Onset of Nucleation for Carbon-Dioxide Gas-Hydrates

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Hydrate formation is a key issue in process engineering, oil industry and the sequestration of carbon dioxide. While generally there has been intense fundamental research on equilibrium properties and structure analysis during the last years, research on hydrate formation has been performed mostly at an applied level. Basic studies focusing on hydrate-formation from a bulk phase or focusing on specific physical or physicochemical aspects are comparatively rare. Therefore, this contribution focuses on the experimental investigation of onset conditions of nucleation of carbon dioxide hydrates from gas-saturated water. To this end we have developed a new high-pressure apparatus (maximum p = 70 MPa) which allows for the preparation of a defined binary mixture of the hydrate forming gas and water under constant temperature. By a rapid change (typically 1-5 ms) of system pressure supersaturation is achieved within the system. Conditions of pressure and temperature can be controlled without a change of the composition of the sample. After attaining a critical supersaturation a first-order phase transition is induced resulting in the formation of gas hydrates in the bulk phase. This process is monitored by light scattering and extinction measurements with high temporal resolution. By a systematic variation of the conditions of hydrate formation the onset of nucleation and the induction time are determined. The experiments indicate a strong variation of the occurrence of hydrate formation and induction times depending on the degree of supersaturation. The results are discussed in terms of classical nucleation theory.