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Formation Behaviors of CO₂ Hydrate in Kaoline and Bentonite Clays with Partially Water Saturated

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Abstract

CO₂ hydrates can be used for long term storage of CO₂ in the deep ocean. Clay minerals are the most important components of the marine sediments. In this study, the formation behaviors of carbon dioxide hydrate in kaoline clay and bentonite clay with partially water saturated were studied in a closed system. The mass ratio of water to Kaoline and Bentonite clays is 20%. The experiments were carried out at the temperature of 279.15 K and the initial formation pressure range of 4.8–3.4 MPa. The liquefaction of the carbon dioxide was observed in the cooling process by setting the temperature of the water bath to the valued experimental temperature. The temperature in the crystallizer begins to rise quickly when the hydrate starts to form and then decreases gradually after reaching the highest value. For the experiments in kaoline clay, the temperature decreases gradually along with the equilibrium hydrate formation temperature, and the hydrate formation rate is controlled by the heat transfer process. The final conversion of the water to hydrate increases with the increase of the initial formation pressure and restrained by the equilibrium hydrate formation pressure. For the experiments in bentonite clay, the temperature during the hydrate formation is much higher than the equilibrium temperature for CO₂ hydrate formation. The final pressure is much higher than the equilibrium pressure of CO₂ hydrate, and increases with the increase of the initial formation pressure, indicating that the equilibrium pressure CO₂ hydrate in bentonite clay is higher than the bulk CO₂ hydrate.

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1. Introduction

CO₂ capture and the geological storage have been considered an effective strategy to mitigate the climate change.[1–3] Gas hydrates have great capacity to store gas since the volume of CO₂ gas stored in 1 m³ of CO₂ hydrate is about 120–160 m³ at standard temperature and pressure.[4] An additional advantage of hydrate storage is the reduced transport of CO₂ in the reservoir compared to CO₂ stored in a fluid state.

In practical situations, storage of CO₂ hydrates is to be carried out in reservoirs under the ocean seafloors, and hence it is essential to understand the CO₂ hydrate formation in porous media. Several researches focusing on the CO₂ hydrate formation behaviors in porous sediments have been presented. Kumar et al.[5] studied the effect of different surfactants on the formation kinetics of CO₂ hydrate in silica gels. They observed that SDS is more effective in reducing the induction time and also in enhancing the rate of hydrate formation. Babu et al.[6] studied the CO₂ hydrate formation kinetics in silica sand and silica gels. They observed that the water to hydrate conversion in the silica sand is much higher than that in silica gels. Kang et al.[7] studied the precombustion capture of CO₂ in silica gel pores from a simulated flue gas. A higher rate of hydrate formation CO₂ hydrates in silica gel matrix was observed. Mohammadi et al.[8] carried out kinetic study of CO₂ hydrate formation in the

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presence of surfactant and silver nanoparticles. They found that the presence of surfactant and silver nanoparticles increases the storage capacity for CO₂ hydrate formation but does not decrease the induction time for hydrate formation. Mekala et al.[9] investigated the effects of the particle size of silica sand on the CO₂ hydrate formation kinetics. They found that the gas consumption of CO₂ in hydrate increases with the decrease of the particle size. The silica sand with particle of 0.46 mm provides an optimum environment for efficient hydrate formation due to the higher average rate of hydrate formation.

Clays are the main components of the marine sediments. However, the information on the formation kinetics of CO₂ hydrate in clays is seldom known in an open literature. In the present study, the formation behaviors of CO₂ hydrate in kaoline and bentonite clays with partially water saturated were studied in a closed system. The mass ratio of water to kaoline and bentonite clays is 20%. The experiments were carried out at the formation temperature of 279.15. The initial temperature is 288.15K and the initial formation pressure range of 4.8-3.4 MPa.

2. Experimental section

2.1 Experimental Apparatus

The experimental apparatus, shown in Figure 1, consists of the high-pressure hydrate crystallizer (CR), the supply vessel (SV), the gas/liquid supply system, the temperature-controlled water bath and the data acquisition system. The effective maximum volume of the crystallizer and the supply vessel is 61.4 cm³ and 250 cm³, respectively. The cell of crystallizer is volume-variable with a movable piston. Two pressure transducers (MBS3000) are employed for pressure measurement, with a maximum certainty of 0.01% of the span (0-25MPa). A Pt100 thermoprobe is used to measure the temperature in the crystallizer within a precision of ± 0.1 K. The crystallizer temperature is controlled by a temperature-controlled water bath within a range of 263.15-303.15K and with an uncertainty of 0.1 K. The signals of the pressure and the temperature are continuously monitored and recorded by a data acquisition system coupled with a computer.

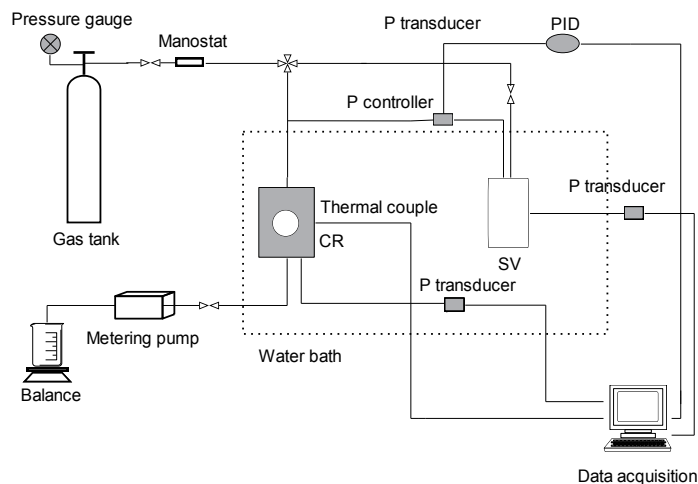


Fig. 1. Schematic design of the experimental apparatus

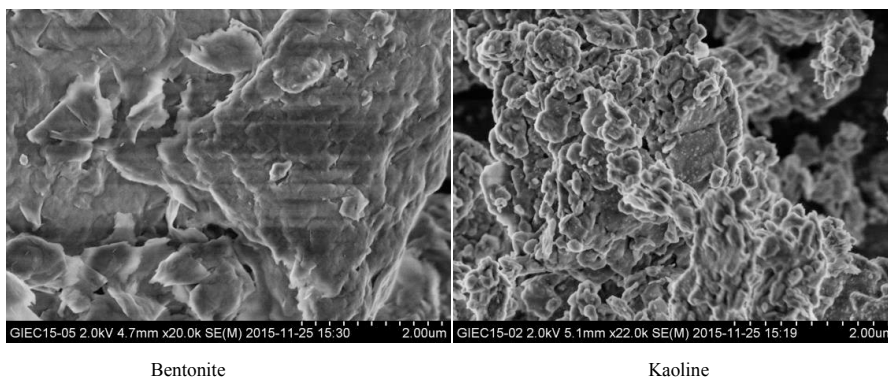


Fig. 2. Electron micrograph of samples

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