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THERMODYNAMIC PROPERTIES OF THE TERNARY SYSTEM MTBE+1-PROPANOL+HEXANE

Application of different group contribution models and empirical methods

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Experimental excess molar enthalpies and excess molar volumes of the ternary system x_1MTBE+$x_2$1-propanol+(1–x_1–x_2) hexane and the involved binary mixtures have been determined at 298.15 K and atmospheric pressure. Excess molar enthalpies were measured using a standard Calvet microcalorimeter, and excess molar volumes were determined from the densities of the pure liquids and mixtures, using a DMA 4500 Anton Paar densimeter. The UNIFAC group contribution model (in the versions of Larsen *et al.*, and Gmehling *et al.*) has been employed to estimate excess enthalpies values. Several empirical expressions for estimating ternary properties from experimental binary results were applied.

Keywords: *empirical expressions, excess molar enthalpies, excess molar volumes, hexane, 1-propanol, ternary mixture, tert-butyl methyl ether (MTBE), UNIFAC group contribution model*

Introduction

Forming part of the scientific project entitled 'Study on physical properties of mixtures hydrocarbon+alcohol +ether like alternative fuels', the present article reports experimental excess molar enthalpies and excess molar volumes for the ternary system x_1MTBE+$x_2$1-propanol +$x_3$hexane and the involved binary mixtures at 298.15 K and atmospheric pressure. The excess molar enthalpies for the binary mixture xMTBE+(1–x)1-propanol and the excess molar volumes for the binary mixture xMTBE+(1–x)hexane were reported in earlier works [1, 2].

Excess molar enthalpies were measured using a standard Calvet microcalorimeter, and excess molar volumes were determined from the densities of the pure liquids and mixtures, using a DMA 4500 Anton Paar densimeter. Attending to the symmetry of the studied mixtures, suitable fitting equations have been used in order to correlate adequately the experimental data.

The UNIFAC group contribution model (in the versions of Larsen *et al.* [3], and Gmehling *et al.* [4]) has been employed to estimate excess molar enthalpies. Several empirical expressions were also applied to estimate ternary properties from binary results.

Experimental

The chemical substances employed, MTBE, 1-propanol, hexane were degassed by ultrasound and dried over molecular sieves (Sigma, type 0.4 nm). The measured densities of the pure liquids displayed good agreement with previously published values [5–7], as shown in Table 1. The handling and disposal of the chemicals used has been done according to the recommendation of the CRC Handbook of Chemistry and Physics [5].

Table 1 Source, purity and densities of chemicals used

Substances	Source	Purity/%	Density/g mL^{-1} exp.	Density/g mL^{-1} references
MTBE	Aldrich	>99.8	0.7356	0.73528[a] 0.7359[b]
1-propanol	Aldrich	>99.5	0.7997	0.79975[a] 0.7995[c]
hexane	Fluka	≥99.5	0.6550	0.6606[a] 0.6551[c]

[a][5], [b][6], [c][7].

The experimental excess molar enthalpies were measured using a Calvet microcalorimeter equipped with a device allowing operation in the absence of vapour phase, and having a calorimeter-cell volume of approximately 10 mL. A Philips PM2535 voltmeter and a data acquisition system were linked to the microcalorimeter. Calibration was performed electrically using a Setaram EJP30 stabilised current source. Further details about the experimental method of operation have been published [8, 9].

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Excess molar volumes were determined from the densities of the pure liquids and mixtures. The measurements of densities were carried out with an DMA 4500 Anton Paar densimeter. The temperature inside the vibrating-tube cell was controlled using digital thermometer and was regulated to better than ±0.01 K. The experimental technique has been described previously [10, 11]. The uncertainty in density measurements was $\pm 5\cdot 10^{-5}$ g mL^{-1}. Before each series of measurements, the apparatus was calibrated at atmospheric pressure using double-distilled and degassed water and heptane (Fluka >0.995); density data were taken from literature: [12] for heptane and [13] for water. The mixtures were prepared by mass using a Mettler AT201 (precision $\pm 1\cdot 10^{-5}$ g) balance, ensuring an accuracy in the mole fraction less than 10^{-4}. All molar quantities are based on the IUPAC relative atomic mass table [14].

Several experimental series of measurements were carried out for the ternary compositions resulting from adding hexane to a binary mixture composed of x_1'MTBE+x_2'1-propanol, where $x_2'=1-x_1'$. The ternary composition point is then a pseudobinary mixture composed of hexane and the mentioned binary mixture. Thus, the ternary excess molar enthalpy can be expressed as

$$H^{\mathrm{E}}_{\mathrm{m},123}=H^{\mathrm{E}}_{\mathrm{m},\phi}+(x_1+x_2)H^{\mathrm{E}}_{\mathrm{m},12} \qquad (1)$$

where $H^{\mathrm{E}}_{\mathrm{m},\phi}$ is the measured excess molar enthalpy for the pseudobinary mixture and $H^{\mathrm{E}}_{\mathrm{m},12}$ is the excess molar enthalpy of the initial binary x_1'MTBE+x_2'1-propanol. Values of $H^{\mathrm{E}}_{\mathrm{m},12}$ at three mole fractions were interpolated by using a spline-fit method. Equation (1) does not involve any approximation.

The excess molar volumes were calculated from the densities of the pure liquids and their mixtures using following equation

$$V^{\mathrm{E}}_{\mathrm{m},123}=\sum_{\mathrm{i}=1}^{\mathrm{n}} x_{\mathrm{i}}\, M_{\mathrm{i}}\,(\rho^{-1}-\rho_{\mathrm{i}}^{-1}) \qquad (2)$$

with x, M and ρ being mole fraction, molar mass, and density, respectively; n is the number of the components in the mixture and the subscript i indicates values for the pure components.

Experimental values for the binary mixtures measured in this work are listed in Table 2. The excess molar enthalpies for the binary mixture xMTBE +(1–x)1-propanol were reported in an earlier paper [1] and the excess molar volumes for xMTBE +(1–x)hexane were taken from [2].

Experimental values of ternary excess molar enthalpies, $H^{\mathrm{E}}_{\mathrm{m},123}$ and ternary excess molar volumes, $V^{\mathrm{E}}_{\mathrm{m},123}$, for the ternary system x_1MTBE+$x_2$1-propanol +(1–x_1–x_2)hexane are listed in Table 3.

Data correlation

The experimental data corresponding to the binary mixtures xMTBE+(1–x)1-propanol, xMTBE+(1–x)hexane were fitted to the variable-degree polynomials suggested by Redlich–Kister [15],

Table 2 Experimental binary excess molar enthalpies, $H^{\mathrm{E}}_{\mathrm{m}}$, and excess molar volumes, $V^{\mathrm{E}}_{\mathrm{m}}$, and densities, ρ, at 298.15 K

x	$H^{\mathrm{E}}_{\mathrm{m}}$ /J mol^{-1}	x	$H^{\mathrm{E}}_{\mathrm{m}}$ /J mol^{-1}	x	$H^{\mathrm{E}}_{\mathrm{m}}$ /J mol^{-1}	x	$H^{\mathrm{E}}_{\mathrm{m}}$ /J mol^{-1}
			xMTBE+(1–x)hexane				
0.0527	61.0	0.2668	304.9	0.5142	380.3	0.8361	209.0
0.1125	146.2	0.3262	341.5	0.5827	372.9	0.9066	127.2
0.1191	151.1	0.3985	368.7	0.6520	340.5	0.9551	66.5
0.1837	224.2	0.4549	383.4	0.7215	305.9		
0.2577	297.8	0.4648	380.6	0.7818	256.6		
			x1-propanol+(1–x)hexane				
0.0987	461.6	0.4342	573.2	0.6835	425.3	0.8841	181.5
0.1984	579.3	0.5034	542.3	0.7510	351.0	0.8913	173.3
0.2684	598.5	0.5702	504.9	0.7967	299.5	0.9326	114.2
0.3635	594.3	0.6427	462.2	0.8418	236.7	0.9659	59.9

x	ρ/g mL^{-1}	$V^{\mathrm{E}}_{\mathrm{m}}$ /mL mol^{-1}	x	ρ/g mL^{-1}	$V^{\mathrm{E}}_{\mathrm{m}}$ /mL mol^{-1}	x	ρ/g mL^{-1}	$V^{\mathrm{E}}_{\mathrm{m}}$ /mL mol^{-1}
				xMTBE+(1–x)1-propanol				
0.0543	0.79576	–0.1433	0.3987	0.77199	–0.6318	0.7856	0.74816	–0.4701
0.1065	0.79198	–0.2560	0.4914	0.76591	–0.6454	0.8957	0.74174	–0.2754
0.1912	0.78604	–0.4156	0.5967	0.75957	–0.6409	0.9446	0.73888	–0.1593
0.2932	0.77896	–0.5425	0.6916	0.75375	–0.5831			
				x1-propanol+(1–x)hexane				
0.0487	0.65847	0.1247	0.4025	0.69380	0.2069	0.7983	0.75489	0.0455
0.0972	0.66243	0.1807	0.4988	0.70625	0.1669	0.9004	0.77602	0.0175
0.1953	0.67135	0.2267	0.6032	0.72136	0.1182	0.9497	0.78734	0.0052
0.2901	0.68100	0.2326	0.6997	0.73697	0.0798			

Table 3 Excess molar enthalpies[a], $H^E_{m,123}$, densities, ρ, and excess molar volumes, $V^E_{m,123}$, at 298.15 K for the ternary mixture x_1MTBE+$x_2$1-propanol+(1–x_1–x_2)hexane

x_1	x_2	$H^E_{m,\phi}$ /J mol^{-1}	$H^E_{m,123}$ /J mol^{-1}	x_1	x_2	$H^E_{m,\phi}$ /J mol^{-1}	$H^E_{m,123}$ /J mol^{-1}
			x_1'=0.2504, $H^E_{m,12}$ =359.7 J mol^{-1}				
0.0441	0.1320	482.8	546.2	0.1673	0.5007	446.0	686.2
0.0679	0.2032	535.6	633.1	0.1814	0.5430	406.5	667.1
0.0833	0.2493	557.7	677.3	0.1948	0.5832	366.4	646.3
0.1026	0.3070	551.5	698.9	0.2067	0.6185	298.7	595.5
0.1193	0.3572	543.6	715.0	0.2192	0.6561	238.4	553.3
0.1340	0.4011	517.0	709.5	0.2294	0.6865	165.8	495.2
0.1528	0.4574	476.7	696.2	0.2400	0.7183	81.6	426.3
			x_1'=0.5017, $H^E_{m,12}$ =557.0 J mol^{-1}				
0.0423	0.0420	247.2	294.1	0.2663	0.2646	544.8	840.5
0.0801	0.0796	382.2	471.2	0.3189	0.3168	492.7	855.4
0.0991	0.0984	412.7	522.7	0.3489	0.3466	448.4	835.8
0.1161	0.1154	462.2	591.2	0.3779	0.3754	396.5	816.1
0.1642	0.1631	528.3	710.6	0.4049	0.4022	323.4	789.1
0.1885	0.1872	546.4	755.7	0.4354	0.4325	237.3	720.7
0.2275	0.2260	552.6	805.2	0.4559	0.4529	169.9	676.1
0.2529	0.2513	546.3	827.1	0.4815	0.4783	83.6	618.2
			x_1'=0.7553, $H^E_{m,12}$ =505.7 J mol^{-1}				
0.0546	0.0177	156.9	193.5	0.4141	0.1342	512.9	790.1
0.1156	0.0374	277.2	354.6	0.4596	0.1489	494.3	802.0
0.1287	0.0417	302.1	388.3	0.5132	0.1663	453.8	797.4
0.1584	0.0513	353.0	459.0	0.5540	0.1795	393.4	764.3
0.1832	0.0594	383.0	505.7	0.5993	0.1942	323.0	724.3
0.2566	0.0832	467.9	639.7	0.6385	0.2069	257.6	685.0
0.3116	0.1010	502.1	710.8	0.6802	0.2204	172.5	627.9
0.3651	0.1183	528.8	773.3	0.7234	0.2344	72.9	557.2

x_1	x_2	ρ/g mL^{-1}	$V^E_{m,123}$/mL mol^{-1}	x_1	x_2	ρ/g mL^{-1}	$V^E_{m,123}$/mL mol^{-1}
0.0230	0.0504	0.6603	0.1428	0.2913	0.3007	0.7097	–0.0036
0.0427	0.9080	0.7848	–0.1004	0.2950	0.4014	0.7243	–0.1238
0.0875	0.1060	0.6699	0.2118	0.2964	0.5085	0.7414	–0.2716
0.0832	0.1976	0.6787	0.1881	0.3046	0.6051	0.7599	–0.4212
0.0935	0.2985	0.6906	0.1539	0.3927	0.1036	0.6944	0.2063
0.0874	0.4225	0.7054	0.0913	0.3941	0.2006	0.7065	0.0448
0.1174	0.4987	0.7192	0.0022	0.4088	0.2897	0.7199	–0.0888
0.1164	0.5916	0.7336	–0.0741	0.4005	0.4046	0.7363	–0.2625
0.0993	0.6996	0.7503	–0.1361	0.4003	0.5035	0.7533	–0.4498
0.1060	0.8008	0.7712	–0.2092	0.4931	0.1018	0.7029	0.1740
0.1968	0.1996	0.6886	0.1477	0.5105	0.2912	0.7306	–0.2167
0.1998	0.2957	0.7003	0.0736	0.5016	0.4028	0.7478	–0.4334
0.1912	0.4060	0.7139	–0.0079	0.5980	0.1955	0.7253	–0.1107
0.2004	0.5004	0.7289	–0.1190	0.6041	0.2970	0.7418	–0.3793
0.1979	0.5986	0.7449	–0.2176	0.6987	0.1026	0.7217	0.0253
0.2014	0.6995	0.7643	–0.3345	0.7956	0.1046	0.7315	–0.1060
0.2957	0.0989	0.6858	0.2346	0.9025	0.0508	0.7339	–0.0632
0.3079	0.1915	0.6975	0.1088				

[a] Three experimental series of measurements were carried out for the ternary compositions resulting from adding hexane to a binary mixture composed of x_1'MTBE+x_2'1-propanol, where x_2'=1–x_1'.

$$Q^E_m = x(1-x)\sum_{i=1}^{n} A_i\,(2x-1)^{i-1} \qquad (3)$$

where Q^E_m is H^E_m (J mol^{-1}) or V^E_m (mL mol^{-1}), while lower deviations for the mixture x1-propanol +(1–x)hexane were obtained by fitting experimental data of excess molar enthalpies to the equation introduced by Myers and Scott [16],

$$H^E_m\ (\text{J mol}^{-1}) = \frac{x(1-x)}{1+k(2x-1)}\sum_{i=1}^{n} A_i\,(2x-1)^{i-1} \qquad (4)$$

and excess molar volumes to the equation suggested by Brandreth *et al.* [17].

$$V^E_m\ (\text{mL mol}^{-1}) = x(1-x)\sum_{i=1}^{n} A_i\,(1-x)^{\frac{i-1}{2}} \qquad (5)$$

The measured values of ternary excess properties $Q^{E}_{m,123}$, were correlated using an equation of the form:

$$Q^{E}_{m,123} = Q^{E}_{m,bin} + x_i x_j (1 - x_i - x_j)\Delta_{123} \qquad (6)$$

where Δ_{123} was correlated using Morris *et al.* equation [18] for excess molar enthalpies.

$$\frac{\Delta_{123}}{RT} = (B_0 + B_1x_1 + B_2x_2 + B_3x_1^2 + B_4x_2^2 + B_5x_1x_2 + B_6x_1^3 + B_7x_2^3 + \cdots) \qquad (7)$$

and the Nagata and Tamura equation [19] has been used in order to correlate adequately the experimental excess molar volumes:

$$\Delta_{123} = (B_0 + B_1x_1 + B_2x_2 + B_3x_1^2 + B_4x_2^2) \qquad (8)$$

The parameters A_i and B_i were calculated using the unweighed least-squares method, with the degree of the polynomial previously optimized through the application of the *F*-test [20]. The uncertainty of the excess molar properties is estimated to be less than 1%. Figure 1 shows the experimental data of H^{E}_{m} and V^{E}_{m} obtained for the three involved binary systems. Isolines of $H^{E}_{m,123}$ and $V^{E}_{m,123}$ are plotted in Figs 2a and b, respectively. Figure 3 represents lines of constant ternary contribution, $x_1x_2x_3\Delta_{123}$, to the excess molar enthalpy and excess molar volume. Table 4 presents the

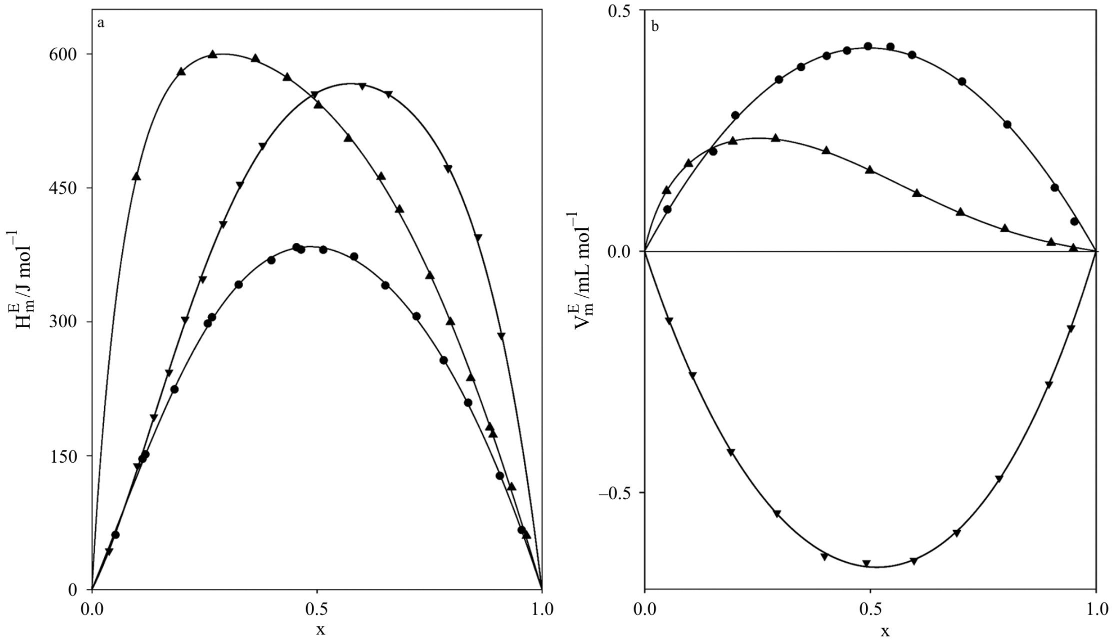

Fig. 1 Excess molar properties, a – H^{E}_{m}, b – V^{E}_{m}, of the binary mixtures: ● – *x*MTBE+(1–*x*)hexane[a], ▼ – *x*MTBE+(1–*x*)1-propanol[b], ▲ – *x*1-propanol+(1–*x*)hexane, [a]experimental data of V^{E}_{m} were taken from [2], [b]experimental data of H^{E}_{m} were taken from [1]

Table 4 Fitting parameters, A_i, B_i and standard deviations, *s*, for all mixtures studied in this work

	A_1	A_2	A_3	A_4	A_5	*k*	*s*
			*x*MTBE+(1–*x*)1-propanol				
[a]H^{E}_{m} /J mol^{-1}	2226.2	533.9	361.3	1073.5	–	–	3.6
V^{E}_{m} /mL mol^{-1}	–2.6182	–0.1305	–0.3518	–	–	–	0.0046
			*x*MTBE+(1–*x*)hexane				
H^{E}_{m} /J mol^{-1}	1534.2	–96.5	–	257.3	–166.3	–	2.8
[b]V^{E}_{m} /mL mol^{-1}	1.6830	–0.0313	–	–	–	–	0.0069
			*x*1-propanol+(1–*x*)hexane				
H^{E}_{m} /J mol^{-1}	2187.1	654.2	453.5	–378.9	–	0.7295	3.6
V^{E}_{m} /mL mol^{-1}	5.4974	–18.4837	32.3331	–30.1311	10.9492	–	0.0017
	B_0	B_1	B_2	B_3	B_4		*s*
			x_1MTBE+$x_2$1-propanol+$x_3$hexane				
H^{E}_{m} /J mol^{-1}	3.6617	5.7921	–12.0485	–7.3426	11.4712	–	5.7
$V^{E}_{m,123}$ /mL mol^{-1}	–10.2491	3.8180	25.2953	3.4983	–20.5942		0.0045

values of the A_i and B_i parameters and their corresponding standard deviations.

Theoretical predictions

The UNIFAC group contribution model (in the versions of Larsen *et al.* [3], and Gmehling *et al.* [4]) has been employed to estimate excess molar enthalpies. Best accuracy for the ternary mixture was yielded by Gmehling version, with a 12% deviation from experimental data, *vs*. a 15% for Larsen version. Table 5 list the average percentage deviations of the excess molar enthalpies predicted by theoretical methods and the experimental values. These results are displayed graphically in Fig. 4.

The empirical expressions of Kohler [21], Jacob and Fitzner [22], Colinet [23], and Knobeloch and Schwartz [24], Tsao and Smith [25], Toop [26], Scatchard *et al.* [27], Hillert [28], and Mathieson and Thynne [29] were applied to estimate ternary properties from binary results. These methods can be divided into symmetric and asymmetric, depending on whether the assumption of the three binaries contributing equally to the ternary mixture magnitude is accepted or not. For the asymmetric equations [25–29],

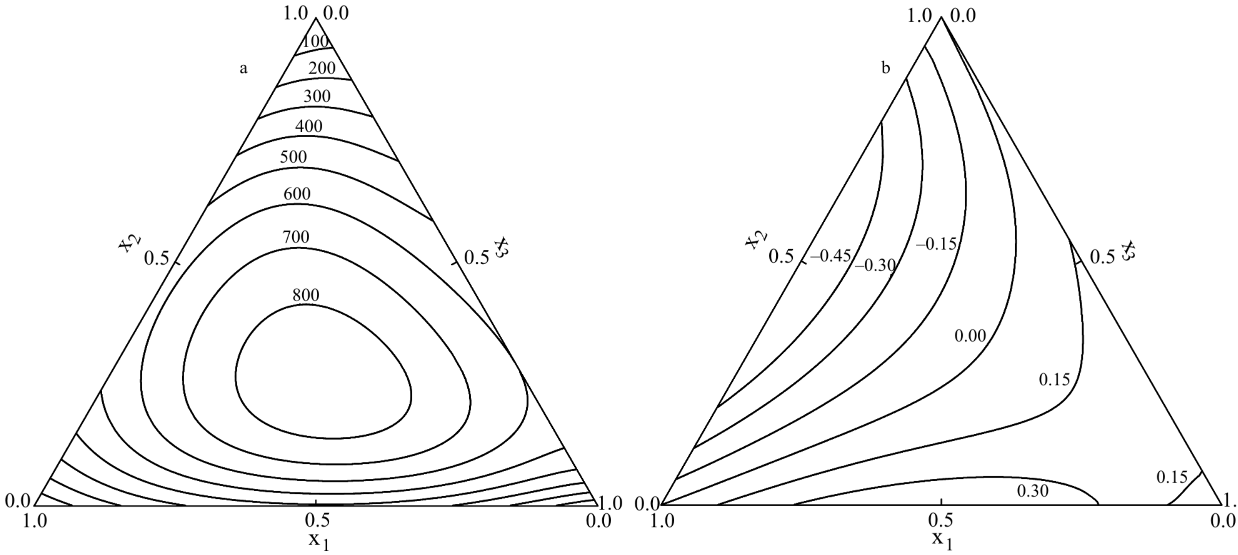

Fig. 2 Isolines for the ternary system x_1MTBE+$x_2$1-propanol+$x_3$hexane at 298.15 K; a – $H^E_{m,123}$ (J mol^{-1}) calculated with Eq. (7), b – $V^E_{m,123}$ (mL mol^{-1}), calculated with Eq. (8)

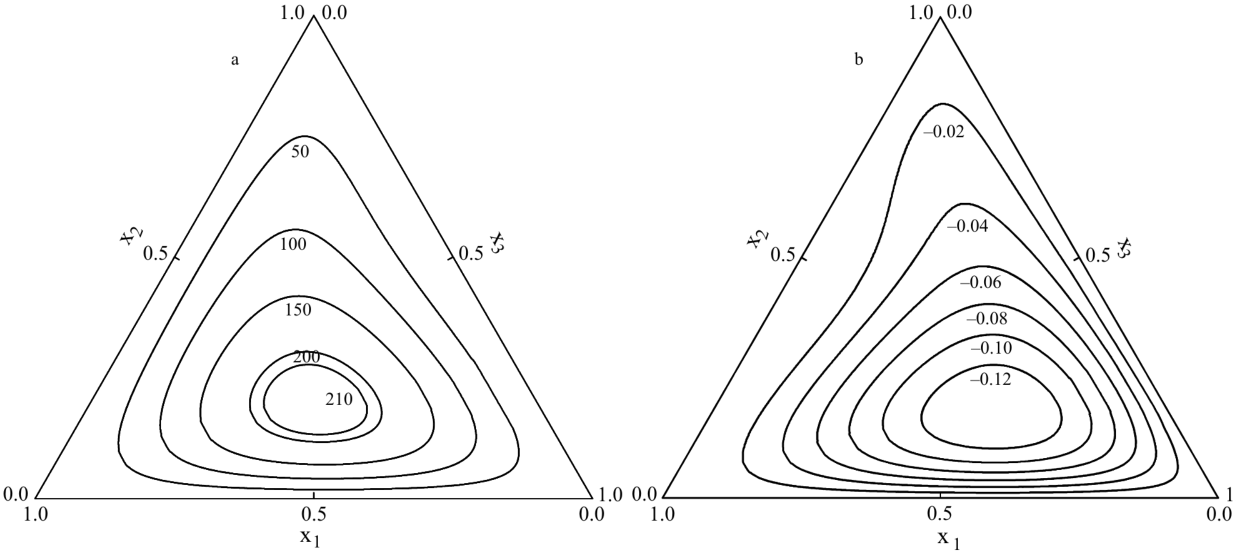

Fig. 3 Curves of constant ternary contribution, $x_1x_2x_3\Delta_{123}$; a – to the excess molar enthalpy $H^E_{m,123}$ (J mol^{-1}), b – to the excess molar volume $V^E_{m,123}$ (mL mol^{-1}), for x_1MTBE+$x_2$1-propanol+$x_3$hexane at 298.15 K, correlated using Eqs (7) and (8), respectively

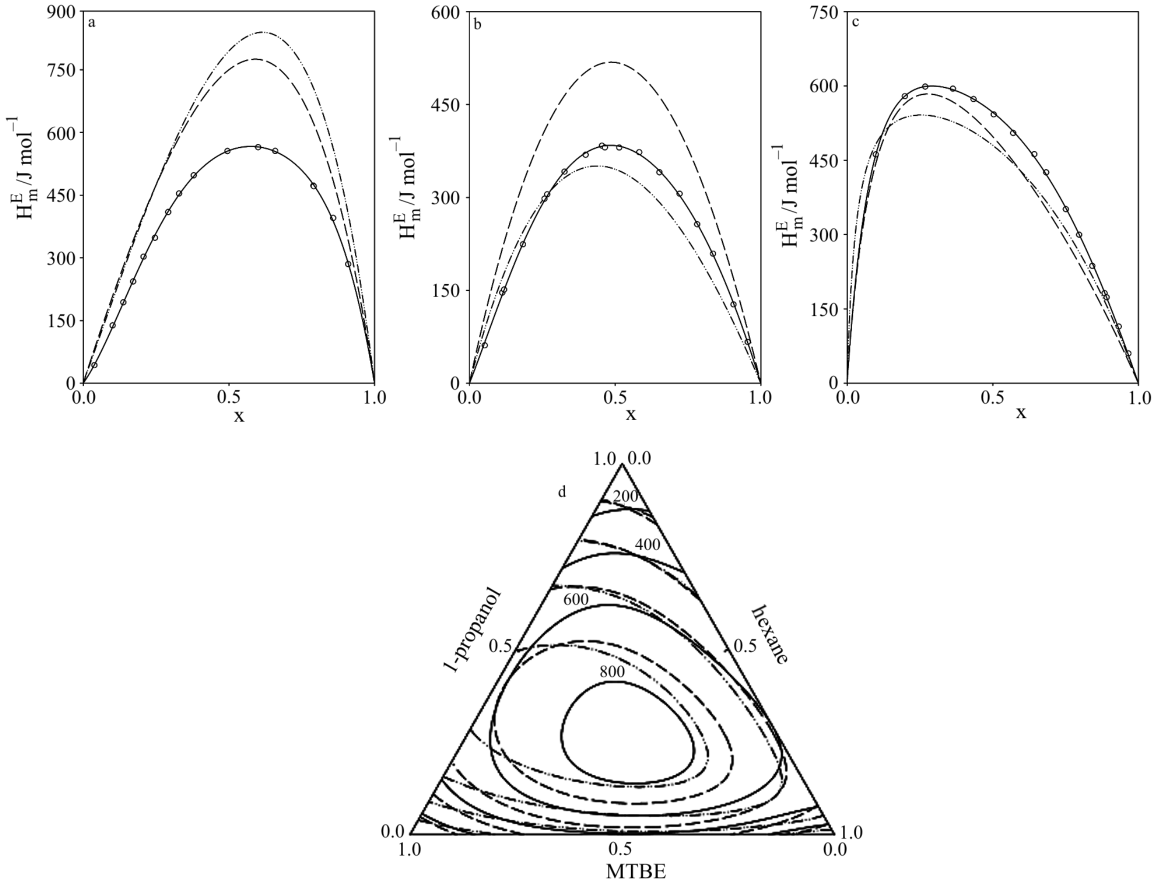

Fig. 4 Excess molar enthalpies for a – MTBE+(1–x)1-propanol, b – MTBE+(1–x)hexane, c – x1-propanol+(1–x)hexane, d – x_1MTBE+$x_2$1-propanol+$x_3$hexane at 298.15 K, —– fit of experimental data, — — – Larsen *et al.* model, —·— – Gmehling *et al.* model

three different numberings of the components have been tested, in order to check the differences in the predicted values, and to find a rule to decide which ordering should be used in each case. MTBE, 1-propanol or hexane were respectively named as 123. Deviations between experimental data and estimated values are shows in Table 6.

Table 5 Average percentage deviations of excess molar enthalpies predicted by the theoretical models from the experimental data

System	Larsen/%	Gmehling/%
MTBE+1-propanol	37	43
MTBE+hexane	39	14
1-propanol+hexane	14	11
MTBE+1-propanol+hexane	15	12

Table 6 Mean deviations from the experimental values obtained with the empirical predictive methods. For the asymmetric equations three numberings of the components have been compared, in this order, 123, 231, 312

	s/J mol^{-1}			s/mL mol^{-1}		
Kohler	85.4			0.0362		
Jacob–Fitner	103.7			0.0343		
Colinet	88.7			0.0358		
Knobeloch–Schwartz	36.0			0.0361		
Tsao–Smith	39.1[a]	19.2[b]	46.5[c]	0.0612[a]	0.1128[b]	0.0343[c]
Toop	101.6[a]	41.3[b]	128.4[c]	0.0381[a]	0.0538[b]	0.0557[c]
Scatchard	120.3[a]	41.6[b]	130.0[c]	0.0587[a]	0.0538[b]	0.0577[c]
Hillert	107.1[a]	42.0[b]	127.5[c]	0.0385[a]	0.0538[b]	0.0553[c]
Mathieson–Thynne	110.8[a]	75.2[b]	118.6[c]	0.0591[a]	0.0430[b]	0.0443[c]

[a]order 123, [b]order 231, [c]order 312

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