**Investigation of Mass Transport and Crystallization Processes of Acoustically Levitated Droplets under Elevated Pressures**

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**Highlights**

* Acoustic Levitation
* Wall-Contact-Free Crystallization
* Gas Hydrates
* Physical Properties & Mass Transport

**1. Introduction**

Mass transport and crystallization experiments are among the most challenging measurements in the field of process engineering and chemistry. The results of these studies provide an important basis for many chemical engineering processes and other technical systems. Mass transport studies of liquids in the presence of dense gases are usually performed with the aid of the pendant-drop-method. Especially, in case of phase-conversion processes, which cause a solidification of the hanging droplet, the disadvantage is the contact between the droplet and the capillary, which is influencing the solidification process. Therefore, a non-contact method would be desirable to observe the mass transport and crystallization processes, e. g. hydrate formation. The acoustic levitation represents a possibility to examine samples without contact to any wall [1] [2].

The modification of a high-pressure-view-cell led to a successful implementation of an acoustic levitator into a pressure- and temperature resistant housing (pressure maximum = 20 MPa, temperature maximum = 453 K). The new measurement device allows the investigation of gas hydrate formation at a contact-free acoustically levitated droplet under elevated pressures.

In preliminary investigations it was possible to show the formation and controlled decomposition of carbon dioxide hydrates. The formation of carbon dioxide hydrates was observed for the first time at acoustically levitated water droplets.

**2. Methods**

The experiments were carried out with a high-pressure view-cell including an integrated acoustic levitation device. The compact laboratory device L800 is well-suited for accurate measurements of mass-transport mechanisms of small samples. The specifically developed software detects the acoustically levitated sample automatically and analyzes the contour of the droplet. With this information the volume of the rotationally symmetric sample can be measured and documented.

**3. Results and discussion**

Previous experiences according to gas hydrate formation measurements show that the exact determination of the transition time of larger test samples cannot be simply done. As a result, it is easier to observe one droplet in order to measure its phase change behaviour and the corresponding test parameters. Furthermore, the small sample has got an almost uniform temperature distribution during the measurement. The images illustrate an example of an experiment regarding the formation of a CO2-hydrate (Fig. 1).

The continuous phase is CO2 with a constant pressure of 2.9 MPa. The temperature of the continuous phase is about 281 K. Shortly after the injection of the water droplet, the cooling of the continuous phase is started. When a temperature of about 279 K is reached the bottom of the droplet begins to solidify. Then the crystal growth covers the entire droplet. The whole process from a liquid droplet to a gas hydrate was in this case about 8 minutes long. The diagram (Fig. 1) contains the measurement results compared with literature data. Each measurement point is a result of the arithmetic average of three measurement points.

**Figure 1**: Contact-free CO2-hydrate formation, p= 2.9 MPa / T = ca. 281 K (start) – 279 K (end) [1] / pT-diagram [3] [4]



**4. Conclusions**

Through the successful combination of a high-pressure measurement cell and an acoustic levitator it was possible for the first time to observe the formation of CO2 gas hydrates contact-free. It was necessary to develop a method for injecting liquid water droplets in the pressurized view-cell without an unwanted crystallization in the pipes. Finally, it was possible to acoustically levitate a water droplet and to observe its phase change from liquid to solid state. The acquired measurement data were compared with literature data. A good agreement with literature data can be confirmed. Consequently, the new apparatus L800 is suitable to measure gas hydrate formation regarding sample volume, composition and morphology at the same time.

**References**

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