

Entrapping CO₂, while Tapping Methane

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Abstract: The injection of carbon dioxide into methane hydrate-bearing sediments causes the release of methane and the formation of carbon dioxide hydrate. This phenomenon known as CH₄-CO₂ replacement creates a unique opportunity to recover an energy resource, methane, while entrapping a greenhouse gas, carbon dioxide. A paper "A comparative analysis of the mechanical behavior of carbon dioxide and methane hydrate-bearing sediments" by Hyodo et al. (2013) investigates stress-strain curves, shear strengths, and the effects of hydrate saturation, effective stress and temperature on the mechanical behaviors of hydrate-bearing sediments that allow us to assessing the feasibility of CH₄-CO₂ replacement method.

Keywords: CH₄-CO₂ replacement, mechanical behavior, carbon dioxide hydrate, methane hydrate.

Methane (CH₄) hydrate is a solid compound in which methane molecules are trapped within cages of hydrogen-bonded lattices of water molecules. Methane hydrate reserves are on the order of 500-to-10,000 Gt of carbon worldwide (Ruppel and Pohlman 2008). Natural methane hydrate, as a potential energy resource because of its vast quantity, is expected to play an important role in supplying natural gases as a part of a national energy portfolio. Methane production from hydrate-bearing sediments requires hydrate dissociation for releasing mobile methane gas in sediments prior to gas production operation. Depressurization, thermal stimulation and chemical injection have been proposed for in-situ gas hydrate production.

In particular, the injection of carbon dioxide, CO₂, into methane hydrate-bearing sediments causes the release of methane, CH₄, and the formation of carbon dioxide (CO₂) hydrate, even if global pressure-temperature conditions remain within the CH₄ hydrate stability field (Ota et al., 2005; Svandal et al., 2006; Zhou et al., 2008). This phenomenon, known as CH₄-CO₂ exchange or CH₄-CO₂ replacement, creates a unique opportunity to recover an energy resource, methane, while entrapping a greenhouse gas, carbon dioxide. CH₄-CO₂ replacement is thermodynamically favorable due to the chemical potential difference between CH₄ and CO₂ hydrate (Seo and Lee, 2001; Svandalet al., 2006). The replacement rates increase near the CH₄ hydrate phase boundary (Ota et al., 2005), with increasing CO₂ pressure (Ota et al., 2007), and higher specific surface of CH₄ hydrate (Kim et al., 1987) (Note. a specific surface represents the total surface area of a material per unit of mass). The CH₄ hydrate cage must separate to release the CH₄ molecule and trap the CO₂ molecule that causes a solid-liquid-solid transformation of hydrate. While CH₄-CO₂ replacement occurs locally and gradually so that the hydrate mass remains solid at pore-scale and no significant change in global stiffness is expected during CH₄-CO₂ replacement at low hydrate saturation (i.e. S_{hyd} = 5-10%) (Espinoza and Santamarina, 2011), pore fluid volumes can still dramatically increase during CH₄-CO₂ replacement at constant pressure in high hydrate saturation (i.e. S_{hyd} > 30%) (Jung et al., 2010). For example, the 390% volume expansion is expected during CH₄-CO₂ replacement for S_{hyd} = 50%. Thus, although the reservoirs with high methane hydrate saturation should be more amenable to CH₄-CO₂ replacement because of high permeability to CO₂, the large interface between CH₄ hydrate and CO₂, and no early CO₂ hydrate clogging, volume-pressure changes associated with CH₄-CO₂ replacement in excess-water reservoirs should be considered. The volume-pressure change may cause increases in fluid pressure, decreases in effective stress and strength loss, volume expansion, and gas-driven fractures if a CH₄ gas phase develops, and the permeability is low enough to prevent pressure dissipation. Therefore, the comparative analysis of the mechanical behavior of carbon dioxide and methane hydrate-bearing sediments investigated by Hyodo et al. (2013) should be the first step to

47 understand the stability of hydrate-bearing sediments during CH₄-CO₂ replacement and assess the
48 feasibility of CH₄-CO₂ replacement method.

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