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## Thermodynamic Conditions and Kinetics of Integrated Methane Recovery and Carbon Dioxide Sequestration

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### Abstract

Today the world is faced with two major energy challenges, i.e., shortage of primary energy sources and climate change. The latter is widely believed to be related to CO<sub>2</sub> released from combustion of fossil fuels. Many options are being considered for reducing the emission of CO<sub>2</sub> to the atmosphere, including using low carbon fuels (e.g., natural gas) and CO<sub>2</sub> capture and storage.

There are vast quantities of methane in the form of gas hydrates in marine sediments and permafrost regions. However, there are many technological challenges in recovering this low carbon fuel. As for CO<sub>2</sub> storage, several techniques have been suggested, including their storage in the form of hydrates in sediments. It might be possible to integrate CO<sub>2</sub> storage with methane gas production.

In this work, we present the results of a series of experiments on the thermodynamic conditions and kinetics of integrated methane recovery and CO<sub>2</sub> sequestration. The preliminary experiments were conducted at different temperature and pressure conditions, in the presence or absence of excess water, in the presence of gaseous or liquid CO<sub>2</sub>. Silica glass beads and a kaolinite-sand mixture were used to simulate marine sediments. Results of the experimental tests show a higher methane recovery rate in the system inside the methane hydrate stability zone (HSZ) and outside the CO<sub>2</sub> HSZ. The presence of excess water noticeably slowed down the CO<sub>2</sub> displacement reaction. It was observed that in the kaolinite-sand mixture methane recovery rate was significantly lower than those observed in the tests with silica glass beads. The experimental results infer that mass transfer plays a crucial role in methane recovery through CO<sub>2</sub> replacement. The study suggests that the thermodynamic conditions inside methane HSZ and outside CO<sub>2</sub> HSZ could be the optimum conditions for integration of methane recovery and CO<sub>2</sub> storage in marine sediments.

### Introduction

It has been identified by seismic survey there are enormous sedimentary deposits of methane hydrates worldwide (Kvenvolden, 1988 and 1993; Milkov, 2004). The methane trapped in the hydrates has been considered as potential energy source in the near future, while the known geological reserves of conventional natural gas and oil are rapidly declining parallel to expanding demands of fossil fuels. In marine sediments under seafloor naturally-occurring gas hydrates may form from either bacterial or thermogenic methane. Bacterial methane, generated by bacteria via either reduction of CO<sub>2</sub> or acetate fermentation, usually forms structure I hydrates, while thermogenic methane is generated from organic matters buried underground (for example, in marine sediments) under high pressure and temperature conditions (Kvenvolden, 1993; Coleman, et al., 1995; Sassen, et al., 1999). Thermogetic methane is always accompanied with non-negligible concentrations of ethane, propane, and butane, and therefore, usually forms structure II hydrates.

Various methods have been suggested for methane recovery from gas hydrates in sediments (Holder, et al., 1984; Sloan, 1998). They include heating, depressurisation, injection of thermodynamic hydrate inhibitors, or their combinations. The principle of these methods is to dissociate gas hydrates by shifting the local thermodynamic conditions in gas hydrate reservoirs out of the hydrate stability zone (HSZ). Apart from the technical difficulties and problems associated with methane recovery from gas hydrates, as described by Holder et al. (1984), methane recovery by these methods may seriously impact seafloor stability (Dawson, et al., 1988; Kayen and Lee, 1991; Booth, et al., 1994), which has been proven by evidence of methane venting through the subsea sediments (Blumier, 2000; Kennett, et al., 2000). Dissociation of gas hydrates also possibly weakens the strength of hydrate-bearing marine sediments, therefore poses hazards to deepwater drilling and gas