

Experimental Indication of Regimes of Heterogeneous and Homogeneous Nucleation of Carbon Dioxide Gas-Hydrates

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The formation of gas hydrates is one of the unwanted issues in plant operation under conditions of elevated pressures, humidity and low temperatures. Industrial applications focusing either on the prevention of hydrate formation or – in case hydrates are formed - on the avoidance of blockages by a suitable flow assurance. Kinetics of hydrate formation as an example of a first order phase transition, involve the classical steps as the formation of a nucleus, growth and coarsening. Depending on the conditions of formation, either heterogeneous or homogeneous nucleation is preferred. In the major cases of industrial processes and experimental scientific studies, heterogeneous nucleation at interfaces is observed.

Unfortunately, basic research on the kinetics of hydrate formation taking into account the different aspects of equilibrium thermodynamics and the kinetics of phase transitions is scarce.

In this work on basis of a pre-characterization of onset-conditions of the formation of gas hydrates from carbon-dioxide saturated water, characteristic times and related nucleation rates for different degrees of supersaturation have been determined.

The experiments have been performed in an experimental set-up, which allows for a spatial separation of the preparation of binary mixtures and the determination of phase transitions induced by fast pressure changes. This concept allows for an independent control of temperature and pressure without a change of the composition of a sample.

Results indicate a strong variation of induction times / nucleation rates of hydrate formation at different degrees of supersaturation. Two different regimes, which are interpreted as heterogeneous and homogeneous nucleation, respectively, are observed.