Thermodynamical Studies on Binary Systems Consisting of Polar and Non-polar Liquids. II. The Measurement of the Heats of Mixing for Binary Systems of Polar and Non-polar Liquids

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The heats of mixing for 22 binary systems of polar and non-polar liquids were measured. Each pair of polar and non-polar liquids had nearly equal molecular volume. This was done in order to see how the magnitude of the dipole moment of the constituent polar molecules relates to the thermodynamical properties of the binary systems.

Samples

Selection of the Pairs of Polar and Non-polar Liquids.—In selecting pairs of polar and non-polar liquids which are suitable for use in investigating the effect of the dipole moments of the constituent molecules on the heats of mixing the following points were taken into account.

1) The molecular volumes of polar and non-polar components in each pair are nearly equal to each other. This eliminates the effect of the difference of molecular size and makes the effect of the dipoles as clear as possible.

2) The polar molecules have various degrees of magnitudes of dipole moment.

The pairs of polar and non-polar components for which the heats of mixing were measured are shown in Table I with their molar volumes and dipole moments.

It is considered that the observed heats of

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TABLE I. PAIRS OF POLAR AND NON-POLAR LIQUIDS FOR WHICH THE HEAT OF MIXING WERE MEASURED, ILLUSTRATING SIMILARITY OF THEIR MOLAR VOLUMES

Non-polar substances	Molar volume at 25°C, cc.	Polar substances	Molar volume at 25°C, cc.	Dipole moment in Debye unit
Cyclohexane	108.09	Chlorobenzene	101.74	1.56
		Bromobenzene	105.51	1.73
		Nitrobenzene	103.16	4.21
		Benzonitrile	103.50	4.39
		n-Butylchloride	105.73	2.07
		n-Butylcyanide	105.15	4.09
		Diethyl ketone	106.40	2.71
		Methyl propyl ketone	106.40	2.71
		Cyclohexanone	104.67	2.75
		(Toluene	106.85	0.37)
Toluene	106.85	Chlorobenzene	101.74	1.56
		Bromobenzene	105.51	1.73
		Nitrobenzene	103.16	4.21
		Benzonitrile	103.50	4.39
		n-Butylchloride	105.73	2.07
		n-Butylcyanide	105.15	4.09
		Cyclohexanone	104.67	2.75
Methylcyclohexane	128.38	Chlorocyclohexane	119.37	2.07
		Bromocyclohexane	124.43	2.15
trans-Dichloroethylene	77.28	*cis*-Dichloroethylene	76.12	1.89
Benzene	89.40	Pyridine	81.31	2.20
1,4-Dioxane	85.80	Pyridine	81.31	2.20

mixing of polar and non-polar liquids contain both contributions from the non-electrostatic dispersion forces and those from the electrostatic dipolar and higher multipolar forces.

The former contribution may be small if the pair of polar and non-polar molecules have molecular structures similar to each other except in the case of the dipole moment. For such binary mixtures, the observed heats of mixing may be considered to be approximately equal to the contribution only from the dipoles of the polar molecules.

Thus at the beginning of the experiment, the measurements were made for the pairs of polar and non-polar molecules not only of similar molecular volume but also of similar molecular structure such as toluene—benzene derivatives (chlorobenzene, bromobenzene, nitrobenzene and benzonitrile), benzene—pyridine, *trans*-dichloroethylene—*cis*-dichloroethylene, methylcyclohexane—halocyclohexanes and cyclohexane—cyclohexanone systems. Although toluene is not strictly non-polar, its polar effect may be small enough to regard it as non-polar.

In the course of the experiment, anomalies were noticed in a group of the systems which contain aromatic compounds.

To make clear whether these anomalies come from the aromatic character of its polar component or of the non-polar component of the pairs of liquids, the experiment was extended to the pairs of aromatic non-polar—aliphatic polar as well as aliphatic non-polar—aromatic polar molecules.

TABLE II. BOILING POINT OF THE SAMPLES

Substance	B. p. (obs.), °C	B. p. (lit. value), °C
Chlorobenzene	131.5∼131.6	131.687
Bromobenzene	155.7∼155.8	155.908
Nitrobenzene	207.9∼208.1	210.80
Benzonitrile	188.7∼189.0	191.10
n-Butylchloride	78.2∼ 78.3	78.44
n-Butylcyanide	140.0∼140.8	142.25
Diethylketone	101.7∼101.8	101.70
Methyl propyl ketone	101.8	—
Cyclohexanone	155.2∼155.3	155.65
Chlorocyclohexane	—	
Bromocyclohexane	—	
Pyridine	115.0	115.58
cis-Dichloroethylene	60.3∼ 60.4	60.36
Cyclohexane	80.4	80.4
Toluene	110.3∼110.4	110.623
Benzene	80.1	80.103
Methlycyclohexane	100.8	—
1,4-Dioxane	101.2	101.32
trans-Dichloroethylene	47.3	47.67

Cyclohexanone, diethyl ketone, and methyl propyl ketone were selected to show the effect of molecular shape and that of the position of the dipole in the molecule.

Purification of the Samples. — Most samples were purified from the commercially available purest products according to the methods described in the literature[1] and were finally distilled under normal pressure, with a concentric vaccum jacketed column of about 40 theoretital plates.

Bromocyclohexane and chlorocyclohexane were kindly donated by Dr. M. L. Huggins of the Eastman Kodak Co. Both compounds decompose at their boiling point under normal pressure, so they were used without further purification.

In Table II the boiling points of the samples are shown with the values given in the literature. The thermometer used for the distillation was not corrected by a standard thermometer. Also the boiling points were not corrected for the atmospheric pressure.

Results and Discussion

The results of the heat of mixing for the 22 binary systems of polar and non-polor liquids are shown in Tables III and IV, in which the following notations are used.

v_2: Volume fraction of the polar liquids.

x_2: Mole fraction of the polar liquid.

w_2: Weight of the polar component taken for the measurement in grams.

w_1: Weight of the non-polar component taken for the measurement in grams.

$\Delta H_{\mathrm{m\ obs}}$: The observed heat of mixing in joule.

ΔH_{m}/cc.: The observed heat of mixing per unit volume of the mixture in joule.

H^{E}: The heat of mixing per mole of the mixture in joule.

α: The coefficient of the van Laar formula for heats of mixing of liquids. It is related to the heat of mixing by the relation

$$\Delta H_{\mathrm{m}} = \alpha \cdot V(1-v_2) \cdot v_2$$

where V is the total volume of the mixture.

The volues of α are plotted against v_2 in Figs. 1 and 2, ΔH_{m}/mol. is plotted against x_2 in Figs. 3 and 4.

For the systems in which the non-polar component is non-aromatic, the coefficient α can be expressed approximately by the following quadratic expressions except for nitrobenzene-cyclohexane systems. For the systems in which the non-polar component is aromatic, α cannot be expressed by a simple formula.

TABLE III. HEATS OF MIXING OF POLAR AND NON-AROMATIC NON-POLAR LIQUIDS

System	w_1	w_2	v_2	x_2	$\Delta H_{\mathrm{m\ obs}}$	ΔH_{m}/cc.	H^{E}	α
	0.126	1.154	0.866	0.873	3.221	2.660	274.1	22.86
Chlorobenzene	0.266	0.862	0.695	0.708	5.831	5.179	538.9	24.43
+	0.454	0.621	0.490	0.506	7.416	6.445	679.6	25.79
Cyclohexane	0.631	0.341	0.275	0.288	6.151	5.468	584.3	27.41
	0.898	0.150	0.105	0.111	3.508	2.705	292.3	28.76
	0.114	1.572	0.878	0.881	3.114	2.587	273.9	24.08
Bromobenzene	0.259	1.125	0.693	0.700	6.188	5.674	604.2	26.68
+	0.439	0.812	0.490	0.498	7.634	6.860	734.9	27.45
Cyclohexane	0.628	0.514	0.299	0.305	7.058	6.101	657.4	29.13
	0.756	0.135	0.085	0.087	2.688	2.518	273.1	32.38
	0.093	0.959	0.889	0.894	4.376	4.057	420.6	40.96
Benzonitrile	0.266	0.706	0.672	0.684	10.864	10.354	1085.6	47.00
+	0.446	0.544	0.485	0.499	14.330	12.800	1355.0	51.52
Cyclohexane	0.635	0.299	0.267	0.278	13.226	11.816	1266.2	60.37
	0.829	0.112	0.095	0.099	7.690	6.500	703.1	75.89
	0.116	1.221	0.872	0.879	7.143	6.112	625.6	54.64
Nitrobenzene	0.265	0.969	0.703	0.715	13.833	11.978	1266.7	58.71
+	0.416	0.625	0.493	0.510	15.497	14.632	1537.1	58.54
Cyclohexane	0.588	0.391	0.300	0.315	14.589	13.432	1430.0	63.92
	0.835	0.135	0.095	0.101	8.359	7.014	757.7	81.90

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[1] A. Weissberger and E. S. Porskauer, "Organic Solvent", Interscience Publishers Inc., New York (1955).

TABLE III. (Continued)

System	w_1	w_2	v_2	x_2	$\Delta H_{\mathrm{m\ obs}}$	ΔH_{m}/cc.	H^{E}	α
	0.114	0.879	0.871	0.875	2.240	1.956	206.2	17.45
n-Butylchloride	0.278	0.693	0.693	0.694	4.687	4.126	434.4	19.38
+	0.433	0.476	0.491	0.500	5.790	5.264	562.8	21.07
Cyclohexane	0.579	0.322	0.328	0.336	5.636	5.061	543.7	22.95
	0.828	0.100	0.096	0.099	2.658	2.246	243.4	25.90
	0.103	0.815	0.885	0.889	4.655	4.019	442.1	39.52
n-Butylcyanide	0.284	0.643	0.688	0.696	10.226	8.696	920.5	40.51
+	0.442	0.442	0.493	0.503	12.558	11.141	1188.2	44.57
Cyclohexane	0.642	0.246	0.272	0.279	11.841	10.396	1118.4	52.54
	0.844	0.107	0.110	0.114	7.028	5.747	621.1	58.64
	0.098	0.785	0.885	0.887	3.771	3.440	366.9	33.67
Diethylketone	0.145	0.261	0.632	0.638	4.135	8.110	869.9	34.90
+	0.265	0.263	0.487	0.492	6.498	9.738	1047.6	38.98
Cyclohexane	0.268	0.138	0.330	0.335	4.967	9.610	1037.6	43.47
	0.815	0.088	0.094	0.095	5.355	4.609	500.2	54.33
	0.097	0.820	0.890	0.892	4.047	3.556	379.2	36.29
Methyl propyl ketone	0.137	0.256	0.641	0.646	4.479	9.082	973.7	39.47
+	0.268	0.256	0.477	0.483	6.841	10.326	1111.2	41.39
Cyclohexane	0.256	0.142	0.346	0.351	5.050	9.951	1073.9	43.99
	0.826	0.098	0.102	0.104	6.113	5.144	558.1	56.84
	0.095	0.859	0.885	0.890	3.318	3.093	328.8	30.51
Cyclohexanone	0.271	0.725	0.687	0.696	7.800	6.965	735.3	32.41
+	0.421	0.503	0.495	0.506	9.479	8.794	935.9	35.18
Cyclohexane	0.629	0.302	0.283	0.292	9.233	8.147	875.9	40.16
	0.806	0.101	0.093	0.097	4.901	4.266	462.0	50.48
	0.103	1.019	0.884	0.891	1.264	1.089	131.1	10.62
Chlorocyclohexane	0.266	0.770	0.690	0.705	2.761	2.459	300.1	11.49
+	0.406	0.584	0.525	0.543	3.473	3.104	383.3	12.44
Methylcyclohexane	0.562	0.382	0.343	0.360	3.403	3.040	380.6	13.48
	0.730	0.144	0.132	0.140	1.922	1.749	222.2	15.26
	0.111	1.333	0.875	0.880	1.518	1.306	163.4	11.94
Bromocyclohexane	0.262	1.007	0.692	0.698	3.060	2.754	346.0	12.92
+	0.443	0.728	0.489	0.497	3.952	3.483	440.2	13.94
Methylcyclohexane	0.625	0.436	0.289	0.296	3.750	3.262	414.8	15.88
	0.822	0.166	0.105	0.108	1.880	1.565	200.2	16.60
	0.816	0.125	0.082	0.084	1.463	1.259	161.2	16.71
	0.141	1.178	0.891	0.893	0.580	0.559	42.63	5.75
cis-Dichloroethylene	0.413	0.897	0.679	0.684	1.458	1.411	108.2	6.47
+	0.694	0.667	0.484	0.490	2.001	1.850	142.5	7.41
trans-Dichloroethylene	0.929	0.391	0.291	0.296	1.883	1.787	128.8	8.65
	1.178	0.128	0.096	0.098	0.883	0.843	65.55	9.72
	0.130	0.992	0.889	0.895	0.217	0.190	15.48	1.93
Pyridine	0.182	0.321	0.650	0.663	0.250	0.495	40.79	2.17
+	0.343	0.329	0.502	0.517	0.418	0.625	51.96	2.50
1,4-Dioxane	0.360	0.157	0.315	0.327	0.357	0.609	58.79	3.24
	1.061	0.116	0.103	0.109	0.422	0.367	31.26	3.93
	0.096	0.857	0.889	0.891	2.342	2.095	224.3	21.25
Toluene	0.265	0.665	0.693	0.696	5.206	4.675	502.2	21.96
+	0.420	0.456	0.494	0.498	6.075	5.670	611.2	22.68
Cyclohexane	0.589	0.267	0.289	0.293	5.464	5.103	552.1	24.83
	0.823	0.110	0.107	0.109	2.796	2.348	254.8	24.55

TABLE IV. HEATS OF MIXING POLAR AND AROMATIC NON-POLAR LIQUIDS

System	w_1	w_2	v_2	x_2	$\Delta H_{\text{m obs.}}$	ΔH_{m}/cc.	H^{E}	α
	0.113	1.107	0.885	0.889	−0.497	−0.437	−44.9	−4.28
Chlorobenzene	0.300	0.799	0.674	0.686	−0.971	−0.904	−93.8	−4.12
+	0.475	0.584	0.491	0.502	−1.346	−1.245	−130.2	−4.98
Toluene	0.643	0.358	0.304	0.313	−1.107	−1.033	−108.9	−4.89
	0.828	0.104	0.090	0.093	−0.438	−0.415	−44.2	−5.09
	0.126	1.463	0.871	0.864	−0.400	−0.354	−37.1	−3.15
Bromobenzene	0.301	0.999	0.658	0.646	−0.776	−0.761	−78.8	−3.38
+	0.481	0.809	0.494	0.480	−0.959	−0.871	−89.4	−3.49
Toluene	0.684	0.474	0.287	0.276	−0.770	−0.693	−70.3	−3.39
	0.900	0.163	0.095	0.090	−0.369	−0.320	−31.9	−3.72
	0.102	1.050	0.899	0.902	0.088	0.076	7.8	0.83
Benzonitrile	0.305	0.758	0.682	0.689	0.080	0.072	7.5	0.33
+	0.481	0.534	0.489	0.498	0.006	0.005	0.5	0.02
Toluene	0.651	0.328	0.303	0.310	0.001_5	0.001	0.1_5	0.00_7
	0.898	0.114	0.099	0.102	0.118	0.104	10.9	1.15
	0.113	1.129	0.878	0.882	1.169	1.089	112.4	10.16
Nitrobenzene	0.289	0.870	0.684	0.693	2.106	1.985	206.4	9.19
+	0.488	0.632	0.482	0.492	2.442	2.234	234.1	8.95
Toluene	0.687	0.455	0.323	0.331	1.929	1.640	173.1	7.50
	0.871	0.106	0.081	0.084	0.849	0.841	82.3	11.35
	0.097	0.817	0.892	0.893	−0.604	−0.581	−61.2	−6.02
n-Butylchloride	0.161	0.291	0.639	0.643	−0.764	−1.477	−156.2	−6.40
+	0.298	0.282	0.481	0.485	−1.085	−1.629	−172.7	−6.54
Toluene	0.297	0.145	0.322	0.327	−0.780	−1.532	−162.8	−7.02
	0.868	0.090	0.092	0.094	−0.672	−0.603	−58.4	−7.21
	0.113	0.707	0.871	0.873	−0.614	−0.599	−63.1	−5.35
n-Butylcyanide	0.155	0.263	0.648	0.653	−0.682	−1.336	−140.8	−5.86
+	0.303	0.263	0.485	0.490	−1.113	−1.631	−172.5	−6.53
Toluene	0.290	0.136	0.337	0.342	−0.761	−1.498	−159.1	−6.71
	0.848	0.075	0.087	0.089	−0.378	−0.351	−37.4	−4.39
	0.094	0.873	0.895	0.897	−0.540	−0.521	−54.5	−5.54
Cyclohexanone	0.290	0.673	0.680	0.685	−1.277	−1.215	−127.6	−5.58
+	0.480	0.499	0.488	0.494	−1.603	−1.475	−155.7	−5.90
Toluene	0.641	0.312	0.308	0.314	−1.461	−1.359	−144.1	−6.38
	0.815	0.086	0.088	0.090	−0.373	−0.359	−38.3	−4.47
	0.114	0.974	0.884	0.894	−0.007	−0.006	−0.5	−0.06
Pyridine	0.301	0.702	0.676	0.697	−0.032	−0.030	−2.5	−0.14
+	0.502	0.532	0.486	0.511	0.007	0.006	0.5	0.03
Benzene	0.715	0.309	0.279	0.299	0.040	0.035	3.1	0.18
	0.918	0.113	0.099	0.108	0.023	0.020	1.7	0.22

Chlorobenzene—Cyclohexane:

$$\alpha=29.53-7.52_7v_2-0.073_7v_2^2$$

Bromobenzene—Cyclohexane:

$$\alpha=32.72-10.02v_2+0.405v_2^2$$

Benzonitrile—Cyclohexane:

$$\alpha=82.60-87.44v_2+46.88v_2^2$$

Nitrobenzene—Cyclohexane:

n-Butylchloride—Cyclohexane:

$$\alpha=27.12-13.50_4v_2+2.876v_2^2$$

n-Butylcyanide—Cyclohexane:

$$\alpha=64.48-54.01v_2+28.75v_2^2$$

Cyclohexanone—Cyclohexane:

$$\alpha=54.91-57.44v_2+34.31v_2^2$$

Diethyl ketone—Cyclohexane:

$$\alpha=59.73-60.62v_2+35.05v_2^2$$

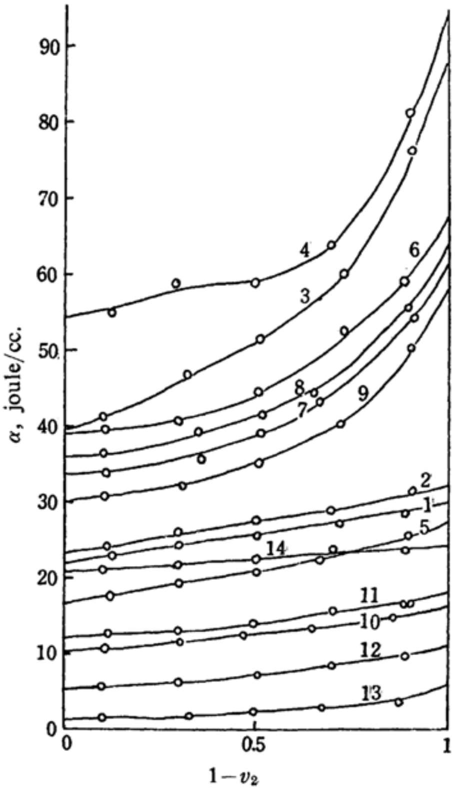

Fig. 1. The plots of van Laar coefficient α vs. volume fraction for the systems of group I.

1. Chlorobenzene—Cyclohexane
2. Bromobenzene—Cyclohexane
3. Benzonitrile—Cyclohexane
4. Nitrobenzene—Cyclohexane
5. *n*-Butylchloride—Cyclohexane
6. *n*-Butylcyanide—Cyclohexane
7. Diethyl ketone—Cyclohexane
8. Methyl propyl ketone—Cyclohexane
9. Cyclohexanone—Cyclohexane
10. Chlorocyclohexane—Methylcyclohexane
11. Bromocyclohexane—Methylcyclohexane
12. *cis*-Dichloroethylene—*trans*-Dichloroethylene
13. Pyridine—1, 4-Dioxane
14. Toluene—Cyclohexane

Methyl propyl ketone—Cyclohexane:

$$\alpha = 62.22 - 61.38 v_2 + 36.92 v_2^2$$

Chlorocyclohexane—Methylcyclohexane:

$$\alpha = 16.01_5 - 7.36_2 v_2 + 1.299 v_2^2$$

Bromocyclohexane—Methylcyclohexane:

$$\alpha = 17.50 - 6.61 v_2 + 0.396 v_2^2$$

cis-Dichloroethylene—

trans-Dichloroethylene:

$$\alpha = 10.438 - 7.172 v_2 + 2.094 v_2^2$$

Toluene—Cyclohexane:

$$\alpha = 24.78 - 3.971 v_2$$

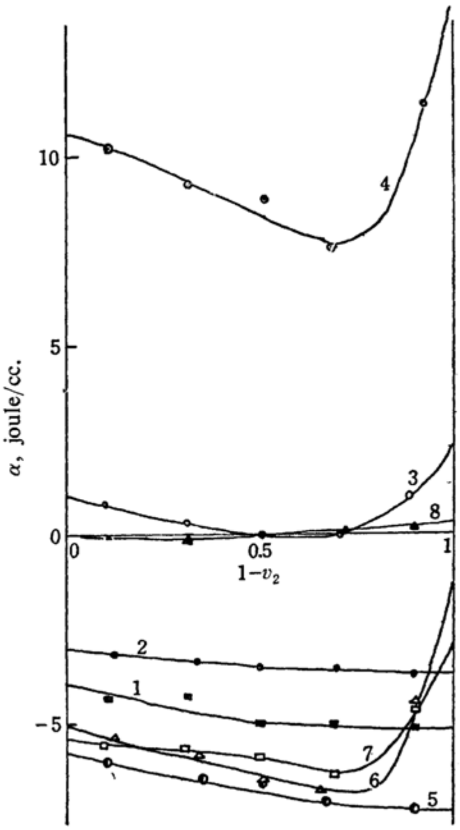

Fig. 2. The plots of van Laar coefficients α vs. volume fraction for the systems of group II.

1. Chlorobenzene—Toluene
2. Bromobenzene—Toluene
3. Benzonitrile—Toluene
4. Nitrobenzene—Toluene
5. *n*-Butylchloride—Toluene
6. *n*-Butylcyanide—Toluene
7. Cyclohexanone—Toluene
8. Pyridine—Benzene

Pyridine—1, 4-Dioxane:

$$\alpha = 4.92_9 - 7.42_5 v_2 + 4.69_2 v_2^2$$

It is obvious from the above figures that for the systems where non-aromatic molecules such as cyclohexane and methyl cyclohexane are selected for the non-polar component (denoted as group I hereafter) the values of α are positive and increase smoothly as the volume fractions of polar component v_2 decrease. It is in sharp contrast to the cases of mixing of two non-polar liquids where the values of α are nearly constant over the whole range of concentrations2).

The larger the dipole moment of the polar molecules the larger the variation of the values of α with concentration. This suggests that

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2) J. H. Hildbrand, "The Solubility of Nonelectrolytes", Reinhold Publishing Corporetion, New York (1950).

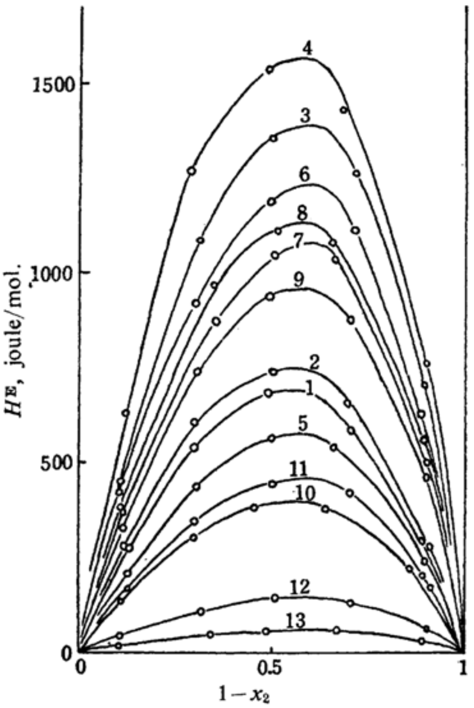

Fig. 3. The plots of H^E vs. mole fraction for the systems of group I.

1. Chlorobenzene—Cyclohexane
2. Bromobenzene—Cyclohexane
3. Benzonitrile—Cyclohexane
4. Nitrobenzene—Cyclohexane
5. *n*-Butylchloride—Cyclohexane
6. *n*-Butylcyanide—Cyclohexane
7. Diethyl ketone—Cyclohexane
8. Methyl propyl ketone—Cyclohexane
9. Cyclohexanone—Cyclohexane
10. Chlorocyclohexane—Methylcyclohexane
11. Bromocyclohexane—Methylcyclohexane
12. *cis*-Dichloroethylene—*trans*-Dichloroethylene
13. Pyridine—1,4-Dioxane

the change of the values of α with concentration originated from the effect of the dipoles of the polar constituents.

However, for the systems in which aromatic compounds such as toluene or benzene are the non-polar components (denoted as group II hereafter), most of the values of α decrease with the decrease of v_2 in most concentration ranges, and their variation seems to have no aparent relation with the dipole moment of the polar components.

Furthermore the values of α are much smaller than the corresponding systems of group I containing the same polar component and cyclohexane and are negative for most of these systems. This is a marked difference in comparison with group I.

It is difficult to interpret these differences by electrostatic energy only, for the dielectric constant of toluene does not differ so much from that of cyclohexane, and the electrostatic energy for both can also not be expected to differ greatly. It is concluded from the experimental results that the structural difference between aliphatic and aromatic non-polar liquids is responsible for these differences, for these anomalies are independent of whether the polar components are aromatic or not. This is in accordance with the fact that benzene shows an anomalous solvent effect in dielectric measurement.

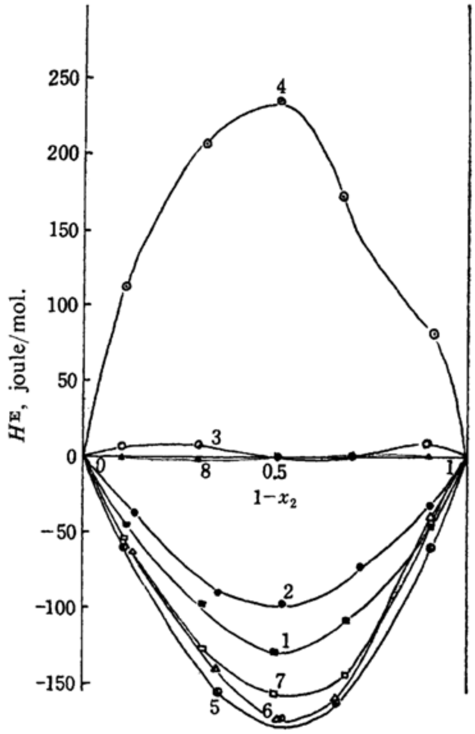

Fig. 4. The plots of H^E vs. mole fraction for the systems of group II.

1. Chlorobenzene—Toluene
2. Bromobenzene—Toluene
3. Benzonitrile—Toluene
4. Nitrobenzene—Toluene
5. *n*-Butylchloride—Toluene
6. *n*-Butylcyanide—Toluene
7. Cyclohexanone—Toluene
8. Pyridine—Benzene

As is seen from Fig. 2, for the systems of group II non-polar components, most of the values of α are negative, and if positive, these are small and do not change greatly with concentration as those systems in group I.

The effect of the dipole of the polar component, as is observed in the systems of group I, is also shown for such systems of group II as contain polar molecules of large dipole moment, in which the values of α begin to increase with the decrease of v_2 at low concentrations of polar molecules. This may be interpreted as follows: in the systems of group II two effects must be taken into account; the one is that governed by the magnitude of the dipole moment of the polar component, and is similar to that observed in the systems of group I and the other is a kind of stabilizing effect, specific, or coherent to the aromatic character of non-polar liquids such as toluene or benzene.

For *cis*-dichloroethylene—*trans*-dichloroethylene and pyridine—dioxane systems the values of α are smaller than that of the other group of the systems of polar-non-aromatic non-polar pairs. This may be due to the quadrupole of their components.

Summary

The heats of mixing were measured for 22 binary systems of polar and non-polar liquids where the pairs of liquids have similar molecular sizes.

For the systems in which the non-polar components are non-aromatic, the van Laar coefficients α are positive and increase smoothly as the concentration of polar liquids decrease. This variation of α with concentration is attributed to the effect of dipole of the polar component.

However, for the systems where the non-polar components are aromatic, the values of α are smaller than that of the corresponding non-aromatic system and are even negative for most of the cases and decrease with concentration of polar component in most concentration ranges.

The sharp difference between two groups of the binary systems, was attributed to the structural difference of aromatic and non-aromatic non-polar components.

The effect presumably due to the quadrupole of the non-polar components was noticed.

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