Time-dependent observation of cage-specific CH_4-CO_2 replacement behaviors in SI hydrate

목정훈, 최원중, 서용원†
UNIST
(ywseo@unist.ac.kr†)

Natural gas hydrates (NGH) are expected to be our future energy resources, and CH_4-CO_2 replacement has been considered as a promising NGH production technology, which can act as both energy recovery and CO_2 sequestration. In this study, the effect of CO_2 injecting pressures on the replacement behavior in sI hydrate was investigated. To identify the replacement efficiency depending on CO_2 injecting pressures, the compositions of the hydrate phase were measured using GC. In addition, Raman spectroscopy, ^{13}C NMR, and PXRD were used to observe the structural information of gas hydrates and the changes in cage occupancies of each guest molecule during the replacement. Raman spectra showed that the cage occupancy ratio of CH_4 molecules in the large and small cages continued to decrease as the replacement proceeded due to the preferential replacement by CO_2 molecules in the large cages. The cage occupancies of CH_4 and CO_2 obtained from PXRD analysis confirmed that more than 70% of the CH_4 molecules in the large cages were replaced by CO2 molecules and CO2 occupation in the large cages was more significant at a higher CO_2 injecting pressure.