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CO_2 injection into submarine, CH_4-hydrate bearing sediments: Parameter studies towards the development of a hydrate conversion technology

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In the recent past, international research efforts towards exploitation of submarine and permafrost hydrate reservoirs have increased substantially. Until now, findings indicate that a combination of different technical means such as depressurization, thermal stimulation and chemical activation is the most promising approach for producing gas from natural hydrates. Moreover, emission neutral exploitation of CH_4-hydrates could potentially be achieved in a combined process with CO_2 injection and storage as CO_2-hydrate.

In the German gas hydrate initiative SUGAR, a combination of experimental and numerical studies is used to elucidate the process mechanisms and technical parameters on different scales. Experiments were carried out in the novel high-pressure flow-through system NESSI (Natural Environment Simulator for $s$ub-Seafloor Interactions). Recent findings suggest that the injection of heated, supercritical CO_2 is beneficial for both CH_4 production and CO_2 retention.

Among the parameters tested so far are the CO_2 injection regime (alternating vs. continuous injection) and the reservoir pressure / temperature conditions. Currently, the influence of CO_2 injection temperature is investigated. It was shown that CH_4 production is optimal at intermediate reservoir temperatures (8 ˚C) compared to lower (2 ˚C) and higher temperatures (10 ˚C). The reservoir pressure, however, was of minor importance for the production efficiency. At 8 ˚C, where CH_4- and CO_2-hydrates are thermodynamically stable, CO_2-hydrate formation appears to be slow. Eventual clogging of fluid conduits due to CO_2-rich hydrate formation force open new conduits, thereby tapping different regions inside the CH_4-hydrate sample volume for CH_4gas. In contrast, at 2 ˚C immediate formation of CO_2-hydrate results in rapid and irreversible obstruction of the entire pore space. At 10 ˚C pure CO_2-hydrates can no longer be formed. Consequently the injected CO_2 flows through quickly and interaction with the reservoir is minimized. Our results clearly indicate that the formation of mixed CH_4-CO_2-hydrates is an important aspect in the conversion process.

The experimental studies have shown that the injection of heated CO_2 into the hydrate reservoir induces a variety of spatial and temporal processes which result in substantial bulk heterogeneity. Current numerical simulators are not able to predict these process dynamics and it is important to improve available transport-reaction models (e.g. to include the effect of bulk sediment permeability on the conversion dynamics).

Our results confirm that experimental studies are important to better understand the mechanisms of hydrate dissociation and conversion at CO_2-injection conditions as a basis towards the development of a suitable hydrate conversion technology. The application of non-invasive analytical methods such as Magnetic Resonance Imaging (MRI) and Raman microscopy are important tools, which were applied to resolve process dynamics on the pore scale. Additionally, the NESSI system is being modified to allow high-pressure flow-through experiments under triaxial loading to better simulate hydrate-sediment mechanics. This aspect is important for overall process development and evaluation of process safety issues.