

# Thermodynamics of Liquid (Xenon + Diborane)

Luís F. G. Martins, Eduardo J. M. Filipe, and Jorge C. G. Calado\*

Centro de Química Estrutural, Instituto Superior Técnico, 1049–001 Lisboa, Portugal

Received: November 13, 2001

The total vapor pressure of liquid mixtures of xenon and diborane has been measured at 161.40 K (the triple point of xenon) and at 182.34 K (the triple point of dinitrogen oxide), as a function of composition. The liquid densities were also measured at 182.34 K. The mixtures show positive deviations from Raoult's law. Both the excess molar Gibbs energy ( $G_m^E$ ) and the excess molar volume ( $V_m^E$ ) are positive. For the equimolar mixture  $G_m^E = 118.8 \text{ J}\cdot\text{mol}^{-1}$  at 161.40 K,  $G_m^E = 91.7 \text{ J}\cdot\text{mol}^{-1}$  at 182.34 K and  $V_m^E = 0.248 \text{ cm}^3\cdot\text{mol}^{-1}$  at 182.34 K. The estimated value of the excess molar enthalpy ( $H_m^E$ ) was found to be  $328 \text{ J}\cdot\text{mol}^{-1}$ . The results were interpreted using the Deiters equation of state. This is the first thermodynamic study of the xenon + diborane system.

## 1. Introduction

The excess thermodynamic properties of mixtures of condensed gases have long been a preferred testing ground of statistical theories of solutions. Thanks to those systems, much has been learned on the effects of molecular size, shape, polarity, and so forth on the overall architecture of phase diagrams. In the 1980s and 1990s, a lot of attention was focused on bicentric, quadrupolar molecules such as ethane and ethylene, which have quadrupole moments of opposite signs. Interesting features, such as negative azeotropy, could sometimes be attributed to the strong mutual attraction between quadrupoles of opposite sign. A case in point is that of the (carbon dioxide + acetylene) system.<sup>1</sup> Shape effects were usually assessed by comparisons with spherical molecules, xenon being a common choice due to its relatively high polarizability (which enhances dispersion forces) and wide liquid range. Nevertheless, the thermodynamic behavior of a mixture is ultimately determined by the various intermolecular forces, to which all the above-mentioned features—shape, polarity, and so forth—contribute.

Among the more interesting systems of the time were the (xenon + ethane) and (xenon + ethylene) mixtures, for they showed very different behaviors. Whereas all four main excess functions,  $G_m^E$ ,  $V_m^E$ ,  $H_m^E$ , and  $S_m^E$  are negative for (xenon + ethane),<sup>2</sup> they are positive for (xenon + ethylene).<sup>3</sup> These results were recently interpreted<sup>2</sup> using the statistical associating fluid theory for potentials of variable range (SAFT-VR) of Gil-Vilegas et al.<sup>4,5</sup>

In the present work, we describe a thermodynamic study of mixtures of xenon and diborane. The molecular structure of diborane resembles, in many ways, that of ethylene (Figure 1), although it is slightly bigger (the B–B distance is 0.177 nm, compared to the C–C distance 0.154 nm in ethane or 0.134 nm in ethylene). In the diborane molecule, the two boron atoms and four of the hydrogen atoms lie on the same plane, in a geometrical arrangement very similar to that of ethylene. The main difference comes from the presence of two extra hydrogen atoms, which form a doubly bridged structure between the two boron atoms. Located in a similar region to that of the  $\pi$ -orbital in ethylene, these hydrogen atoms are electron deficient because they are bonded to both boron atoms. This feature brings an

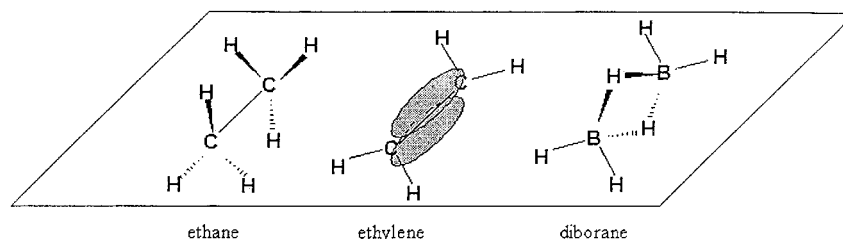
extra interest to the study of (xenon + diborane), the potential for an incipient association between the two components because xenon can be considered an extremely weak Lewis base.

Vapor–liquid equilibria and density measurements were carried out at 161.40 and 182.34 K. The corresponding excess properties, the excess Gibbs energy ( $G_m^E$ ) and the excess volume ( $V_m^E$ ), were calculated from the experimental data and the excess molar enthalpy ( $H_m^E$ ) estimated from the temperature dependence of ( $G_m^E$ ). The results were interpreted using the Deiters equation of state (DEOS).<sup>6</sup> As far as we are aware, this is the first thermodynamic study of this mixture.

## 2. Experimental Section

The apparatus and techniques for the vapor pressure and density measurements have been described elsewhere.<sup>7</sup> A triple point cryostat was used, the working temperatures being 161.40 K (the triple point of xenon) and 182.34 K (the triple point of nitrous oxide). The mixtures were prepared by condensing known amounts of each component into a calibrated pycnometer. The vapor pressures were measured using a quartz-spiral gauge (Texas Instruments, model PPG 149) with 500 kPa full range and a resolution of 2 Pa, which had been calibrated against a Budenberg dead weight gauge. The pycnometer used for the density determinations was calibrated at 182.34 K with liquid ethylene, using the data of McCarty et al.<sup>8</sup> ( $V_{\text{pyk}} = (2.2367 \pm 0.0003) \text{ cm}^3$ ). The amount of substance of each component was calculated from  $pVT$  measurements. Pressures were measured using another quartz bourdon manometer (Texas Instruments, model 145) with 130 kPa full range and a resolution of 0.5 Pa, which had been calibrated against mercury manometers.

Xenon (from Linde, 99.99% purity) and nitrous oxide (from Air Liquide, 99.9% purity) were further purified by fractionation in a low-temperature column. The final purity of the samples was checked by measuring the constancy of the triple point during melting. The values obtained were the following: for xenon,  $(81.669 \pm 0.02) \text{ kPa}$ , to be compared with the recommended value of  $(81.674 \pm 0.011) \text{ kPa}$ ;<sup>9</sup> for nitrous oxide,  $(87.869 \pm 0.008) \text{ kPa}$ , to be compared with  $(87.865 \pm 0.012) \text{ kPa}$ .<sup>9</sup> The purity of xenon can also be assessed from the comparison between the measured values of its vapor pressure



**Figure 1.** Comparing the ethane, ethylene, and diborane molecular structures.

and molar volume and the recommended values in the literature at the working temperatures. At 182.34 K, the vapor pressure of xenon was found to be 248.19 kPa, which compares favorably with 248.24 kPa,<sup>10</sup> whereas the average value of six determinations obtained for the molar volume was  $(46.480 \pm 0.009) \text{ cm}^3 \text{ mol}^{-1}$  (to be compared with a literature value<sup>11</sup> of  $46.485 \text{ cm}^3 \text{ mol}^{-1}$ ).

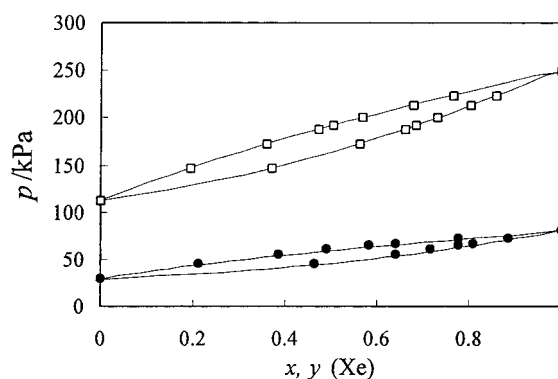
Diborane was synthesized through the reaction of sodium borohydride with iodine in diglyme solution, following the procedure of Freeguard and Long.<sup>12</sup> A 24 g portion of sodium borohydride (from Riedel de Häen, 95% purity) was placed in a three-necked flask fitted with an inlet tube, a solid reagent addition bulb, and an outlet tube leading to an oil pump via a series of traps. These were cooled in liquid nitrogen, except the first one, which was cooled in an acetone/dry ice bath to remove traces of volatilized solvent. A 500  $\text{cm}^3$  portion of previously dried and distilled diglyme (from Riedel de Häen, 99% purity) was then added to the flask, the system swept with dry nitrogen and the pressure reduced to near the solvent vapor pressure. A 80.6 g portion of solid iodine was slowly added to the solution over a period of 2 h. Pressure was kept below 2600 Pa by means of a valve connected to the oil vacuum pump. Pumping was continued for 45 min after the final addition to ensure complete transference of the diborane to the liquid nitrogen traps. Diborane was purified by fractionation in a low-temperature column and the purity checked by measuring its vapor pressure at the working temperatures. At 161.40 K, the vapor pressure of diborane was found to be  $(30.28 \pm 0.06) \text{ kPa}$  (which compares favorably with  $30.22 \text{ kPa}$ <sup>13</sup>) and at 182.34 K,  $(111.89 \pm 0.03) \text{ kPa}$  (against  $111.83 \text{ kPa}$ <sup>14</sup>). Due to its chemical instability, diborane had to be stored at liquid nitrogen temperatures.

With our apparatus and technique, only the overall composition of the mixture is directly measured. To calculate the individual compositions of the liquid and vapor phases, some ancillary data are needed. As usual, the nonideality of the vapor phase was taken into account in the calculations of the phase compositions. Because the involved pressures stay below 300 kPa, second virial coefficients are sufficient. For xenon, at room temperature, they were taken from Dymond and Smith<sup>15</sup> ( $B = -130.2 \text{ cm}^3 \text{ mol}^{-1}$  at 298.15 K) and for diborane from Daubert et al.<sup>16</sup> ( $B = -191 \text{ cm}^3 \text{ mol}^{-1}$  at 298.15 K). At low temperatures, second virial coefficient data are much scarcer. In the case of xenon, the values were taken from Brewer<sup>17</sup> ( $B = -424 \text{ cm}^3 \text{ mol}^{-1}$  at 161.40 K and  $B = -330 \text{ cm}^3 \text{ mol}^{-1}$  at 182.34 K); as for diborane because there are no reported measurements, they were estimated from the correlation of Leland and Chappelaar,<sup>18</sup> with the following results,  $B = -775 \text{ cm}^3 \text{ mol}^{-1}$  at 161.40 K and  $B = -561 \text{ cm}^3 \text{ mol}^{-1}$  at 182.34 K. Cross virial coefficients were estimated using the correlation of Van Ness and Abbott, as discussed by Prausnitz et al.<sup>19</sup> ( $B_{12} = -550 \text{ cm}^3 \text{ mol}^{-1}$  at 161.40 K,  $B_{12} = -430 \text{ cm}^3 \text{ mol}^{-1}$  at 182.34 K and  $B_{12} = -158 \text{ cm}^3 \text{ mol}^{-1}$  at 298.15 K).

**TABLE 1: Total Vapor Pressure,  $p$ , and Excess Molar Gibbs Energy,  $G_m^E$ , of Xenon + Diborane Mixtures at 161.40 and 182.34 K<sup>a</sup>**

$x$	$y$	$p/\text{kPa}$	$\delta p/\text{kPa}$	$G_m^E/(\text{J} \cdot \text{mol}^{-1})$
161.40 K				
0	0	30.28	-	0
0.21264	0.46451	45.82	-0.028	82.70
0.38654	0.64094	55.61	+0.11	115.66
0.49025	0.71628	60.44	-0.12	117.06
0.58286	0.77559	64.73	-0.054	114.20
0.64016	0.80943	67.37	+0.078	110.16
0.77673	0.88312	72.98	+0.0091	82.66
1	1	81.67	-	0
182.34 K				
0	0	111.89	-	0
0.19342	0.37180	146.72	-0.042	5.93
0.35974	0.56311	172.56	+0.089	11.79
0.47223	0.66053	188.28	+0.31	17.88
0.50486	0.68426	191.65	-0.52	20.86
0.56834	0.73124	200.31	+0.11	28.76
0.67830	0.80419	213.32	-0.0079	32.45
0.76628	0.85898	223.36	+0.056	26.25
1	1	248.19	0	0

<sup>a</sup> The variables  $x$  and  $y$  are the liquid and the vapor phase mole fractions of xenon, respectively.  $\delta p$  values are the pressure residuals.



**Figure 2.** Vapor-liquid equilibrium for xenon + diborane mixtures at: ●, 161.40 K; □, 182.34 K. The lines are the predictions by the DEOS.

### 3. Results

The total vapor pressures,  $p$ , for xenon + diborane mixtures at 161.40 and 182.34 K are recorded in Table 1 and plotted in Figure 2 as a function of the liquid mole fraction of xenon,  $x$ . The xenon mole fractions in the vapor phase,  $y$ , were evaluated using Barker's method<sup>20</sup> which minimizes the pressure residuals  $\delta p = p - p_{\text{calc}}$ . The  $G_m^E$  values were then calculated at zero pressure and fitted to a Redlich-Kister-type equation

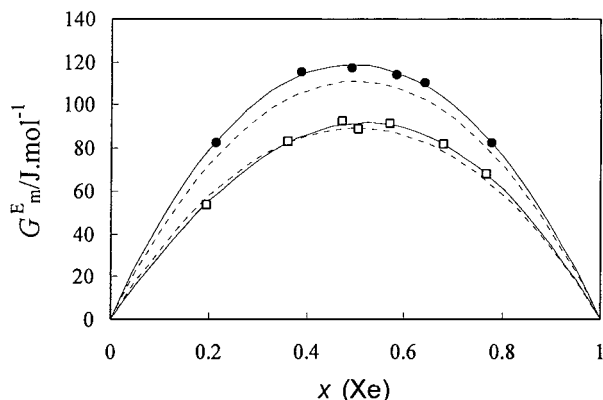
$$G_m^E/RT = x(1-x)[A + B(1-2x) + C(1-2x)^2] \quad (1)$$

The fitting parameters  $A$ ,  $B$ , and  $C$  together with their standard deviations and  $G_m^E(x = 0.5)$  at both temperatures are recorded

**TABLE 2: Orthobaric Molar Volumes,  $V_m$ , and Excess Molar Volumes,  $V_m^E$ , of Xenon + Diborane Liquid Mixtures at 182.34 K<sup>a</sup>**

$x$	$V_m/(\text{cm}^3\text{mol}^{-1})$	$V_m^E/(\text{cm}^3\text{mol}^{-1})$	$\delta V/(\text{cm}^3\text{mol}^{-1})$
0	65.054	0	
0.22929	60.882	0.0869	+0.0040
0.34278	58.856	0.169	-0.0044
0.43835	57.131	0.219	-0.010
0.58652	54.418	0.258	+0.010
0.68782	52.491	0.213	+0.0043
0.79695	50.379	0.128	-0.0038
1	46.480	0	

<sup>a</sup>  $\delta V$  values are the volume residuals, i.e., the differences between experimental and calculated (from eq 2) excess molar volumes.



**Figure 3.** Excess molar Gibbs energy for xenon + diborane mixtures at: ●, 161.40 K; □ 182.34 K. The solid lines are given by eq 1. The dashed lines are the DEOS predictions.

in Table 3. The  $G_m^E$  vs  $x$  curves, which are positive and fairly symmetric, are shown in Figure 3. From the temperature dependence of  $G_m^E$ , an average value of the excess molar enthalpy,  $H_m^E$ , could be estimated using the Gibbs–Helmholtz equation. For the equimolar mixture a value of  $(328 \pm 24)$  J mol<sup>-1</sup> was obtained. The corresponding value of the excess molar entropy was found to be  $S_m^E = (1.29 \pm 0.14)$  J.K<sup>-1</sup>.mol<sup>-1</sup>.

The orthobaric molar volumes,  $V_m$ , and excess molar volumes,  $V_m^E$ , of the same mixture at 182.34 K are shown in Table 2. Given the low pressures involved, these values can be taken to be, within experimental error, as equal to those at zero pressure.

The excess molar volume results were fitted to a Redlich–Kister equation

$$V_m^E/(\text{cm}^3 \text{mol}^{-1}) = x(1-x)[D + E(1-2x) + F(1-2x)^2] \quad (2)$$

where, as before,  $x$  is the liquid mole fraction of xenon, yielding  $D = (0.993 \pm 0.027)$  cm<sup>3</sup>.mol<sup>-1</sup>,  $E = (-0.360 \pm 0.044)$  cm<sup>3</sup>.mol<sup>-1</sup> and  $F = (-1.123 \pm 0.14)$  cm<sup>3</sup>.mol<sup>-1</sup>. The molar volume residuals, defined as  $\delta V = V_m^E - V_m^E(\text{calc})$ , are also given in Table 2. The experimental and fitted  $V_m^E$  results are plotted in Figure 4.  $V_m^E$  is positive for this system, contrarily to the behavior most commonly found in mixtures of simple fluids; for the equimolar mixture,  $V_m^E(x = 0.5) = (0.248 \pm 0.0068)$  cm<sup>3</sup>.mol<sup>-1</sup>.

#### 4. Discussion

In Table 4 the results for the present system are compared with those of the (xenon + ethane) and (xenon + ethylene) mixtures, previously studied.<sup>1,2</sup> Owing to its molecular structure, one could expect diborane to behave much like ethylene when

mixed with xenon. However, as discussed before, the different nature of some of the chemical bonds in diborane (and resulting electron density), will considerably alter its intermolecular forces. As can be observed in Table 4, the excess functions for (xenon + diborane) mimic those of (xenon + ethylene), both in terms of sign and order of magnitude. This could indicate that in these two similarly shaped molecules, the tricentric bonds in diborane interact much like the double bond in ethylene.

Xenon + diborane is a simple (in a molecular sense) system, whose behavior should be well described by any moderately sophisticated equation of state. A good example is the Deiters<sup>6</sup> equation of state (DEOS)—a semiempirical equation based on the square-well potential model which usually performs well, even with mixtures of molecules differing considerably in shape (as is the present case)

$$p = \frac{RT}{V_m} \left[ 1 + cc_0 \frac{a_1\xi + a_2(3a_2 - a_1 + 2)\xi^2 - a_2\xi^3}{(1 - \xi)^3} \right] - \frac{RT^*b}{V_m^2} \tilde{T} [e^{1/\tilde{T}} - 1] I_1(\xi) \quad (3)$$

with

$$\tilde{T} = \left( \frac{cT}{T^*} + \lambda\xi \right) \frac{1}{y(\xi)} \text{ and } T^* = \frac{\epsilon}{k}$$

In this equation  $V_m$  denotes the molar volume,  $b$  is a volumetric parameter (covolume,  $b = N_A\sigma^3/\sqrt{2}$ ),  $\xi$  is the reduced density, ( $\xi = \sqrt{2} \pi b/6V$ ) and  $c$  is a shape parameter which corrects for the nonsphericity of the molecules ( $c = 1$  for spherical molecules);  $c_0 = 0.6887$  is a universal constant which accounts for the deviation of the real pair potential from the rigid core model.  $\lambda = -0.09333$  is another universal constant which accounts for the influence of three-body interactions.  $I_1(\xi)$  and  $y(\xi)$  are complicated functions of density (and generally of  $c$ ) derived from statistical mechanics.<sup>6</sup> The coefficients  $a_1$  and  $a_2$ , derive from the Mansoori–Leland–Carnahan–Starling theory (which is an extension of the Carnahan–Starling theory to mixtures), and are given by

$$a_1 = \frac{3\bar{r}\bar{s}}{\bar{v}}, a_2 = \frac{\bar{s}^3}{\bar{v}^2} - 1 \quad (4)$$

with

$$\bar{r} = \sum x_i r_i, \bar{s} = \sum x_i r_i^2, \bar{v} = \sum x_i r_i^3$$

and

$$r_i = \sqrt[3]{b_{ii}}$$

For the composition dependence of the mixture parameters, a density dependent relation for the averaged attraction parameter was used

$$\epsilon\sigma^\gamma = \sum_i \sum_k x_i x_k \epsilon_{ik} \sigma_{ik}^\gamma \quad (5)$$

with

$$\gamma = 3(1 - \xi^2)$$

where  $\gamma$  is the structure exponent of the square well potential. Furthermore, quantum and quasichemical corrections were

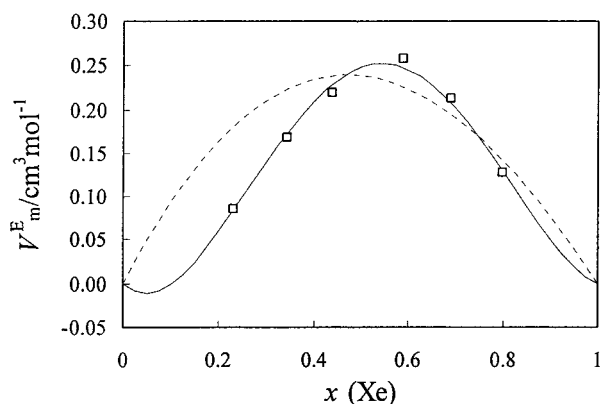
**TABLE 3: Values of the Redlich–Kister Coefficients (eq 1) and of the Excess Molar Gibbs Energy,  $G_{1/2}^E$ , for the Equimolar Mixture of Xenon + Diborane at 161.40 and 182.34 K**

T (K)	A	B	C	$G_{1/2}^E/(J \cdot mol^{-1})$
161.40	$0.354 \pm 0.0045$	$0.013 \pm 0.012$	$0.025 \pm 0.020$	$118.8 \pm 1.5$
182.34	$0.2419 \pm 0.0036$	$-0.018 \pm 0.0099$	$-0.053 \pm 0.018$	$91.7 \pm 1.4$

**TABLE 4: Values of the Excess Molar Functions for the Equimolar Mixtures of (xenon + diborane), (xenon + ethylene) and (xenon + ethane)**

system	T/K	$G_{1/2}^E/J \cdot mol^{-1}$	$H_{1/2}^E/J \cdot mol^{-1}$	$V_{1/2}^E/cm^3 \cdot mol^{-1}$
xenon + diborane	161.40 K	118.8	328 <sup>a</sup>	
	182.34 K	91.7		0.248
xenon + ethylene	161.40 K	145.4	284.7 <sup>b</sup>	0.346
xenon + ethane	161.40 K	-28.5	-51.7 <sup>c</sup>	-0.115
	182.34 K	-28.0		

<sup>a</sup> Average value for 161–182 K interval. <sup>b</sup> At 162.9 K. <sup>c</sup> At 163.0 K.

**Figure 4.** Excess molar volumes for xenon + diborane mixtures at 182.34 K. The solid lines are given by eq 2. The dashed line is the DEOS prediction.

added to these equations which are explained in detail in ref 22. The binary attraction parameter  $\epsilon_{12}$  was defined as

$$\epsilon_{12} = \theta(\epsilon_{11}\epsilon_{22})^{1/2} \quad (6)$$

$\theta$  is an adjustable interaction parameter and was calculated from the experimental results for the mixture.

The size parameter ratio is given by

$$\frac{\sigma_2}{\sigma_1} = \psi \left( \frac{c_{11}}{c_{22}} \right)^2 \left( \frac{b_{22}}{b_{11}} \right)^{1/3} \quad (7)$$

where  $\psi$  is a second adjustable interaction parameter.

The composition dependence of  $b$  and  $c$  is given by

$$b = x_1^2 b_{11} + 2x_1 x_2 b_{12} + x_2^2 b_{22} \quad (8)$$

$$c = x_1 c_{11} + x_2 c_{22} \quad (9)$$

with

$$b_{12} = \frac{(b_{11} + b_{22})}{2} \quad (10)$$

The parameters for the pure components were determined from critical, vapor-pressure, and saturated liquid density data and are listed in Table 5. The binary interaction parameters were calculated from one experimental VLE data point for a mixture at 182.34 K. No experimental molar volumes for the mixtures were included. All parameters are listed in Table 5.

**TABLE 5: Deiters EOS Parameters for Xenon and Diborane and Binary Interaction Parameters for Xenon + Diborane Mixture**

	$a/K$	$b/dm^3 \cdot mol^{-1}$	$c$	$\theta$	$\psi$
xenon	199.24	0.02614	1.000		
diborane	249.08	0.03697	1.098		
Xe+B <sub>2</sub> H <sub>6</sub>				0.969967	1.00283

The calculated curves are plotted in Figures 2, 3, and 4. The experimental phase diagram is very well reproduced by the equation of state at both temperatures (Figure 2) and so are the  $G_m^E$  vs  $x$  curves (Figure 3). For the equimolar composition the theoretical values of  $G_m^E$  are 111 J·mol<sup>-1</sup> at 161.40 K and 89 J·mol<sup>-1</sup> at 182.34 K, which basically coincide with the experimental data, 118.8 J·mol<sup>-1</sup> at 161.40 K and 91.7 J·mol<sup>-1</sup> at 182.34 K. As observed in Figure 4, the predictions for  $V_m^E$  can also be considered quite good (although the shapes of the curves are fairly different). The experimental value of  $V_m^E$  ( $x = 0.5$ ) = 0.248 cm<sup>3</sup>·mol<sup>-1</sup> is reproduced by the calculated one,  $V_m^E$  ( $x = 0.5$ ) = 0.238 cm<sup>3</sup>·mol<sup>-1</sup> almost within experimental error. The predicted values of  $H_m^E$  are 302 J·mol<sup>-1</sup> at 161.40 K and 257 J·mol<sup>-1</sup> at 182.34 K, in reasonable agreement with the average indirect experimental value of 328 J·mol<sup>-1</sup>. It should be emphasized that no experimental volumetric or calorimetric data for the mixtures were used in the calculation of the binary interaction parameters.

## References and Notes

- (1) Rowlinson, J. S.; Swinton, F. L. *Liquids and Liquid Mixtures*; Butterworth: London, 1982.
- (2) Filipe, E. J. M.; Gomes de Azevedo, E. J. S.; Martins, L. F. G.; Soares, V. A. M.; Calado, J. C. G.; McCabe, C.; Jackson, G. *J. Phys. Chem. B* **2000**, *104*, 1315.
- (3) Nunes da Ponte, M.; Chokappa, D.; Calado, J. C. G.; Zollweg, J.; Streett, W. B. *J. Phys. Chem. B* **1986**, *90*, 1147.
- (4) Gil-Villegas, A.; Galindo, A.; Whitehead, P. J.; Mills, S. J.; Jackson, G.; Burgess, A. N. *J. Chem. Phys.* **1997**, *106*, 4168.
- (5) Galindo, A.; Davies, L. A.; Gil-Villegas, A.; Jackson, G. *Mol. Phys.* **1998**, *93*, 241.
- (6) Deiters, U. *Chem. Eng. Sci.* **1981**, *36*, 1139.
- (7) Calado, J. C. G.; Gomes de Azevedo, E. J. S.; Soares, V. A. M. *Chem. Eng. Commun.* **1980**, *5*, 149.
- (8) McCarty, R. D.; Jacobsen, R. T., *NBS Technical Note 1045*, National Bureau of Standards, Boulder, USA 1981.
- (9) Staveley, L. A. K.; Lobo, L. Q.; Calado, J. C. G. *Cryogenics* **1981**, *21*, 131.
- (10) Calado, J. C. G.; Azevedo, E. J. S. G.; Soares, V. A. M.; Lucas, K.; Shukla, K. *Fluid Phase Equilibria* **1984**, *16*, 171.
- (11) Calado, J. C. G.; Rebelo, L. P. N.; Streett, W. B.; Zollweg, J. A. *J. Chem. Thermodyn.* **1986**, *18*, 931.
- (12) Freeguard, G. F.; Long, L. H. *Chem. & Ind.* **1965**, 471.
- (13) Rifkin, E. B.; Kerr, E. C.; Johnston, H. L. *J. Am. Chem. Soc.* **1953**, *75*, 785.
- (14) Wirth, H. E.; Palmer, E. D. *J. Phys. Chem.* **1956**, *60*, 911.

- (15) Dymond, J. B.; Smith, E. B. *The Virial Coefficients of Pure Gases and Mixtures*; Clarendon Press: Oxford, 1980.
- (16) Daubert, T. E.; Danner, R. P.; Sibul, H. M.; Stebbins, C. C., *Physical and Thermodynamic Properties of Pure Chemicals*, Department of Chemical Engineering, The Pennsylvania State University, Pennsylvania, Taylor and Francis (Diborane Sheet, 1987).
- (17) Brewer, J. Rep. No. MRL-2915-C, AFOSR-67-2795, 1968.

- (18) Leland, T. W., Chappelaar, P. S. *Ind. Eng. Chem.* **1968**, 60, 15.
- (19) Prausnitz, J. M.; Lichtenthaler, R. N.; Gomes de Azevedo, E. J. *Molecular Thermodynamics of Fluid-Phase Equilibria*; Prentice-Hall: Englewood Cliffs, New Jersey, 1986.
- (20) Barker, J. A. *Aust. J. Chem.* **1953**, 6, 207.
- (21) Deiters, U. *Fluid Phase Equilibria* **1987**, 33, 267.