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# Liquid Mixtures Involving Cyclic Molecules: Xenon + Cyclopropane

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The total vapor pressure of liquid mixtures of xenon and cyclopropane has been measured at 161.39 K (the triple-point of xenon) and at 182.33 K (the triple-point of dinitrogen oxide), as a function of composition. At 182.33 K the liquid densities were also measured. The mixtures show positive deviations from Raoult's law. Both the excess molar Gibbs energy ( $G_m^E$ ) and the excess molar volume ( $V_m^E$ ) were calculated from the experimental data. For the equimolar mixture,  $G_m^E = 90.6 \text{ J mol}^{-1}$  at 161.39 K,  $G_m^E = 124.1 \text{ J mol}^{-1}$  at 182.33 K, and  $V_m^E = -0.758 \text{ cm}^3 \text{ mol}^{-1}$  at 182.33 K. The excess molar enthalpy ( $H_m^E$ ) could be estimated from the temperature dependence of  $G_m^E$  and found to be  $-168 \text{ J mol}^{-1}$ . The results were interpreted using the 1cLJ perturbation theory of Fisher et al.

## 1. Introduction

Molecular shape cannot be ignored in any quantitative description of the thermodynamic properties of liquid mixtures of nonspherical molecules. With this in mind, we have recently started a systematic study on mixtures involving triangular molecules and reported results on the xenon + propane system.<sup>1</sup>

Cyclopropane, the simplest three-membered ring molecule, provides a good model of a triangular molecule and the study of its mixtures with xenon, when compared to the xenon + propane mixture, should contribute to the elucidation of the role played by a cyclic structure in intermolecular forces. The differences and similarities between propane and cyclopropane can be used to understand the nature of the intermolecular interactions. For instance, in cyclopropane there is no rotation around the C–C bond, the CH<sub>2</sub> groups are all in an eclipsed configuration, and owing to the distortion of the bonding angles, there is an enormous strain pushing the electronic density out of the ring, probably interfering with the hydrogen atom interactions. As a result, it is likely that the electric field created by each CH<sub>2</sub> group in cyclopropane is different from the corresponding one in propane. A similar type of comparison, but for a polar group, can be made between dimethyl ether and ethylene oxide. The results for both systems, xenon + dimethyl ether and xenon + ethylene oxide, have been reported.<sup>2,3</sup> As far as we are aware no other work has been carried out on the xenon + cyclopropane mixture. Moreover, within thermodynamic studies of mixtures involving cyclic molecules, only cycloalkanes higher than cyclopentane have been studied.

Lustig<sup>4</sup> modeled both propane and cyclopropane with a 3cLJ potential and calculated the thermodynamic properties of the pure substances using the perturbation theory of Fischer et al. The results were in excellent agreement with experiment, but the extension of the theory to mixtures is still to be developed. For this reason our experimental results were interpreted on the basis of a simple 1cLJ+1cLJ model.

## 2. Experimental Section

The vapor pressure and density measurements were carried out in an apparatus described elsewhere,<sup>5</sup> using similar experimental procedures. As usual, a triple-point cryostat was used, the working temperatures being 161.39 K (the triple-point of

xenon) and 182.33 K (the triple-point of dinitrogen oxide, N<sub>2</sub>O). The mixtures were prepared by condensing known amounts of each component into a calibrated pycnometer.

Samples of xenon (99.995% purity from Air Liquide or 99.99% purity from Linde), dinitrogen oxide (99.99% purity from Air Liquide), and cyclopropane (99.0% purity from Merck) were further purified by fractional distillation in the laboratory low-temperature column. In the case of xenon and dinitrogen oxide, the final purity was checked by measuring the constancy of the triple-point pressure during melting. The values obtained were the following: for xenon,  $81.669 \pm 0.007 \text{ kPa}$ , to be compared with the recommended value of  $81.674 \pm 0.011 \text{ kPa}$ ;<sup>6</sup> for dinitrogen oxide,  $87.815 \pm 0.010 \text{ kPa}$ , to be compared with  $87.865 \pm 0.012 \text{ kPa}$ .<sup>6</sup> In the case of cyclopropane, the triple-point vapor pressure is too low to be measured accurately by this method; however, experiments with other liquefied gases (including low hydrocarbons) show that the purity obtained using our distillation technique is never less than 99.99%.

A further purity check was provided by the measured values of the vapor pressure and/or molar volume of the pure components at the triple-point temperature of dinitrogen oxide. In the case of xenon these values were  $p(\text{Xe}) = 247.78 \text{ kPa}$  and  $V_m(\text{Xe}) = 46.468 \pm 0.006 \text{ cm}^3 \text{ mol}^{-1}$ , whereas in the case of cyclopropane  $p(\text{C}_3\text{H}_6) = 3.192 \text{ kPa}$ . These results compare favorably with the values reported by Calado et al.<sup>7</sup> at the same temperature:  $p(\text{Xe}) = 247.55 \pm 0.21 \text{ kPa}$ ,  $V_m(\text{Xe}) = 46.485 \pm 0.049 \text{ cm}^3 \text{ mol}^{-1}$  and  $p(\text{C}_3\text{H}_6) = 3.163 \text{ kPa}$ .<sup>8</sup>

The vapor pressures and densities of the Xe + C<sub>3</sub>H<sub>6</sub> mixtures were measured at the triple-point temperatures of N<sub>2</sub>O (182.33 K) and Xe (161.39 K). Most of the vapor pressure data were obtained with a fused quartz bourdon manometer (Texas Instrument model 145) with 130 kPa full range and 0.5 Pa resolution. In the case of the higher vapor pressures at 182.33 K, a similar manometer with 420 kPa full range and 1.4 Pa resolution was used instead. The density measurements were performed using a pycnometer ( $V_{\text{pyk}} = 2.2383 \text{ cm}^3$ ), calibrated with liquid ethane (Air Liquide, 99.995%) at 182.33 K on the basis of the experimental molar volumes of Haynes and Hiza.<sup>9</sup>

The ancillary data necessary for the evaluation of  $G_m^E$  and  $V_m^E$  from vapor pressure and molar volume values have already been given as far as xenon is concerned.<sup>7</sup> For cyclopropane, the second virial coefficient at room temperature was taken from the compilation of Dymond and Smith<sup>10</sup> ( $B = -360 \text{ cm}^3 \text{ mol}^{-1}$ ).

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**TABLE 1: Total Vapor Pressure and Excess Molar Gibbs Energy of Xenon + Cyclopropane Mixtures at 161.39 and 182.33 K<sup>a</sup>**

<i>x</i>	<i>y</i>	<i>p</i> /kPa	<i>R<sub>p</sub></i> /kPa	<i>G<sub>m</sub><sup>E</sup></i> /J mol <sup>-1</sup>
161.39 K				
0	0	0.467		0
0.420 90	0.992 09	37.14	-0.09	87.5
0.429 30	0.992 36	38.00	0.12	91.0
0.464 25	0.993 20	40.51	-0.05	89.6
0.478 27	0.993 53	41.61	-0.01	90.3
0.606 70	0.995 88	51.53	0.22	89.6
0.633 42	0.996 24	53.16	-0.15	81.4
0.751 27	0.997 70	61.96	-0.19	64.6
0.846 16	0.998 67	69.62	0.23	51.2
1	1	81.67		0
182.33 K				
0	0	3.192		0
0.198 98	0.956 21	60.94	+0.28	72.9
0.303 98	0.972 65	87.37	-0.60	96.1
0.346 16	0.973 72	98.45	+0.12	108.6
0.515 20	0.984 84	137.48	+0.54	127.5
0.625 74	0.989 29	160.60	-0.23	116.8
0.726 77	0.993 63	182.45	-0.33	98.6
0.847 84	0.996 66	210.61	+0.31	66.8
1	1	247.78		0

<sup>a</sup> *R<sub>p</sub>* are the pressure residuals defined as  $R_p = p_{\text{exp}} - p_{\text{calc}}$ .

There are no experimental values for the second virial coefficient of cyclopropane at low temperatures. They were, therefore, estimated using the equation of Leland.<sup>11</sup> The values obtained were  $B = -1667 \text{ cm}^3 \text{ mol}^{-1}$  at 161.39 K and  $B = -1264 \text{ cm}^3 \text{ mol}^{-1}$  at 182.33 K. The cross virial coefficients for the xenon + cyclopropane mixtures, were also estimated using the correlation of Van Ness and Abbott, as discussed by Prausnitz<sup>12</sup> ( $B_{12} = -777 \text{ cm}^3 \text{ mol}^{-1}$  at 161.39 K,  $B_{12} = -601 \text{ cm}^3 \text{ mol}^{-1}$  at 182.33 K, and  $B_{12} = -222 \text{ cm}^3 \text{ mol}^{-1}$  at 298.15 K).

### 3. Results

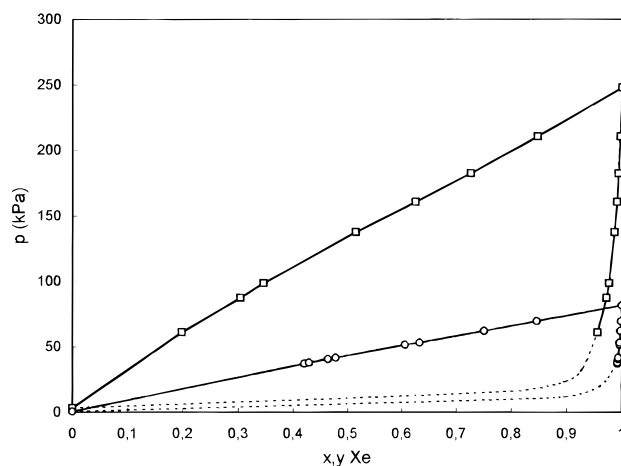
The vapor pressure, *p*, of xenon + cyclopropane mixtures at 161.39 and 182.33 K are given in Table 1 as a function of the liquid mole fraction of xenon, *x*. The excess molar Gibbs energy of each mixture, *G<sub>m</sub><sup>E</sup>*, and the mole fraction of xenon in the vapor phase, *y*, were calculated from the total vapor pressure measurements using the Redlich–Kister model and the method of Barker<sup>13</sup> for the minimization of the pressure residuals, *R<sub>p</sub>* = *p* - *p<sub>calc</sub>*.

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

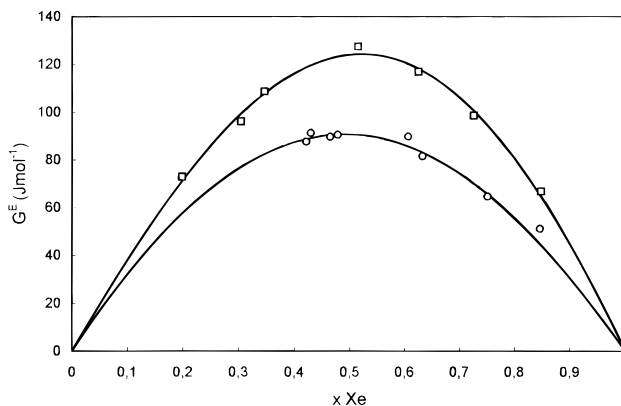
The values of *R<sub>p</sub>*, which are also given in Table 1, are a good indication of the self-consistency of the experimental data. *p<sub>calc</sub>* is the calculated vapor pressure from eq 1 and Barker's method.

The *p*(*x*,*y*) diagrams for both temperatures are presented in Figure 1. At 161.39 K, cyclopropane exhibits a tendency to condense into small droplets in the pyknometer stem, making the measurements unreliable. As a result of this, only mixtures with xenon mole fractions above 0.4 could be studied. Several experiments were carried out around the 0.5 mole fraction, to ensure a better defined *G<sub>m</sub><sup>E</sup>* curve and a more accurate value for the equimolar mixture.

The *G<sub>m</sub><sup>E</sup>* results are plotted in Figure 2 for both temperatures as a function of *x*. As can be seen, the curves are fairly symmetric. The corresponding Redlich–Kister expansion coefficients, as well as the values of *G<sub>m</sub><sup>E</sup>* for the equimolar mixture, are recorded in Table 2. It should be noted that the uncertainty in *G<sub>m</sub><sup>E</sup>* at 161.39 K is much larger than usual, since no



**Figure 1.** Vapor–liquid equilibrium for xenon + cyclopropane mixtures at 161.39 (circles) and 182.33 K (squares).



**Figure 2.** Excess molar Gibbs energy for xenon + cyclopropane mixtures at 161.39 (squares) and 182.33 K (circles).

experimental data could be obtained for *x* < 0.4. The excess molar enthalpy, *H<sub>m</sub><sup>E</sup>*, could be estimated within the limits of uncertainty of *G<sub>m</sub><sup>E</sup>*(*x* = 0.5) at both temperatures. For the equimolar composition a value of *H<sub>m</sub><sup>E</sup>* = -168 J mol<sup>-1</sup> was found. The excess molar entropy is thus also negative, *S<sub>m</sub><sup>E</sup>* = -1.6 J mol<sup>-1</sup>.

The orthobaric molar volumes, *V<sub>m</sub>*, of xenon + cyclopropane mixtures at 182.33 K are given in Table 3. The excess molar volumes, *V<sub>m</sub><sup>E</sup>*, were not corrected to zero pressure since, owing to the low pressures involved, the corrections would be within experimental error.

The molar volume of pure cyclopropane, *V<sub>m</sub>*(C<sub>3</sub>H<sub>6</sub>), could not be determined experimentally with our technique because the substance condenses easily in the pyknometer inlet tube, forming droplets and bubbles. It was estimated through the extrapolation of the measured values for the mixtures

$$V_m(\text{C}_3\text{H}_6) = \lim_{x \rightarrow 0} \frac{V_m - xV_m(\text{Xe})}{(1-x)} \quad (2)$$

A value of  $56.544 \pm 0.040 \text{ cm}^3 \text{ mol}^{-1}$  was obtained. A similar procedure applied to xenon leads to a value of  $46.434 \pm 0.040 \text{ cm}^3 \text{ mol}^{-1}$ , in good agreement with the measured value of  $46.486 \text{ cm}^3 \text{ mol}^{-1}$ . The excess molar volume results were fitted to a Redlich–Kister type equation

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

with  $D = -3.0315 \pm 0.0663 \text{ cm}^3 \text{ mol}^{-1}$  and  $E = 0.1538 \pm 0.0619 \text{ cm}^3 \text{ mol}^{-1}$ . The molar volume residuals, *R<sub>V</sub>* = *V<sub>m</sub><sup>E</sup>* -

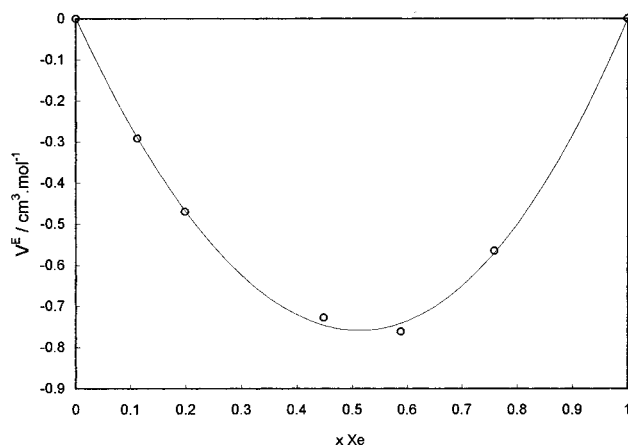
**TABLE 2: Excess Molar Gibbs Energy of Xenon + Cyclopropane Mixtures at 161.39 and 182.33 K: Values for the Equimolar Mixture,  $G_{1/2}^E$ , and Coefficients for Eq 1**

	A	B	C	$G_{1/2}^E/\text{J mol}^{-1}$
161.39 K	$0.2700 \pm 0.0912$	$0.0083 \pm 0.0999$	$0.0157 \pm 0.0736$	$90.6 \pm 30.6$
182.33 K	$0.3274 \pm 0.0108$	$-0.0312 \pm 0.0171$	$-0.0363 \pm 0.0226$	$124.1 \pm 4.1$

**TABLE 3: Molar Volumes and Excess Molar Volumes of Xenon + Cyclopropane Mixtures at 182.33 K and under the Saturation Vapor Pressure<sup>a</sup>**

$x$	$V_m/\text{cm}^3 \text{ mol}^{-1}$	$V_m^E/\text{cm}^3 \text{ mol}^{-1}$	$R_V/\text{cm}^3 \text{ mol}^{-1}$
0	56.544	0	0
0.112 57	55.118	-0.291	-0.002
0.198 55	54.074	-0.470	-0.004
0.448 56	51.297	-0.727	0.016
0.588 33	49.855	-0.761	-0.023
0.758 58	48.335	-0.565	0.002
1	46.468	0	0

<sup>a</sup>  $R_V$  are the volume residuals, defined as  $R_V = V_m^E - V_m^E(\text{eq 3})$ .

**Figure 3.** Excess molar volume for xenon + cyclopropane mixtures at 182.33 K.**TABLE 4: Excess Molar Functions for the Equimolar Mixtures of Xenon + Propane and Xenon + Cyclopropane at 182.33 K**

system	$G_{1/2}^E/\text{J mol}^{-1}$	$H_{1/2}^E/\text{J mol}^{-1}$	$V_{1/2}^E/\text{cm}^3 \text{ mol}^{-1}$
xenon + propane	-33.8	200	-0.308 <sup>a</sup>
xenon + cyclopropane	124.1	-168	-0.758

<sup>a</sup> At 161.39 K.

$V_m^E$  (eq 3), are also given in Table 3. The experimental values and the fitted  $V_m^E$  curve are presented in Figure 3. For  $x = 0.5$ ,  $V_m^E = -0.758 \pm 0.013 \text{ cm}^3 \text{ mol}^{-1}$ .

#### 4. Discussion

The influence of a cyclic structure on the thermodynamic behavior of mixtures can be assessed comparing the results for the present system with those for the xenon + propane mixtures. This is done in Table 4. The xenon + propane system follows the usual behavior for xenon + *n*-alkane mixtures, with negative  $G_m^E$  and  $V_m^E$  and positive  $H_m^E$ . With cyclopropane, the picture is entirely different. Although there is an increase in  $G_m^E$  relative to that for the xenon + propane mixture,  $H_m^E$  and  $S_m^E$  are now negative. This suggests a stronger interaction between xenon and cyclopropane and an ordering of the liquid mixture. The effect on  $V_m^E$  is difficult to interpret since this property is very sensitive to structure and packing in the condensed state.

Systematic studies involving the higher cycloalkanes,<sup>14,15</sup>  $C_5$ – $C_{10}$ , have shown that, as a rule, these molecules behave as quasi-spherical ones and can act as structure breakers when mixed

**TABLE 5: Comparison of Experimental and Calculated Excess Functions for the Equimolar Mixture of Xenon + Cyclopropane at 182.33 K**

	$\xi$	$G_m^E/\text{J mol}^{-1}$	$H_m^E/\text{J mol}^{-1}$	$V_m^E/\text{cm}^3 \text{ mol}^{-1}$
exptl		124	-168	-0.758
LB	1	34	-82	-0.59
KOE	0.9861	138	64	-0.47
$\xi(G_m^E)$	0.9879	(124)	45	-0.49
$\xi(H_m^E)$	1.0082	-28	(-168)	-0.66

with linear alkanes. This is reflected, for instance, in positive values for the excess molar volumes. Mixtures with cyclopentane constitute an exception to this behavior, exhibiting smaller (negative) excess enthalpies and entropies than those involving the higher cycloalkanes. To explain this, Patterson proposed a “condensation effect”, according to which the *n*-alkane molecules show a tendency to “condense” onto rigid, highly branched or platelike molecules (such as 3,3-diethylpentane and substituted cyclohexanes, respectively), resulting in negative contributions to  $H_m^E$ . At the same time, the condensation effect corresponds to a restriction of molecular motion, thermodynamically equivalent to the creation of order in the liquid, therefore contributing negatively to  $S_m^E$  and positively to  $C_p^E$ . Cyclopentane seemed to be the only cyclic molecule with this kind of behavior, which was attributed to its platelike molecular structure, whereas the other (higher) cycloalkanes are more globular. The anisotropic forces around the cyclopentane molecule are said to favor its condensation, whereas rotation is hindered.

We believe that a similar situation can be found in cyclopropane and cyclobutane since both molecules are also platelike, anisotropic rotors. The effect might even be strengthened since in these molecules all the  $\text{CH}_2$  groups are now in a rigid eclipsed conformation, whereas cyclopentane still shows conformational changes. The results found for the xenon + cyclopropane mixture, namely negative  $H_m^E$  and  $S_m^E$ , are probably a consequence of the shape of the cyclopropane molecule, which enhances the interaction with the xenon atom, restricts its own rotation, and brings order to the solution. It might be argued that xenon is not a big highly branched molecule, but on the other hand, the cyclopropane molecule is also much smaller than cyclopentane and xenon has a high polarizability, which enhances dispersion forces. If this is true, a similar effect should also appear with cyclobutane, and the study of the xenon + cyclobutane mixture will be undertaken soon.

For a more theoretical quantitative interpretation of the results the perturbation theory of Fischer et al.<sup>16</sup> was used. The adequate description of this system would be a 1cLJ + 3cLJ model, but since the corresponding statistical mechanics formalism has not been developed so far, the mixture was treated as 1cLJ+1cLJ. The calculations were performed using the correlation formulas proposed by Kohler.<sup>17</sup> The intermolecular potential parameters  $\epsilon$  and  $\sigma$  were obtained adjusting the LJ equation of state of Fischer et al.<sup>18</sup> to the experimental vapor pressures and to the orthobaric densities of the pure components. We obtained for xenon  $\epsilon/k = 227.86 \text{ K}$ ,  $\sigma = 0.39478 \text{ nm}$  and for cyclopropane  $\epsilon/k = 343.88 \text{ K}$ ,  $\sigma = 0.43971 \text{ nm}$ . In calculating the properties of the mixture we used the so-called “Kohler old extended rule” (KOE), that is, we used the arithmetic mean rule for  $\sigma_{12}$ .

$$\sigma_{12} = (\sigma_{11} + \sigma_{22})/2 \quad (4)$$

and for  $\epsilon_{12}$

$$\epsilon_{12} = \xi (\epsilon_{11}\epsilon_{22})^{1/2} \quad (5)$$

with

$$\xi = \left( \frac{(\sigma_{11}\sigma_{22})^{1/2}}{(\sigma_{11} + \sigma_{22})/2} \right)^6 \frac{(h\nu_1 h\nu_2)^{1/2}}{(h\nu_1 + h\nu_2)/2} \quad (6)$$

where  $h\nu_i$  was taken as twice the experimental ionization energy of molecule  $i$ ,<sup>19</sup> (12.13 eV for xenon<sup>19</sup> and 9.86 eV for cyclopropane<sup>20</sup>). Alternatively we have adjusted  $\xi$  to fit  $G_m^E$  and calculated  $H_m^E$ , and vice-versa. This procedure has been suggested by Bohn et al.<sup>19</sup> as a possible way of including shape effects in fitted  $\xi$ -values. The results are shown in Table 5. Surprisingly the Lorentz–Berthelot rule predicts the correct sign for all three excess functions. The KOE rule gives values for  $G_m^E$  and  $V_m^E$  which are in good agreement with the experimental ones, but the sign for  $H_m^E$  is wrong. Fitting  $\xi$  to the value of either  $G_m^E$  or  $H_m^E$  fails to predict even the sign of the other excess function. It should also be noted that the KOE rule gives an excellent prediction of  $\xi$ , as fitted to  $G_m^E$ . This might indicate that shape effects, not taken into account since we are using a spherical model, are essential to describe this system adequately.

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