

# Diffusion Model for Gas Replacement in an Isostructural CH<sub>4</sub>-CO<sub>2</sub> Hydrate System

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## Abstract

© 2017 American Chemical Society. Guest exchange in clathrates is a complex activated phenomenon of the guest-host cage interaction on the molecular-scale level. To model this process, we develop a mathematical description for the nonequilibrium binary permeation of guest molecules during gas replacement based on the microscopic "hole-in-cage-wall" diffusive mechanism. The transport of gas molecules is envisaged as a series of jumps between occupied and empty neighboring cages without any significant lattice restructuring in the bulk. The gas exchange itself is seen as two-stage swapping initiated by almost instantaneous formation of a mixed hydrate layer on the hydrate surface followed by a much slower permeation-controlled process. The model is constrained by and validated with available time-resolved neutron diffraction data of the isostructural CH<sub>4</sub> guest replacement by CO<sub>2</sub> in methane hydrate, a process of possible importance for the sequestration of CO<sub>2</sub> with concomitant recovery of CH<sub>4</sub> in marine gas hydrates. (Graph Presented).

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