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EXCESS ENTHALPIES OF SEVEN BINARY LIQUID SYSTEMS *

WEI-DONG YAN, RUI-SEN LIN and WEN-HSING YEN
Department of Chemistry, Zhejiang University, Hangzhou (P.R. China)

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ABSTRACT

A Martin-type isothermal displacement calorimeter was constructed for the determination of excess enthalpies of binary mixtures. Seven binary liquid systems of cyclohexane + benzene, + *n*-hexane, ethanol + benzene, *n*-butanol + methyl acetate, + ethyl acetate, + *n*-propyl acetate and + *n*-butyl acetate were investigated.

INTRODUCTION

Solution chemistry has been and will be extensively studied because of its scientific importance as well as its industrial applications. Binary solutions though simple in content possess properties strongly influenced by the nature of their individual components, especially if they are associated when pure or if they become associated when mixed.

Association in solutions may be investigated by vapour–liquid equilibrium studies and by changes in enthalpy or volume observed during mixing. Ideally, solutions with excess enthalpy but no volume change are classified as regular solutions, while solutions with excess volume but no enthalpy change are classified as athermal solutions.

In this investigation, enthalpy changes were measured by mixing a number of liquid pairs, simple and associated. Mixing of associated liquid or liquids takes place usually with changes both in enthalpy and volume.

The mixing of simple liquids was studied not only for the evaluation of the calorimeter constructed but also for a better understanding of their simpler excess enthalpy–mole fraction relationships. Mixing of an associated liquid with a simple liquid will furnish characteristics for further insights into the mixing of associated liquids.

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EXPERIMENTAL RESULTS

A Martin-type isothermal displacement calorimeter [1] was constructed in this laboratory.

The calorimeter chamber was first filled with one component and the other was added by displacement. The solution was diluted stepwise with successive displacements measured by increases in height in the mercury column. Due to absorption of heat during each successive mixing, the calorimeter was kept isothermal by adding electrical energy at known heating rates.

The excess enthalpy of mixing for a particular solution composition was determined by summing the electrical energy added during the successive dilutions for making up the final composition attained.

Effective dilution is possible only with small displacements, amounting to a total volume fraction of 0.500, which is equivalent to a mixture of one to one by volume. In order to obtain solutions of volume fractions of more than 0.500 for the second component, the filling of the calorimeter chamber and the addition by displacement were exchanged with one component substituting for the other.

Binary systems of cyclohexane and benzene at 289.15, 293.15 and 298.15 K, cyclohexane and *n*-hexane at 298.15 K, and ethanol and benzene at 303.15 K were investigated.

The physical properties of benzene, cyclohexane and *n*-hexane are given in Table 1. The normal boiling points are compared with those reported by Pitzer and Scott [2], Ruehrwein and Huffman [3], and Kobe and Lynn [4], and the refractive indices with those reported in Weast's Handbook [5].

The excess enthalpies of binary mixtures of cyclohexane and benzene at 289.15, 293.15 and 298.15 K are given in Tables 2–5, while those for cyclohexane and *n*-hexane at 298.15 K are given in Table 6.

TABLE 1

Physical properties of benzene, cyclohexane and *n*-hexane

		Benzene	Cyclohexane	*n*-Hexane
Purity		99.85	99.84	99.90
t_B (°C)	exptl.	80.08	80.70	68.73
	lit.	80.1 [a]	80.7 [b]	68.74 [c]
n_D^{20}	exptl.	1.5010	1.4262	1.3722
	lit.	1.5011 [d]	1.4262 [d]	1.3723 [d]

[a] Ref. 2.
[b] Ref. 3.
[c] Ref. 4.
[d] Ref. 5.

TABLE 2

Excess enthalpies of binary mixtures of cyclohexane (1) and benzene (2) at 289.15 K

x_1	H^E (J mol^{-1})	x_1	H^E (J mol^{-1})
0.00744	25.43	0.49821	866.98
0.02331	78.48	0.55026	854.58
0.04476	148.01	0.55746	850.66
0.07701	246.76	0.56842	843.24
0.11295	350.37	0.58832	830.22
0.14991	450.66	0.62235	802.67
0.18186	529.24	0.65896	764.80
0.21001	592.04	0.68949	727.76
0.23749	646.30	0.71916	685.19
0.26171	689.78	0.74989	634.15
0.28601	728.18	0.79380	555.40
0.30714	759.87	0.81964	504.84
0.33344	788.74	0.85059	436.56
0.34963	805.99	0.88912	362.17
0.36726	821.63	0.91783	265.66
0.40560	848.23	0.95342	158.23
0.45023	867.48	0.98475	54.04

TABLE 3

Excess enthalpies of binary mixtures of cyclohexane (1) and benzene (2) at 293.15 K

x_1	H^E (J mol^{-1})	x_1	H^E (J mol^{-1})
0.00922	31.67	0.50230	828.21
0.02782	94.55	0.54552	823.19
0.06115	203.66	0.54788	821.40
0.08353	272.00	0.56174	809.07
0.10174	322.09	0.58321	793.67
0.12166	374.12	0.62534	762.99
0.15616	466.55	0.64923	739.54
0.18836	538.25	0.67786	708.52
0.21763	596.48	0.70790	669.67
0.24686	646.96	0.74230	619.23
0.27304	689.44	0.77987	557.84
0.29365	720.29	0.82093	479.73
0.31592	749.08	0.86527	389.06
0.34405	779.15	0.90473	292.40
0.36352	790.35	0.93143	218.67
0.36869	794.01	0.96483	116.39
0.37942	801.48	0.98735	43.44
0.45034	828.53		

TABLE 4

Excess enthalpies of binary mixtures of cyclohexane (1) and benzene (2) at 298.15 K (I)

x_1	H^E (J mol^{-1})	x_1	H^E (J mol^{-1})
0.02305	77.18	0.50143	797.75
0.04989	165.36	0.52457	794.45
0.07724	245.87	0.55327	787.24
0.10503	321.87	0.57878	774.24
0.15304	439.32	0.60603	757.78
0.19726	532.02	0.63868	732.75
0.23751	602.77	0.67765	692.82
0.27387	657.47	0.71664	643.42
0.30789	699.25	0.73906	610.24
0.33984	733.84	0.76169	574.02
0.36532	756.76	0.78734	529.42
0.38852	771.60	0.81411	478.51
0.41607	785.91	0.87366	348.61
0.43682	795.29	0.90744	262.56
0.45849	798.48	0.94401	164.47
0.47051	798.87	0.97998	62.01

Experimental values for cyclohexane and benzene at three temperatures with values reported at 298.15 K by Stokes et al. [6] are plotted in Fig. 1, while those for cyclohexane and *n*-hexane with values reported by Marsh

TABLE 5

Excess enthalpies of binary mixtures of cyclohexane (1) and benzene (2) at 298.15 K (II)

x_1	H^E (J mol^{-1})	x_1	H^E (J mol^{-1})
0.02795	94.56	0.42604	790.52
0.05503	181.23	0.50143	797.75
0.07833	250.32	0.52457	794.45
0.10215	315.22	0.55327	787.24
0.12481	373.24	0.57878	774.24
0.14704	427.53	0.60603	757.78
0.16576	468.06	0.63868	732.75
0.18507	508.45	0.67765	692.82
0.20349	544.36	0.71664	643.42
0.23538	600.59	0.73906	610.24
0.26756	648.55	0.76169	574.02
0.29753	688.18	0.78734	529.42
0.32362	716.54	0.81411	478.51
0.34609	738.49	0.87366	348.61
0.37047	759.12	0.90744	262.56
0.39374	771.62	0.94401	164.47
0.41383	782.62	0.97998	62.01

TABLE 6

Excess enthalpies of binary mixtures of cyclohexane (1) and *n*-hexane (2) at 298.15 K

x_1	H^E (J mol^{-1})	x_1	H^E (J mol^{-1})
0.02294	15.88	0.57890	221.33
0.05703	38.45	0.59137	221.74
0.09339	61.38	0.60293	221.98
0.12969	82.77	0.61767	220.57
0.18488	112.63	0.64206	216.36
0.23066	134.76	0.65803	213.34
0.27536	154.12	0.68753	208.79
0.31425	168.78	0.71551	203.82
0.35176	180.67	0.74654	192.64
0.38367	191.95	0.78111	178.79
0.41378	200.34	0.81868	160.23
0.43377	204.97	0.85918	133.85
0.45926	210.31	0.87996	119.30
0.47785	213.53	0.89872	104.46
0.49328	216.01	0.92521	79.75
0.52314	218.89	0.95136	55.08
0.56308	221.57	0.97607	28.17

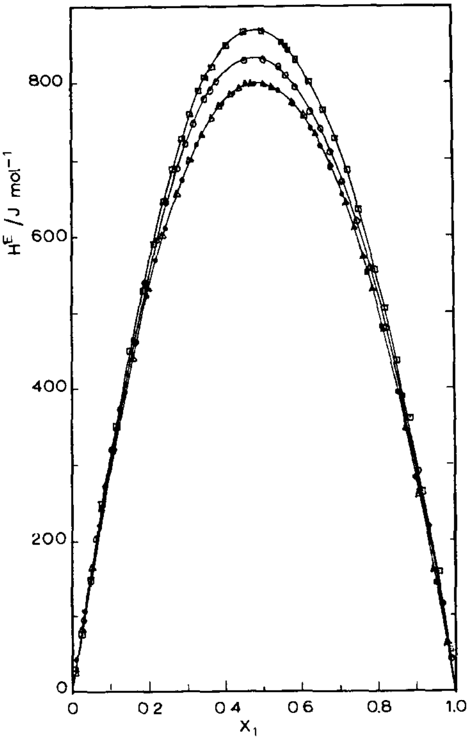

Fig. 1. Excess enthalpies of binary mixtures of cyclohexane (1) and benzene (2). □, 289.15 K; ○, 293.15 K; △, 298.15 K; ●, Stokes et al. [6].

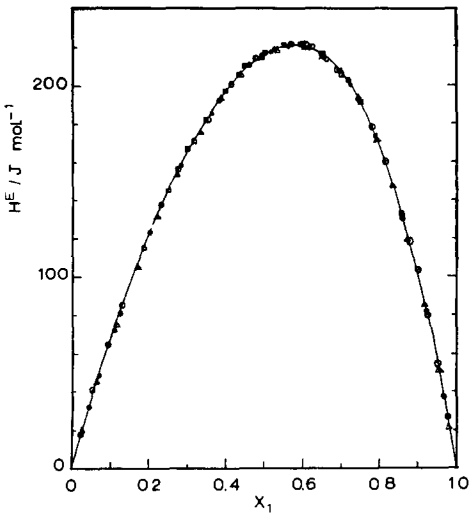

Fig. 2. Excess enthalpies of binary mixtures of cyclohexane (1) and *n*-hexane (2) at 298.15 K. •, Present work; ○, Marsh and Stokes [7]; □, Murakami and Benson [8]; △, Tanaka et al. [9].

and Stokes [7], Murakami and Benson [8], and Tanaka et al. [9] are plotted in Fig. 2.

Numerical equations were established for the excess enthalpies given in Tables 2–6. The empirical constants calculated are listed in Table 7.

With the empirical constants obtained, the excess enthalpies at successive 0.10 mole fraction intervals of cyclohexane for the above two binary mixtures were enumerated and compared with values cited in the literature. For binary mixtures of cyclohexane and benzene, additional values reported by Van Ness [10], Murakami and Benson [8], Ewing et al. [11], Nagata and

TABLE 7

Empirical constants of

$H^E = x_1 x_2 \Sigma A_i (x_2 - x_1)^i$

for binary mixtures of cyclohexane–benzene and cyclohexane–*n*-hexane

Temp. (K)	A_0	A_1	A_2	A_3	σ
Cyclohexane (1)–benzene (2)					
289.15	3474.3	288.8	23.09	−436.8	0.95
293.15	3331.2	326.87	172.44	−350.38	2.15
298.15 (I)	3201.6	145.34	124.23	41.54	1.67
(II)	3199.5	137.82	146.31	80.77	1.29
Cyclohexane (1)–*n*-hexane (2)					
298.15	862.97	−257.41	111.18	−2.61	0.67

TABLE 8

Comparison of excess enthalpies (J mol^{-1}) of binary mixtures of cyclohexane (1) and benzene (2) at 298.15 K with values cited in the literature

x_1	Present work		Literature					
	I	II	Ref. 10	Ref. 6	Ref. 8	Ref. 11	Ref. 12	Ref. 13
0.10	310.0	310.0	307.3	308.1	308.3	308.7	308.3	307.8
0.20	537.4	536.4	534.2	535.3	535.7	536.4	535.6	534.8
0.30	691.1	689.5	688.2	689.5	690.4	690.6	690.0	688.7
0.40	777.3	776.1	774.6	776.3	777.6	776.9	777.0	775.0
0.50	800.1	799.9	797.3	799.3	800.0	799.3	800.1	797.5
0.60	761.9	762.5	758.5	760.7	762.2	760.0	761.1	758.3
0.70	663.4	664.1	659.4	661.5	662.6	660.0	661.1	658.5
0.80	504.4	504.3	500.2	501.8	502.2	500.7	500.3	498.7
0.90	283.7	282.7	280.6	281.4	281.2	280.8	279.0	279.0

Kazuma [12], and Murray and Martin [13] are included while for cyclohexane and *n*-hexane, additional values reported by McGlashan and Stoeckli [14] are included. Agreements among the tabulated excess enthalpies in Tables 8 and 9 are much more evident than among the plotted points in Figs. 1 and 2.

As a preliminary study on associated liquids, binary mixtures of ethanol and benzene, an associated liquid and a simple liquid, were selected for investigation. The physical properties of ethanol are: purity = 99.95%; t_B = 78.30 °C, 78.55 °C (Fiock et al. [15]); $n_D^{20} = 1.3616$. Their excess enthalpies at 303.15 K are given in Table 10. The values are also plotted in Fig. 3 with those for the binary mixtures of ethanol and *n*-heptane reported by Shatas et al. [16] included for comparison.

TABLE 9

Comparison of excess enthalpies (J mol^{-1}) of binary mixtures of cyclohexane (1) and *n*-hexane (2) at 298.15 K with values cited in the literature

x_1	Present work	Literature			
		Ref. 7	Ref. 8	Ref. 14	Ref. 9
0.10	65.42	64.77	64.35	65.71	63.94
0.20	119.68	119.67	119.17	121.02	118.05
0.30	163.30	163.93	163.50	165.50	162.13
0.40	195.82	196.55	196.25	198.18	195.21
0.50	215.94	216.10	215.91	217.50	215.50
0.60	220.54	220.38	220.28	220.70	220.36
0.70	206.62	206.20	206.15	205.70	206.25
0.80	169.28	169.07	169.06	167.00	168.67
0.90	102.73	102.96	102.97	100.10	102.10

TABLE 10

Excess enthalpies of binary mixtures of ethanol (1) and benzene (2) at 303.15 K

x_1	H^E (J mol^{-1})	x_1	H^E (J mol^{-1})
0.01101	228.85	0.66062	621.34
0.07932	641.20	0.68450	579.55
0.16175	841.36	0.70585	546.60
0.23489	906.66	0.73071	502.31
0.29611	927.63	0.75756	454.93
0.34830	919.71	0.78599	402.34
0.39536	900.44	0.81549	347.07
0.43394	875.32	0.85010	280.57
0.46773	847.22	0.88398	214.27
0.60887	702.66	0.92143	144.04
0.62129	684.40	0.96502	60.77
0.64058	654.28		

For deeper insights into the interactions of molecules of associated liquids, care was exercised in the selection of the liquid pair components so that information could be gained by comparing the magnitudes of the maximum excess enthalpies as well as the shapes of the excess enthalpy–mole fraction curves.

n-Butanol was chosen as one component. It is associated due to its hydroxyl content, but its hydrogen-bonding activity is weakened with the extension of its alkyl chain length compared with ethanol. For the other component, an homologous series of four acetates (methyl acetate, ethyl acetate, *n*-propyl acetate and *n*-butyl acetate) was selected. These four

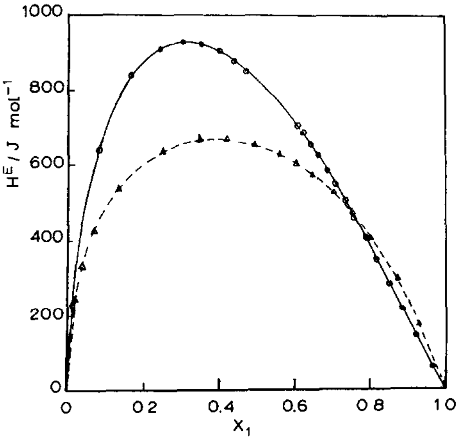

Fig. 3. Excess enthalpies of binary mixtures of ○, ethanol (1) and benzene (2) at 303.15 K and △, ethanol (1) and *n*-heptane at 303.15 K [16].

TABLE 11

Excess enthalpies of binary mixtures of *n*-butanol (1) and methyl acetate (2) at 303.15 K

x_1	H^E (J mol^{-1})	x_1	H^E (J mol^{-1})
0.05967	468.14	0.52705	1950.71
0.12876	925.59	0.53933	1947.66
0.19050	1244.73	0.55719	1941.16
0.24356	1474.06	0.58184	1906.68
0.29089	1636.60	0.61117	1855.14
0.33168	1751.40	0.64438	1784.02
0.36812	1831.48	0.67992	1690.97
0.40204	1886.16	0.72020	1565.88
0.43151	1919.56	0.76733	1397.43
0.45599	1939.14	0.81745	1162.89
0.48376	1952.27	0.87602	851.23
0.50683	1956.32	0.94472	415.80

acetates are associated, and their molecular association decreases with the length of their alkyl chain.

The excess enthalpies of binary mixtures of *n*-butanol and each of the four acetates at 303.15 K are given in Tables 11–14. Among these four liquid pairs, *n*-butanol and ethyl acetate at 298.15 K was studied by Nagata et al. [17], and *n*-butanol + *n*-butyl acetate at 298.15 K was studied by Bravo [18]. Numerical equations were established for the excess enthalpies given in Tables 11–14, with empirical constants calculated and listed in Table 15.

TABLE 12

Excess enthalpies of binary mixtures of *n*-butanol (1) and ethyl acetate (2) at 303.15 K

x_1	H^E (J mol^{-1})	x_1	H^E (J mol^{-1})
0.05764	466.10	0.53161	1641.04
0.11917	843.08	0.54414	1632.86
0.17409	1084.36	0.56519	1631.44
0.22284	1266.00	0.58602	1586.81
0.26956	1402.84	0.61282	1546.70
0.30723	1495.38	0.63647	1507.11
0.34215	1556.33	0.66456	1449.81
0.37352	1600.93	0.69479	1376.48
0.40338	1629.00	0.72888	1285.61
0.42943	1644.15	0.76319	1175.64
0.45457	1654.24	0.80513	1015.42
0.47744	1654.99	0.84962	820.04
0.49833	1648.95	0.89957	579.34
0.50836	1645.71	0.95414	284.00

TABLE 13

Excess enthalpies of binary mixtures of *n*-butanol (1) and *n*-propyl acetate (2) at 303.15 K

x_1	H^E (J mol^{-1})	x_1	H^E (J mol^{-1})
0.06670	444.10	0.57604	1369.47
0.13869	808.61	0.58757	1361.33
0.20105	1048.24	0.60772	1338.24
0.25613	1208.89	0.62733	1310.95
0.30378	1306.83	0.65410	1267.51
0.34422	1366.84	0.67556	1227.56
0.38684	1397.34	0.70191	1175.03
0.41659	1415.12	0.72851	1113.50
0.44487	1427.66	0.75944	1031.75
0.47026	1430.67	0.79267	931.97
0.49648	1430.56	0.82565	820.24
0.52189	1420.71	0.87008	642.29
0.54050	1406.49	0.91290	453.30
0.55206	1396.79	0.95897	226.22

TABLE 14

Excess enthalpies of binary mixtures of *n*-butanol (1) and *n*-butyl acetate (2) at 303.15 K

x_1	H^E (J mol^{-1})	x_1	H^E (J mol^{-1})
0.06290	434.50	0.59883	1246.34
0.13908	809.60	0.61029	1231.72
0.20441	1036.09	0.62950	1202.98
0.25618	1159.90	0.65109	1167.71
0.30529	1250.90	0.67543	1121.95
0.34784	1306.20	0.69688	1078.30
0.38476	1340.17	0.72259	1022.24
0.41953	1357.08	0.74927	955.75
0.44973	1357.33	0.77919	875.43
0.47377	1356.43	0.81029	775.14
0.50122	1338.96	0.84620	655.77
0.52346	1327.31	0.88307	518.91
0.54434	1310.50	0.92307	358.14
0.55819	1297.52	0.96695	162.67

TABLE 15

Empirical constants of

$H^E = x_1 x_2 \Sigma A_i (x_2 - x_1)^i$

for binary mixtures of *n*-butanol and four acetates at 303.15 K

	A_0	A_1	A_2	A_3	A_4	A_5	σ
Methyl acetate	7844.3	34.01	−118.4	660.4	872.0	−539.9	4.9
Ethyl acetate	6618.9	580.5	752.3	430.9	484.4	515.2	2.9
n-Propyl acetate	5698.5	637.2	1265.2	593.5	−388.4	−644.5	3.2
n-Butyl acetate	5371.5	1034.3	836.0	−98.9	372.3	710.8	1.9

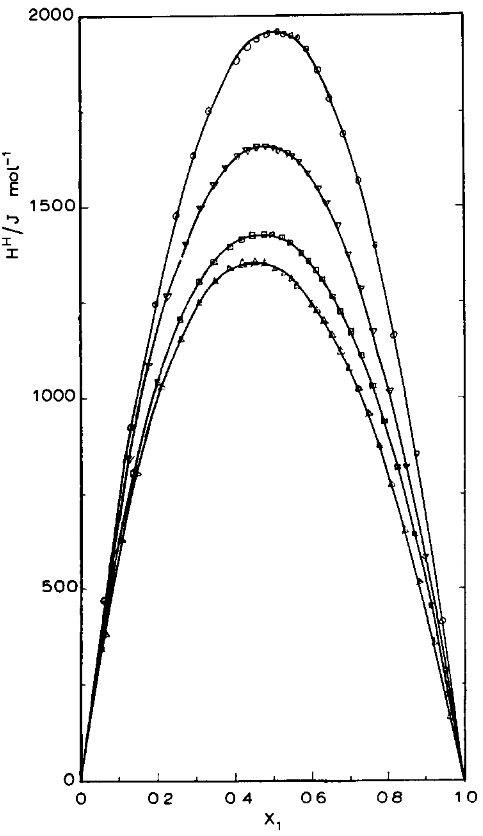

Fig. 4. Excess enthalpies of binary mixtures of *n*-butanol (1) and methyl acetate (□), ethyl acetate (▽), *n*-propyl acetate (□), and *n*-butyl acetate (△) at 303.15 K.

The experimental values for *n*-butanol and each of the four acetates are plotted in Fig. 4.

DISCUSSION

For excess enthalpies and mole fractions of the mixtures at their maxima, the numerical equations for six liquid pairs were solved. That for ethanol and benzene will be considered separately. Maximum excess enthalpies and their corresponding mole fractions of the mixtures are listed in Table 16. The results clearly show that the liquid pairs may be arranged in three groups: cyclohexane and *n*-hexane at 220 J mol^{-1}, cyclohexane and benzene at 800–870 J mol^{-1} for three temperatures and *n*-butanol and four acetates at 1360–1960 J mol^{-1}.

It should be pointed out that the maximum excess enthalpy can not be considered as the sole criterion for classification, especially if associated

TABLE 16

Excess enthalpies and mole fractions of six binary mixtures at maxima

	H^E (J mol^{-1})	x_1
Cyclohexane (1) and *n*-hexane (2)		
298.15 K	220.90	0.58557
Cyclohexane (1) and benzene (2)		
289.15 K	870.07	0.47935
293.15 K	834.89	0.47455
298.15 K (I)	800.82	0.48821
(II)	800.27	0.48872
n-Butanol (1) and acetates (2) at 303.15 K		
Methyl acetate	1961.08	0.49893
Ethyl acetate	1657.58	0.47531
n-Propyl acetate	1430.33	0.46425
n-Butyl acetate	1357.21	0.44533

liquids are involved. Interactions between molecules of the same liquid as well as between molecules of different liquids must be given due attention.

In Table 17, normal boiling points and heats of vaporization for the nine liquids involved are given. The heats of vaporization may be helpful in estimating the relative strengths of interaction between molecules of the same liquid. The strength of the interaction decreases in the order ethanol > ethyl acetate > benzene ≈ cyclohexane ≈ *n*-hexane. Inclinations of the excess enthalpy–mole fraction curves do appear: very strongly toward benzene with ethanol–benzene mixtures, toward benzene with cyclohexane–benzene

TABLE 17

Normal boiling points and heats of vaporization of eight liquids

	T_b (K)	ΔH_v (kJ mol^{-1})
n-Hexane [a]	341.89	28.85
Benzene [a]	353.25	30.76
Cyclohexane [a]	353.88	30.08
Ethanol [a]	351.70	38.58
n-Butanol [a]	390.88	43.14
Methyl acetate [b]	330.95	31.20
Ethyl acetate [a]	349.15	32.30
n-Propyl acetate [b]	374.95	34.74

[a] Ref. 19.

[b] Ref. 20.

mixtures and significantly toward cyclohexane with cyclohexane–*n*-hexane mixtures.

However, the above rule is not applicable to an homologous series of compounds at increasing normal boiling points. If the compounds in the homologous series possess similar entropies of vaporization, i.e. they obey Trouton's rule, the molecules of the higher boiling compounds do not interact with each other any more strongly, even though the compounds have larger heats of vaporization. Actually, their entropies of vaporization become slightly smaller with the higher boiling acetates, thus possessing weaker strengths of interaction.

Comparing the mixing of *n*-butanol with the four acetates, the maximum excess enthalpies decrease at a decreasing rate with the ascending acetates. The excess enthalpy–mole fraction of the mixture curves incline with regular increases toward the acetates on ascending the acetate series. These phenomena are in good agreement with the assumption that the lengthening of the alkyl chain of the acetates tends to weaken the association between acetate molecules and between molecules of *n*-butanol and acetates.

The influence of temperature on the mixing of cyclohexane and benzene is apparent both in appearance and theory. The mixing of cyclohexane and benzene is endothermic. The energies of endothermic reactions, as exemplified by the vaporization of liquids, always decrease thermodynamically with increases in temperature. The inclination of the excess enthalpy–mole fraction of the mixture curves toward benzene clearly indicates that interactions between molecules of both benzene and cyclohexane become weaker at higher temperatures but at a slightly slower rate for the former.

The mixing of ethanol, an associated liquid, and benzene, a simple liquid, deserves special consideration. Its excess enthalpy–mole fraction of the mixture curve is highly inclined. The regular polynomial equation introduced in Tables 7 and 15 would not suffice for the numerical representation of such a distorted curve so that a modified polynomial equation which

TABLE 18

Empirical constants of

$$H^{E} = \frac{x_1 x_2}{1 - k(x_2 - x_1)} \Sigma A_i (x_2 - x_1)^i$$

and excess enthalpy and mole fraction at maximum for a binary mixture of ethanol (1) and benzene (2)

Empirical constants					
A_0	A_1	A_2	A_3	A_4	k
3295.7	−1259.7	−497.64	240.57	−662.81	0.9617

Properties at maximum

$H^{E} = 925.21$ kJ mol^{-1} $x_1 = 0.30573$

included an acentric factor k was adopted instead. Empirical constants for the numerical equation and the excess enthalpy and mole fraction of the mixture at maximum by solving the equation are given in Table 18. When ethanol and benzene were mixed at low ethanol content, polymeric ethanol molecules would be dispersed and dissociated into monomeric ethanol molecules simultaneously. This additional dissociation requires energy, thus rendering the benzene-rich leg of the distorted curve much steeper than the ethanol-rich leg where benzene molecules remain unchanged.

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