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Excess volumes, isentropic and isothermal compressibilities and isochoric heat capacities of the mixtures of 2,2,2-trifluoroethan-1-ol + benzene, benzene + dimethyl sulfoxide, and 2,2,2-trifluoroethan-1-ol + dimethyl sulfoxide at 298.15 K

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Abstract

Densities (ρ) and speeds of sound (u) in $x\mathrm{CF_3CH_2OH}+(1-x)\mathrm{C_6H_6}$, $x\mathrm{C_6H_6}+(1-x)\mathrm{(CH_3)_2SO}$ and $x\mathrm{CF_3CH_2OH}+(1-x)\mathrm{(CH_3)_2SO}$ were measured at 298.15 K. Excess volumes (V_m^E) and isentropic compressibilities (κ_S^E) were obtained from the above measurements, and excess isothermal compressibilities (κ_T^E) and isochoric heat capacities ($C_{V,\mathrm{m}}^\mathrm{E}$) were estimated using the thermodynamic relationships with isobaric heat capacities which have been reported in a previous paper. For the last-named mixture V_m^E is positive, and κ_S^E, κ_T^E and $C_{V,\mathrm{m}}^\mathrm{E}$ are negative. The first mixture shows a positive value for all the excess properties and is consistent with the positive H_m^E, except for $C_{V,\mathrm{m}}^\mathrm{E}$, which is anomalous in being similar to $C_{p,\mathrm{m}}^\mathrm{E}$. For the second mixture $C_{V,\mathrm{m}}^\mathrm{E}$ changes its sign from negative to positive with the mole fraction x, similarly to $C_{p,\mathrm{m}}^\mathrm{E}$, but the other excess properties are negative, and this mixture is anomalous if one considers that it consists of a polar component and a non-polar one, like the first mixture. The relationships between the excess thermodynamic functions are not simple and reflect the characteristics of the component molecules.

INTRODUCTION

We have previously reported H_m^E and $C_{p,\mathrm{m}}^\mathrm{E}$ for the various binary mixtures of 2,2,2-trifluoroethan-1-ol (TFE), dimethyl sulfoxide (DMSO) and benzene [1]. TFE and DMSO themselves have a "cluster structure" in the pure liquids [2,3], and the structures are retained even in the mixture [4]. The resultant "non-randomness" effect [5] is appreciable in the mixture.

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We will report here the effect that the structures in the pure liquids and the mixtures have on the excess volume, isothermal and isentropic compressibilities, and the isochoric heat capacity of the mixture.

EXPERIMENTAL

The materials were purified by the same procedure as previously [1]. The densities were measured at 298.15 K by a vibrational tube densimeter (Anton-Paar DM602) with the bath controlled to within ±0.0005 K; details of the preparation of the sample mixtures are given in our previous paper [6]. Our densimeter is found to have a non-linear term of the square of the period of resonance vibration τ^2, ascribed to the mechanical strain on the vibrating tube, besides the usual relationship (1):

$$\rho = a + b\tau^2 \tag{1}$$

We have therefore calibrated our densimeter by adding a term of the double square of τ:

$$\rho = a + b\tau^2 + c\tau^4 \tag{2}$$

and the coefficient c is estimated from fitting values for five pycnometrically defined samples (widely selected from $\rho = 0.7–1.4$) by means of the least-squares method. The coefficient c is regarded as an apparatus constant and the coefficients a and b are then defined by means of the usual routine procedures. In this work the value c is confirmed again by using the other six samples ($\tau = 1.1–1.58$). The accuracy of the density measurement is $\pm 10^{-5}$ g cm^{-3}. The speed of sound was measured by a sing-around technique (Chou-Onpa Kogyo, SVM-2), details of which are in our previous paper [7]. The accuracy was better than ± 0.3 m s^{-1}.

RESULTS AND DISCUSSION

The physical properties of the materials are listed in Table 1. The densities ρ and speeds of sound u observed in the mixtures consisting of $x\mathrm{CF_3CH_2OH} + (1-x)\mathrm{C_6H_6}$, $x\mathrm{C_6H_6} + (1-x)\mathrm{(CH_3)_2SO}$ and $x\mathrm{CF_3CH_2OH} + (1-x)\mathrm{(CH_3)_2SO}$ are given in Table 2. The isentropic compressibilities κ_S and isothermal ones κ_T are estimated from the following relations:

$$\kappa_S = (\rho u^2)^{-1} \tag{3}$$

$$\kappa_T = \kappa_S + \alpha^2 V_\mathrm{m} T / C_{p,\mathrm{m}} \tag{4}$$

The isochoric heat capacity $C_{V,\mathrm{m}}$ is also estimated from the above quantities through the relation

$$C_{V,\mathrm{m}} = C_{p,\mathrm{m}}(\kappa_S/\kappa_T) \tag{5}$$

TABLE 1

Physical properties of materials at 298.15 K

Property	2,2,2-Trifluoro-ethan-1-ol	Dimethyl sulfoxide	Benzene
ρ (g cm^{-3})	1.38280	1.09569	0.87365
u (m s^{-1})	833.1	1485.0	1299.8
κ_S (TPa^{-1})	1042.1	413.8	677.5
κ_T (TPa^{-1})	1444.4	556.8	974.2
$C_{p,\mathrm{m}}$ (J K^{-1} mol^{-1})	177.78	148.25	135.75
$C_{V,\mathrm{m}}$ (J K^{-1} mol^{-1})	128.26	110.19	94.42
α (K^{-1})	0.001821	0.000998	0.001229

where α, V_m, and $C_{p,\mathrm{m}}$ are the thermal expansion coefficient, molar volume and molar isobaric heat capacity of the liquid or mixture respectively. The induced properties κ_S, κ_T and $C_{V,\mathrm{m}}$ are also given in Table 2. The excess values are obtained by subtracting the following ideal values from the above observed ones for the mixtures

$$\begin{aligned} V_\mathrm{m}^\mathrm{id} &= x_1 V_{\mathrm{m},1}^0 + x_2 V_{\mathrm{m},2}^0 \\ C_{p,\mathrm{m}}^\mathrm{id} &= x_1 C_{p,\mathrm{m},1}^0 + x_2 C_{p,\mathrm{m},2}^0 \\ \kappa_T^\mathrm{id} &= \phi_1 \kappa_{T,1}^0 + \phi_2 \kappa_{T,2}^0 \\ \alpha^\mathrm{id} &= \phi_1 \alpha_1^0 + \phi_2 \alpha_2^0 = \alpha \end{aligned} \tag{6}$$

where

$$\phi_i = x_i V_{\mathrm{m},i}^0 / V_\mathrm{m}^\mathrm{id}$$

$$\kappa_S^\mathrm{id} = \kappa_T^\mathrm{id} - \alpha^2 V_\mathrm{m}^\mathrm{id} T / C_{p,\mathrm{m}}^\mathrm{id}$$

$$C_{V,\mathrm{m}}^\mathrm{id} = C_{p,\mathrm{m}}^\mathrm{id}\left(\kappa_S^\mathrm{id}/\kappa_T^\mathrm{id}\right)$$

where the superscript 0 means the value for the pure substance. The value of α of the mixture is assumed here to be equal to the ideal value. The excess values of the properties V_m^E, κ_S^E, κ_T^E, and $C_{V,\mathrm{m}}^\mathrm{E}$ are given with the corresponding mole fraction x in Table 3.

The observed points of excess volumes, V_m^E, excess isentropic compressibilities, κ_S^E, excess isothermal compressibilities, κ_T^E, and excess isochoric heat capacities, $C_{V,\mathrm{m}}^\mathrm{E}$, are plotted in Figs. 1, 2, 3 and 4 respectively, and the curves calculated by using the parameters in Table 4 are also drawn there. The parameters are fitted to the following Redlich–Kister or Myers–Scott equation by the least-squares method:

$$F^\mathrm{E}(\text{per unit}) = x(1-x)[1 - G(1-2x)]^{-1} \Sigma A_i (1-2x)^{i-1} \tag{7}$$

where, if $G = 0$, the equation is the Redlich–Kister type equation.

TABLE 2

Density, speed of sound and isentropic and isothermal compressibilities of the mixtures at 298.15 K

x	ρ (g cm^{-3})	u (m s^{-1})	κ_S (TPa^{-1})	κ_T (TPa^{-1})	$C_{V,\mathrm{m}}$ (J K^{-1} mol^{-1})
$x\mathit{CF_3CH_2OH} + (1-x)\mathit{C_6H_6}$					
0.00000	0.873631	1299.84	677.47	974.1	94.41
0.10298	0.912081	1228.28	726.72	1024.3	102.51
0.20895	0.954786	1159.71	778.74	1076.2	111.64
0.30250	0.994834	1106.17	821.50	1123.1	117.79
0.41592	1.046250	1047.56	870.98	1182.2	122.97
0.52003	1.096610	998.77	914.14	1237.8	125.90
0.62740	1.151562	952.56	957.03	1296.5	127.56
0.72497	1.205410	914.64	991.66	1347.2	128.22
0.83105	1.268958	878.54	1021.01	1395.0	128.35
0.93655	1.337967	848.28	1038.67	1430.8	128.27
1.00000	1.382822	833.08	1041.98	1444.3	128.26
0.00000	0.873637	1299.87	677.44	974.1	94.41
0.05135	0.892416	1264.84	700.43	999.3	97.61
0.15563	0.932908	1192.81	753.38	1050.3	107.32
0.26447	0.978325	1127.32	804.30	1103.8	115.51
0.36407	1.022331	1073.75	848.40	1154.6	120.88
0.47147	1.072643	1021.01	894.30	1211.8	124.72
0.56881	1.120994	977.28	934.02	1264.6	126.80
0.67296	1.176487	934.51	973.30	1320.0	127.94
0.76790	1.230894	899.63	1003.81	1366.6	128.30
0.87323	1.295833	865.71	1029.69	1411.0	128.33
0.98080	1.369062	837.46	1041.47	1440.8	128.25
1.00000	1.382819	833.07	1042.01	1444.3	128.26
$x\mathit{C_6H_6} + (1-x)\mathit{(CH_3)_2SO}$					
0.00000	1.095609	1484.93	413.94	556.9	110.19
0.06534	1.078420	1470.49	428.83	580.8	108.56
0.17079	1.051751	1447.89	453.54	619.9	106.48
0.27768	1.026028	1426.98	478.64	659.9	104.66
0.37510	1.003581	1409.01	501.90	697.1	103.10
0.48254	0.979680	1389.74	528.50	739.5	101.52
0.58458	0.957803	1372.39	554.33	780.6	100.17
0.68922	0.936069	1354.93	581.91	824.1	98.98
0.79285	0.915047	1337.96	610.48	868.9	97.86
0.89890	0.893884	1319.85	642.20	918.6	96.48
0.97578	0.878523	1305.23	668.14	959.5	95.01
1.00000	0.873662	1299.67	677.63	974.2	94.42
0.00000	1.095614	1484.96	413.91	556.9	110.19
0.02228	1.089708	1479.94	418.99	565.0	109.59
0.12916	1.062109	1456.56	443.79	604.5	107.25
0.23511	1.036135	1435.12	468.61	643.9	105.36
0.32778	1.014361	1417.44	490.68	679.1	103.85
0.43355	0.990454	1398.34	516.34	720.1	102.23
0.53114	0.969168	1381.38	540.72	758.9	100.86
0.63771	0.946674	1363.34	568.32	802.6	99.55
0.84766	0.904073	1328.91	626.33	893.8	97.20
0.94793	0.884098	1310.72	658.38	944.1	95.61
1.00000	0.873661	1299.65	677.65	974.3	94.42

TABLE 2 (continued)

x	ρ (g cm^{-3})	u (m s^{-1})	κ_S (TPa^{-1})	κ_T (TPa^{-1})	$C_{V,\mathrm{m}}$ (J K^{-1} mol^{-1})
$x CF_3CH_2OH + (1-x)(CH_3)_2SO$					
0.00000	1.095769	1485.19	413.73	556.7	110.18
0.10192	1.122151	1395.36	457.69	627.7	107.49
0.21042	1.150610	1307.56	508.33	709.5	105.19
0.31374	1.178151	1230.51	560.57	793.6	103.36
0.42167	1.207608	1156.64	618.98	887.4	101.90
0.50048	1.230391	1106.05	664.37	959.1	101.34
0.63376	1.269167	1030.90	741.39	1079.4	101.82
0.73053	1.298933	979.24	802.84	1167.9	104.38
0.83646	1.333078	925.01	876.71	1263.1	110.43
0.94759	1.366926	866.26	974.89	1372.1	122.04
1.00000	1.382771	833.07	1042.04	1444.4	128.26
0.00000	1.095778	1485.11	413.77	556.7	110.18
0.05386	1.108919	1439.39	435.25	592.4	108.57
0.15675	1.136475	1350.23	482.64	668.1	106.26
0.26463	1.165030	1266.01	535.54	753.1	104.20
0.36731	1.192681	1192.76	589.34	839.7	102.58
0.47613	1.222970	1121.81	649.74	936.4	101.43
0.56397	1.248358	1069.66	700.12	1016.0	101.22
0.77208	1.312417	957.91	830.39	1205.0	106.27
0.87668	1.345969	903.83	909.47	1301.1	113.96
0.98619	1.379078	841.19	1024.76	1423.9	127.35
1.00000	1.382773	833.01	1042.20	1444.5	128.26

The $x CF_3CH_2OH + (1-x)C_6H_6$ *system*

The H_m^E value of this mixture is very large and positive because of the breaking of clusters in pure TFE by mixing, as shown in Fig. 5; this has been reported by Cooney and Morcom [8]. The V_m^E value of the mixture is also large as a result of the large positive H_m^E value, as shown in Fig. 1. The V_m^E value of this system was also reported by Cooney and Morcom [8], although their values are slightly larger than ours (approximately 0.05 cm^3 mol^{-1} at $x = 0.5$). The κ_S^E and κ_T^E values in Figs. 2 and 3 are positive. The TFE-rich mixtures are more compressible than that with less TFE. Figure 4, however, shows a large positive $C_{V,\mathrm{m}}^\mathrm{E}$ value, consistent with the previous results for the thermal properties H_m^E and $C_{p,\mathrm{m}}^\mathrm{E}$ [1] in Figs. 5 and 6. The properties concerned with volume, i.e. V_m^E, κ_S^E and κ_T^E, are consistent with the previous conclusions that the cluster structure or hydrogen-bonding structure is left unbroken to some extent even in the mixture.

The $x C_6H_6 + (1-x)(CH_3)_2SO$ *system*

The H_m^E of the system in Fig. 5 is reasonably positive, owing to the breaking of the dipolar interaction in pure liquid DMSO by mixing with

TABLE 3

Excess volume, isentropic and isothermal compressibilities and excess isochoric heat capacity of mixtures at 298.15 K

x	V_m^E (cm^3 mol^{-1})	κ_S^E (TPa^{-1})	κ_T^E (TPa^{-1})	$C_{V,m}^E$ (J K^{-1} mol^{-1})
$xCF_3CH_2OH+(1-x)C_6H_6$				
0.05135	0.2567	7.56	5.5	1.463
0.10298	0.4639	18.02	10.3	4.607
0.15563	0.6332	28.27	15.2	7.642
0.20895	0.7650	36.63	19.3	10.158
0.26447	0.8731	44.18	23.7	12.147
0.30250	0.9367	48.78	26.8	13.142
0.36407	1.0171	54.93	31.7	14.145
0.41592	1.0631	59.62	36.2	14.481
0.47147	1.0954	63.37	40.6	14.351
0.52003	1.0930	65.73	44.0	13.884
0.56881	1.1039	67.71	47.7	13.133
0.62740	1.0745	68.73	51.2	11.914
0.67296	1.0111	67.51	52.2	10.747
0.72497	0.9507	65.50	53.0	9.267
0.76790	0.8333	60.48	50.2	7.901
0.83105	0.6886	51.86	45.1	5.815
0.87323	0.5476	42.85	38.2	4.362
0.93655	0.3025	24.70	22.8	2.162
0.98080	0.0918	8.02	7.5	0.641
$xC_6H_6+(1-x)(CH_3)_2SO$				
0.02228	−0.0170	−3.59	−3.4	−0.400
0.06534	−0.0468	−10.07	−9.7	−1.016
0.12916	−0.0898	−18.25	−17.8	−1.616
0.17079	−0.1197	−22.98	−22.6	−1.878
0.23511	−0.1649	−29.32	−29.1	−2.149
0.27768	−0.1935	−32.86	−32.7	−2.261
0.32778	−0.2249	−36.16	−36.1	−2.329
0.37510	−0.2550	−38.86	−39.0	−2.360
0.43355	−0.2818	−40.84	−41.2	−2.313
0.48254	−0.3020	−41.82	−42.4	−2.221
0.53114	−0.3174	−42.08	−42.9	−2.074
0.58458	−0.3291	−41.58	−42.7	−1.848
0.63771	−0.3327	−40.01	−41.5	−1.545
0.68922	−0.3309	−37.87	−39.8	−1.207
0.79285	−0.2910	−30.69	−33.3	−0.446
0.84766	−0.2477	−25.29	−28.1	−0.092
0.89890	−0.1910	−18.65	−21.3	0.146
0.94793	−0.1122	−10.85	−12.7	0.200
0.97578	−0.0559	−5.64	−6.6	0.124
$xCF_3CH_2OH+(1-x)(CH_3)_2SO$				
0.05386	0.1628	−15.38	−12.8	−3.002
0.10192	0.2069	−25.44	−20.6	−5.228
0.15675	0.3045	−37.31	−29.5	−7.687
0.21042	0.3882	−47.17	−36.2	−9.893
0.26463	0.4614	−55.63	−41.0	−11.969
0.31374	0.5205	−62.46	−44.3	−13.751
0.36731	0.5700	−68.28	−46.1	−15.534
0.42167	0.6058	−73.22	−46.8	−17.184

TABLE 3 (continued)

x	V_m^E (cm^3 mol^{-1})	κ_S^E (TPa^{-1})	κ_T^E (TPa^{-1})	$C_{V,m}^E$ (J K^{-1} mol^{-1})
xCF$_3$CH$_2$OH + (1−x)(CH$_3$)$_2$SO				
0.47613	0.6161	−76.96	−46.3	−18.608
0.50048	0.5871	−77.51	−45.1	−19.108
0.56397	0.5932	−81.40	−44.6	−20.289
0.63376	0.5363	−82.98	−42.9	−20.825
0.73053	0.4059	−80.27	−39.8	−19.802
0.77208	0.3118	−77.80	−39.5	−18.564
0.83646	0.1800	−69.68	−37.9	−15.384
0.87668	0.0998	−60.81	−35.3	−12.466
0.94759	0.0534	−36.75	−26.4	−5.440
0.98619	−0.0112	−9.45	−8.6	−0.713

benzene as reported in a previous paper [9]. The $C_{p,m}^E$ in Fig. 6 which is reported in our previous paper [1] is, however, unusual, and the curve changes sign from negative to positive depending on the mole fraction of benzene. However, V_m^E has a small negative value, and the negative κ_S^E and κ_T^E values are consistent with the result for V_m^E. This suggests that in the mixture a new good and hard-packed structure is formed from tetrahedral molecules of DMSO and flatter ellipsoidal molecules of benzene. It can be considered, however, that the clusters of DMSO in the pure state are left unbroken to some extent even in the mixture, and the smooth benzene

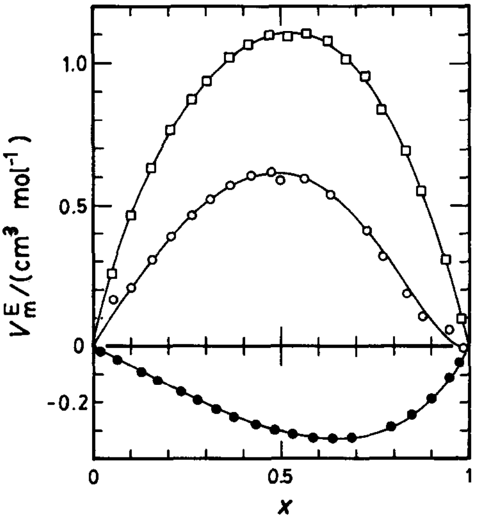

Fig. 1. Excess molar volumes of the mixtures at 298.15 K; □, xCF$_3$CH$_2$OH+(1−x)C$_6$H$_6$; ●, xC$_6$H$_6$+(1−x)(CH$_3$)$_2$SO; ○, xCF$_3$CH$_2$OH+(1−x)(CH$_3$)$_2$SO.

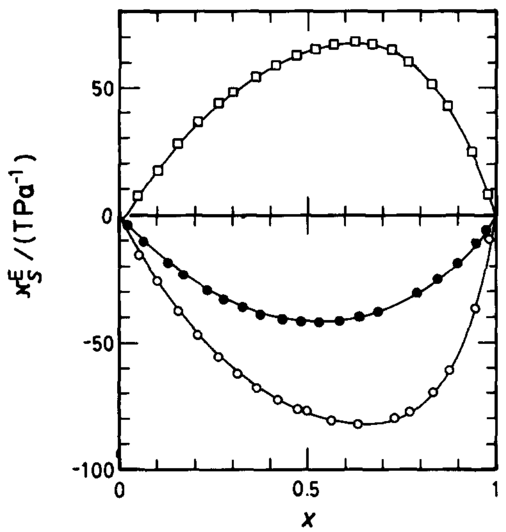

Fig. 2. Excess isentropic compressibilities of the mixtures at 298.15 K. Symbols as in Fig. 1.

molecules slip in to occupy the spaces among the clusters. This situation is analogous to the mixing of components with very different molecular size, and in such a case V_m^E is very often negative. The positive $C_{p,m}^E$ and $C_{V,m}^E$ values observed for the benzene-rich mixture are regarded as due to a non-random mixing effect [5] originating from clusters that remain unbroken in the mixture, in support of the latter explanation.

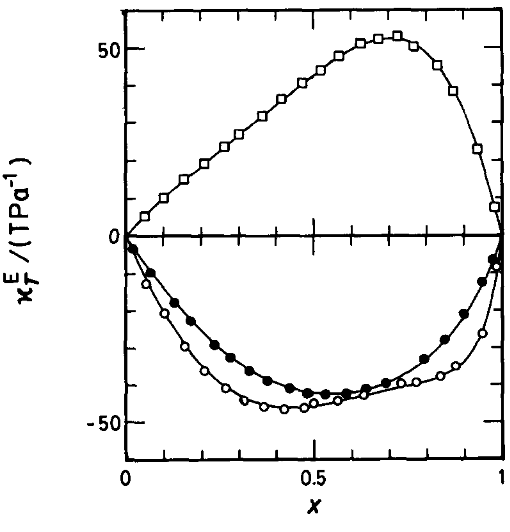

Fig. 3. Excess isothermal compressibilities of the mixtures at 298.15 K. Symbols as in Fig. 1.

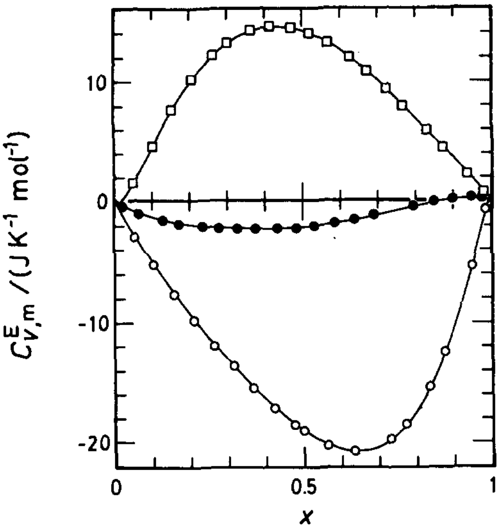

Fig. 4. Excess isochoric heat capacities of the mixtures at 298.15 K. Symbols as in Fig. 1.

The $xCF_3CH_2OH + (1 - x)(CH_3)_2SO$ *system*

It is anomalous that V^E_m is large and positive, in contrast to the huge negative H^E_m value in Fig. 5 which was reported previously [1]. Another anomalous point is that κ^E_S and κ^E_T are negative in spite of a positive value of V^E_m. Such an observation is rare in the cases of non-polar mixtures. These anomalous relationships seem to be characteristic of a case in which strong dipolar interactions and a highly directional interaction such as hydrogen bonding act simultaneously between the component molecules. Some of the DMSO and TFE molecules clustered in the pure state by dipolar interaction and intermolecular and intramolecular hydrogen-bonding will be liberated from the clusters by mixing, and newly establish intermolecular hydrogen-bonding between them. The first step affects V^E_m in the same way as in a non-polar solvent, and the second step, more reasonably, has a positive affect on V^E_m because of the poor packing that results when the bulky tetrahedral DMSO molecule replaces the linear TFE molecule in the hydrogen-bonded structure [4].

In spite of the positive V^E_m value, the mixture is less compressible than the pure components and κ^E_S and κ^E_T are negative, since the new structure of hydrogen bonding between the TFE and DMSO molecules is more stable in energy and more rigid so that the space produced by new hydrogen bonding cannot be compressed.

As seen in Fig. 4, $C^E_{V,m}$ is large and negative as is the $C^E_{p,m}$ [1] in Fig. 6. All these three mixtures show the existence of non-random effects due to the clustering of dipolar molecules or to directional hydrogen-bonding

TABLE 4

Parameters of smoothed curves represented by eqn. (7)

Property	G	A_1	A_2	A_3	A_4	A_5	s
$xCF_3CH_2OH+(1-x)C_6H_6$							
V_m^E (g cm^{-3})	0.55874	4.4167	−2.8013	0.9614	–	–	0.0077
κ_S^E (TPa^{-1})	0.97147	260.49	−357.64	184.12	−94.31	–	0.29
κ_T^E (TPa^{-1})	–	171.6	−155.1	103.9	–	–	0.3
$C_{V,m}^E$ (J K^{-1} mol^{-1})	0.90896	56.469	−31.129	−26.268	−2.575	–	0.006
$xC_6H_6+(1-x)(CH_3)_2SO$							
V_m^E (g cm^{-3})	−0.43233	−1.2335	0.1456	0.0418	0.0911	–	0.0012
κ_S^E (TPa^{-1})	−0.75376	−167.99	−116.68	−5.03	–	–	0.09
κ_T^E (TPa^{-1})	−0.58734	−170.7	−76.2	−12.2	14.3	–	0.07
$C_{V,m}^E$ (J K^{-1} mol^{-1})	–	−8.709	−5.758	3.636	−7.509	−1.150	0.004
$xCF_3CH_2OH+(1-x)(CH_3)_2SO$							
V_m^E (g cm^{-3})	−0.60332	2.4327	1.8041	−0.8115	–	–	0.0196
κ_S^E (TPa^{-1})	−0.69159	−313.53	−106.88	−67.69	–	–	0.73
κ_T^E (TPa^{-1})	−0.78704	−183.7	−181.5	−115.7	52.8	–	0.5
$C_{V,m}^E$ (J K^{-1} mol^{-1})	−0.98622	−76.598	−32.306	20.089	−26.148	–	0.030

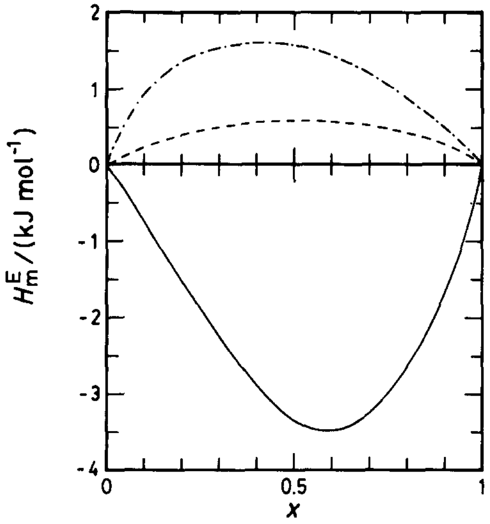

Fig. 5. Excess enthalpies of the mixtures at 298.15 K: ·–·–·, $x\text{CF}_3\text{CH}_2\text{OH}+(1-x)\text{C}_6\text{H}_6$ [8]; – – –, $x\text{C}_6\text{H}_6+(1-x)(\text{CH}_3)_2\text{SO}$ [9]; ——, $x\text{CF}_3\text{CH}_2\text{OH}+(1-x)(\text{CH}_3)_2\text{SO}$ [1].

interaction. The relationships between the excess functions are not simple and reflect the characteristics of the component molecules.

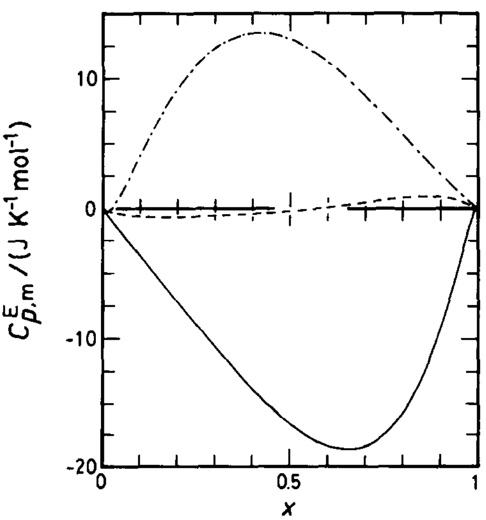

Fig. 6. Excess isobaric heat capacities of the mixtures at 298.15 K [1]: ·–·–·, $x\text{CF}_3\text{CH}_2\text{OH}+(1-x)\text{C}_6\text{H}_6$; – – –, $x\text{C}_6\text{H}_6+(1-x)(\text{CH}_3)_2\text{SO}$; —— $x\text{CF}_3\text{CH}_2\text{OH}+(1-x)(\text{CH}_3)_2\text{SO}$.

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