Storing CO_2 in marine gas hydrate reservoirs: Resolving the sediment microstructure

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The reaction of CO_2 and water to CO_2 hydrate in the ocean at water depths of 300 mbsl or more opens the possibility of using engineered injection to immobilize and trap anthropogenic CO_2 long-term within deep marine sediments. The presence of natural methane hydrates in sediments targeted for such CO_2 storage adds further appeal, since the liberated methane could be used as a carbon-neutral hydrocarbon energy resource. The exchange of methane for CO_2 in hydrates is a spontaneous process provided that both hydrate species and binary mixtures thereof are thermodynamically stable. Recent field tests have shown that this strategy is a promising approach. Industrial-scale applications of this method require identification of fluid-flow characteristics as well as thermodynamic and kinetic controls on the reactions, since these affect, for example, the hydraulic conductivity and geomechanic stability of the seabed. Due to experimental limitations, localized reactions and fluid-flow phenomena responsible for these effects remain unresolved thus far at both microscale (1 μm – 1 mm) and near-natural, reservoir conditions. We address this deficit by showing results from high-resolution, two-dimensional Raman spectroscopy mappings of a natural hydrate reservoir analogue before, during and after injection of liquid CO_2 under realistic reservoir conditions. Macroscale properties of the sample were inferred from the mapped distribution of sand grains, pore water, hydrate phases and CO_2. Moreover, the time-resolved experiments allow us to 1) determine hydrate conversion rate and efficiency and 2) reveal effects of shifting fluid pathways on CO_2 retention and methane yield. The Raman data reveal that the conversion of single hydrate grains follows a shrinking core model controlled by diffusion of exchanged hydrate guests through a thickening shell of converted hydrate growing toward grain cores. We show that the conversion can be modeled simply by using 1) published permeation coefficients for CO_2 and methane in hydrates and 2) hydrate grain size as the only input parameters.