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Experimental Quantification and Modeling of Dissolved Gas during Hydrate Crystallization: CO_2 Hydrate Case

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Abstract : Gas hydrates have long been considered as problematic for flow assurance in natural gas and oil transportation. On the other hand, they are now seen as future promising materials for various applications (i.e. desalination of seawater, natural gas and hydrogen storage, gas sequestration, gas combustion separation and cold storage and transport). Nonetheless, a better understanding of the crystallization mechanism of gas hydrate and of their formation kinetics is still needed for a better comprehension and control of the process. To that purpose, measuring the real-time evolution of the dissolved gas concentration in the aqueous phase during hydrate formation is required. In this work, CO_2 hydrates were formed in a stirred reactor equipped with an Attenuated Total Reflection (ATR) probe coupled to a Fourier Transform InfraRed (FTIR) spectroscopy analyzer. A method was first developed to continuously measure in-situ the CO_2 concentration in the liquid phase during solubilization, supersaturation, hydrate crystallization and dissociation steps. Thereafter, the measured concentration data were compared with those of equilibrium concentrations. It was observed that the equilibrium is instantly reached in the liquid phase due to the fast consumption of dissolved gas by the hydrate crystallization. Consequently, it was shown that hydrate crystallization kinetics is limited by the gas transfer at the gas-liquid interface. Finally, we noticed that the liquid-hydrate equilibrium during the hydrate crystallization is governed by the temperature of the experiment under the tested conditions.

Keywords : gas hydrate, dissolved gas, crystallization, infrared spectroscopy
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