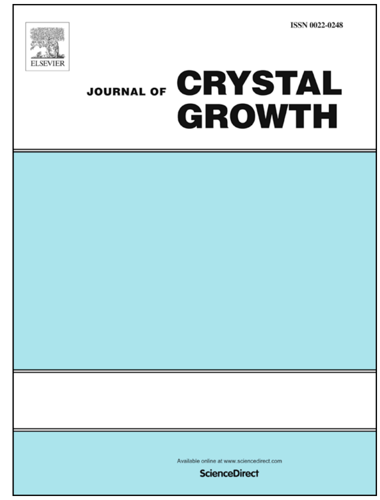

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Observation of the growth characteristics of gas hydrate in the quiescent-type formation method using surfactant

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Abstract
We have observed the growth behavior of gas hydrate with addition of surfactants by several observation methods, and we discuss the mechanism of gas hydrate formation along the wall surface. From observation using colored water, the water moves up through the porous hydrate structure and unreacted water remains in the middle of the porous hydrate. In contrast to previous studies which observed growth from the side, we observed the growth behavior from the top. The hydrate height agrees well with that of water pulled up by capillary action. These results confirm that the driving force for hydrate growth in the vertical direction is capillary action. We discuss the driving force for hydrate growth in the horizontal direction. We suggest that the driving force in the horizontal direction is different because the initial concentration of the surfactant solution affects the growth behavior in the horizontal direction while it does not affect the hydrate height.

Keywords: A1. Growth models, A1. Mass transfer, A1. Heat transfer, A2. Growth from solutions, B1. Gas hydrate

1. Introduction

Natural gas hydrate has great potential in natural gas storage and transportation. The storage and transportation costs of natural gas can be decreased by using natural gas hydrate, because natural gas hydrate is stable at higher temperature and lower pressure conditions than liquid natural gas. Gas hydrates are solid-state materials composed of gas and water, which are called clathrate hydrates. There are many types of gas hydrates depending on the guest gas, such as methane hydrate, ethane hydrate, and propane hydrate. In the present work, we used propane hydrate because of its manageability in ambient conditions.

It is difficult to continuously form gas hydrates at a high production rate because the formed hydrate covers the surface of water, which inhibits the reaction between the gas and water. Expanding/renewing the gas–liquid interface is necessary to increase the production rate. In industrial fields, splaying [1] and bubbling methods [2] are commonly used, which can be classified as stirred systems. Quiescent systems, in which mechanical agitation or pumping of gas/water is not required, have some advantages. For example, the facility can be simplified and the operation/maintenance costs can be reduced. Various attempts have also been made to enhance the production rate in quiescent systems. Kumano et al. [3] used a fiber layer to increase the gas–liquid interface. Kuhs et al. [4] and Teraoka et al. [5] used fine ice particles. Addition of a surfactant is an effective and convenient method because it can be operated with a simple facility.

Addition of surfactant results in continuous formation of gas hydrate without mechanical power, because the gas hydrate grows up along the side wall of the container. A number of researchers have investigated the mechanism of gas hydrate formation [6-12]. They suggest that the driving force for gas hydrate formation should be capillary action, which continuously transports water/surfactant solution to the surface of the gas hydrate. To further investigate the mechanism, Veluswamy et al. [13], Lim et al. [14], Yoslim et al. [15], and Okutani et al. [11] observed the growth of gas hydrates along the wall surface using surfactant solution. However, the gas hydrate covered the observation window and prevented observation of its growth. Hayama et al. [16, 17] observed gas hydrate formation from droplets of surfactant solution and investigated the effect of the surfactant concentration. Lee et al. [18] observed the growth of hydrate around gas bubbles