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# High-Pressure Phase Diagrams of Propane + Decahydronaphthalene and Propane + Naphthalene Mixtures

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Vapor−liquid equilibrium data for the mixtures propane + decahydronaphthalene (decalin) at (392, 439, and 415.5) K and propane + naphthalene at (373, 415, and 443) K are reported. The experiments have been conducted in a high-pressure cell at pressures up to 9 MPa. The molar concentrations have been measured by a spectroscopic method described earlier, which allows us to eliminate the effect of the baseline shifts. $P-x$ coexistence curves are determined up to the mixtures critical points. The unusual shape of the bubble curve of the propane + naphthalene mixture at 373 K is probably due to the existence of a critical end point at a temperature close to this one.

## Introduction

Vapor−liquid data for binary hydrocarbon systems are needed to calibrate and test thermodynamic models that can be used in process simulators. Binary interactions between two molecules or between two chemical groups (if one uses group contribution) have an important effect on the binary phase diagram and must be taken into account in models. Hence, binary mixture data are necessary to determine the parameters of models.

This paper presents data on two mixtures of propane + heavy hydrocarbons obtained with an in situ method using infrared spectroscopy, which was described previously.[1,7]

The mixtures previously investigated using this method are listed hereafter: methane + 1-methylnaphthalene,[1] methane + hexane and methane + benzene,[2] methane + squalane and methane + hexatriacontane,[3] methane + 1,2,3,4-tetrahydronaphthalene and methane + 9,10-dihydrophenanthrene,[4] methane + decahydronaphthalene and methane + acenaphthene,[5] and propane + 1,2,3,4-tetrahydronaphthalene and propane + 1-methylnaphthalene.[6]

## Experimental Section

***Materials.*** The purity of propane was 99.95%. *cis*-Decalin was provided by Aldrich with a purity of 99%. Note that, for this kind of compound, materials of higher purity are not provided by the different manufacturers.

Naphthalene also obtained from Aldrich was labeled 99+%. No further purification was attempted.

***Apparatus and Procedures.*** Since the apparatus[1,2] and the method[1,7] for calculating concentrations from spectra have been described in previous work, only a brief description will be provided here. The measurements have been performed in a medium-pressure equilibrium cell (up to 50 MPa) described earlier.[2] This is a variable volume cell with two sapphire windows allowing the light beam through the analyzed fluid. The cell can be rotated around the optical axis in order to stir the fluid toward equilibrium. The rotation of the cell also allows us to measure the liquid or the vapor phase, depending on the position of the cell. A known amount of solute is introduced in the cell. Then the cell is evacuated, and the solvent is introduced until an appropriate pressure is reached. Then the cell is sealed and heated until the temperature of interest. The temperature of the furnace is maintained constant by a regulator. The volume of the cell is then varied, producing different vapor−liquid equilibria at different pressures. The spectra were recorded with a Fourier transform spectrometer (BOMEM MB155), yielding better accuracy of the spectra in this work than what was obtained in our previous work with a grating monochromator.[1−5] The spectra are analyzed by a method which has been fully described in previous papers. The calculations involve calibration spectra obtained on pure species and integration of the spectra on five different spectral bands. It has been shown[1,7] that the resulting concentrations are not dependent on the baseline of the spectra. It has even been demonstrated[1,7] that the results are independent of the baselines of the calibration spectra. Readers more interested in the details of this method are referred to our previous papers,[1,7] where it has been described in full detail.

***Accuracy.*** As already stated in previous papers,[3−5] the method gives direct access to the concentrations of both compounds $C_1$ and $C_2$ (solute and propane respectively). The mole fractions may be calculated as $x_i = C_i/(C_1 + C_2)$. The uncertainty of the concentrations is estimated to be 1%. As in our previous work, the main source of uncertainty is the pressure gauge with an uncertainty of 0.05 MPa. The temperature uncertainty is estimated to be 0.5 °C. The accuracy, the validity, and the reproducibility of the method has been tested several times. One previous paper is only devoted to the subject. In this article, the necessity of a treatment to get rid of the errors introduced by baselines is shown. Once the calibration is done, the reproducibility

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**Table 1. Vapor−Liquid Equilibrium Data for Decalin (1) + Propane (2) + at (392, 415.5, and 439) K**

| | gas phase | | liquid phase | | | gas phase | | liquid phase | |
|---|---|---|---|---|---|---|---|---|---|
| $P$/MPa | $C_1$/mol L$^{-1}$ | $C_2$/mol L$^{-1}$ | $C_1$/mol L$^{-1}$ | $C_2$ /mol L$^{-1}$ | $P$/MPa | $C_1$/mol L$^{-1}$ | $C_2$/mol L$^{-1}$ | $C_1$/mol L$^{-1}$ | $C_2$ /mol L$^{-1}$ |
| $T$ = 392 K | | | | | | | | | |
| 1.72 | | | 4.852 | 2.393 | 4.37 | | | 1.612 | 7.319 |
| 2.04 | | | 4.499 | 3.001 | 4.37 | | | 1.618 | 7.301 |
| 2.26 | 0.006 | 0.813 | 4.276 | 3.411 | 4.40 | 0.00 | 2.182 | | |
| 2.54 | 0.005 | 0.946 | 3.977 | 3.903 | 4.40 | 0.00 | 2.178 | | |
| 2.85 | 0.00 | 1.083 | 3.666 | 4.418 | 4.45 | 0.00 | 2.291 | 1.485 | 7.352 |
| 3.00 | | | 3.349 | 4.931 | 4.47 | | | 1.522 | 7.399 |
| 3.11 | 0.00 | 1.252 | | | 4.47 | | | 1.528 | 7.386 |
| 3.35 | 0.00 | 1.375 | 3.050 | 5.380 | 4.50 | 0.00 | 2.263 | | |
| 3.38 | 0.00 | 1.501 | | | 4.50 | 0.00 | 2.261 | | |
| 3.66 | 0.00 | 1.563 | 2.683 | 5.939 | 4.55 | | | 1.421 | 7.489 |
| 3.85 | 0.00 | 1.703 | 2.431 | 6.300 | 4.55 | | | 1.366 | 7.469 |
| 4.05 | 0.00 | 1.866 | 2.163 | 6.667 | 4.56 | 0.00 | 2.381 | | |
| 4.14 | | | 1.99 | 6.782 | 4.59 | 0.00 | 2.340 | 1.311 | 7.592 |
| 4.21 | | | 1.955 | 6.903 | 4.68 | | | 1.209 | 7.616 |
| 4.21 | | | 1.953 | 6.914 | 4.71 | 0.00 | 2.576 | 1.148 | 7.632 |
| 4.26 | | | 1.774 | 7.123 | 4.80 | 0.00 | 2.721 | 1.002 | 7.732 |
| 4.26 | | | 1.774 | 7.120 | 4.90 | 0.00 | 2.854 | 0.869 | 7.796 |
| 4.28 | | | 1.765 | 7.060 | 4.95 | | | 0.768 | 7.803 |
| 4.31 | 0.00 | 2.076 | | | 5.05 | | | 0.705 | 7.836 |
| 4.31 | 0.00 | 2.073 | | | 5.06 | | | 0.661 | 7.805 |
| $T$ = 415.5 K | | | | | | | | | |
| 2.09 | | | 4.656 | 1.724 | 5.18 | 0.014 | 2.380 | 2.252 | 5.231 |
| 2.22 | | | 4.568 | 1.862 | 5.25 | 0.013 | 2.405 | 2.200 | 5.358 |
| 2.37 | | | 4.477 | 2.005 | 5.28 | 0.017 | 2.466 | 2.160 | 5.337 |
| 2.53 | | | 4.374 | 2.176 | 5.38 | 0.018 | 2.543 | 2.094 | 5.415 |
| 2.66 | | | 4.289 | 2.313 | 5.43 | 0.016 | 2.542 | 2.044 | 5.543 |
| 2.76 | 0.006 | 0.966 | 4.178 | 2.466 | 5.48 | 0.022 | 2.661 | 1.951 | 5.584 |
| 2.98 | 0.006 | 1.064 | 4.029 | 2.696 | 5.52 | 0.018 | 2.668 | 1.924 | 5.695 |
| 3.08 | 0.005 | 1.101 | 3.981 | 2.782 | 5.57 | 0.023 | 2.752 | 1.867 | 5.672 |
| 3.29 | | | 3.843 | 3.021 | 5.63 | | | 1.806 | 5.826 |
| 3.42 | 0.00 | 1.273 | 3.754 | 3.156 | 5.66 | | | 1.761 | 5.877 |
| 3.60 | | | 3.615 | 3.369 | 5.69 | 0.026 | 2.866 | 1.762 | 5.784 |
| 3.76 | | | 3.468 | 3.615 | 5.72 | | | 1.663 | 5.991 |
| 3.88 | 0.005 | 1.485 | | | 5.80 | 0.030 | 2.983 | 1.615 | 6.027 |
| 3.98 | 0.00 | 1.520 | 3.315 | 3.731 | 5.80 | | | 1.686 | 5.865 |
| 4.07 | 0.00 | 1.568 | 3.251 | 3.821 | 5.92 | 0.034 | 3.110 | 1.568 | 5.976 |
| 4.22 | 0.00 | 1.659 | 3.130 | 4.014 | 6.02 | 0.040 | 3.256 | 1.442 | 6.081 |
| 4.37 | 0.005 | 1.754 | 3.002 | 4.214 | 6.13 | 0.049 | 3.418 | 1.318 | 6.160 |
| 4.48 | 0.007 | 1.827 | | | 6.23 | 0.055 | 3.554 | 1.223 | 6.228 |
| 4.54 | 0.006 | 1.858 | 2.868 | 4.414 | 6.31 | 0.061 | 3.706 | 1.132 | 6.254 |
| 4.64 | 0.007 | 1.939 | 2.735 | 4.603 | 6.41 | 0.072 | 3.889 | 1.017 | 6.284 |
| 4.82 | 0.008 | 2.061 | 2.610 | 4.785 | 6.49 | | | 0.910 | 6.298 |
| 4.98 | 0.010 | 2.174 | 2.465 | 4.980 | 6.58 | | | 0.790 | 6.253 |
| 5.05 | 0.012 | 2.272 | 2.380 | 5.075 | 6.60 | | | 0.733 | 6.208 |
| 5.11 | 0.011 | 2.276 | 2.341 | 5.161 | | | | | |
| $T$ = 439 K | | | | | | | | | |
| 2.43 | 0.021 | 0.804 | | | 5.21 | 0.051 | 2.185 | 2.940 | 4.448 |
| 2.46 | | | 4.499 | 1.897 | 5.40 | 0.054 | 2.308 | 2.821 | 4.631 |
| 2.46 | | | 4.494 | 1.899 | 5.60 | 0.061 | 2.450 | 2.690 | 4.824 |
| 2.55 | 0.022 | 0.850 | | | 5.80 | 0.067 | 2.585 | 2.562 | 5.015 |
| 2.66 | | | 4.393 | 2.075 | 6.00 | 0.075 | 2.740 | 2.418 | 5.191 |
| 2.73 | 0.022 | 0.923 | | | 6.20 | 0.084 | 2.919 | 2.270 | 5.380 |
| 2.85 | | | 4.301 | 2.245 | 6.42 | 0.091 | 3.091 | 2.111 | 5.539 |
| 2.98 | 0.024 | 1.012 | 4.220 | 2.383 | 6.62 | 0.108 | 3.283 | 1.967 | 5.685 |
| 3.13 | 0.025 | 1.091 | | | 6.81 | 0.128 | 3.501 | 1.795 | 5.841 |
| 3.15 | 0.026 | 1.091 | 4.134 | 2.525 | 6.96 | 0.145 | 3.709 | 1.675 | 5.927 |
| 3.35 | | | 4.040 | 2.687 | 7.00 | 0.141 | 3.714 | | |
| 3.44 | 0.028 | 1.226 | | | 7.10 | 0.155 | 3.834 | | |
| 3.46 | 0.028 | 1.227 | | | 7.15 | 0.165 | 3.938 | 1.516 | 6.033 |
| 3.51 | | | 3.958 | 2.823 | 7.25 | 0.179 | 4.072 | | |
| 3.66 | | | 3.855 | 2.997 | 7.32 | 0.200 | 4.172 | | |
| 3.78 | 0.030 | 1.363 | 3.787 | 3.103 | 7.32 | 0.201 | 4.169 | | |
| 3.95 | 0.032 | 1.456 | 3.702 | 3.243 | 7.33 | 0.202 | 4.193 | | |
| 4.14 | 0.034 | 1.555 | 3.602 | 3.412 | 7.35 | 0.197 | 4.187 | | |
| 4.29 | | | 3.514 | 3.567 | 7.41 | 0.219 | 4.334 | | |
| 4.41 | 0.037 | 1.727 | 3.426 | 3.695 | 7.44 | 0.231 | 4.423 | | |
| 4.43 | 0.037 | 1.694 | | | 7.52 | 0.257 | 4.589 | | |
| 4.50 | | | 3.385 | 3.765 | 7.56 | 0.284 | 4.700 | | |
| 4.55 | 0.040 | 1.751 | | | 7.60 | 0.274 | 4.671 | | |
| 4.60 | 0.040 | 1.825 | 3.315 | 3.873 | 7.63 | 0.313 | 4.837 | | |
| 4.65 | | | 3.284 | 3.926 | 7.67 | 0.313 | 4.848 | 1.014 | 6.121 |
| 4.74 | 0.042 | 1.858 | | | 7.67 | 0.379 | 5.015 | | |
| 4.80 | 0.043 | 1.943 | 3.186 | 4.092 | 7.70 | 0.352 | 5.012 | 0.939 | 6.082 |
| 5.00 | 0.046 | 2.056 | 3.070 | 4.268 | 7.74 | 0.477 | 5.366 | 0.877 | 5.936 |

**Table 2. Vapor−Liquid Equilibrium Data for Naphthalene (1) + Propane (2) at (373, 415, and 443) K**

| | gas phase | | liquid phase | | | gas phase | | liquid phase | |
|---|---|---|---|---|---|---|---|---|---|
| $P$/MPa | $C_1$/mol L$^{-1}$ | $C_2$/mol L$^{-1}$ | $C_1$/mol L$^{-1}$ | $C_2$/mol L$^{-1}$ | $P$/MPa | $C_1$/mol L$^{-1}$ | $C_2$/mol L$^{-1}$ | $C_1$/mol L$^{-1}$ | $C_2$/mol L$^{-1}$ |
| $T$ = 373 K | | | | | | | | | |
| 0.72 | 0.00 | 0.178 | 7.504 | 0.385 | 3.55 | | | 3.781 | 5.372 |
| 1.29 | 0.00 | 0.356 | 7.169 | 0.853 | 3.60 | 0.00 | 1.670 | | |
| 1.84 | 0.00 | 0.617 | 6.724 | 1.578 | 3.73 | 0.00 | 1.841 | 3.109 | 6.152 |
| 2.10 | 0.00 | 0.742 | 6.491 | 1.904 | 3.88 | 0.00 | 2.009 | 2.479 | 6.852 |
| 2.39 | 0.00 | 0.890 | 6.115 | 2.359 | 3.96 | | | 2.193 | 7.147 |
| 2.56 | 0.00 | 0.970 | 5.798 | 2.723 | 4.03 | 0.00 | 2.191 | | |
| 2.64 | 0.00 | 1.036 | | | 4.14 | 0.012 | 2.347 | 0.974 | 7.802 |
| 3.05 | 0.00 | 1.261 | 5.159 | 3.556 | 4.24 | 0.013 | 2.544 | 0.715 | 7.785 |
| 3.28 | 0.00 | 1.442 | | | 4.32 | 0.013 | 2.691 | 0.563 | 7.733 |
| 3.49 | 0.00 | 1.630 | | | 4.37 | | | 0.477 | 7.658 |
| $T$ = 415 K | | | | | | | | | |
| 0.60 | 0.00 | 0.108 | 7.498 | 0.186 | 6.16 | 0.065 | 3.137 | | |
| 1.68 | 0.00 | 0.430 | 6.868 | 0.747 | 6.16 | 0.070 | 3.136 | | |
| 2.37 | 0.007 | 0.730 | 6.600 | 1.266 | 6.26 | 0.068 | 3.211 | 3.139 | 5.260 |
| 2.77 | 0.006 | 0.882 | 6.382 | 1.519 | 6.42 | 0.104 | 3.534 | | |
| 3.30 | 0.008 | 1.114 | 6.089 | 1.926 | 6.43 | 0.098 | 3.428 | | |
| 4.29 | | | 5.348 | 2.777 | 6.44 | 0.102 | 3.481 | | |
| 4.35 | 0.014 | 1.645 | | | 6.46 | 0.104 | 3.556 | | |
| 5.03 | 0.025 | 2.114 | 4.586 | 3.711 | 6.47 | 0.087 | 3.518 | | |
| 5.19 | 0.028 | 2.244 | | | 6.55 | 0.118 | 3.650 | | |
| 5.20 | 0.030 | 2.241 | | | 6.55 | 0.120 | 3.717 | | |
| 5.46 | 0.038 | 2.451 | | | 6.66 | 0.141 | 3.851 | | |
| 5.47 | 0.034 | 2.449 | | | 6.66 | 0.140 | 3.923 | | |
| 5.70 | 0.040 | 2.612 | 3.920 | 4.472 | 6.72 | 0.168 | 4.072 | | |
| 5.71 | | | 3.959 | 4.444 | 6.76 | 0.173 | 4.153 | | |
| 5.79 | 0.045 | 2.747 | | | 6.83 | 0.165 | 4.253 | 1.957 | 6.140 |
| 5.80 | 0.049 | 2.748 | | | 6.87 | 0.215 | 4.373 | 0.208 | 4.407 |
| 5.96 | 0.053 | 2.841 | | | 6.97 | 0.243 | 4.530 | | |
| $T$ = 443 K | | | | | | | | | |
| 1.39 | 0.032 | 0.369 | | | 7.14 | 0.121 | 3.208 | 3.862 | 4.372 |
| 1.72 | | | 6.997 | 0.677 | 7.61 | 0.164 | 3.627 | 3.430 | 4.810 |
| 3.70 | 0.030 | 1.218 | 6.065 | 1.801 | 7.64 | 0.168 | 3.686 | | |
| 4.26 | 0.035 | 1.454 | 5.804 | 2.134 | 8.10 | 0.219 | 4.136 | 2.891 | 5.304 |
| 5.06 | 0.042 | 1.841 | 5.381 | 2.660 | 8.12 | 0.244 | 4.181 | | |
| 5.95 | 0.064 | 2.360 | 4.803 | 3.357 | 8.46 | 0.355 | 4.682 | 2.395 | 5.664 |
| 6.18 | 0.068 | 2.533 | | | 8.51 | 0.357 | 4.723 | | |
| 6.53 | 0.082 | 2.736 | 4.409 | 3.804 | 8.55 | | | 2.255 | 5.745 |
| 6.95 | 0.106 | 3.086 | | | 8.72 | | | 1.986 | 5.873 |

of the concentration results is shown to be excellent. The reproducibility has been tested in different respects: reproducibility of the spectra, of the mole fraction of the same mixture under different pressures, and of the calculated concentrations.[7] The results have also been compared for a few selected points to those obtained with other methods (synthetic method) and have been shown to fall within the experimental error.[5,7]

## Results

***Propane* + *Decalin Mixture.*** Molar concentrations $C_1$ (decalin) and $C_2$ (propane) are reported in Table 1 at different temperatures. Other quantities such as mole fractions or molar volumes may be easily calculated from these data. Note that for the gas phase and especially at 392 K many values of $C_1$ were set to zero because values lower than 0.005 mol·L$^{-1}$ were found to have no significance. Such a zero value does not mean that there is no decalin in the gas phase; it only means that the concentration was not measurable in the present state of our instrumentation. Plots of $C_1$ vs pressure and $C_2$ vs pressure are presented in Figures 1 and 2, respectively. Coexistence curves are also presented in Figure 3 as a plot of pressure vs mole fraction of propane for the different isotherms investigated.

As already noted,[6] the critical pressure increases with temperature since the temperature is only a few degrees higher than the critical temperature of propane (369 K).

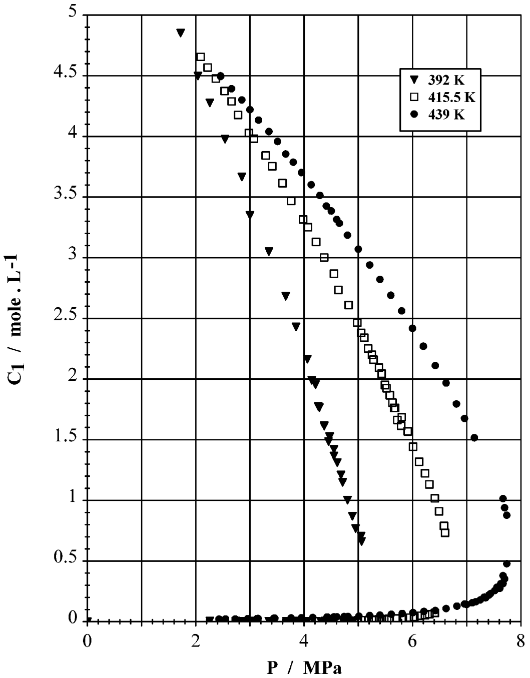

**Figure 1.** Concentration of decalin as a function of pressure in the gas and liquid coexisting phases for the mixture decalin (1) + propane (2): ▼, 392 K; □, 415.5 K; ●, 439 K.

The critical line originating from the critical point of the lighter component generally goes upward according to the

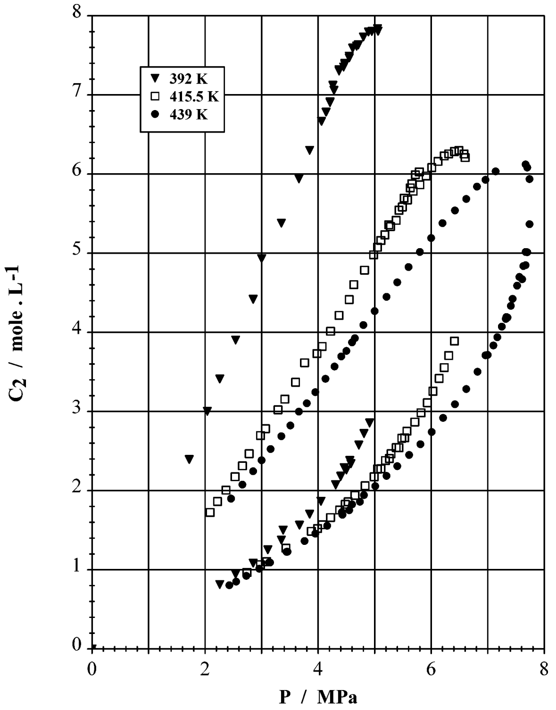

**Figure 2.** Concentration of propane as a function of pressure in the gas and liquid coexisting phases for the mixture decalin (1) + propane (2): ▼, 392 K; □, 415.5 K; ●, 439 K.

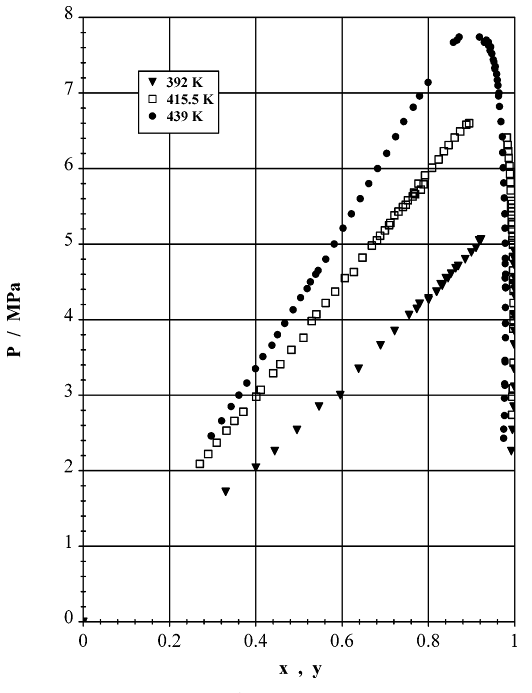

**Figure 3.** Phase diagrams of decalin (1) + propane (2): ▼, 392 K; □, 415.5 K; ●, 439 K. $x,y = C_2/(C_1 + C_2)$ = mole fractions of propane in liquid and gas phases.

known behavior of binary phase diagrams classified by Van Konynenburg and Scott.[8]

There were no data in the literature for comparison with this work.

***Propane* + *Naphthalene Mixture.*** Molar concentrations $C_1$ (naphthalene) and $C_2$ (propane) are reported in Table 2 at different temperatures. Here again, values lower than 0.005 mol·L$^{-1}$ were set to zero for the same reason as above. Plots of $C_1$ vs pressure and $C_2$ vs pressure are presented in Figures 4 and 5, respectively. Coexistence curves are also presented in Figure 6 as a plot of pressure vs propane mole fraction for the different isotherms investigated. Again, the critical pressure increases with temperature for the same reason as above.

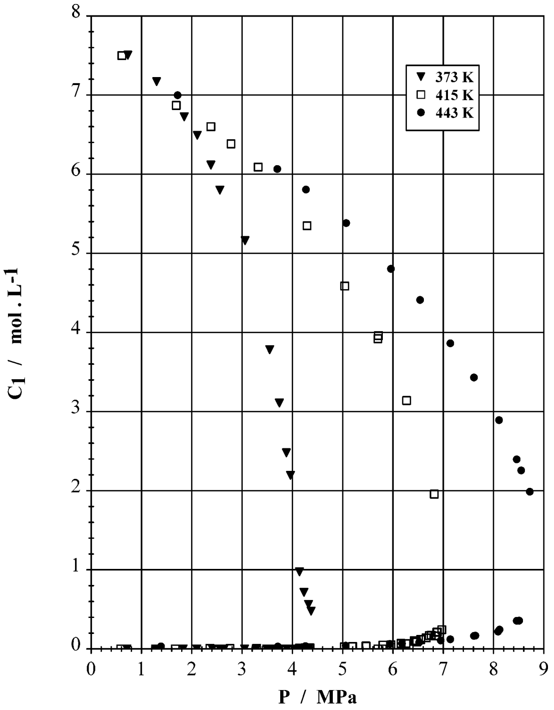

**Figure 4.** Concentration of naphthalene as a function of pressure in the gas and liquid coexisting phases for the mixture naphthalene (1) + propane (2): ▼, 373 K; □, 415 K; ●, 443 K.

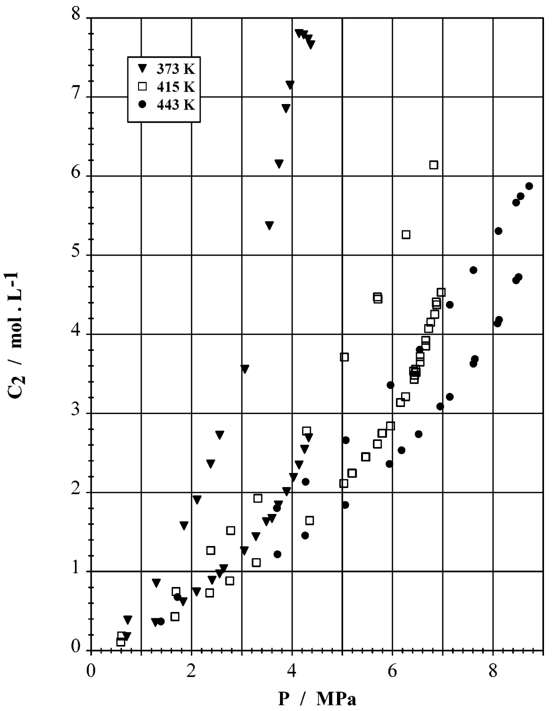

**Figure 5.** Concentration of propane as a function of pressure in the gas and liquid coexisting phases for the mixture naphthalene (1) + propane (2): ▼, 373 K; □, 415 K; ●, 443 K.

Note the unusual shape of the bubble curve at 373 K. Similarly shaped bubble curves have been reported by Peters et al.[9-12] in their studies on different mixtures of propane with heavy hydrocarbons. This may be understood if we remember that the critical temperature of propane is 369 K. It seems that the critical line beginning at the

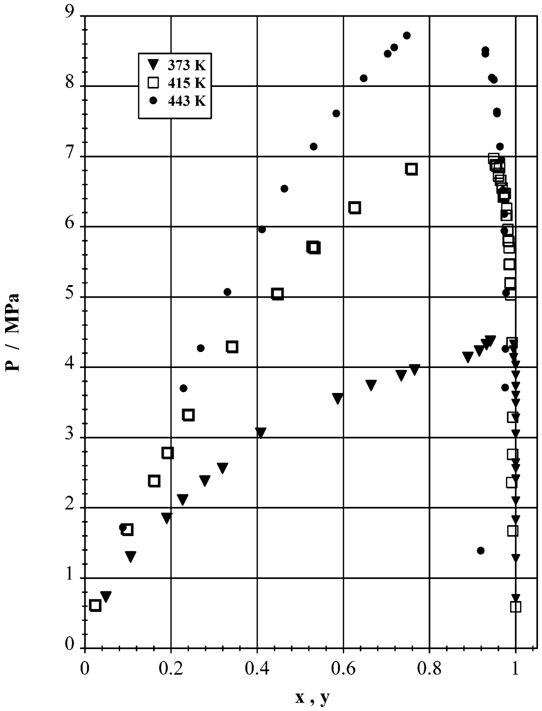

**Figure 6.** Phase diagram of naphthalene (1) + propane (2): ▼, 373 K; □, 415 K; ●, 443 K. $x,y = C_2/(C_1 + C_2)$ = mole fractions of propane in the liquid and gas phases.

critical point of propane is a short one ending with a critical end point at a temperature between 373 K and 415 K. Thus, the binary phase diagram may be either a type III or IV.[8,13] At this temperature (373 K), there is probably a third-phase equilibrium line around 4.1 MPa, although we could not see it during the measurements. It is also likely that a second-phase equilibrium (i.e., liquid−liquid) is present in a narrow range of compositions around 0.85 mole fraction of propane. See, for example, Figure 5c of ref 13. We did not observe this second equilibrium probably because the overall mole fraction did not fall in the narrow range where this equilibrium is present.

There were no experimental data in the literature on this system. Only Peters et al.[9−12] have mentioned a previous study on this mixture but do not give experimental data to which we could compare. Note, however, that these authors do not find a critical end point for this system while they report one for other propane + polyaromatic hydrocarbon binary mixtures.

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