Thermodynamic Properties of Ternary Mixtures. 2. The Excess Volumes of Mixing of Ternary Mixtures of Cyclohexane, Aromatics, and Halomethanes

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The excess volumes of mixing of seven ternary mixtures, viz., (i) carbon tetrachloride (CCl_4) + benzene + *p*-xylene, (ii) cyclohexane + CCl_4 + toluene, (iii) cyclohexane + CCl_4 + *p*-xylene, (iv) cyclohexane + chloroform ($CHCl_3$) + toluene, (v) cyclohexane + $CHCl_3$ + *p*-xylene, (vi) cyclohexane + methylene dichloride (CH_2Cl_2) + toluene, and (vii) cyclohexane + CH_2Cl_2 + *p*-xylene, have been measured over a wide concentration range. The results have been fitted to an equation of the type $V^E_{123} = V^E_{12}{}^* + V^E_{23}{}^* + V^E_{31}{}^* + x_1x_2x_3[A + Bx_1(x_2 - x_3) + Cx_1^2(x_2 - x_3)^2]$ where V^E_{123} is the excess volume of mixing per mole of the ternary mixture in which mole fractions of components 1, 2, and 3 are x_1, x_2, and x_3, respectively. A, B, and C are constants which are characteristic of a ternary system and the quantities $V^E_{12}{}^*$, $V^E_{23}{}^*$, and $V^E_{31}{}^*$ are given by $V^E_{12}{}^* = x_1x_2[A_{12} + B_{12}(x_1 - x_2) + C_{12}(x_1 - x_2)^2]$, $V^E_{23}{}^* = x_2x_3[A_{23} + B_{23}(x_2 - x_3) + C_{23}(x_2 - x_3)^2]$, and $V^E_{31}{}^* = x_1x_3[A_{31} + B_{31}(x_1 - x_3) + C_{31}(x_1 - x_3)^2]$, where A_{12}, B_{12}, C_{12}, etc. are constants which are determined from the data on excess volumes of mixing for the binaries. The deviations, ΔV^E, of the experimental values of V^E_{123} from the appropriate quantities of the three binary systems have been calculated by using the relation $\Delta V^E = V^E_{123} - 1/2[(X_1 + X_2)V^E_{12} + (X_2 + X_3)V^E_{23} + (X_3 + X_1)V^E_{31}]$, where X_1, X_2, and X_3 refer to the number of moles of components 1, 2, and 3 in the ternary system, and the quantities V^E_{12} etc. refer to the excess volumes of mixing per mole of a binary mixture in which mole fraction of component 1 is $X_1/(X_1 + X_2)$ and of component 2 is $X_2/(X_1 + X_2)$ etc. The values of ΔV^E have been found to be positive in the case of all the ternary systems. It is suggested that this is due to weakening of halomethane–aromatic interaction by the third component.

Thermodynamic properties of ternary mixtures have been studied by very few workers (*1, 2, 5, 7*). Thermodynamic interpretation of vapor pressure of ternary liquid mixtures was reported by Srivastava and Rastogi (*7*) several years ago. Quite recently Rastogi, Nath, and Yadava (*3*) have reported the excess volume of mixing for two ternary systems, viz., (i) quinoline + CCl_4 + cyclohexane and (ii) benzene + CCl_4 + cyclohexane. The results were fitted to an analytical equation which could not be considered to be a general equation for ternary mixtures. In this paper the results have been fitted by a more general equation and the results are interpreted in terms of ternary interactions and the influence of one component on the binary interaction of the remaining components.

Experimental Section

(*a*) ***Purification of Materials.*** Benzene (A.R.), toluene (A.R.), and chloroform (A.R.) were subjected to further purification by the method described by Rastogi, Nath, and Misra (*4*), whereas carbon tetrachloride (A.R.) and methylene dichloride (G.R., S. Merck) were purified by the method described by Vogel (*9*). *p*-Xylene (Riedel) was purified by fractional crystallization. The sample thus obtained was subjected to fractional distillation. The spectroscopically pure sample of cyclohexane (S. Merck) was used without further purification. The densities of the purified components were found to be in good agreement with the values available in literature (*8*).

(*b*) ***Procedure.*** The excess volumes of mixing were determined by a three-limbed dilatometer shown in Figure 1. Known amounts of the three liquid components were filled over mercury in the limbs K, L, and M of the dilatometer with the help of a hypodermic syringe having a bent needle. The stopper S was then tightly inserted in the mouth of the side tube T. The dilatometer (mounted on a wooden stand) was placed in a water thermostat maintained at the required temperature to better than ±0.01 °C. After the contents of the dilatometer had attained the temperature of the thermostat, which was indicated by the constancy of the mercury level in the capillary D, the three liquids were mixed by repeatedly tilting the mixing cell. The volume change on mixing was estimated by noting the change in the height of the mercury level in the capillary before and after mixing by using a cathetometer which could read correct to ±0.001 cm. The measurements of V^E for binary mixtures were made with the dilatometer using only two limbs K and M. In the present design of the dilatometer, an error in the measurements could be on account of the compressibility of liquid mixtures due to varying pressure of the mercury column in the dilatometer. Calculations showed that the uncertainty due to this factor would be of the order of 1.0×10^{-5} cm^3 mol^{-1} which can be neglected, since the values of V^E are correct to ±0.002 cm^3 mol^{-1}. Also the error on account of dilatation of the vessel would be much less.

The accuracy of the dilatometer was tested using the method of Rastogi, Nath, and Yadava (*3*). The experimental results of V^E for the binary systems benzene + *p*-xylene, cyclohexane +

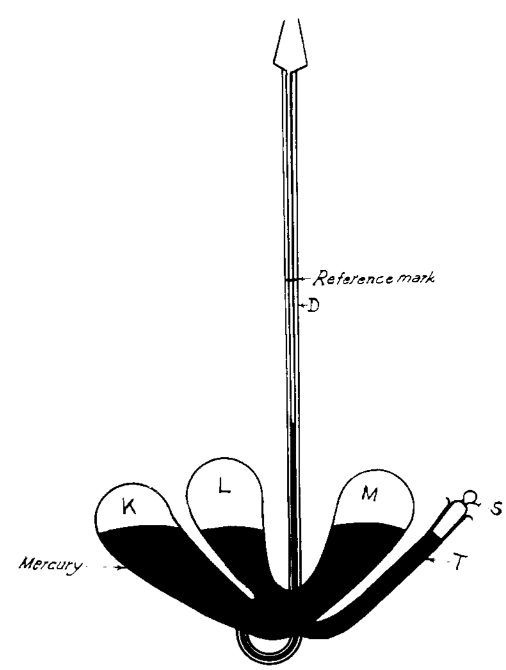

Figure 1. The dilatometer.

Table I. Excess Volumes of Mixing for the Various Binary Systems[a] at 30 °C

x	V^E, cm^3 mol^{-1}	x	V^E, cm^3 mol^{-1}	x	V^E, cm^3 mol^{-1}
Benzene + *p*-Xylene		CCl_4 + *p*-Xylene		Cyclohexane + *p*-Xylene	
0.2479	0.140	0.0773	−0.002	0.2198	0.359
0.3682	0.183	0.1132	−0.005	0.2747	0.417
0.3814	0.184	0.2710	−0.007	0.3383	0.494
0.4741	0.201	0.3610	−0.009	0.4262	0.557
0.5494	0.208	0.3908	−0.007	0.4268	0.566
0.5496	0.207	0.4595	−0.009	0.5230	0.619
0.6091	0.201	0.4970	−0.008	0.5972	0.618
0.6975	0.180	0.5336	−0.008	0.7436	0.514
0.8097	0.137	0.7084	−0.006	0.8049	0.455
0.8584	0.109	0.8429	−0.002	0.8293	0.442
		0.8554	−0.005	0.8363	0.406
Cyclohexane + Toluene		Cyclohexane + CH_2Cl_2		Cyclohexane + $CHCl_3$	
0.2737	0.390	0.1415	0.538	0.1313	0.235
0.3668	0.479	0.2128	0.642	0.1953	0.336
0.4667	0.543	0.3209	0.820	0.2733	0.423
0.5208	0.560	0.3734	0.885	0.4032	0.522
0.5226	0.567	0.4846	0.931	0.4747	0.535
0.5708	0.549	0 5050	0.935	0.5479	0.541
0.6945	0.492	0.5350	0.937	0.6039	0.526
0.8182	0.388	0.5910	0.917	0.6683	0.490
0.8529	0.312	0.6810	0.812	0.7469	0.427
		0.7868	0.623	0.8121	0.346
		0.8520	0.470	0.8401	0.309
		0.8763	0.422	0.8495	0.284
				0.8672	0.266
				0.8774	0.254
				0.9020	0.204

[a] x refers to the mole fraction of the first component.

Table II. Excess Volumes of Mixing for Ternary Systems at 30 °C

x_2	x_3	V^E_{123}, cm^3 mol^{-1}	V^E_{123}(a), cm^3 mol^{-1}	ΔV^E, cm^3 mol^{-1}	V^E_{123}(b), cm^3 mol^{-1}	ΔV^{E*}, cm^3 mol^{-1}	δ^a, cm^3 mol^{-1}
			CCl_4 (1) + Benzene (2) + *p*-Xylene (3)				
0.1063	0.6231	0.047	0.031	0.016	0.044	0.003	0.000
0.1562	0.5862	0.071	0.044	0.027	0.066	0.005	0.000
0.1802	0.5536	0.079	0.050	0.029	0.074	0.005	−0.001
0.2846	0.4613	0.115	0.069	0.046	0.103	0.012	0.004
0.2991	0.4487	0.114	0.071	0.043	0.106	0.008	0.000
0.3694	0.3779	0.121	0.077	0.044	0.115	0.006	−0.003
0.4310	0.3441	0.135	0.080	0.055	0.124	0.011	0.002
0.4644	0.2976	0.126	0.078	0.048	0.119	0.007	−0.002
0.5464	0.2146	0.112	0.068	0.044	0.104	0.008	0.000
0.6240	0.1701	0.106	0.060	0.046	0.097	0.009	0.003
0.6761	0.1508	0.096	0.056	0.040	0.093	0.003	−0.002
			Cyclohexane (1) + CCl_4 (2) + Toluene (3)				
0.0878	0.4110	0.497	0.274	0.223	0.486	0.011	0.000
0.0895	0.1387	0.313	0.171	0.142	0.310	0.003	0.000
0.0900	0.4078	0.496	0.273	0.223	0.484	0.012	0.001
0.1198	0.3197	0463	0.264	0.199	0.454	0.009	−0.001
0.1236	0.3200	0.460	0.263	0.197	0.452	0.008	−0.002
0.2369	0.4475	0.346	0.228	0.118	0.334	0.012	0.001
0.2984	0.4115	0.304	0.214	0.090	0.293	0.011	0.002
0.3547	0.3721	0.271	0.203	0.068	0.263	0.008	0.001
0.4517	0.1912	0.249	0.200	0.049	0.245	0.004	0.001
0.5940	0.2384	0.129	0.133	−0.004	0.128	0.001	−0.002
			Cyclohexane (1) + CCl_4 (2) + *p*-Xylene (3)				
0.0565	0.5567	0.512	0.275	0.237	0.504	0.008	0.000
0.1070	0.1852	0.432	0.242	0.190	0.421	0.011	0.000
0.1389	0.7009	0.256	0.155	0.101	0.245	0.011	−0.001
0.1608	0.6092	0.340	0.206	0.134	0.323	0.017	0.000
0.1753	0.1439	0.374	0.220	0.154	0.356	0.018	0.004
0.2057	0.3329	0.466	0.287	0.179	0.441	0.025	0.000
0.3211	0.2467	0.382	0.262	0.120	0.357	0.025	−0.004
0.4674	0.3034	0.252	0.197	0.055	0.227	0.025	−0.003
0.5491	0.0752	0.219	0.164	0.055	0.205	0.014	0.000
0.6233	0.1519	0.190	0.166	0.024	0.170	0.020	0.001

Table II (continued)

x_2	x_3	V^E_{123}, $cm^3\ mol^{-1}$	$V^E_{123}(a)$, $cm^3\ mol^{-1}$	ΔV^E, $cm^3\ mol^{-1}$	$V^E_{123}(b)$, $cm^3\ mol^{-1}$	ΔV^{E*}, $cm^3\ mol^{-1}$	δ^a, $cm^3\ mol^{-1}$
			Cyclohexane (1) + CH_2Cl_2 (2) + *p*-Xylene (3)				
0.1552	0.6191	0.450	0.388	0.062	0.502	−0.052	−0.001
0.2127	0.5004	0.566	0.485	0.081	0.636	−0.070	0.006
0.2670	0.4769	0.542	0.496	0.046	0.636	−0.094	−0.008
0.3310	0.4307	0.557	0.513	0.044	0.654	−0.097	−0.002
0.3596	0.3959	0.587	0.530	0.057	0.681	−0.094	0.006
0.4016	0.3640	0.583	0.534	0.049	0.684	−0.101	0.001
0.4207	0.3247	0.621	0.557	0.064	0.729	−0.108	−0.001
0.4566	0.2992	0.623	0.554	0.069	0.730	−0.107	−0.001
0.5423	0.2128	0.662	0.552	0.110	0.758	−0.096	0.002
0.6178	0.1577	0.656	0.517	0.139	0.736	−0.080	−0.001
			Cyclohexane (1) + $CHCl_3$ (2) + Toluene (3)				
0.1610	0.6575	0.295	0.240	0.055	0.316	−0.021	0.000
0.2523	0.5559	0.312	0.269	0.043	0.340	−0.028	0.005
0.2939	0.4461	0.382	0.335	0.047	0.433	−0.051	−0.006
0.3468	0.4478	0.321	0.298	0.023	0.372	−0.051	−0.006
0.3970	0.3736	0.361	0.321	0.040	0.405	−0.044	0.007
0.4893	0.2627	0.376	0.330	0.046	0.425	−0.049	0.003
0.5231	0.2568	0.341	0.307	0.034	0.392	−0.051	−0.002
0.5789	0.1817	0.367	0.308	0.059	0.410	−0.043	0.000
0.6103	0.1548	0.363	0.297	0.066	0.402	−0.039	0.001
0.6487	0.0543	0.435	0.278	0.157	0.453	−0.018	0.000
			Cyclohexane (1) + $CHCl_3$ (2) + *p*-Xylene (3)				
0.1646	0.5799	0.437	0.333	0.104	0.466	−0.029	0.002
0.1695	0.6366	0.359	0.286	0.073	0.390	−0.031	−0.004
0.2495	0.5591	0.380	0.313	0.067	0.411	−0.031	0.004
0.3297	0.4776	0.387	0.332	0.055	0.430	−0.043	−0.003
0.4605	0.3714	0.371	0.317	0.054	0.407	−0.036	0.000
0.6154	0.2054	0.374	0.304	0.070	0.399	−0.025	0.000
0.7199	0.0669	0.377	0.263	0.114	0.387	−0.010	−0.001
0.7479	0.0779	0.334	0.241	0.093	0.342	−0.008	0.002
			Cyclohexane (1) + CH_2Cl_2 (2) + Toluene (3)				
0.1358	0.6511	0.386	0.333	0.053	0.423	−0.037	−0.008
0.1996	0.5823	0.422	0.385	0.037	0.478	−0.056	−0.006
0.2924	0.4910	0.461	0.437	0.024	0.540	−0.079	0.001
0.3597	0.4185	0.499	0.471	0.028	0.590	−0.091	0.006
0.3833	0.3930	0.507	0.480	0.027	0.607	−0.100	0.003
0.4743	0.3087	0.541	0.492	0.049	0.641	−0.100	0.009
0.5316	0.2432	0.574	0.501	0.073	0.677	−0.103	0.004
0.6124	0.1643	0.596	0.485	0.111	0.694	−0.098	−0.011
0.6872	0.0909	0.634	0.448	0.186	0.701	−0.067	−0.011

[a] δ is the deviation in the experimental value of V^E_{123} from that calculated according to eq 2.

toluene, CCl_4 + *p*-xylene, cyclohexane + CH_2Cl_2, cyclohexane + *p*-xylene, and cyclohexane + $CHCl_3$ are recorded in Table I, and have been fitted by the method of least squares to the equations given in Table III. The experimental values of the excess volume of mixing for the various ternary systems are recorded in the third column of Table II.

Results and Discussion

The excess volumes of mixing, V^E_{123}, for a ternary mixture containing X_1, X_2, and X_3 moles of components 1, 2, and 3, respectively, may be expressed as

$$V^E_{123} = \frac{1}{2}\left[(X_1 + X_2)V^E_{12} + (X_2 + X_3)V^E_{23} + (X_3 + X_1)V^E_{31}\right] \quad (1)$$

where V^E_{12} is the excess volume of mixing per mole of a binary mixture in which mole fraction of component 1 is $X_1/(X_1 + X_2)$ and of component 2 is $X_2/(X_1 + X_2)$, V^E_{23} is the excess volume of mixing per mole of a binary mixture in which mole fraction of component 2 is $X_2/(X_2 + X_3)$ and of component 3 is $X_3/(X_2 + X_3)$, and V^E_{31} is the excess volume of mixing per mole of a binary mixture in which mole fraction of component 3 is $X_3/(X_3 + X_1)$ and of component 1 is $X_1/(X_3 + X_1)$. The values of V^E_{123} obtained from eq 1 by making use of the equations of the excess volumes of mixing for the various binary systems (see Table III) have been represented as $V^E_{123}(a)$ in fourth column of Table II. The deviations $\Delta V^E = V^E_{123} - V^E_{123}(a)$, in the experimental values of the excess volumes of mixing for the ternary systems from those of $V^E_{123}(a)$, are given in the fifth column of Table II.

The experimental results of V^E_{123} for the various ternary systems have been fitted by the method of least squares to the equation

$$V^E_{123} = V^E_{12}{}^* + V^E_{23}{}^* + V^E_{31}{}^* + x_1x_2x_3[A + Bx_1(x_2 - x_3) + Cx_1^2(x_2 - x_3)^2] \quad (2)$$

where x_1, x_2, and x_3 are the mole fractions of components 1, 2, and 3, respectively, in the ternary mixture and A, B, and C are constants for a ternary system. The quantities $V^E_{12}{}^*$, $V^E_{23}{}^*$, and $V^E_{31}{}^*$ are given by

$$V^E_{12}{}^* = x_1x_2\left[A_{12} + B_{12}(x_1 - x_2) + C_{12}(x_1 - x_2)^2\right] \quad (3)$$

$$V^E_{23}{}^* = x_2x_3\left[A_{23} + B_{23}(x_2 - x_3) + C_{23}(x_2 - x_3)^2\right] \quad (4)$$

$$V^E_{31}{}^* = x_1x_3\left[A_{31} + B_{31}(x_1 - x_3) + C_{31}(x_1 - x_3)^2\right] \quad (5)$$

where A_{12}, B_{12}, C_{12}, etc. are constants which are determined

Table III. Equations Fitting the Experimental Data of the Excess Volumes of Mixing for the Various Binary Systems at 30 °C

System	Equation fitting the data[g]: V^E ($cm^3\ mol^{-1}$)
Benzene + *p*-xylene[a]	$x_2x_3[0.8187 + 0.1226(x_2 - x_3) - 0.0379(x_2 - x_3)^2]$
Cyclohexane + toluene[a]	$x_1x_3[2.2044 + 0.4999(x_1 - x_3) - 0.0322(x_1 - x_3)^2]$
Cyclohexane + *p*-xylene[a]	$x_1x_3[2.3983 + 0.7161(x_1 - x_3) + 0.2693(x_1 - x_3)^2]$
CCl_4 + *p*-xylene[a]	$x_1x_3[-0.03224 + 0.0064(x_1 - x_3) - 0.00325(x_1 - x_3)^2]$
Cyclohexane + $CHCl_3$[a]	$x_1x_2[2.1704 + 0.1377(x_1 - x_2) + 0.05035(x_1 - x_2)^2]$
Cyclohexane + CH_2Cl_2[a]	$x_1x_2[3.7153 - 0.2605(x_1 - x_2) + 0.5771(x_1 - x_2)^2]$
CCl_4 + benzene[b]	$0.043x_1x_2$
Cyclohexane + CCl_4[c]	$0.638x_1x_2$
CCl_4 + toluene[d]	$x_2x_3[-0.1524 - 0.0204(x_2 - x_3) + 0.0243(x_2 - x_3)^2]$
$CHCl_3$ + toluene[e]	$x_2x_3[0.180 - 0.016(x_2 - x_3) - 0.040(x_2 - x_3)^2]$
$CHCl_3$ + *p*-xylene[e]	$x_2x_3[0.585 + 0.085(x_2 - x_3) - 0.165(x_2 - x_3)^2]$
CH_2Cl_2 + toluene[f]	$x_2x_3[0.653 + 0.320(x_2 - x_3) - 0.048(x_2 - x_3)^2]$
CH_2Cl_2 + *p*-xylene[f]	$x_2x_3[0.922 + 0.398(x_2 - x_3) + 0.020(x_2 - x_3)^2]$

[a] Equations based on the values of V^E obtained during this program. [b] Rastogi, R. P., Nath, J., *Indian J. Chem.*, **5**, 249 (1967). [c] See ref 3. [d] Rastogi, R. P., Nath, J., Misra, J., *J. Phys. Chem.*, **71**, 1277 (1967). [e] See ref 4. [f] Nigam, R. K., Mahl, B. S., *Indian J. Chem.*, **9**, 1250 (1971). [g] x_1, x_2, and x_3 refer to the mole fractions of cyclohexane, halomethanes, and aromatics, respectively, in the various binary systems except in the case of CCl_4 + benzene, CCl_4 + *p*-xylene, and benzene + *p*-xylene where x_1, x_2, and x_3 are the mole fractions of CCl_4, benzene, and *p*-xylene, respectively.

Table IV. Values of the Constants A, B, C, and σ at 30 °C

System	*A*, $cm^3\ mol^{-1}$	*B*, $cm^3\ mol^{-1}$	*C*, $cm^3\ mol^{-1}$	σ, $cm^3\ mol^{-1}$
CCl_4 + benzene + *p*-xylene	0.2652	0.3432	−1.9167	0.002
Cyclohexane + CCl_4 + toluene	0.196	−1.621	6.026	0.002
Cyclohexane + CCl_4 + *p*-xylene	0.832	0.688	−1.363	0.002
Cyclohexane + $CHCl_3$ + toluene	−1.479	−3.399	10.282	0.005
Cyclohexane + $CHCl_3$ + *p*-xylene	−1.284	1.386	13.045	0.003
Cyclohexane + CH_2Cl_2 + toluene	−3.078	10.829	28.721	0.009
Cyclohexane + CH_2Cl_2 + *p*-xylene	−2.931	−5.987	−6.288	0.005

from the data on excess volume of mixing for the binaries. The quantities $V^E_{12}{}^*$, $V^E_{23}{}^*$, and $V^E_{31}{}^*$ can be estimated by making use of the equations (see Table III) for the excess volumes of mixing for the three binaries. As for example, the quantities $V^E_{12}{}^*$, $V^E_{23}{}^*$, and $V^E_{31}{}^*$, in the case of the ternary system cyclohexane + $CHCl_3$ + toluene, as ascertained from the equations (see Table III) for the excess volumes of mixing for the three binary systems cyclohexane + $CHCl_3$, $CHCl_3$ + toluene and cyclohexane + toluene, are represented as

$$V^E_{12}{}^* = x_1x_2[2.1704 + 0.1377(x_1 - x_2) + 0.05035(x_1 - x_2)^2] \quad (6)$$

$$V^E_{23}{}^* = x_2x_3[0.180 - 0.016(x_2 - x_3) - 0.040(x_2 - x_3)^2] \quad (7)$$

$$V^E_{31}{}^* = x_1x_3[2.2044 + 0.4999(x_1 - x_3) - 0.0322(x_1 - x_3)^2] \quad (8)$$

The mole fractions x_1, x_2, and x_3 of components 1, 2, and 3, respectively, used to calculate $V^E_{12}{}^*$, $V^E_{23}{}^*$, and $V^E_{31}{}^*$ from eq 3–5 or eq 6–8 have been kept the same (see Table II) as those in the ternary mixtures. The values of $V^E_{12}{}^* + V^E_{23}{}^* + V^E_{31}{}^*$ for the various ternary systems are represented as V^E_{123}(b) in the sixth column of Table II. Accordingly the deviations, $\Delta V^{E*} = V^E_{123} - (V^E_{12}{}^* + V^E_{23}{}^* + V^E_{31}{}^*)$, of the experimental values of V^E_{123} from those of the sum, $V^E_{12}{}^* + V^E_{23}{}^* + V^E_{31}{}^*$, are given in the seventh column of Table II. The values of the constants *A*, *B*, and *C* along with the standard deviations, σ, in the experimental values of V^E_{123} from those obtained from eq 2 are given in Table IV. Table II shows that eq 2 fits the experimental values of V^E_{123} accurately in the case of the various ternary systems.

The ternary data on the eight systems studied by us show that ΔV^E is positive for (i) benzene + CCl_4 + cyclohexane (*3*), (ii) CCl_4 + benzene + *p*-xylene, (iii) cyclohexane + CCl_4 + toluene and (iv) cyclohexane + CCl_4 + *p*-xylene, (v) cyclohexane + $CHCl_3$ + toluene, (vi) cyclohexane + $CHCl_3$ + *p*-xylene, (vii) cyclohexane + CH_2Cl_2 + toluene, and (viii) cyclohexane + CH_2Cl_2 + *p*-xylene. This can be explained to be due to the weakening of donor–acceptor interaction between halomethanes and aromatics by cyclohexane, since the molecular charge distribution is bound to be affected by the surrounding molecules as is experimentally found to be true in many cases (*6*).

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