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Densities, Viscosities, and Excess Properties of Trifluoroethanol–Water, Tetraethylene Glycol Dimethylether–Water, and Trifluoroethanol–Tetraethylene Glycol Dimethylether at 303.15 K

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Densities and kinematic viscosities of trifluoroethanol + water, tetraethylene glycol dimethylether + water, and trifluoroethanol + tetraethylene glycol dimethylether have been measured at 303.15 K and atmospheric pressure over the entire range of composition. Dynamic viscosities, excess volumes, excess viscosities, and excess Gibbs energies of activation of flow were obtained from the experimental results. The excess volumes were negative, whereas excess viscosities and energies of activation were positive, presenting the three thermodynamic properties asymmetric composition dependence. The kinematic viscosities were used to test McAllister, Stephan, and Soliman and Marshall correlations.

KEY WORDS: density; excess properties; fluorinol; kinematic viscosity; trifluoroethanol; tetraethylene glycol dimethylether.

1. INTRODUCTION

In recent years, investigations concerning working fluid systems for absorption processes have increased considerably. Stephan and Seher [1] have proposed the mixture trifluoroethanol (TFE) and tetraethylene glycol dimethylether (E181) as a working fluid in absorption heat pumps and

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heat transformers. Subsequently, to improve the general characteristic of the cycle, Stephan and Hengerer [2] have proposed adding water using a mixture of TFE and water as refrigerant and E181 as absorbant.

The thermodynamic properties of the ternary mixture TFE–H_2O–E181 and the corresponding binary mixtures are needed to study the performance of the cycle. Among the thermodynamic properties necessary to evaluate the performance of the working mixtures are the densities and the viscosities. We have started an investigation of viscosities, densities, and excess enthalpies of the ternary and corresponding binaries to provide data for these thermodynamic properties.

Heat-of-mixing data for TFE–E181 have been reported recently by Svejda et al. [3] at 293.15 K and atmospheric pressure. They also reported the excess molar volumes at 293.15 and 313.15 K and atmospheric pressure together with the isothermal compressibilities at 293.15 K and at pressures ranging between 0 and 10 MPa. Excess molar enthalpies of TFE–H_2O have been measured by Denda et al. [4] and Cooney and Morcom [5] at 298.15, 318.15, and 338.15 K. Krumbeck and Schulz [6] have reported heat-of-mixing data for TFE–H_2O, E181–H_2O, and TFE–E181 at six temperatures ranging between 298.15 and 448.15 K and at pressures of 0.4 MPa and saturation.

Isothermal vapor–liquid equilibria at 298.15 for TFE–H_2O have been measured by Cooney and Morcom [5] and by Smith et al. [7]. Jadot and Fraiha [8] have measured isobaric vapor–liquid equilibria of the same mixture.

Kinematic viscosities for TFE–H_2O have been measured by Kaiser et al. [9], but they have not reported numerical values. Kobayashi and Nagashima [10] have presented experimental dynamic viscosities for two composition of aqueous solutions of TFE in the temperature range 273.15 to 453.15 K and pressures up to 40 MPa and kinematic viscosities at atmospheric pressure over the whole range of compositions at 293.15 and 313.15 K.

In this paper we report densities, viscosities, excess molar volumes, dynamic viscosities, and Gibbs activation energies at 303.15 K and atmospheric pressure of the binary mixtures TFE–H_2O, E181–H_2O, and TFE–E181. The viscosity data were correlated by the method of McAllister [11], Stephan and Heckenberger [12], and Soliman and Marshall [13].

2. EXPERIMENTS

2.1. Materials

TFE (Fluka; >99%) was distilled; E181 (Fluka; >98%) was used without further purification. H_2O was deionized and distilled once from alkaline potassium permanganate and twice from itself. TFE and E181 were carefully dried with molecular sieves type 4A (Fluka). All chemicals were partially degassed before used. Densities and viscosities of the pure components at 303.15 K and atmospheric pressure are in agreement with literature values [14, 15].

2.2. Equipment

A digital densimeter (Anton Paar DMA602H-DMA60) was employed for determination of the densities of the pure components and the binary mixtures. The vibrating-tube temperature was regulated to better than 0.01 K with a Julabo F20 thermostat together with a digital precision thermometer (Anton Paar MKT100). Immediately prior to each series of measurements, water and air were used to calibrate the densimeter. Binary mixtures were prepared using a Mettler AE260 balance. The possible error in the mole fractions is estimated to be less than 0.0001.

Kinematic viscosities of the pure liquids and their mixtures were measured with a Model AVS 300 Schott apparatus, consisting of a basic control unit, a measuring stand, a thermostat, and a calibrated Ubbelohde suspended-level viscometer. Hagenbach corrections were applied for the Ubbelohde viscometer supplied by Schott–Geräte. Time measurements were made automatically using two TC sensors across the viscometer and an electronic timer. The temperature was kept constant at ±0.01 K. The accuracy of the kinematic viscosity data was estimated to be better than $5 \times 10^{-4}\ \mathrm{mm^2 \cdot s^{-1}}$.

3. RESULTS

3.1. Excess Properties

Excess thermodynamic properties were calculated with the following expression:

$$Q^{E} = Q_{m} - (x_1 Q_1 + x_2 Q_2) \tag{1}$$

where either $Q^E = V^E$ ($\mathrm{cm^3 \cdot mol^{-1}}$) or $Q^E = \eta^E$ (mPa · s), Q_m, Q_1, and Q_2 are the corresponding molar properties of the mixture and the pure liquids, respectively, and x_1 and x_2 are the mole fractions. Table I displays the experimental densities and the excess molar volumes at 303.15 K for the three mixtures studied.

The excess Gibbs energies of activation for viscous flow were obtained using the Reed–Taylor [16] equation:

$$\ln[\eta v] = x_1 \ln[\eta_1 v_1] + x_2 \ln[\eta_2 v_2] + \frac{{}^{*}G^{E}}{RT} \tag{2}$$

Table I. Density and Excess Volume at 303.15 K

x_1	ρ $(g \cdot cm^{-3})$	V^E $(cm^3 \cdot mol^{-1})$
	TFE–H_2O	
0.0000	0.99564	—
0.0198	1.03122	−0.1330
0.0302	1.04733	−0.1806
0.0499	1.07645	−0.2870
0.0985	1.13347	−0.4629
0.1458	1.17428	−0.5475
0.1597	1.18442	−0.5641
0.1918	1.20531	−0.5916
0.2373	1.23027	−0.6161
0.2497	1.23623	−0.6188
0.2818	1.25072	−0.6311
0.3180	1.26488	−0.6327
0.3577	1.27852	−0.6302
0.4370	1.30028	−0.5873
0.4986	1.31405	−0.5472
0.5375	1.32150	−0.5142
0.6012	1.33237	−0.4620
0.6940	1.34529	−0.3675
0.7471	1.35180	−0.3196
0.8068	1.35776	−0.2376
0.8396	1.36081	−0.1944
0.8987	1.36580	−0.1126
0.9237	1.36789	−0.0842
0.9609	1.37089	−0.0441
1.0000	1.37382	—
	E181–H_2O	
0.0000	0.99564	—
0.0301	1.01863	−0.5044
0.0495	1.02660	−0.7807
0.0600	1.02920	−0.9050
0.0982	1.03424	−1.2858
0.1197	1.03370	−1.3980
0.1431	1.03267	−1.4954
0.2025	1.02806	−1.5889
0.2522	1.02446	−1.6011
0.2918	1.02192	−1.5843
0.3380	1.01899	−1.5165
0.3870	1.01635	−1.4288
0.4741	1.01273	−1.2634
0.4986	1.01191	−1.2170
0.5426	1.01045	−1.1147

Table I. *(Continued)*

x_1	ρ $(\mathrm{g \cdot cm^{-3}})$	V^E $(\mathrm{cm^3 \cdot mol^{-1}})$
0.5889	1.00913	−1.0081
0.6568	1.00753	−0.8570
0.7092	1.00643	−0.7317
0.7563	1.00551	−0.6105
0.8029	1.00474	−0.4985
0.8634	1.00373	−0.3290
0.9201	1.00299	−0.1897
0.9571	1.00255	−0.0985
1.0000	1.00211	—
	TFE–E181	
0.0000	1.00211	—
0.0499	1.00842	−0.0020
0.1007	1.01533	−0.0082
0.1499	1.02249	−0.0078
0.2006	1.03044	−0.0072
0.2489	1.03862	−0.0070
0.3023	1.04845	−0.0087
0.3524	1.05849	−0.0065
0.4055	1.07017	−0.0081
0.4536	1.08178	−0.0089
0.5017	1.09452	−0.0094
0.5511	1.10898	−0.0118
0.6018	1.12548	−0.0123
0.6484	1.14242	−0.0148
0.7023	1.16459	−0.0249
0.7531	1.18855	−0.0423
0.7975	1.21259	−0.0750
0.8521	1.24680	−0.1170
0.8760	1.26369	−0.1342
0.9013	1.28303	−0.1492
0.9211	1.29922	−0.1509
0.9474	1.32235	−0.1403
0.9540	1.32837	−0.1292
0.9652	1.33876	−0.1004
0.9743	1.34762	−0.0822
1.0000	1.37382	—

Table II. Density, Viscosity, and Activation Energy at 303.15 K for Binary Mixtures

x_1	ρ (g·cm^{-3})	ν (mm^2·s^{-1})	η (mPa·s)	η^E (mPa·s)	$^*G^E$ (J·mol^{-1})
			TFE–H_2O		
0.0000	0.99564	0.8022	0.7987	—	—
0.0431	1.06640	1.0626	1.1332	0.3020	936.7
0.0716	1.10191	1.1719	1.2913	0.4387	1295.2
0.1072	1.14098	1.2593	1.4368	0.5573	1582.5
0.1526	1.17924	1.3156	1.5514	0.6377	1785.3
0.2127	1.21678	1.3268	1.6144	0.6555	1872.3
0.2506	1.23664	1.3186	1.6306	0.6431	1872.0
0.2959	1.25624	1.2918	1.6228	0.6012	1817.2
0.3507	1.27611	1.2593	1.6070	0.5441	1723.6
0.4187	1.29526	1.2125	1.5705	0.4564	1559.5
0.5051	1.31529	1.1602	1.5260	0.3468	1320.5
0.5574	1.32490	1.1350	1.5038	0.2852	1169.7
0.6184	1.33476	1.1185	1.4929	0.2283	982.6
0.6718	1.34220	1.1002	1.4767	0.1719	844.3
0.7137	1.34771	1.0855	1.4629	0.1265	708.9
0.7718	1.35427	1.0845	1.4687	0.0885	557.7
0.8534	1.36198	1.0914	1.4865	0.0448	350.1
0.9382	1.36906	1.1093	1.5187	0.0132	143.2
1.0000	1.37382	1.1298	1.5521	—	—
			E181–H_2O		
0.0000	0.99564	0.8022	0.7987	—	—
0.0511	1.02700	3.3428	3.4331	2.5247	4258.1
0.0747	1.03114	4.1339	4.2626	3.3035	4961.1
0.1080	1.03399	4.7948	4.9578	3.9272	5483.9
0.1586	1.03147	5.1462	5.3082	4.1690	5754.3
0.1950	1.02864	5.0361	5.1803	3.9630	5696.9
0.2442	1.02504	4.7886	4.9085	3.5856	5501.0
0.3140	1.02051	4.3315	4.4203	2.9475	5058.1
0.3509	1.01830	4.1576	4.2337	2.6817	4821.6
0.4210	1.01494	3.8669	3.9247	2.2222	4338.9
0.5176	1.01128	3.5274	3.5672	1.6573	3617.2
0.6055	1.08874	3.3343	3.3634	1.2648	2972.4
0.6597	1.00747	3.2483	3.2726	1.0577	2575.6
0.7231	1.00616	3.1639	3.1834	0.8323	2105.3
0.7983	1.00482	3.0881	3.1030	0.5905	1545.2
0.8819	1.00349	3.0268	3.0374	0.3454	918.8
0.9251	1.00293	2.9902	2.9990	0.2143	583.0
1.0000	1.00211	2.9393	2.9455	—	—

Table II. *(Continued)*

x_1	ρ (g·cm^{-3})	ν (mm^2·s^{-1})	η (mPa·s)	η^E (mPa·s)	$^*G^E$ (J·mol^{-1})
		TFE–E181			
0.0000	1.00211	2.9393	2.9455	—	—
0.0590	1.00986	2.9068	2.9354	0.0721	149.7
0.0993	1.01492	2.8727	2.9155	0.1084	239.8
0.1483	1.02197	2.8422	2.9046	0.1658	356.7
0.2135	1.03262	2.7716	2.8620	0.2140	481.2
0.2619	1.03981	2.7331	2.8419	0.2614	582.8
0.3014	1.04732	2.6952	2.8227	0.2972	657.5
0.3421	1.05641	2.6522	2.8018	0.3330	728.4
0.3991	1.06864	2.5840	2.7614	0.3720	815.6
0.4583	1.08316	2.5106	2.7194	0.4125	897.4
0.4994	1.09359	2.4559	2.6858	0.4362	946.3
0.5676	1.11451	2.3557	2.6255	0.4710	1009.1
0.6111	1.12738	2.2854	2.5765	0.4826	1035.8
0.6515	1.14347	2.2087	2.5255	0.4879	1042.6
0.6993	1.16441	2.1231	2.4721	0.5011	1048.9
0.7517	1.18926	2.0251	2.4084	0.5104	1040.8
0.7973	1.21258	1.9461	2.3598	0.5253	1032.0
0.8487	1.24422	1.8358	2.2842	0.5214	982.1
0.8997	1.28031	1.7064	2.1847	0.4929	887.1
0.9499	1.31825	1.4811	1.9525	0.3307	610.7
1.0000	1.37382	1.1298	1.5521	—	—

where v, v_1, and v_2 represent the molar volumes of the mixtures and the pure liquids.

Table II gives the density (ρ), kinematic viscosity (ν) together with the dynamic viscosity (η), excess dynamic viscosity (η^E), and excess molar Gibbs free energy of activation of flow ($^*G^E$). The density values in Table II have been calculated from the experimental data in Table I.

Excess molar properties presenting asymmetrical composition dependence are difficult to fit. Different smoothing equations have been proposed for H^E and V^E of aqueous mixtures, mainly for alcohol–H$_2$O [17–19]. We have found that for η^E and V^E the best fits were obtained with the equation given by Ott et al. [17], which includes an exponential switching factor:

$$V^E \quad \text{or} \quad \eta^E = x_1 x_2 \Big[\exp(-\alpha x_1) \sum_{i=0}^{1} A_i (1-2x_1)^i + (1-\exp(-\alpha x_1)) \sum_{i=0}^{3} B_i (1-2x_1)^i\Big] \qquad (3)$$

Table III. Parameters of Eq. (3) at 303.15 K for Excess Volume and Excess Viscosity

System	α	A_0	A_1	B_0	B_1	B_2	B_3	s
TFE-H_2O								
V^E	21.3881	−27.1699	20.8985	−2.1885	−1.4213	−1.0442	−0.8360	0.0044
η^E	8.7687	5.0581	3.5131	1.3683	2.2468	1.6564	0.7486	0.0034
E181-H_2O								
V^E	17.8222	−70.9822	54.9607	−4.8272	−4.2349	−3.6788	−2.2086	0.0059
η^E	0.5281	10.4704	59.5910	−4.8194	−169.0970	−139.5087	−55.6582	0.0396
TFE-E181								
V^E	−0.03003	−0.2582	−0.4254	−13.9467	−28.2255	−51.5140	−171.7506	0.0072
η^E	1.6803	1.8342	0.3919	1.6815	−1.5378	−0.3208	−7.0138	0.0116

Table IV. Parameters of Eq. (4) at 303.15 K for Excess Activation Energy

System	a_0	a_1	a_2	a_3	a_4	a_5	a_6	s
TFE-H_2O	5336.1015	5873.5197	4685.5655	1856.3496	648.8080	4989.2036	4821.1299	5.54
E181-H_2O	14897.7920	15642.7099	18597.7892	2468.3156	−11156.310	37992.0410	47742.2358	44.82
TFE-E181	3792.2704	−2153.2876	1208.5562	515.4543	2114.3138	−4725.1446	3097.9337	3.77

The parameters A_i, B_i, and α calculated by unweighted least-squares using a Marquard algorithm [7] are shown in Table III, together with the standard deviations s.

In the case of $*G^E$ the following Redlich–Kister equation has yielded the best correlation:

$$*G^E = x_1 x_2 \sum_{i=1}^{n} a_i (x_2 - x_1)^{i-1} \tag{4}$$

where the coefficients a_i have been determined by a linear least-squares fitting procedure. The values of the parameters a_i and the standard deviations s are listed in Table IV. Figures 1, 2, and 3 show, respectively, the excess molar volumes, the excess viscosities, and the excess Gibbs energies of activation for the three studied mixtures together with the smoothing curves.

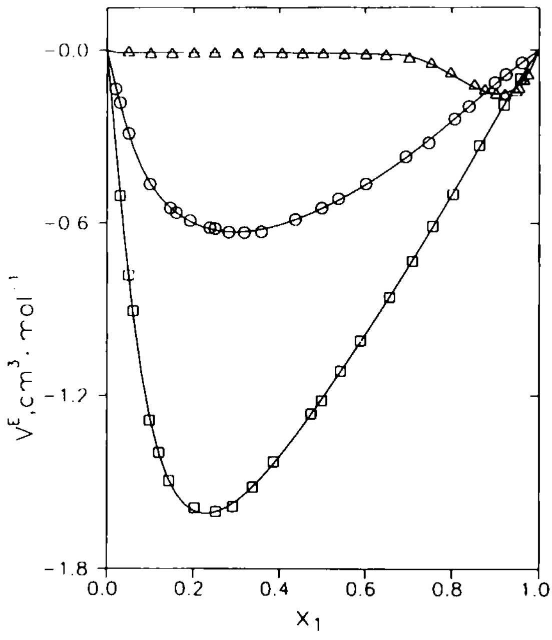

Fig. 1. Excess molar volumes at 303.15 K: (○) TFE–H_2O, (□) E181–H_2O, and (△) TFE–E181.

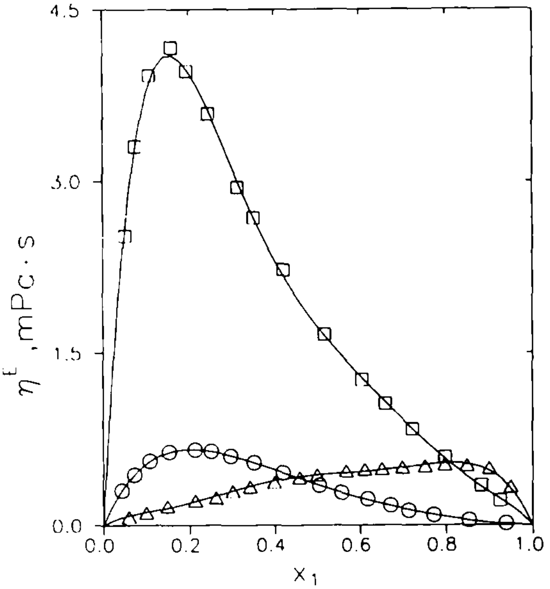

Fig. 2. Excess viscosities at 303.15 K: (○) TFE–H_2O, (□) E181–H_2O, and (△) TFE–E1812.

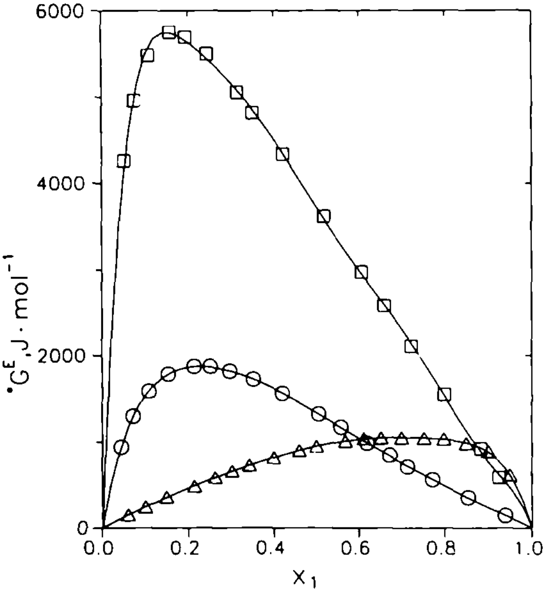

Fig. 3. Excess energies for activation at 303.15 K: (○) TFE–H_2O, (□) E181–H_2O, and (△) TFE–E181.

3.2. Viscosity Correlations as a Function of Concentration

The following equations were used to describe the composition dependence of the kinematic viscosity.

3.2.1. Correlation of McAllister—Two Parameters [11]

$$\begin{aligned}\ln[\gamma_m M_m] &= x_1^3 \ln[\gamma_1 M_1] + x_2^3 \ln[\gamma_2 M_2] \\ &\quad + 3x_1^2 x_2 \ln[\gamma_{12} M_{12}] + 3x_2^2 x_1 \ln[\gamma_{21} M_{21}] \\ M_m = x_1 M_1 + x_2 M_2; &\qquad M_{12} = \frac{2M_1 + M_2}{3}; \qquad M_{21} = \frac{2M_2 + M_1}{3}\end{aligned} \tag{5}$$

where γ_{12} and γ_{21} are the fitted parameters.

3.2.2. Correlation of McAllister—Three Parameters [11]

$$\begin{aligned}\ln[\gamma_m M_m] &= x_1^4 \ln[\gamma_1 M_1] + x_2^4 \ln[\gamma_2 M_2] + 4x_1^3 x_2 \ln[\gamma_{1112} M_{1112}] \\ &\quad + 6x_1^2 x_2^2 \ln[\gamma_{1122} M_{1122}] + 4x_1 x_2^3 \ln[\gamma_{2221} M_{2221}] \\ M_m &= x_1 M_1 + x_2 M_2; \qquad M_{1112} = \frac{3M_1 + M_2}{4} \\ M_{1122} &= \frac{M_1 + M_2}{2}; \qquad M_{2221} = \frac{M_1 + 3M_2}{4}\end{aligned} \tag{6}$$

In this case γ_{1112}, γ_{1122}, and γ_{2221} are the three adjustable parameters.

3.2.3. Correlation of Stephan and Heckenberger [12]

$$\ln \gamma_m = x_1 \ln \gamma_1 + x_2 \ln \gamma_2 + \ln\left[1 + \frac{x_1 x_2}{A + B x_1^2}\right] \tag{7}$$

where A and B are the fitted parameters.

Table V. Comparison of the Values of the Standard Deviation for Soliman, Stephan, and McAllister's Equations of Viscosities

	σ (10^{-6} m$^2 \cdot$ s^{-1})			
System	Soliman	Stephan	McAllister—2 p	McAllister—3 p
		303.15 K		
TFE-H_2O	0.034	0.019	0.10	0.048
E181-H_2O	0.300	0.025	1.42	0.830
TFE-E181	0.066	0.033	0.10	0.057

Table VI. Coefficients of the Stephan and Heckenberger Correlation

System	*A*	*B*	Conc. range
TFE–H_2O	0.13591	3.89441	0.043–0.938
E181–H_2O	0.01488	0.68198	0.051–0.925
TFE–E181	0.95240	−0.87218	0.099–0.950

3.2.4. Correlation of Soliman and Marshall [13]

$$\ln \gamma_m = x_1^3 \ln \gamma_1 + x_2^3 \ln \gamma_2 + 3x_1x_2 \ln \gamma_{12} + \frac{B_{12}x_1x_2}{(M_1/M_2)^2 x_1 + x_2}$$

where M_1 and M_2 are the molecular weight of the compounds, and γ_{12} and B_{12} the adjustable parameters.

For the four correlations the parameters were determined from experimental data using a nonlinear least-squares fit. Table V shows the standard deviations for the studied systems. In all cases the best correlations were found with the Stephan and Heckenberger equation and the coefficients are given in Table VI. Neither McAllister equation represents our results well. Similar fitting deviations were obtained by Kaiser et al. [9] for TFE–water mixtures.

4. DISCUSSION

Excess molar volumes can be explained in terms of positive contributions due to breaking of like interactions of the pure liquids and negative contributions due to the formation of unlike interactions and to the packing effect. In aqueous mixtures the solute enhances structure and close packing in the disordered regions, which results in a volume contraction [21]. Positive excess viscosities and excess Gibbs energies of activation for viscous flow indicate that there are interaction between unlike molecules.

For the three mixtures, excess volumes were negative, whereas excess viscosities and energies of activation were positive, presenting the three thermodynamic properties' asymmetric composition dependence.

The negative values of TFE–H_2O are due mainly to packing effects, because for this mixture excess molar enthalpies are positive for $x_1 < 0.8$. Therefore, there are no strong interactions between the fluoroalkanol and water since the η^E and $*G^E$ values are positive. This is in agreement with the analysis of excess enthalpies and excess Gibbs energies of various authors [4, 5, 8].

The mixture E181–H_2O presents large negative V^E values and large positive η^E and $*G^E$ values. This indicates very strong interactions between ether and water. This is due to the capability of the oxygens of the polyether to hydrogen-bond with the water molecules. McGee et al. [22] have reported dynamic viscosities of the same mixture at 298.15 K at various compositions. Since the temperature of the present data is higher, our results are lower.

Svejda et al. [3] have reported V^E values for TFE–E181 at 293.15 and 313.15 K. Our results at 303.15 K are consistent with theirs, presenting also the same strongly peaked curve with a minimum at $x_1 \approx 0.9$. This characteristic curve, together with the large exothermic heat of mixing and the positive η^E and $*G^E$ values, indicates complex formation between the fluoroalkanol and the polyether.

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