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Phase behavior of the binary system CO₂ + indane from 293.15 K to 360.15 K: measurements and modelling

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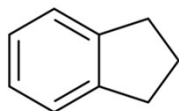
1 - Introduction and objectives

- Binary systems containing CO₂ and common hydrocarbons have been extensively measured: a large amount of data is available
- A lack of data is observed for systems containing uncommon compounds such as heterocyclic hydrocarbons
- Naphtenoaromatic compounds (tetralin, indane...) can be found in significant proportion in some crude oils
- **The aim of this work is to study the phase behavior of the binary mixture CO₂ (1) + indane (2)**
- To do so, two experimental devices were used:
 - A classical variable volume high-pressure cell (for visual observation of phase transitions)
 - A small volume high-pressure microscopy cell
- The Peng-Robinson equation of state was used to correlate the experimental results obtained

2 - Experimental section

Materials

- ❖ CO₂ (CAS 124-38-9) was supplied by Linde with a mass fraction purity higher than 0.999
- ❖ Indane, C₉H₁₀ (CAS 496-11-7) was supplied by TCI with a mass fraction purity of 0.997 (information provided by the manufacturer and confirmed by gas chromatography analyses performed in our lab)



Experimental device

- ❖ First, a thermo-regulated variable volume high-pressure cell comprising two sapphire windows was employed to visually determine phase transitions. The cell was equipped with an optical fiber to illuminate the mixture and an endoscope for the visualization of the fluid. However, in some parts of the phase diagram, it was not possible to accurately measure saturation pressures due to the cloudy appearance of the mixture and the absence of a marked transition phenomenon.
 - ❖ In a second time, a high-pressure microscopy (HPM) cell was connected to the equilibrium cell previously described and another one to enable the fluid to flow in the system. Figure 1 schematically exhibits the full experimental device whereas Figure 2 presents in detail the HPM cell.
- The HPM cell is mainly composed of a stainless-steel block in which two transparent windows are positioned face-to-face. The gap between the windows is limited to 1 mm, thus creating a thin volume in which the studied mixture is flowing. Thanks to this small sample thickness, the observation of complex mixtures behavior can be significantly improved. The HPM cell is equipped with an optical system composed of a video microscope unit and an objective lens (50× magnification).

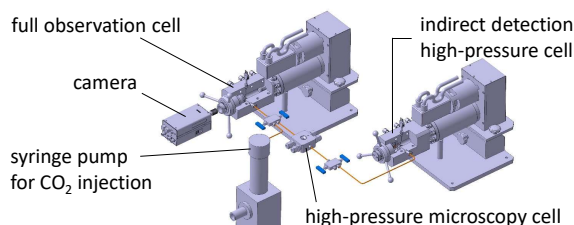


Figure 1. schematic description of the experimental device used in this work

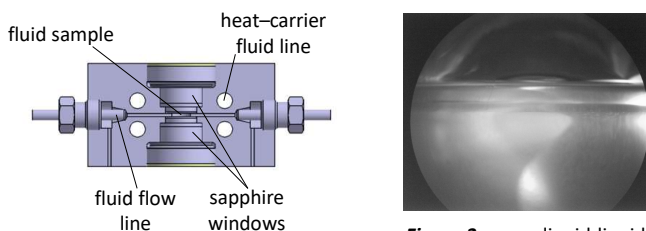


Figure 2. schematic description of the high-pressure microscopy cell

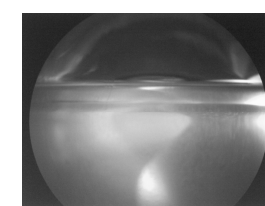


Figure 3. vapor-liquid-liquid equilibrium of the CO₂ + indane mixture in the UCEP vicinity ($T = 313.1 \text{ K}$; $P = 80.2 \text{ bar}$)

3 - Experimental results and modeling

- ❖ 55 bubble points were measured at 12 different compositions ranging from 0.200 to 0.975 CO₂ mole fraction. The phase behavior has been studied at temperatures ranging from 293.15 K to 360.15 K
- ❖ Vapor-liquid-liquid phase transitions were detected (see Figure 3), coordinates of the three-phase line were thus measured including the upper critical end point (UCEP). $T_{UCEP} \approx 313.1 \text{ K}$; $P_{UCEP} \approx 80.2 \text{ bar}$
- ❖ The studied binary system probably belongs to type III binary systems according to the classification of van Konynenburg and Scott
- ❖ In some parts of the phase diagram, it was not possible to accurately measure saturation pressures. Indeed, in the liquid-liquid phase splitting region as well as for rich-CO₂ compositions, precise observation of the fluid using the HPM cell revealed that the phase transition was not an abrupt phenomenon but rather a continuous process with, at the beginning, the appearance of hardly detectable thin droplets
- ❖ The Peng-Robinson equation of state was used to correlate the experimental points. van der Waals mixing rules were employed and a binary interaction parameter (k_{ij}) of 0.1 was found to correctly describe the phase behaviour
- ❖ Experimental data are partly depicted in Figure 4 with isothermal calculated curves

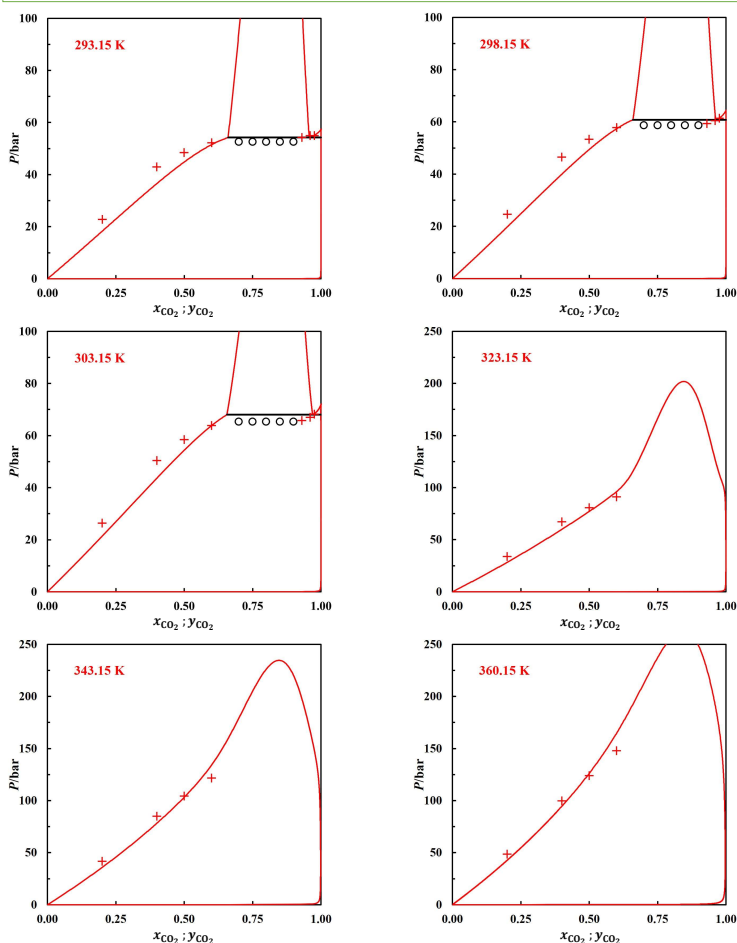


Figure 4. isothermal phase equilibria of the binary system CO₂ + indane
+ : experimental bubble points ; o : experimental points located on the three-phase line (vapor-liquid-liquid phase transitions) ; red curve: calculated saturation curve ; black horizontal line: calculated three-phase line

4 - Conclusions

- A high-pressure device and a synthetic method were employed to measure new saturation pressures for the CO₂ (1) + indane (2) binary system
- The system CO₂ (1) + indane (2) was studied for the first time in this work
- Bubble points were measured from 293.15 K to 360.15 K
- A three-phase line was detected and measured from 293.15 K to the UCEP, pointing out that the mixture likely belongs to type III binary systems
- Experimental data obtained in this work were fitted by means of the Peng-Robinson equation of state with classical mixing rules

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