Storage Performance and Structural Characteristics of Natural Gas Hydrate

Natural gas hydrate (NGH) pellets are one candidate for a natural gas storage and transportation medium due to their high gas density and much easier storage method compared to liquefied natural gas (LNG) and compressed natural gas (CNG). More than 80% of natural gas stored within NGH pellets with different particle sizes from 0.50 mm to 30 mm was maintained for two weeks at 253 K under ambient pressure. The internal texture of the NGH pellets was investigated using phase contrast X-ray CT by diffraction enhanced imaging (DEI), revealing that the outer surface of the pellet was covered with an ice film and NGH was well preserved inside.

The production of natural gas from both conventional and unconventional fields is increasing because of the expanding worldwide demand for natural gas. Therefore, energy transportation systems have diversified not only to pipelines and LNG, but also new means such as gas to liquids (GTL), natural gas liquids (NGL), natural gas hydrate (NGH) [1-6]. NGH is a clathrate crystal that consists of water and natural gases such as CH₄, C₂H₆, and C₄H₁₀, with natural compositions. NGHs are usually stable at high pressure and low temperature, and can contain natural gas of approximately 170 times their own volume. It is known that CH₄ and CO₂ hydrates are preserved just below the freezing point of water under atmospheric pressure, though the conditions are outside the thermodynamically stable zone of CH₄ and CO₂ hydrates [7-11]. This is called the self-preservation phenomenon [7]. The surfaces of self-preserved CH₄ and CO₂ hydrates are covered with ice [9-12], so the distribution of ice film is thought to be important for the preservation of gas hydrate. NGH also has lower NGH fractions at the start of the storage test and smallest. This result showed that smaller NGH samples had lower NGH fractions at the start of the storage test and were likely to dissociate during the storage period. This is because the smaller NGH particles have larger specific surface areas. Thus, larger particles of over approximately 10 mm in diameter of NGH pellets are more favorable than smaller ones from the viewpoint of gas storage.

Non-destructive measurement by DEI was performed on the NGH pellets to visualize the internal distribution of NGH and ice with a spatial resolution of 0.040 mm. A 38-keV monochromatic synchrotron X-ray at a vertical wiggler beam was irradiated to an NGH pellet that was immersed in methyl acetate at 193 K ± 1 K to eliminate artifacts caused by the outer surface of the pellet [12, 13]. Phase map images were obtained by detecting the X-ray beam that diffracted after passing through the sample using the analyzer crystal. The sample was scanned at 11 positions with the scanning time of one second for each position, and the number of projections was 500. An internal image of the NGH pellet is shown in Fig. 2. The difference in density of the NGH pellet is reflected in the gray scale [Fig. 2(a)]. The white region shows ice whose density was lower than that of the surroundings, and the gray and black areas were NGH and methyl acetate, respectively. The observed result showed that only some part of the surface was covered with ice film [Fig. 2(b)]. But this is due to the sample that was shaved at a temperature below 150 K, a condition which suppressed NGH dissociation, to suitable size for the DEI measurement. Considering the good storage performance as shown in Fig. 1 and its similarity with CH₄ hydrates, the NGH pellet in common state was undoubtedly thought to be fully covered with ice film. An ice film with a thickness of approximately 0.30 mm was observed on the pellet, and the inside of the pellet was dense with NGH without ice grains or pore spaces. Consequently, the formed NGH pellet that maintained natural gases under mild storage conditions was proved to be stabilized by the surface thin ice film.

REFERENCES

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