

Measurement of clathrate hydrate precipitation from CO₂ solution by a nondestructive method

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ABSTRACT

Using recorded data of pressure and temperature, we developed a nondestructive method to estimate the precipitation rate of carbon dioxide (CO₂) hydrate by considering the CO₂ solution density equation in association with the mass conservation equation. We applied this method to the investigation of the dynamic process of clathrate hydrate precipitation from CO₂ solution using a high-pressure dissolution system consisting of a high-pressure vessel and optical detecting instruments. The role of stirring was examined. The temperatures studied in the experiments were from 275 to 288 K over a pressure range of 4–8 MPa. Experimental results showed that our method can quantitatively monitor this dynamic process. The volume ratio of precipitated hydrate to that of the pressure-vessel approached 0.09 when a steady state was reached, which took about 100 seconds; more than 60% of the total clathrate hydrate precipitated within 10 seconds at the cooling rates of this experiment. For precipitation, stirring enhanced the cooling and led to a large hydrate precipitation rate from 0.004 to 0.008 in volume ratio per second. For nucleation, however, the residual structure of the solution decreased the amount of hysteresis in the formation of hydrate nuclei during supercooling, of which induction times were reduced by 4–9 minutes.