

# Study on Excess Molar Enthalpies and Excess Molar Volumes of the Binary Systems 1,2-Dichlorobenzene + (Benzene, Hexane, 1-Chlorohexane) and 1,3-Dichlorobenzene + (Benzene, Hexane, 1-Chlorohexane) at 298.15 K

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Excess molar enthalpies,  $H_m^E$ , and excess molar volumes,  $V_m^E$ , have been measured at  $T = 298.15$  K and normal atmospheric pressure for the binary 1,2-dichlorobenzene + (benzene, hexane, or 1-chlorohexane) and 1,3-dichlorobenzene + (benzene, hexane, or 1-chlorohexane) systems using a Calvet microcalorimeter and a vibrating tube densimeter, respectively. The experimental values of  $H_m^E$  and  $V_m^E$  of the mixtures are found to be positive or negative depending on the type and magnitude of the interactions involved in each process. The results are compared with those found in the literature.

## Introduction

This work gives information about the excess molar enthalpies and excess molar volumes of 1,2-dichlorobenzene + (benzene, hexane, or 1-chlorohexane) and 1,3-dichlorobenzene + (benzene, hexane, or 1-chlorohexane) mixtures at the temperature of 298.15 K and atmospheric pressure. The measurement of excess molar enthalpies was carried out using a Calvet microcalorimeter, and the excess molar volumes were determined from the densities of the pure liquids and mixtures by using an Anton Paar DMA 60/602 vibrating-tube densimeter. The experimental results are compared with those found in the literature for the excess molar enthalpies of 1,2-dichlorobenzene + benzene and 1,3-dichlorobenzene + benzene, as well as for the excess molar volumes of the same mixtures.<sup>1–3</sup>

## Experimental Section

1,2-Dichlorobenzene, 1,3-dichlorobenzene, hexane, and 1-chlorohexane were provided by Fluka (>0.99 mole fraction), and benzene was provided by Aldrich (>0.999 mole fraction). The products were subjected to no further purification other than being dried with Union Carbide 0.4 nm molecular sieves. Gas chromatographic analysis detected no significant peaks of impurities for any the components. The experimental densities, given in Table 1, are in close agreement with the literature values. The handling and disposal of the chemicals have been done according to the recommendations of the *CRC Handbook of Chemistry and Physics*.<sup>4</sup>

The experimental excess molar enthalpies were determined by Calvet microcalorimetry using the calibration and operating conditions described by Paz Andrade et al.<sup>5</sup> and

Table 1. Data for the Pure Liquids at 298.15 K

substance	$\rho/\text{g}\cdot\text{cm}^{-3}$	
	present work	literature
1,2-dichlorobenzene	1.2830	1.2828 <sup>a</sup>
1,3-dichlorobenzene	1.3003	1.3003 <sup>a</sup>
benzene	0.87345	0.87347 <sup>b</sup>
hexane	0.65528	0.6552 <sup>c</sup>
1-chlorohexane	0.87338	0.87333 <sup>d</sup>

<sup>a</sup> Reference 8. <sup>b</sup> Reference 22. <sup>c</sup> Reference 23. <sup>d</sup> Reference 24.

Paz Andrade.<sup>6</sup> This apparatus works with a calorimeter-cell volume of  $\sim 10$  cm<sup>3</sup>, and it is equipped with a device allowing operation in the absence of the vapor phase; a Philips PM2535 voltmeter and a data acquisition system are linked to the microcalorimeter. The uncertainty of the excess molar enthalpies is estimated as better than 1%.

Density measurements were carried out with an Anton-Paar DMA 60/602 vibrating-tube densimeter operating under static mode. The temperature inside the vibrating-tube cell was measured using an Anton Paar DT 100-30 digital thermometer and was regulated to better than  $\pm 0.01$  K using a Haake F3 thermostat. The uncertainty of the densities was  $\pm 2 \times 10^{-5}$  g·cm<sup>3</sup>. The apparatus was calibrated at atmospheric pressure before each series of measurements using bidistilled and degassed water and *n*-heptane; density data were taken from the literature for *n*-heptane<sup>7</sup> and for water.<sup>8</sup> Binary mixtures were prepared by weight using a Mettler H51 (precision,  $\pm 1 \times 10^{-5}$  g) balance, with a probable error in the mole fraction of  $< 10^{-4}$ . All molar quantities are based on the IUPAC relative atomic mass table.<sup>9</sup>

## Results and Discussion

Experimental values of  $H_m^E$  and  $V_m^E$  for all of the mixtures are listed in Tables 2 and 3, respectively. The experimental results for each mixture were fitted by the

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**Table 2. Excess Molar Enthalpies  $H_m^E$  at 298.15 K**

$x$	$H_m^E/\text{J}\cdot\text{mol}^{-1}$	$x$	$H_m^E/\text{J}\cdot\text{mol}^{-1}$	$x$	$H_m^E/\text{J}\cdot\text{mol}^{-1}$
1,2-Dichlorobenzene + Benzene					
0.0875	47.6	0.4569	154.4	0.8211	94.3
0.1296	69.9	0.4776	156.3	0.8693	71.5
0.1822	91.4	0.5582	153.1	0.9120	49.7
0.2476	113.6	0.6066	149.9		
0.2749	122.6	0.6506	139.7		
0.3346	138.3	0.6599	141.1		
0.3976	147.0	0.7405	118.0		
1,2-Dichlorobenzene + Hexane					
0.0709	220.9	0.4792	680.5	0.8332	389.9
0.1347	368.2	0.5271	679.6	0.8787	287.0
0.1807	456.2	0.6484	629.7	0.8957	259.8
0.2203	509.8	0.6936	587.9	0.9382	156.9
0.3259	632.4	0.7410	537.7		
0.3865	665.1	0.7755	484.6		
0.4253	674.5	0.8274	396.3		
1,2-Dichlorobenzene + 1-Chlorohexane					
0.0635	-19.4	0.4045	-67.6	0.7489	-49.3
0.0697	-21.5	0.4719	-71.4	0.7862	-43.0
0.1363	-35.8	0.5041	-71.1	0.8262	-35.0
0.1858	-46.5	0.5718	-70.4	0.8703	-26.2
0.2349	-52.9	0.6145	-67.6	0.9216	-15.8
0.2796	-58.5	0.6536	-65.3	0.9591	-8.6
0.3539	-65.1	0.6979	-59.1		
1,3-Dichlorobenzene + Benzene					
0.0482	49.1	0.3708	177.3	0.7369	118.1
0.0783	74.6	0.3835	175.6	0.7583	111.2
0.1124	97.3	0.4379	175.5	0.8402	79.2
0.1516	121.8	0.4924	171.7	0.8867	59.1
0.2025	143.9	0.5411	166.5	0.9289	39.1
0.2380	156.0	0.5935	156.1		
0.2835	166.0	0.6788	136.1		
1,3-Dichlorobenzene + Hexane					
0.0696	169.7	0.4352	579.7	0.7790	418.5
0.1228	273.4	0.4899	586.9	0.8164	363.0
0.1714	355.5	0.5472	582.8	0.8704	279.6
0.2110	412.7	0.5983	569.3	0.9090	199.9
0.2760	486.2	0.6479	539.8	0.9575	103.6
0.3241	530.3	0.6868	513.7		
0.3667	558.2	0.7453	450.3		
1,3-Dichlorobenzene + 1-Chlorohexane					
0.0602	-15.2	0.4436	-59.5	0.7942	-30.8
0.1218	-25.4	0.4853	-60.5	0.8313	-26.4
0.1883	-36.7	0.5402	-58.5	0.8771	-21.6
0.2166	-39.3	0.5868	-55.6	0.9095	-16.2
0.2766	-47.4	0.6430	-49.8	0.9535	-9.5
0.3349	-53.7	0.6887	-44.8		
0.3959	-58.6	0.7251	-40.1		

following variable-degree polynomials of the form

$$H_m^E/\text{J}\cdot\text{mol}^{-1} = x(1-x) \sum_{i=1}^n A_i(2x-1)^{i-1} \quad (1)$$

$$V_m^E/\text{cm}^3\cdot\text{mol}^{-1} = x(1-x) \sum_{i=1}^n A_i(2x-1)^{i-1} \quad (2)$$

Equations 1 and 2 are Redlich–Kister fitting polynomials,<sup>10</sup> and the parameters  $A_i$  have been obtained by a fitting computer program that uses the least-squares procedure and a Marquard algorithm.<sup>11</sup> The number of parameters used in eqs 1 and 2 for each mixture was determined by applying an  $F$  test.<sup>12</sup>

The parameters  $A_i$  and the standard deviations for  $H_m^E$  are given in Table 4. The same parameters for  $V_m^E$  are shown in Table 5. Plots of the experimental values with their corresponding smoothing curves for  $V_m^E$  and  $V_m^E$  are shown in Figures 1 and 2, respectively.

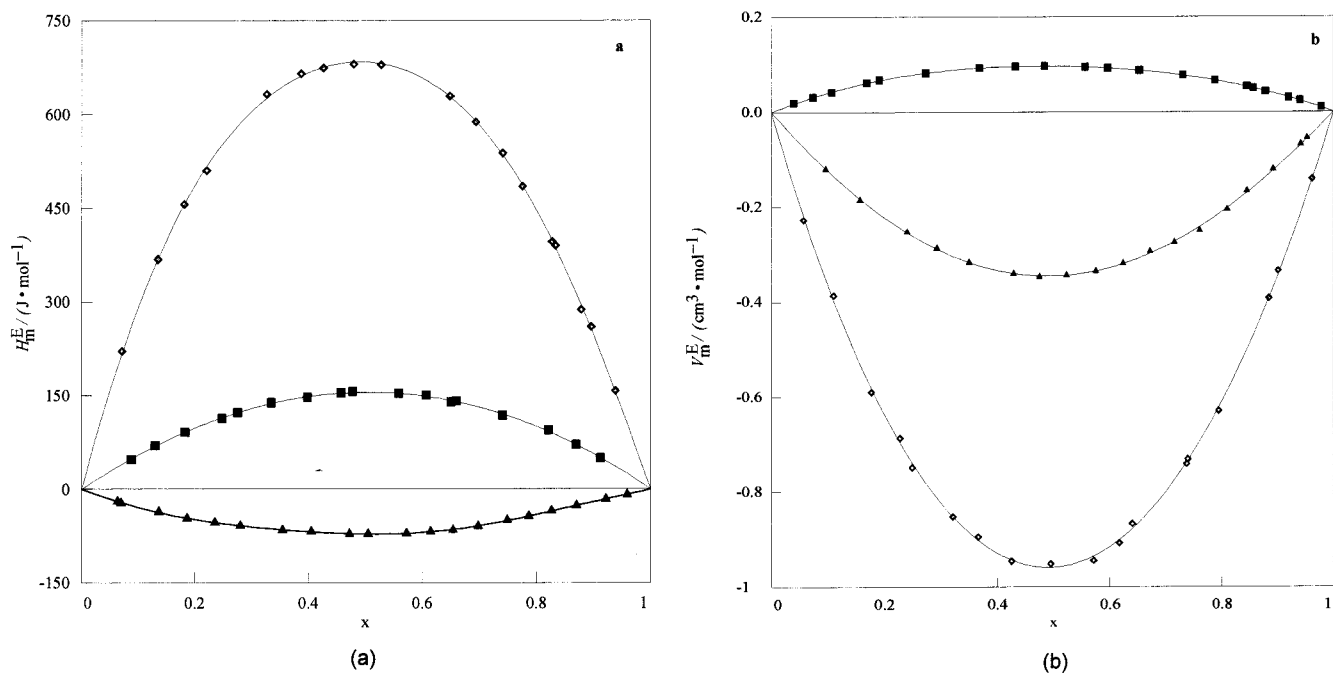
**Table 3. Excess Molar Volumes  $V_m^E$  at 298.15 K**

$x$	$V_m^E/\text{cm}^3\cdot\text{mol}^{-1}$	$x$	$V_m^E/\text{cm}^3\cdot\text{mol}^{-1}$	$x$	$V_m^E/\text{cm}^3\cdot\text{mol}^{-1}$
(x)1,2-Dichlorobenzene + (1-x)Benzene					
0.0398	0.0196	0.4323	0.0953	0.8449	0.0535
0.0739	0.0310	0.4839	0.0963	0.8565	0.0504
0.1066	0.0417	0.5562	0.0948	0.8781	0.0426
0.1688	0.0618	0.5964	0.0928	0.9196	0.0299
0.1910	0.0676	0.6528	0.0878	0.9400	0.0242
0.2731	0.0826	0.7302	0.0782	0.9779	0.0105
0.3681	0.0930	0.7877	0.0667		
(x)1,2-Dichlorobenzene + (1-x)Hexane					
0.0567	-0.2277	0.4247	-0.9457	0.7940	-0.6292
0.1097	-0.3861	0.4946	-0.9521	0.8841	-0.3925
0.1766	-0.5900	0.5709	-0.9445	0.9005	-0.3345
0.2274	-0.6869	0.6169	-0.9079	0.9618	-0.1414
0.2485	-0.7483	0.6400	-0.8668		
0.3208	-0.8519	0.7365	-0.7409		
0.3657	-0.8947	0.7387	-0.7313		
(x)1,2-Dichlorobenzene + (1-x)1-Chlorohexane					
0.0958	-0.1199	0.5227	-0.3428	0.8447	-0.1667
0.1564	-0.1855	0.5751	-0.3348	0.8916	-0.1212
0.2400	-0.2527	0.6235	-0.3188	0.9411	-0.0689
0.2928	-0.2867	0.6715	-0.2938	0.9523	-0.0552
0.3498	-0.3165	0.7150	-0.2745		
0.4286	-0.3400	0.7601	-0.2497		
0.4745	-0.3471	0.8093	-0.2055		
(x)1,3-Dichlorobenzene + (1-x)Benzene					
0.0438	0.0481	0.3654	0.1777	0.7292	0.1184
0.0631	0.0659	0.4111	0.1772	0.7885	0.0950
0.0980	0.0916	0.4863	0.1723	0.8566	0.0703
0.1611	0.1285	0.5398	0.1673	0.9137	0.0461
0.2045	0.1476	0.5911	0.1546	0.9738	0.0162
0.2736	0.1710	0.6359	0.1457	0.8566	0.0703
0.3141	0.1750	0.7061	0.1230	0.9137	0.0461
(x)1,3-Dichlorobenzene + (1-x)Hexane					
0.0029	-0.0125	0.4366	-0.7862	0.8007	-0.4924
0.1167	-0.3486	0.4931	-0.7893	0.8225	-0.4554
0.1725	-0.4852	0.5413	-0.7916	0.8493	-0.3923
0.2330	-0.5859	0.5961	-0.7662	0.9202	-0.2150
0.2853	-0.6631	0.6422	-0.7156	0.9665	-0.0902
0.3311	-0.7301	0.6913	-0.6698		
0.3830	-0.7622	0.7590	-0.5633		
(x)1,3-Dichlorobenzene + (1-x)1-Chlorohexane					
0.0921	-0.0841	0.5348	-0.2403	0.8505	-0.1204
0.1526	-0.1280	0.5784	-0.2344	0.8998	-0.0874
0.2436	-0.1833	0.6256	-0.2244	0.9275	-0.0663
0.2932	-0.2075	0.6795	-0.2124	0.9544	-0.0406
0.3470	-0.2217	0.7220	-0.1963		
0.4267	-0.2438	0.7676	-0.1747		
0.4772	-0.2473	0.8110	-0.1470		

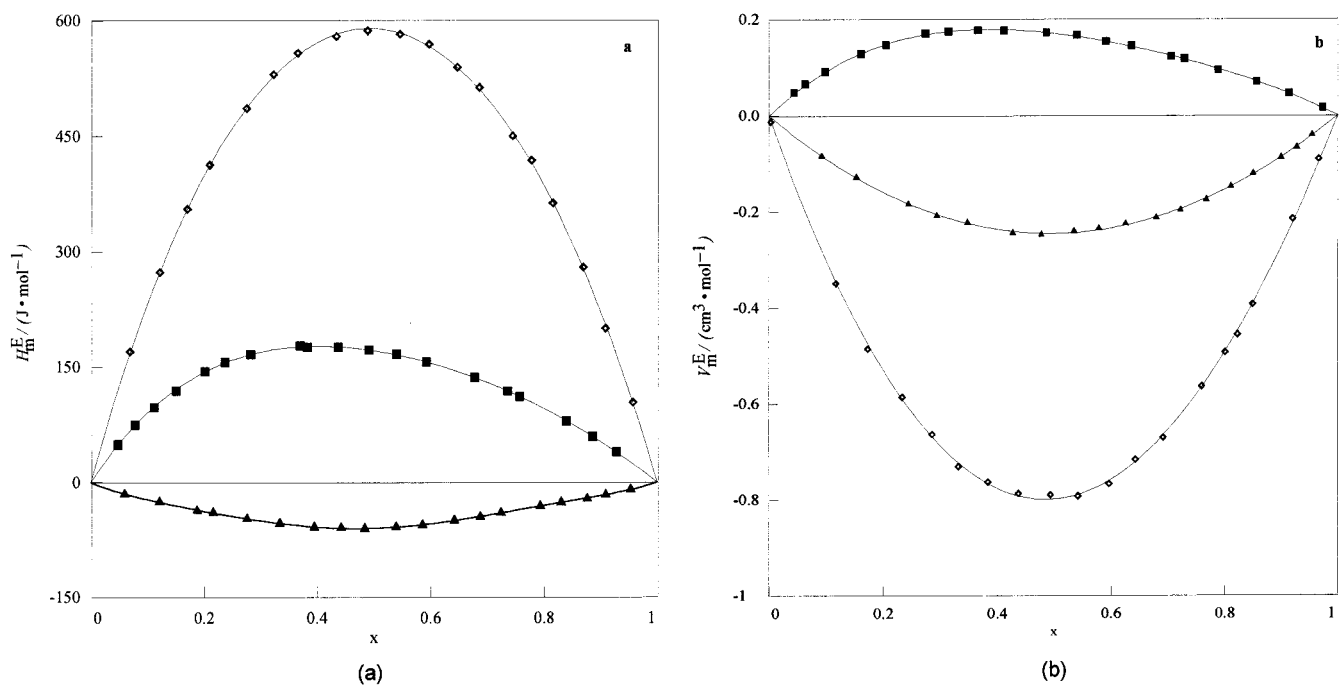
**Table 4. Fitting Parameters,  $A_i$ , for Equation 1 and Standard Deviations,  $s$ , for  $H_m^E$** 

$A_1$	$A_2$	$A_3$	$A_4$	$A_5$	$A_6$	$s$
1,2-Dichlorobenzene + Benzene						
620.7	10.9					1.3
1,2-Dichlorobenzene + Hexane						
2736.7	-80.0	632.3	-363.5	-373.1		4.0
1,2-Dichlorobenzene + 1-Chlorohexane						
-286.0	-22.3	24.0	237.3		-168.6	0.6
1,3-Dichlorobenzene + Benzene						
684.4	-226.0	171.6	-52.2			0.6
1,3-Dichlorobenzene + Hexane						
2359.5	-56.2	256.3				2.7
1,3-Dichlorobenzene + 1-Chlorohexane						
-240.2	44.0	141.0	-19.6	-178.5		0.5

The discussion of the experimental results will be on the basis of the positive and negative contributions to  $H_m^E$  and  $V_m^E$  due to the rupture or the formation of molecular interactions during the mixing process. Most of the com-



**Figure 1.** (a) Experimental excess molar enthalpies,  $H_m^E$ , and (b) experimental excess molar volumes,  $V_m^E$ , at 298.15 K of (x)1,2-dichlorobenzene + (1 - x)(benzene, hexane, 1-chlorohexane): ■, benzene; ◇, hexane; ▲, 1-chlorohexane. Values calculated from eqs 1 and 2 are shown by the straight line.



**Figure 2.** (a) Experimental excess molar enthalpies,  $H_m^E$ , and (b) experimental excess molar volumes,  $V_m^E$ , at 298.15 K of (x)1,3-dichlorobenzene + (1 - x)(benzene, hexane, 1-chlorohexane): ■, benzene; ◇, hexane; ▲, 1-chlorohexane. Values calculated from eqs 1 and 2 are shown by the straight line.

ponents used in the systems described in this work are believed to have quite strong interactions in their pure states. This is the case with benzene, which may present weak pairwise interactions, described in previous works by Hobza et al.<sup>13</sup>

For 1,2-dichlorobenzene + benzene,  $H_m^E$  and  $V_m^E$  are positive over the whole composition range, which indicates a predominance in the rupture of previous interactions over the formation of new interactions during the mixing process. The same facts occur for 1,3-dichlorobenzene + benzene, and in this case both magnitudes are more

positive compared to those of 1,2-dichlorobenzene + benzene. The greater dipole moment of 1,2-dichlorobenzene makes more important the dipole-dipole interaction and lowers the maximum values of  $H_m^E$  and  $V_m^E$  compared to those of 1,3-dichlorobenzene. The experimental values of  $H_m^E$  and  $V_m^E$  for these mixtures are in close agreement with the literature values.<sup>1-3</sup>

Tanaka et al.<sup>1</sup> and Tanaka and Benson<sup>2</sup> measured  $H_m^E$  for (x)1,2-dichlorobenzene + (1 - x)benzene at 298.15 K and obtained  $H_m^E(x = 0.5) = 163.4 \text{ J} \cdot \text{mol}^{-1}$ , ~5% higher

**Table 5. Fitting Parameters,  $A_i$ , for Equation 2 and Standard Deviations,  $s$ , for  $V_m^E$** 

$A_1$	$A_2$	$A_3$	$s$
0.3869	1,2-Dichlorobenzene + Benzene −0.0252	0.0765	0.0009
−3.8357	1,2-Dichlorobenzene + Hexane −0.1475	−0.1659	0.0078
−1.3814	1,2-Dichlorobenzene + 1-Chlorohexane 0.0614	0.0800	0.0030
0.6836	1,3-Dichlorobenzene + Benzene −0.2819	0.1876	0.0017
−3.1932	1,3-Dichlorobenzene + Hexane 0.1997		0.0061
−0.9795	1,3-Dichlorobenzene + 1-Chlorohexane 0.0276		0.0023

than ours,  $H_m^E(x = 0.5) = 155.2 \text{ J}\cdot\text{mol}^{-1}$ . Tanaka et al.<sup>1</sup> and Tanaka and Benson<sup>3</sup> measured  $H_m^E$  for (x)1,3-dichlorobenzene + (1 - x)benzene at 298.15 K; their value of  $H_m^E(x = 0.5) = 171.4 \text{ J}\cdot\text{mol}^{-1}$  agrees satisfactorily with ours,  $H_m^E(x = 0.5) = 171.1 \text{ J}\cdot\text{mol}^{-1}$ . Tanaka and Benson<sup>2</sup> measured  $V_m^E$  for (x)1,2-dichlorobenzene + (1 - x)benzene at 298.15 K and obtained  $V_m^E(x = 0.5) = 0.1004 \text{ cm}^3\cdot\text{mol}^{-1}$ , ~4% higher than ours,  $V_m^E(x = 0.5) = 0.0967 \text{ cm}^3\cdot\text{mol}^{-1}$ . Finally, Tanaka and Benson<sup>3</sup> obtained  $V_m^E(x = 0.5) = 0.1843 \text{ cm}^3\cdot\text{mol}^{-1}$  for (x)1,3-dichlorobenzene + (1 - x)benzene at 298.15 K, ~7% higher than ours,  $V_m^E(x = 0.5) = 0.1709 \text{ cm}^3\cdot\text{mol}^{-1}$ .

Some  $H_m^E$  and  $V_m^E$  data have been published in the literature for 1,2-dichlorobenzene + benzene and 1,3-dichlorobenzene + benzene at different temperatures. Turner et al.<sup>14</sup> reported measurements of  $H_m^E$  for the system 1,1,1-trichloroethane + benzene + 1,2-dichlorobenzene at  $T = 293.15 \text{ K}$ . Mahl et al.<sup>15</sup> measured  $H_m^E$  for 1,2-dichlorobenzene + benzene at 308.15 K. Otin et al.<sup>16</sup> determined excess enthalpies of 1,2-dichlorobenzene, 1,3-dichlorobenzene + benzene at 303.15 K, and Dhillon<sup>17</sup> published excess volumes of 1,2-dichlorobenzene + benzene at 303.15 K and 308.15 K.

$H_m^E$  values for 1,2-dichlorobenzene + hexane and 1,3-dichlorobenzene + hexane are positive for all of the compositions, and the maximum values are slightly higher for 1,2-dichlorobenzene + hexane. A different situation is found for  $V_m^E$ . The experimental values are negative over the whole range of composition and agree with those of some authors who have studied excess properties of several binary mixtures containing polar–nonpolar pairs<sup>18</sup> and found that the nonideality of the mixtures increases with the dipole moment of the polar component. In our case, 1,2-dichlorobenzene, which has a greater dipole moment than 1,3-dichlorobenzene, will produce a higher deviation of ideality, and the values of  $V_m^E$  are more negative for the mixture containing 1,2-dichlorobenzene.

The experimental results obtained for 1,2-dichlorobenzene + 1-chlorohexane and 1,3-dichlorobenzene + 1-chlorohexane are quite interesting. Both  $H_m^E$  and  $V_m^E$  are negative for all compositions, most probably because the dipole–dipole interactions are very important in this case, and they produce a negative contribution to  $H_m^E$  and  $V_m^E$ . The experimental values of  $H_m^E$  and  $V_m^E$  are slightly more negative for the mixtures containing 1,2-dichlorobenzene.

As far as we know, there are no previous measurements on these mixtures. We found only a few data sources for the other systems:  $H_m^E$  for chlorobenzene, 1,2-dichloroben-

zene, 1,3-dichlorobenzene + *n*-alkane ( $n = 7, 9, 14$ ) at 298.15 K (Wilhelm et al.<sup>19</sup>),  $H_m^E$  for 1,2,4-trichlorobenzene + *n*-alkane ( $n = 6, 7, 9, 10, 14$ ) (Wilhelm<sup>20</sup>), and  $V_m^E$  for 1,2-dichlorobenzene, 1,3-dichlorobenzene + *n*-octane at 298.15 K (López et al.<sup>21</sup>).

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