Experimental study on the phase equilibrium condition of carbon dioxide hydrate with sediments

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Gas hydrate has been found in marine sediments around Japan and been attracting attention as an unconventional natural gas resource. It forms under the environment where enough amounts of gas-water coexist and temperature-pressure satisfy the stability condition, which may change easily in response to the surrounding physicochemical environment. A number of experimental studies have been conducted to examine the effects of sediments on the gas hydrate stability, little quantitative research has, however, reported the physicochemical effects of particle-scale phenomena. Detailed examinations of such fundamental effects lead to further understanding of the nature of gas hydrates. In this research, we carried out artificial gas hydrate experiment using carbon dioxide in order to discuss the formation/dissociation conditions under the presence of natural sediments.

Carbon dioxide hydrate was formed using a pressure vessel with sediment collected from the seafloor where shallow gas hydrate accumulated in the Japan Sea. The formation pressure of the hydrate with 10wt% sediment was higher than that without any sediments.This is probably due to the increased liquid-solid boundaryby fine sedimentsand to the adsorption of water molecules with structures similar to hydrate lattice on clay minerals, which may promote the nucleation of gas hydrate.The dissolution temperature of hydrate with sediment was lower than that for the sediment-free condition, which reduced the stability region slightly.This indicates that the depression of solidifying points due to the leached ions from the sediments, the influence of adsorption by clay minerals, and the increase of solid-liquid contact area due to the presence of clay minerals. These microscopic effects may constrain the stability of gas hydrate, which largely control the distribution and occurrence of natural gas hydrates.