Excess Molar Volumes and Enthalpies of Mixing for the Ternary System (Butyl Butyrate + 1-Octanol + Dodecane) at the Temperature 308.15 K†

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This work continues our studies about the excess thermodynamic properties for ternary mixtures containing esters, 1-alkanols, and alkanes. We present excess volume and excess enthalpy data for the ternary system (butyl butyrate + 1-octanol + dodecane) at the temperature 308.15 K and atmospheric pressure. Excess molar enthalpy measurements were made using a Calvet microcalorimeter. The accuracy in the excess enthalpy measurements was estimated to be better than 1%. Excess molar volumes were determined from the densities of pure liquids and their corresponding mixtures using an Anton Paar digital densimeter with a resolution of $\pm 2 \times 10^{-6}$ g·cm^{-3}. The binary experimental data were fitted using a Redlich−Kister variable-degree polynomial equation. The ternary system results were correlated using the Cibulka equation. Experimental values were compared with the predictions obtained by several group contribution models. The prediction of some empirical equation was also tested. These equations offer reliable estimations of excess properties for a multicomponent mixture using the experimental data of the binary systems. There are the symmetric equations introduced by Kohler; Jacob and Fitzner; and Colinet and the asymmetric ones due to Tsao and Smith; Toop; Scatchard et al.; and Hillert.

Introduction

In previous papers[1−5] we reported experimental excess thermodynamic properties for ternary mixtures containing esters, 1-alkanol, and *n*-alkanes as components. The present article continues this work, reporting excess molar volumes and excess molar enthalpies at 308.15 K and normal atmospheric pressure of (butyl butyrate + 1-octanol + dodecane) and of the constituent binary systems (butyl butyrate + 1-octanol), (butyl butyrate + dodecane), and (1-octanol + dodecane). The study of these properties gives us information of the behavior of the mixture from structural and energetic considerations.

The binary experimental data were fitted using a variable-degree polynomial due to Redlich−Kister.[6] The Cibulka equation[7] has been used to correlate the experimental values of ternary mixtures. Experimental values were compared with the predictions obtained by applying the group contribution model of Nitta−Chao.[8] In addition, the excess molar enthalpies were compared with the predictions of UNIFAC group contribution models considered by Larsen et al.,[9] by Dang and Tassios,[10] and by Gmehling.[11]

In addition, the experimental values obtained were used to test the empirical methods. These equations offer reliable estimation of excess properties for a multicomponent mixture using the involved binary experimental data. The symmetric equations used were those introduced by Kohler,[12] Jacob−Fitzner,[13] and Colinet,[14] while the asym-

Table 1. Experimental Excess Molar Enthalpies for Binary Mixtures at 308.15 K

x	H_m^E / J·mol^{-1}	x	H_m^E / J·mol^{-1}	x	H_m^E / J·mol^{-1}
(x)Butyl Butyrate + (1 − x)1-Octanol[a]					
0.0793	434	0.3756	1462	0.6880	1509
0.1229	663	0.4232	1547	0.7472	1391
0.1965	951	0.4562	1588	0.7902	1263
0.2223	1044	0.5388	1621	0.8451	1042
0.2762	1218	0.5782	1599	0.8911	795
0.3239	1347	0.6402	1573		
(x)1-Octanol + (1 − x)Dodecane					
0.1345	721	0.4591	812	0.7661	504
0.1898	782	0.5302	777	0.8263	390
0.2658	828	0.5667	749	0.8566	327
0.3122	836	0.6587	663	0.8983	240
0.4077	831	0.6595	668	0.9353	153
0.4433	821	0.7509	529	0.9700	71
(x)Butyl Butyrate + (1 − x)Dodecane					
0.0894	295	0.3260	764	0.6779	777
0.1261	392	0.3509	787	0.7202	727
0.1734	516	0.4179	851	0.8233	537
0.2050	573	0.4544	869	0.8287	525
0.2668	689	0.5170	866	0.9006	338
0.3014	736	0.5526	864	0.9307	240

[a] From ref 1.

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Table 2. Experimental Excess Molar Volumes for Binary Mixtures at 308.15 K

x	V_m^E / cm³·mol⁻¹	x	V_m^E / cm³·mol⁻¹	x	V_m^E / cm³·mol⁻¹
		(x)Butyl Butyrate + (1 − x)1-Octanol[a]			
0.0568	0.0878	0.3164	0.2318	0.5963	0.2390
0.1175	0.1354	0.3305	0.2305	0.6471	0.2353
0.1746	0.1678	0.3644	0.2426	0.7092	0.2133
0.1951	0.1761	0.4371	0.2466	0.7702	0.1792
0.2347	0.1958	0.5100	0.2494	0.8247	0.1500
0.2688	0.2097	0.5449	0.2528	0.8626	0.1153
0.2901	0.2247	0.5698	0.2441	0.9502	0.0479
		(x)1-Octanol + (1 − x)Dodecane			
0.1048	0.2039	0.4390	0.2636	0.7934	0.1318
0.1681	0.2344	0.5235	0.2545	0.8325	0.1098
0.2366	0.2548	0.6646	0.1953	0.8701	0.0847
0.3292	0.2684	0.7097	0.1748	0.9093	0.0595
0.3848	0.2648	0.7531	0.1531		
		(x)Butyl Butyrate + (1 − x)Dodecane			
0.0860	0.2250	0.5057	0.6525	0.7625	0.4759
0.1642	0.4085	0.5458	0.6501	0.7821	0.4581
0.2326	0.4990	0.6564	0.5883	0.8186	0.4073
0.3151	0.5957	0.6617	0.5830	0.8623	0.3308
0.4238	0.6386	0.6962	0.5604	0.9593	0.1150
0.4435	0.6409	0.7365	0.5311		

[a] From ref 1.

metric ones were those of Tsao−Smith,[15] Toop,[16] Scatchard et al.,[17] and Hillert.[18]

Experimental Section

Butyl butyrate (Fluka, purity > 99%), 1-octanol (Fluka, purity> 99.5%), and dodecane (Aldrich, purity > 99%) were degassed by ultrasound and dried over 0.4 nm molecular sieves. The measured densities of the pure liquids published previously[1−5] present good agreement with literature values.

Excess molar volumes were determined from the densities of the pure liquids and of the mixtures measured with an Anton Paar DMA 60/602 densimeter with a resolution of $\pm 2 \times 10^{-6}$ g·cm^{-3}, thermostated at (308.15 K ± 0.01 K) in a Haake F3 circulating water bath. The excess molar enthalpies were measured using a Calvet microcalorimeter connected to a Philips PM 2535 voltmeter. The uncertainty of excess molar enthalpies is better than 1%. Paz Andrade[19] and López et al.[20] have described the details of the calibration and experimental procedure. In all cases, the samples were prepared covering the whole composition range of the mixtures using a Mettler AT 201 balance with a precision of 1×10^{-8} kg. The precision of the mole fraction is estimated to around $\pm 1 \times 10^{-4}$.

Three experimental runs were carried out for ternary mixtures formed by adding dodecane to a binary mixture composed of (x_1')butyl butyrate and (1 − x_1')1-octanol, where x_1' is the mole fraction of butyl butyrate in the

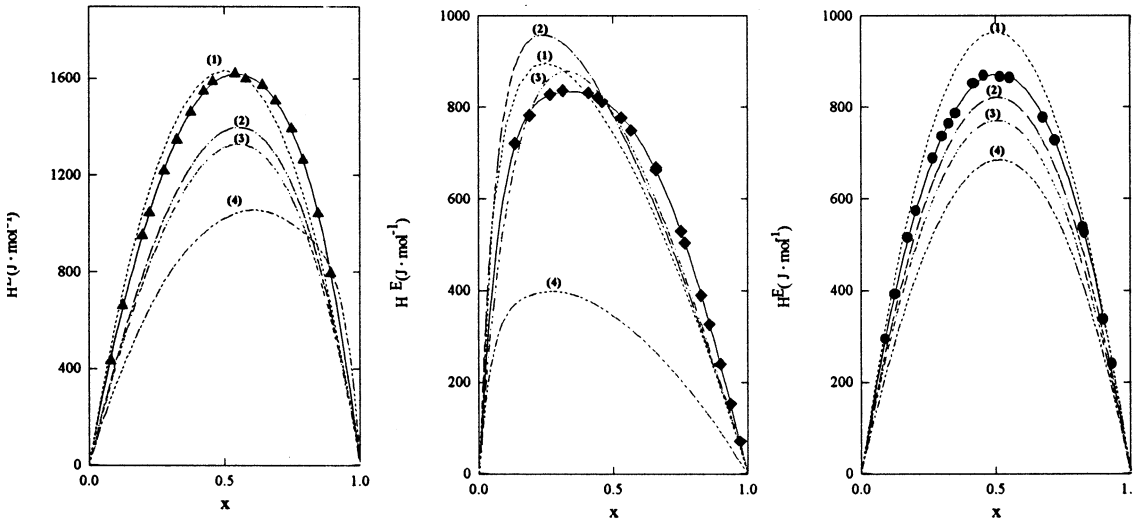

Figure 1. Excess molar enthalpies for (a) {(x)butyl butyrate + (1 − x)1-octanol}; (b) {(x)1-octanol + (1 − x)dodecane}; and (c) {(x)butyl butyrate + (1 − x)dodecane}, where symbols and solid lines represent experimental data and dashed lines represent the following models: (1) Nitta−Chao; (2) UNIFAC (Gmehling); (3) UNIFAC (Larsen); (4) UNIFAC (Tassios).

Table 3. Coefficients A_p and B_p of Eqs 2 and 3 and Standard Deviations, s

	A_0	A_1	A_2	A_3	A_4	A_5	s
			(x)Butyl Butyrate + (1 − x)1-Octanol[a]				
V_m^E/cm³·mol⁻¹	1.0041	0	0.2381	−0.3200			0.0047
H_m^E/J·mol⁻¹	6433.8	904.6	1139.5	2075.9		−2088.1	8
			(x)1-Octanol + (1 − x)Dodecane				
V_m^E/cm³·mol⁻¹	1.0247	−0.4632	0	0	1.0914	−1.1360	0.0024
H_m^E/J·mol⁻¹	3188.9	−1007.5	1152.9	−1853.3	2122.9	−1342	4
			(x)Butyl Butyrate + (1 − x)Dodecane				
V_m^E/cm³·mol⁻¹	2.6078	−0.1132	0.4915				0.0077
H_m^E/J·mol⁻¹	3486.5	102.2	315.3				4

	B_0	B_1	B_2	s
	(x_1)Butyl Butyrate + (x_2)1-Octanol + (1 − x_1 − x_2)Dodecane			
$V_{m,123}^E$/cm³·mol⁻¹	−0.3771	2.5273	0.1398	0.0056
$H_{m,123}^E$/J·mol⁻¹	14474.6	2332.0	−20185.0	29

[a] From ref 1.

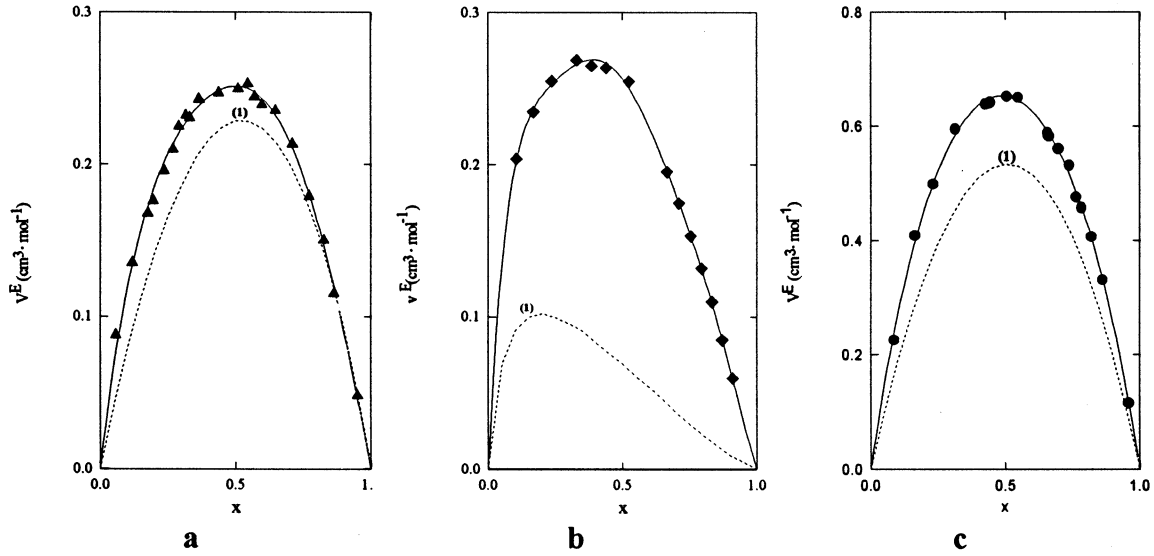

Figure 2. Excess molar volumes for (a) {(x)butyl butyrate + (1 − x)1-octanol}; (b) {(x)1-octanol + (1 − x)dodecane}; and (c) {(x)butyl butyrate + (1 − x)dodecane}, where symbols and solid lines represent experimental data and dashed lines represent the (1) Nitta−Chao model.

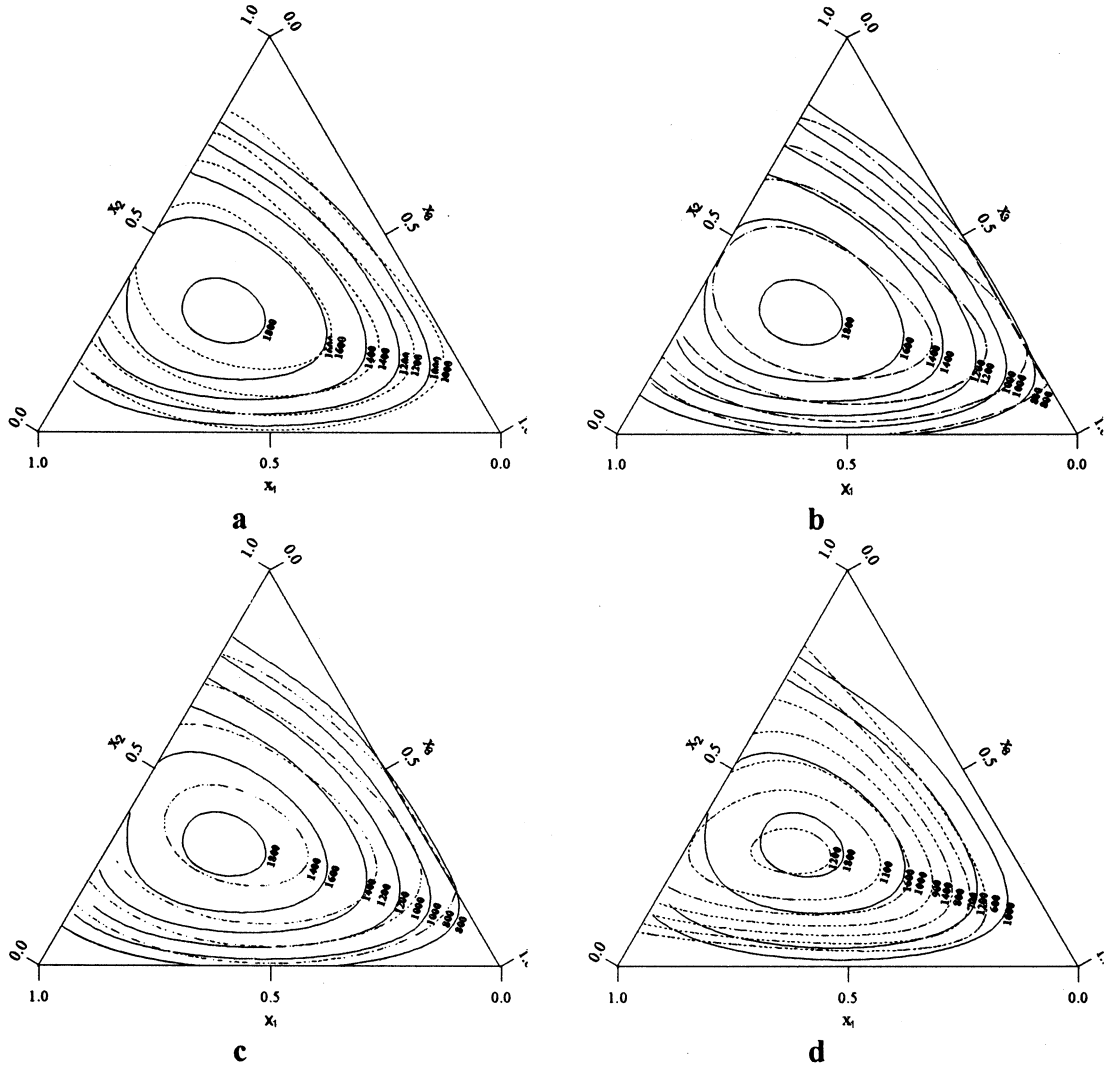

Figure 3. Curves of constant $H^E_{m,123}$ (J·mol^{-1}) for {(x_1)butyl butyrate + (x_2)1-octanol + (x_3)dodecane} at 308.15 K; solid lines show experimental data and dashed lines show data for the following models: (a) Nitta−Chao; (b) Gmehling; (c) Larsen; (d) Tassios.

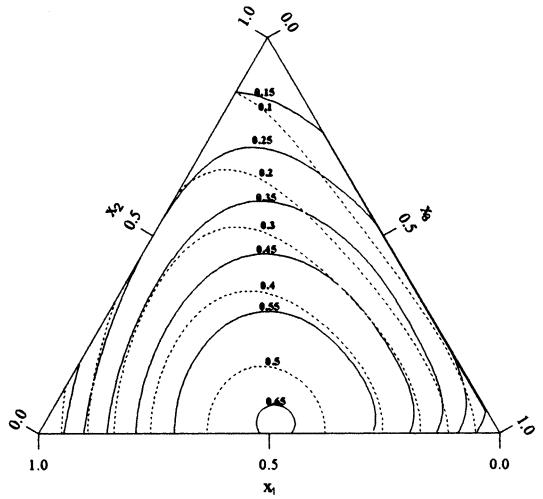

Figure 4. Curves of constant $V^E_{m,123}$ ($cm^3 \cdot mol^{-1}$) for {(x_1)butyl butyrate + (x_2)1-octanol + (x_3)dodecane} at 308.15 K; solid lines show experimental data and dashed lines show the Nitta−Chao model.

Table 4. Experimental Excess Molar Enthalpies for Ternary Mixture {x_1 butyl butyrate + x_2 1-octanol + (1-x_1-x_2) Dodecane} at 308.15 K

x_1	x_2	$H^E_{m,\phi}$ / J·mol^{-1}	$H^E_{m,123}$ / J·mol^{-1}
x'_1 = 0.2463		$H^E_{m,12}$ = 1126 J·mol^{-1}	
0.1309	0.4007	695	1294
0.1226	0.3752	728	1289
0.1004	0.3073	752	1212
0.1039	0.3180	738	1213
0.0767	0.2349	762	1113
0.0720	0.2202	749	1078
0.0485	0.1486	712	934
0.1574	0.4818	628	1348
0.1731	0.5298	561	1353
0.1786	0.5466	531	1347
0.1950	0.5966	429	1320
0.2025	0.6198	383	1309
0.2094	0.6409	329	1287
0.2203	0.6743	239	1246
0.2368	0.7247	95	1178
x'_1 = 0.4981		$H^E_{m,12}$ = 1608 J·mol^{-1}	
0.2753	0.2774	749	1638
0.2594	0.2617	752	1589
0.2331	0.2349	771	1523
0.2157	0.2173	791	1487
0.1921	0.1935	792	1412
0.1542	0.1554	766	1264
0.3209	0.3233	671	1707
0.3452	0.3479	625	1739
0.3651	0.3679	570	1748
0.3901	0.3930	493	1752
0.4109	0.4140	430	1757
0.4243	0.4276	362	1732
0.4468	0.4502	274	1716
0.4662	0.4697	173	1678
0.4782	0.4818	112	1655
x'_1 = 0.7476		$H^E_{m,12}$ = 1387 J·mol^{-1}	
0.4108	0.1387	825	1588
0.3790	0.1280	851	1555
0.3526	0.1191	860	1514
0.4800	0.1621	808	1699
0.5106	0.1724	768	1716
0.5529	0.1867	711	1737
0.5701	0.1925	672	1730
0.6208	0.2096	543	1695
0.6259	0.2114	538	1699
0.6659	0.2249	391	1626

binary mixture. A ternary mixture may be considered as a pseudobinary mixture composed of that binary mixture and dodecane.

Table 5. Experimental Excess Molar Volumes for the Ternary Mixture {(x_1)Butyl Butyrate + (x_2)1-Octanol + (1 − x_1 − x_2)Dodecane} at 308.15 K

x_1	x_2	$V^E_{m,123}$ / $cm^3 \cdot mol^{-1}$	x_1	x_2	$V^E_{m,123}$ / $cm^3 \cdot mol^{-1}$
0.0329	0.1466	0.2737	0.1057	0.0602	0.3601
0.0608	0.2707	0.3497	0.2074	0.1183	0.5247
0.0867	0.3858	0.3739	0.3751	0.2138	0.6051
0.1092	0.4862	0.3647	0.4041	0.2304	0.5963
0.0910	0.4049	0.3785	0.4501	0.2566	0.5651
0.1299	0.5781	0.3370	0.5126	0.2922	0.4895
0.1490	0.6630	0.2840	0.0714	0.0216	0.2471
0.0528	0.1407	0.3119	0.1320	0.0400	0.3849
0.0904	0.2409	0.3984	0.2451	0.0742	0.5583
0.1273	0.3393	0.4410	0.3450	0.1044	0.6305
0.1629	0.4343	0.4237	0.4502	0.1363	0.6429
0.1750	0.4665	0.4228	0.4852	0.1469	0.6297
0.2045	0.5451	0.3887	0.6122	0.1853	0.5044
0.2212	0.5895	0.3468	0.6955	0.2105	0.3616
0.2487	0.6627	0.2854	0.0804	0.0096	0.2374
0.0822	0.0915	0.3415	0.1523	0.0182	0.3908
0.1529	0.1702	0.4677	0.2721	0.0325	0.5568
0.2767	0.3080	0.5495	0.4064	0.0485	0.6467
0.3019	0.3360	0.5277	0.6235	0.0744	0.5868
0.3811	0.4243	0.4482	0.7139	0.0851	0.4830
0.4260	0.4742	0.3641	0.8088	0.0965	0.3124

Table 6. Mean Deviations, *s*, of the Experimental Excess Molar Volumes from the Values Obtained with the Group Contribution Model of Nitta−Chao

	s / $cm^3 \cdot mol^{-1}$
butyl butyrate + 1-octanol	0.03
1-octanol + dodecane	0.14
butyl butyrate + dodecane	0.10
butyl butyrate +1-octanol + dodecane	0.12

The ternary excess molar enthalpies at compositions x_1, x_2, and x_3 can be expressed as

$$H^E_{m,123} = H^E_{m,\phi} + (x_1 + x_2)H^E_{m,12} \quad (1)$$

where $H^E_{m,\phi}$ are the measured excess molar enthalpies and $H^E_{m,12}$ are the excess mole enthalpies of the initial binary mixture (butyl butyrate + 1-octanol). Values of $H^E_{m,12}$ at different mole fractions were interpolated by a spline-fit method.

Results and Discussion

Experimental excess molar enthalpies and excess molar volumes for binary mixtures are reported in Tables 1 and 2, respectively. A variable-degree Redlich−Kister polynomial in mole fractions x_i was fitted to the results for each binary system by the unweighted least-squares method. This expression has the form

$$Q^E(x_i,x_j) = x_i x_j \sum_{p=0}^{q} A_p (x_i - x_j)^p \quad (2)$$

where Q^E represents excess molar enthalpies, H^E_m, or excess molar volumes, V^E_m. The degree of the polynomial, q, was optimized by applying the F-test.[21] The coefficients A_P and the standard deviations are given in Table 3. Figures 1 and 2 present the experimental binary H^E_m and V^E_m values plotted against mole fraction of the first component of the binary mixture in each case and the curves calculated from the smoothing equation (eq 2).

The experimental excess molar volumes and excess molar enthalpies of the ternary system $H^E_{m,123}$ and $V^E_{m,123}$ are shown in Tables 4 and 5. The Cibulka equation has

Table 7. Mean Deviations, *s*, of the Experimental Excess Molar Enthalpies from the Values Obtained with the Group Contribution Models Used in This Work

	s/J·mol^{-1}			
	Nitta−Chao	Larsen	Tassios	Gmehling
butyl butyrate + 1-octanol	84	253	437	201
1-octanol + dodecane	75	46	358	85
butyl butyrate + dodecane	65	85	151	48
butyl butyrate +1-octanol + dodecane	60	250	580	174

Table 8. Standard Deviations, *s*, of Models for (a) (x_1)Butyl Butyrate + (x_2)1-Octanol + (1 − x_1 − x_2)Dodecane; (b) (x_1)1-Octanol + (x_2)Dodecane + (1 − x_1 − x_2)Butyl Butyrate; and (c) (x_1)Dodecane + (x)Butyl Butyrate + (1 − x_1 − x_2)1-Octanol

	a	b	c
$s(V^E_{m,123})$/cm^3 mol^{-1}			
Kohler		0.014	
Jacob-Fitzner		0.022	
Colinet		0.011	
Tsao-Smith	0.027	0.025	0.019
Toop	0.019	0.023	0.020
Scatchard	0.019	0.023	0.020
Hillert	0.020	0.023	0.019
$s(H^E_{m,123})$/J·mol^{-1}			
Kohler		221	
Jacob-Fitzner		245	
Colinet		207	
Tsao-Smith	132	144	91
Toop	227	188	259
Scatchard	228	198	269
Hillert	225	184	249

been fitted to the experimental values of the ternary mixtures:

$$Q^E_{123} = Q^E_{bin} + x_i x_j (1 - x_i - x_j)(B_0 + B_1 x_i + B_2 x_j) \quad (3)$$

where

$$Q^E_{bin} = Q^E_{12} + Q^E_{13} + Q^E_{23} \quad (4)$$

The symbol Q^E_{123} represents $V^E_{m,123}$ or $H^E_{m,123}$, and Q^E_{ij} values were fitted by means of eq 2. Table 3 also presents the parameters B_p and the standard deviation between experimental and fitted values.

Our results for binary and ternary mixtures were compared with those of the Nitta−Chao theory using the interaction parameters given in refs 8 and 22−24. The curves obtained are represented in Figures 1−4 by dashed lines. Tables 6 and 7 show mean deviations between experimental data and the behavior predicted from the Nitta−Chao model.

Results for excess molar enthalpies were also compared with the predictions of UNIFAC group-contribution models considered by Larsen et al.,[9] Tassios et al.,[10] and Gmehling et al.[11] Table 7 shows the mean deviation percentage obtained in each case. Figure 1 shows by dashed lines the curves obtained by application of those models. By applying the Nitta−Chao model, we observe that the predicted values of excess molar volumes are in all cases better than 0.14 cm^3·mol^{-1}. For the excess molar enthalpies data, the overall best prediction comes from the Nitta−Chao model, although for the binary systems the best prediction is obtained for the Larsen version for 1-octanol + dodecane and the Gmehling version for butyl butyrate + dodecane.

Values of $V^E_{m,123}$ and $H^E_{m,123}$ have also been calculated using the empirical equations proposed by Kohler,[12] Jacob and Fitzner,[13] Colinet,[14] Tsao and Smith,[15] Toop,[16] Scatchard,[17] and Hillert,[18] which take only the binary contribution into account. For the asymmetric methods (Toop; Scatchard; Tsao−Smith; and Hillert), we must indicate the order of components in the mixtures. Table 8 shows the standard deviation between experimental and predicted values. For the application of empirical expressions, we observe that the symmetric empirical expressions show a better correlation for the excess molar volumes; on the other hand, for the excess molar enthalpies, the better results are offered by the asymmetric empirical expression of Tsao−Smith, choosing the alkane as the first component.

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