American Mineralogist, Volume 100, pages 1203–1208, 2015

Insights into the structure of mixed CO_2/CH_4 in gas hydrates

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Abstract

The exchange of carbon dioxide for methane in natural gas hydrates is an attractive approach to harvesting CH_4 for energy production while simultaneously sequestering CO_2. In addition to the energy and environmental implications, the solid solution of clathrate hydrate $(CH_4)_{1-x}(CO_2)_x \cdot 5.75H_2O$ provides a model system to study how the distinct bonding and shapes of CH_4 and CO_2 influence the structure and properties of the compound. High-resolution neutron diffraction was used to examine mixed CO_2/CH_4 gas hydrates. CO_2-rich hydrates had smaller lattice parameters, which were attributed to the higher affinity of the CO_2 molecule interacting with H_2O molecules that form the surrounding cages, and resulted in a reduction in the unit-cell volume. Experimental nuclear scattering densities illustrate how the cage occupants and energy landscape change with composition. These results provide important insights on the impact and mechanisms for the structure of mixed CH_4/CO_2 gas hydrate.

Keywords: Neutron diffraction, methane hydrate, carbon dioxide/methane exchange, Fourier density maps