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TERNARY HEAT EFFECTS IN TERNARY MIXTURES

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ABSTRACT

Excess enthalpies of three ternary mixtures consisting of benzene + carbon tetrachloride + cyclohexane at 303.15 K, acetone + carbon tetrachloride + cyclohexane, and acetone + cyclohexane + chloroform at 298.15 K have been measured over the whole composition range using an adiabatic calorimeter. Ternary effects determined as the differences of the experimental excess heats from those of the sum of the binary contributions by the Nissema method are found to be positive for the first two systems and both positive and negative for the last system.

INTRODUCTION

The need of binary mixtures acting as the building blocks for prediction of properties of ternary mixtures has led to the publication of quite a number of papers in the last fifteen years [1–8]. In continuation of our work on excess functions of ternary systems [19], we report excess heats of three ternary systems measured over the entire concentration range (for the first system at 303.15 K and for the other two at 298.15 K). The systems are (i) benzene + carbon tetrachloride + cyclohexane; (ii) acetone + carbon tetrachloride + cyclohexane; (iii) acetone + cyclohexane + chloroform.

The systems contain those components which present an assortment of interactions. An attempt has been made to find the differences between the experimentally measured values and those of the statistical sums of the corresponding binary contributions employing the Nissema method [1]. The differences, termed ternary heat effects, have been illustrated as equivalue contours on equilateral triangles. The plots resemble those of the corresponding volume effects, and thus point to definite ternary effects and almost identical behaviour for volume and heat effects.

EXPERIMENTAL

The solvents used were all of A.R. grade except cyclohexane, which was of L.R. grade, and were purified by methods described elsewhere [9]. Purities

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TABLE 1

Comparison of H^E data with literature values at 303.15 K

x	H^E (J mol^{-1})	
	This work	lit. [a]
	$(x)\text{c-C}_6\text{H}_{12}+(1-x)\text{C}_6\text{H}_6$	
0.2001	472.4	468.4
0.3100	619.3	615.7
0.4735	711.8	714.7
0.7018	619.4	618.6
	$(x)\text{C}_6\text{H}_6+(1-x)\text{CCl}_4$	
0.3075	92.5	94.9
0.4752	113.4	115.6
0.6105	109.1	107.2
0.6473	100.8	101.2

[a] From ref. 14.

were checked by comparing the densities and refractive indices of the solvents with literature values [9]. The comparison was found to be good.

Heats of mixing of binary and ternary mixtures were determined by using an adiabatic calorimeter as reported by Nigam and Mahl [10] and Lark and Banipal [11] and is a modified version of those used by Larkin and McGlashan [12]. The calorimeter was checked by measuring excess heats of benzene + cyclohexane mixtures over the entire composition range at 298.15 K. The agreement with various literature values is very good and is reported elsewhere [13]. Excess heats of a few binary mixtures of this system and of the benzene + carbon tetrachloride system were also determined at 303.15 K and are compared with corresponding literature values in Table 1. The agreement is good.

Heat of mixing of a ternary mixture H_t^E was obtained by diluting one of its component binaries with a suitable amount of the third component and the value per mole of the ternary mixture was obtained from the expression

$$H_t^E = \frac{H^E + H_b^E x_i}{x_i^1} \tag{1}$$

where H_b^E is the molar heat of mixing of the component binary selected, say (1,2) at mole fraction x_i^1 of the component $i (i = 1$ or $2)$, x_i is the mole fraction of the same component in the resultant ternary mixture and H^E is given by

$$H^E = \left(c^2 Rt/\Sigma\eta\right)\left(\theta_1/\theta_2\right) \tag{2}$$

where c is the current in amperes, R is the resistance of the heater in ohms, and t is the time in s. θ_1, θ_2 are the deflections obtained from the deflection–decay curves obtained during mixing and heating.

All computer calculations were carried out on system DEC-20.

RESULTS AND DISCUSSION

Measured excess heats of the three ternary systems as a function of x_1 and x_2 are summarized in Table 2. To compare the results with those predicted from the sum of the corresponding binary contributions, excess heats for the component binary mixtures (seven in all) from the literature were fitted to the equation

$$H^E = x_1 x_2 \left[A + B(2x-1) + C(2x-1)^2 + \ldots \right] \tag{3}$$

For some of the systems where reported deviations of the measured values appeared to be larger, fresh measurements were carried out. The fitting constants A, B, C of eqn. (3) for all systems are summarized in Table 3. It may be seen that H^E values for equimolar mixtures of these systems agree very well with those reported in the literature, except for the carbon tetrachloride + acetone system. However, the values measured in this work with smaller standard deviations have been used in further calculations.

A summing procedure given by Nissema [1] has been used to determine the sum of the contributions of the component binaries. In this method it is assumed that the partial molal heat of a component in a ternary mixture remains the same, and in the absence of ternary effects, the only effect is dilution by the third component and thus the partial molal heat of a component i in the (ijk) mixture is given as

$$H_i^E = \left(\frac{x_j}{x_j + x_k} \right) \left(H_i^E \right)_{ij} + \left(\frac{x_k}{x_j + x_k} \right) \left(H_i^E \right)_{ik} \tag{4}$$

where $i = 1, 2, 3$, $j = 2, 3, 1$ and $k = 3, 1, 2$. $(H_i^E)_{ij}$ is the partial molal excess heat of the ith component in the (ij) binary mixture at mole fraction $x_i/(x_i + x_j)$ of the ith component. Excess heat of a ternary mixture as a sum of binary contributions is given by

$$H_{123}^E = \Sigma x_i H_i^E \tag{5}$$

The heat effects in excess of the statistical sum of the binary contributions, termed ternary heat effects (ΔH^E) are given as

$$\Delta H^E = H_t^E - H_{123}^E \tag{6}$$

The ΔH^E values obtained for the three systems have been least square fitted to the general expression

$$\begin{aligned} \Delta H^E = x_1 x_2 x_3 \Big[D_1 + \Sigma \big\{ & D_{i+1} x_i + D_{i+4}(x_i - x_{i+1}) + D_{i+7}(x_i - x_{i+1})^2 \\ & + D_{i+10} x_{i+2}(x_i - x_{i+1}) + D_{i+13}\left(x_{i+2}(x_i - x_{i+1})\right)^2 \big\} \Big] \end{aligned} \tag{7}$$

where $x_4 = x_1$ and $x_5 = x_2$. These parameters $D_1 - D_{16}$ are the regression coefficients for sixteen functions of mole fractions x_1 and x_2. The maximum of two parameters from D_2 to D_7 and maximum two from each of the sets

TABLE 2

Excess heats of ternary mixtures and corresponding ternary heat effects

x_1	x_2	H_t^E (J mol^{-1})	ΔH^E (J mol^{-1})	$\delta(\Delta H^E)$ (J mol^{-1})
$(x_1)C_6H_6 + (x_2)CCl_4 + (1 - x_1 - x_2)c\text{-}C_6H_{12}$ at 303.15 K				
0.1676	0.4701	283.7	30.3	1.3
0.1500	0.4206	293.7	23.2	−3.0
0.1253	0.3515	306.1	24.3	4.2
0.1333	0.3740	300.5	20.5	−1.7
0.1586	0.4447	288.8	25.8	−2.0
0.1009	0.2831	290.4	15.8	2.4
0.0686	0.1925	235.1	4.1	−1.7
0.5208	0.1541	511.8	38.5	0.5
0.5991	0.1772	425.9	36.3	−0.4
0.6233	0.1841	394.7	35.5	0.9
0.4243	0.1255	562.0	28.0	−2.8
0.1358	0.1083	343.0	6.4	−0.8
0.1761	0.1405	392.8	11.2	−2.3
0.4167	0.3325	365.8	46.9	0.2
0.2076	0.1654	427.4	24.9	5.3
0.3682	0.2938	408.8	46.8	−1.2
0.3533	0.2819	421.6	48.3	1.3
0.3224	0.2573	438.6	45.6	2.5
0.3515	0.1039	565.1	20.8	−1.5
0.3899	0.0863	602.6	25.1	4.5
0.2620	0.0579	528.6	4.0	−4.8
0.4138	0.0916	602.8	27.0	4.1
0.6810	0.1507	370.9	25.6	−2.4
0.4601	0.1018	586.2	23.0	−4.1
0.3898	0.0863	602.8	25.3	4.7
0.5563	0.3371	266.4	26.1	−0.2
0.3598	0.4864	254.1	29.1	−1.5
0.6129	0.2941	256.5	24.0	0.8
0.5276	0.3589	269.8	28.0	0.4
0.7639	0.1794	195.4	17.0	4.3
0.1491	0.6465	187.9	16.0	−1.7
0.7797	0.1674	184.8	14.8	3.4
0.0961	0.6867	167.6	8.8	−2.1
0.3849	0.3426	358.9	50.0	2.5
0.5829	0.2323	364.0	32.3	−5.1
0.4859	0.2863	375.5	40.9	−3.6
0.7876	0.1183	258.0	15.9	0.6
0.7703	0.1279	275.6	20.7	3.5
0.1010	0.5006	250.4	17.4	0.4
				$\sigma = 2.9$
$(x_1)(CH_3)_2CO + (x_2)CCl_4 + (1 - x_1 - x_2)c\text{-}C_6H_{12}$ at 298.15 K				
0.6418	0.1173	902.6	10.5	−3.1
0.2939	0.0538	1253.1	9.1	−6.1
0.4476	0.0819	1265.6	13.3	4.2

TABLE 2 (continued)

x_1	x_2	H_t^E (J mol^{-1})	ΔH^E (J mol^{-1})	$\delta(\Delta H^E)$ (J mol^{-1})
0.3781	0.0692	1288.8	4.5	−7.5
0.6276	0.1149	948.5	17.5	4.1
0.3726	0.0682	1293.4	8.9	−3.3
0.6443	0.1179	897.3	13.0	−1.6
0.4088	0.2184	889.1	26.9	3.0
0.4533	0.2420	802.4	25.0	0.0
0.4699	0.2508	765.1	24.2	−1.9
0.4464	0.2383	811.0	19.1	−5.5
0.2921	0.1561	1014.1	33.4	3.0
0.2480	0.1517	977.4	41.1	6.0
0.3737	0.1995	952.1	36.6	11.9
0.3339	0.1782	979.4	21.1	−6.0
0.3492	0.1864	973.2	29.1	3.0
0.1617	0.1847	768.9	44.2	−0.1
0.1968	0.2247	785.5	49.1	3.4
0.1616	0.1845	763.8	39.1	−5.2
0.3121	0.3464	646.3	41.0	0.6
0.3271	0.3734	618.1	40.9	0.4
0.3270	0.3733	611.1	33.7	−6.8
0.1871	0.2135	781.7	45.0	−0.7
0.2192	0.2502	777.2	49.4	4.4
0.1517	0.4858	465.8	46.2	−1.7
0.1230	0.3941	525.6	54.3	6.6
0.1735	0.5559	418.7	50.2	3.3
0.1151	0.3686	524.3	44.5	−2.6
0.0923	0.2958	526.4	44.4	1.2
				$\sigma = 4.9$
$(x_1)(CH_3)_2CO + (x_2)c\text{-}C_6H_{12} + (1 - x_1 - x_2)CHCl_3$ at 298.15 K				
0.4551	0.0779	−1236.1	20.8	5.0
0.5715	0.0976	−836.5	28.9	4.4
0.3196	0.0547	−1378.3	5.7	0.0
0.3048	0.0522	−1372.5	0.1	−4.6
0.4052	0.0693	−1334.8	15.6	3.5
0.3452	0.0590	−1385.3	6.3	−0.9
0.3684	0.0629	−1389.1	−3.5	−12.4
0.4204	0.0872	−1188.1	26.9	12.0
0.5287	0.3487	770.0	13.2	−1.9
0.3946	0.4479	779.3	2.4	2.9
0.6320	0.2722	753.6	35.0	15.3
0.7602	0.1774	620.6	7.3	−8.2
0.5259	0.3507	776.8	19.8	4.9
0.4011	0.4431	770.1	−6.3	−6.8
0.4508	0.4063	775.4	4.3	−2.8
0.4592	0.4001	768.1	−1.9	−10.0
0.6938	0.2264	703.4	25.3	6.4
0.4335	0.2105	−372.0	22.6	2.9
0.4155	0.1544	−748.9	17.6	−0.6

TABLE 2 (continued)

x_1	x_2	H_t^E (J mol^{-1})	ΔH^E (J mol^{-1})	$\delta(\Delta H^E)$ (J mol^{-1})
0.3434	0.2440	−313.4	7.4	−0.0
0.7005	0.1119	−231.8	32.6	6.7
0.3499	0.2415	−313.2	15.3	6.9
0.6338	0.1358	−317.9	27.2	−2.1
0.4653	0.0870	−1155.5	19.9	2.3
0.6202	0.0617	−1012.8	9.2	−10.0
0.2970	0.1138	−983.0	12.9	6.7
0.7122	0.0468	−805.5	4.6	−11.0
0.4589	0.0896	−1160.7	3.0	−4.6
0.3291	0.1091	−1058.1	−0.4	−9.3
0.7170	0.0458	−792.9	6.5	−8.7
				$\sigma = 7.7$

of three subsequent parameters were evaluated by stepwise regression analysis. After each addition of a parameter a statistical test (increase in $\overline{R}^2$) was applied and the new parameter was retained only if it stood the test. The D_i parameters obtained for the presently studied three ternary systems along with the standard deviations are summarized in Table 4. Using these parameters, ΔH^E for any composition may be evaluated from eqn. (7) and the corresponding total H_t^E value from eqn. (6). The differences between the

TABLE 3

Parameters of eqn. (3)

Systems	A	B	C	$10^4\sigma$ (J mol^{-1})	Ref.	H^E (J mol^{-1}) at $x = 0.5$
$C_6H_6 + CCl_4$	462.9	−18.7	−161.9	1	14	115.7
					[a]	113.5
$CCl_4 + c\text{-}C_6H_{12}$	672.5	79.0	−84.5	2	14	168.1
	666.8	40.5	20.3	3	[a]	166.7
$c\text{-}C_6H_{12} + C_6H_6$	2868.3	91.0	313.3	3	14	717.1
					[a]	720.5
$(CH_3)_2CO + CCl_4$	706.3	−903.6	392.5	16	15	176.6
	736.6	−1076.4	−617.3	6	[a]	184.1
$(CH_3)_2CO + c\text{-}C_6H_{12}$	6292.3	−681.8	1630.1	12	16	1573.1
	6301.4	−694.1	1376.7	4	[a]	1575.3
$c\text{-}C_6H_{12} + CHCl_3$	2565.9	109.3	−43.1	3	17	641.5
$CHCl_3 + (CH_3)_2CO$	−7676.6	−2163.6	1750.4	24	18	−1919.1
	−7636.9	−2328.2	1946.7	18	[a]	−1909.2

[a] This work.

TABLE 4

Parameters of eqn. (7)

System	T (K)		σ (J mol^{-1})
$C_6H_6+CCl_4+c\text{-}C_6H_{12}$	303.15	1 = 899.3, 2 = 1132.0, 9 = −883.3, 16 = −7262.8	3
$(CH_3)_2CO+CCl_4+c\text{-}C_6H_{12}$	298.15	1 = 1018.5, 11 = −5201.7, 9 = 2665.2, 10 = 2425.1	5
$(CH_3)_2CO+c\text{-}C_6H_{12}+CHCl_3$	298.15	1 = −520.3, 5 = 2034.8, 2 = 1554.1	8

experimental ΔH_t^E values and those recalculated from eqn. (7) are included in the last column of Table 2.

A general survey of ternary effects reveals that these are positive and appreciable for the first two systems and both positive and negative and comparatively small for the last system. To recognize their composition dependence, it is necessary to present H_t^E and ΔH^E values on equivalue contour diagrams which have been made for the three systems in Figs. 1–3. The location of equivalue points in triangular plots was computed by a method in which mole fraction of one component was kept constant while those of the other two were varied in steps of 0.05 and the final refinement was attained through a binary search method. The values of the maximum ternary effects together with the corresponding compositions of the mixtures are summarized in Table 5. For comparison, corresponding data for volume effects are also included [19]. Systemwise findings are as follows.

Benzene (1) + carbon tetrachloride (2) + cyclohexane (3)

H_t^E values for the three component binaries of this system are all positive and possess almost symmetric plots [14] (not illustrated). The system (3 + 1)

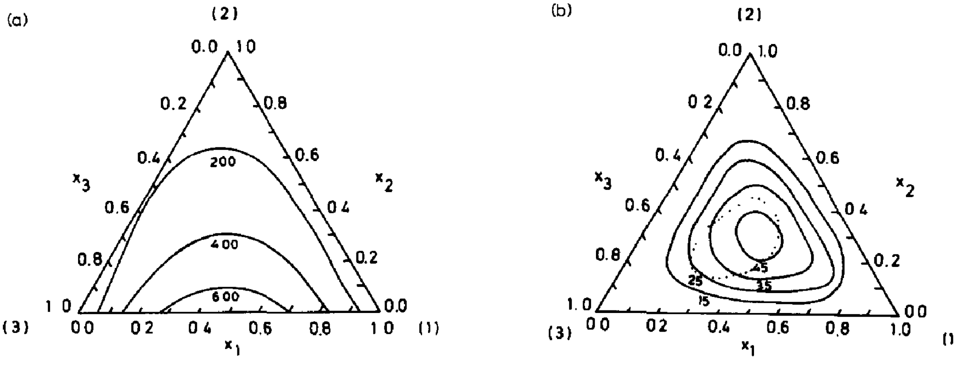

Fig. 1. Plot of equivalue contours of (a) H_t^E and (b) ΔH^E for $C_6H_6+CCl_4+c\text{-}C_6H_{12}$ at 303.15 K.

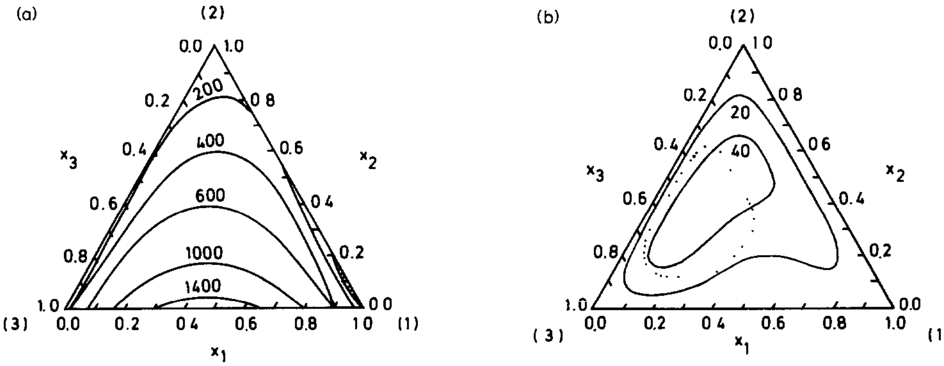

Fig. 2. Plot of equivalue contours of (a) H_t^E and (b) ΔH^E for $(CH_3)_2CO + CCl_4 + c\text{-}C_6H_{12}$ at 298.15 K.

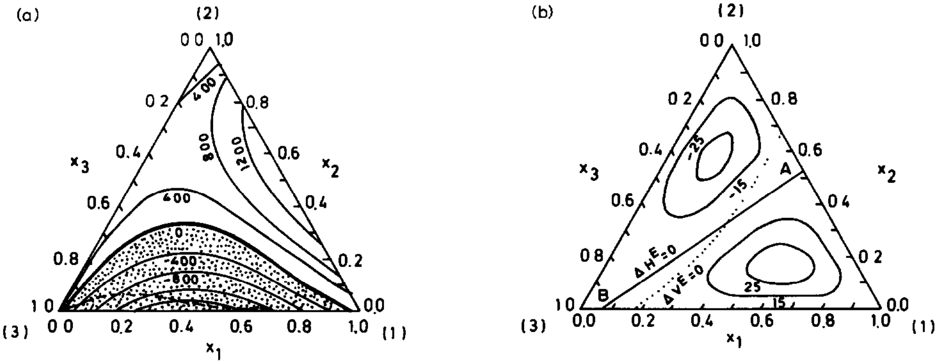

Fig. 3. Plot of equivalue contours of (a) H_t^E and (b) ΔH^E for $(CH_3)_2CO + c\text{-}C_6H_{12} + CHCl_3$ at 298.15 K.

TABLE 5

Values of maximum ternary effects and corresponding properties at 298.15 K

System	Property	x_1	x_2	X_t^E	ΔX^E
$C_6H_6 + CCl_4 + c\text{-}C_6H_{12}$	H^E (J mol^{-1}) [a]	0.41	0.30	400	50
	V^E (cm^3 mol^{-1})	0.44	0.30	0.35	0.07
$(CH_3)_2CO + CCl_4 + c\text{-}C_6H_{12}$	H^E (J mol^{-1})	0.17	0.35	580	48
	V^E (cm^3 mol^{-1})	0.15	0.40	0.48	0.11
$(CH_3)_2CO + c\text{-}C_6H_{12} + CHCl_3$	H^E (J mol^{-1})	0.15	0.58	650	−27
		0.57	0.17	−360	25
	V^E (cm^3 mol^{-1})	0.15	0.40	0.50	−0.06
		0.60	0.16	0.60	0.18

[a] At 303.15 K.

possesses the largest values, with the result that the equivalue contours for the ternary system (Fig. 1(a)) run along this system. Thus dilution of this binary by (2) gives decreasing H_t^E values, while the dilution of (1 + 2) or (2 + 3) by the respective third component gives each value twice. V_t^E values for this system display similar behaviour [19]. The ternary effects, ΔH^E, of this system (Fig. 1(b)) are all positive and symmetrically placed in the triangle, with a maximum of 50 J mol^{-1} which is ca. 12% of the corresponding H_t^E value lying at x_1, $x_2 \simeq 0.4$, 0.3. Lying at nearly the same position, the maximum of ΔV^E (0.07 cm^3 mol^{-1}) is around 20% of the corresponding V_t^E value. One contour of $V^E = 0.06$ cm^3 mol^{-1}, in dots is included in Fig. 1(b) for comparison.

Acetone (1) + carbon tetrachloride (2) + cyclohexane (3)

This system contains one polar component i.e. acetone, while the other two are non-polar. System (1 + 2) shows sigmoid H^E vs. x dependence with slightly negative values at low concentrations of carbon tetrachloride. This may be attributed to two competitive contributions — exothermic n–π interactions and endothermic breaking up of dipole–dipole interactions and also of H-bonds owing their existence to the presence of the enol form of acetone molecules. The strong attractive interaction is more explicit in the volumetric behaviour. In the illustration of equienthalpic contours (Fig. 2(a)), the exothermic H_t^E region does make its appearance but, being small, has little effect on the symmetrical disposition of contours around the (3 + 1) binary, which is highly endothermic.

Ternary effects (ΔH^E) of this system also are all positive (Fig. 2(b)), although the maximum is unsymmetrically placed in the cyclohexane-rich region and the magnitude (8%) is a little smaller than that observed in the previous system, where all the components are non-polar. The positive sign of ΔH^E values over the entire composition range shows that in the case of an interacting mixture (acetone + carbon tetrachloride) being diluted by a non-polar component, the reduction in interaction is more than that expected on a statistical basis. This fact is rather more evident in ΔV^E values (Table 5). One contour of $\Delta V^E = 0.06$ cm^3 mol^{-1} (in dots) displayed in Fig. 2(b) prints out the similarity between the volumetric and enthalpic behaviour.

Acetone (1) + cyclohexane (2) + chloroform (3)

This ternary system may be distinguished from the other two by the fact that one of the component binaries i.e. chloroform + acetone (3 + 1) is characterized by appreciable exothermic mixing over the entire concentration range because of the formation of a heterohydrogen-bonded complex. H^E vs. a composition plot of this system is almost symmetrical [18]. H_t^E

values for the various mixtures of this ternary system studied are either positive or negative. The equiexcess enthalpic contours run into two regions. The shaded area (Fig. 3(a)) is characterized by exothermic mixing separated from the unshaded endothermic mixing region by the zero excess enthalpic contour shown by a thick line. The negative region runs symmetric to the (3 + 1) mixture and the zero H_t^E curve extends almost to an equimolar ternary mixture. In the positive region, as expected, the contours run around the (1 + 2) system as this is greater than the (2 + 3) system. The dilution of the (2 + 3) equimolar binary mixture by (1) is quite interesting. The (2,3) interactions are progressively replaced by (1,2) and (1,3) interactions. As the first is more endothermic, this more than compensates for the exothermic (1,3) interactions up to $x_1 \simeq 0.2$, beyond which the addition of component (1) is practically athermal. This athermal mixing does not indicate that the mixtures represented by the zero enthalpy contour are ideal but rather that the athermicity results from the cancellation of two opposing contributions. This is further supported by the quite different placement of the zero V_t^E value contour, shown by the dashed line in Fig. 3(a).

In the first instance ΔH^E values of this system appear to be negligible, but the plotting of these as equivalue contours in Fig. 3(b) suggests that they may have significance. The triangle is almost equally divided into two regions of positive and negative ΔH^E values by the zero value contour AB. Similar findings have been recorded by us in corresponding ΔV^E values. The regions of negative and positive ΔH^E and ΔV^E values are placed almost identically in the triangle. The zero value contours signifying the concentrations at which the additivity of the binary contributions is obeyed are almost overlapping within the experimental uncertainties and show that the dilution of an equimolar (1 + 2) mixture with (3) is quite predictable from the sum of the binary contributions. Interestingly, mixtures rich in cyclohexane have negative ΔH^E and ΔV^E values arising from further strengthening of interactions. In the acetone-rich region, the results are opposite, indicating weakening interactions.

An overall general conclusion is that in the case of non-interacting ternary mixtures, the mutual interactions are weakened more than in the case of weakly-interacting mixtures, and in the case of strongly-interacting mixtures, the opposite, i.e. stronger interactions, may even result when the concentration of the non-polar component is large. It may be that the solvent effect favours the less polar products. Data from more such systems are needed to confirm these findings.

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