11,14-Pentaoxapentadecane (2)	0.3291 ± 0.004	0.0559 ± 0.007	0.058 ± 0.017	0.0016	0.0823

$V_m^E(x = 0.5)$ is the excess molar volume calculated from Eq. (1) at equimolar composition.

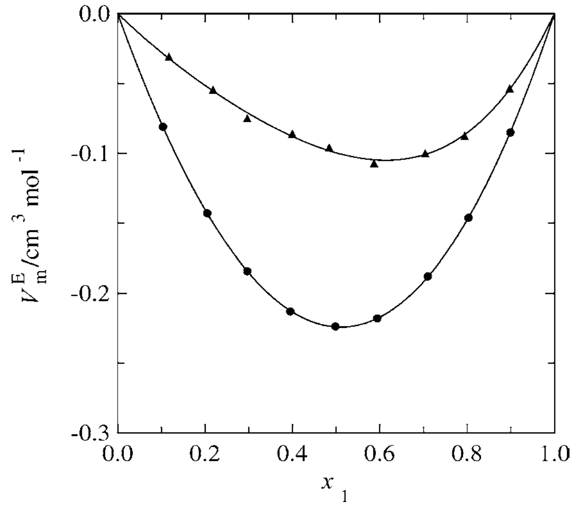

Fig. 3. Excess molar volume, V_m^E at 298.15 K and atmospheric pressure for 2-butoxyethanol (1) + n-polyether (2) mixtures. Solid lines, results from the smoothing Eq. (1) with the coefficients listed in Table 3. Points, experimental data: +2,5-dioxahexane (●); +2,5,8,11,14-pentaoxapentadecane (▲).

molar volume (Figs. 1 and 3). Nevertheless V_m^E is small in absolute value, due to the positive contribution to V_m^E from the breaking of dipole–dipole interactions between ether molecules.

(iii) In systems with 2,5,8-trioxanonane, V_m^E varies in the sequence: 2-methoxyethanol $<$ 2-ethoxyethanol $<$ 2-butoxyethanol. This is probably due to interactions between unlike molecules become weaker in the same order; as well as the difference in size between components increases in the opposite order.

(iv) On the contrary, in solutions with 5,8,11-trioxapentadecane, V_m^E varies as follows: 2-methoxyethanol $>$ 2-ethoxyethanol $>$ 2-butoxyethanol. As already mentioned, the oxygen atoms here are more screened, and it leads to larger number of broken dipole–dipole interactions between alkoxyethanol molecules. Note that H_m^E for 2-alkoxyethanol + given n-alkane varies in the same way [7].

4. Conclusions

Data on V_m^E at 298.15 K and atmospheric pressure for 2-methoxyethanol or 2-butoxyethanol + 2,5-dioxahexane, +2,5,8-trioxanonane, +3,6,9-trioxaundecane, +5,8,11-trioxapentadecane or +2,5,8,11,14-pentaoxapentadecane, or for 2-ethoxyethanol + 2,5,8-trioxanonane, +3,6,9-trioxaundecane and +5,8,11-trioxapentadecane are reported. The excess molar volumes are usually small in absolute value due to a compensation between positive and negative contributions to the excess molar volume.

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