

The Effect of Swapping Speed on Methane Recovery in Gas Hydrate-Bearing Unconsolidated Sediments

*Dong-Gun Lee¹⁾, Jong-Se Lim²⁾, Joo Yong Lee³⁾, Jaehyoung Lee⁴⁾

^{1), 2)} Department of Energy Resources Engineering, Korea Maritime University,
Busan 606-791, Korea

¹⁾ donggun@hhu.ac.kr, ²⁾ jslim@hhu.ac.kr

^{3), 4)} Petroleum & Marine Research Division, Korea Institute of Geoscience & Mineral
Resources, Daejeon 305-350, Korea

³⁾ jyl@kigam.re.kr, ⁴⁾ ljh1155@kigam.re.kr

ABSTRACT

The aims of this study are to describe the behavior of swapping production in the gas hydrate-bearing unconsolidated sediments, and to analyze how the swapping speed affects CH₄ recovery by constructing an experimental system. The results of the swapping experiment indicate that the soaking process and swapping speed play a critical role in an additional CH₄ recovery mechanism. The swapping efficiency decreases and thus CH₄ recovery decreases as the swapping speed increases. This phenomenon reflects that the swapping speed shows a kind of inhibition effect on the swapping efficiency and the minimum reaction time required for swapping between the CH₄ hydrate and the flue gas. Additional CH₄ recovery was also possible with soaking process. The results of this study can provide an insight into swapping phenomenon in gas hydrate-bearing sediment.

1. INTRODUCTION

Typically methods to recover hydrates include thermal injection, depressurization, and the injection of chemical additives such as CH₃OH or HO(CH₂)₂OH to change the conditions of dissociation (Bayles *et al.* 1986; Kmath and Godbole 1987; Selim and Sloan 1990; Verigin *et al.* 1980; Yousif *et al.* 1990, 1991). The basic mechanism of this production technology is the separation of CH₄ gas by solid hydrate dissociation, so it changes geo-mechanical properties and affects formation stability (Lee *et al.* 2010). The recently proposed swapping method has received attention as an alternative that can resolve the limitation of the existing production technology. We can recover CH₄ gas safely through a swapping phenomenon in which in-situ CH₄ hydrate is replaced by hydrate that has CO₂ and N₂ as guest molecules once the flue gas (CO₂+N₂, CO₂:N₂=2:8) is injected into the gas hydrate-bearing sediment (Ota *et al.* 2005; Yeon *et al.* 2006; Park *et al.* 2006; Shin *et al.* 2008). This production technology is beneficial not

¹⁾ Graduate Student

²⁾ Professor

³⁾ Senior Researcher

⁴⁾ Senior Researcher

only to the production of energy resources, but also to the storage of the greenhouse gases that cause global warming.

In production of gas from gas hydrate-bearing sediments, two things are crucial: (1) investigation of the fluid flow characteristics of the gas hydrate-bearing sediment, and (2) analysis of the production efficiency in relation to the production method (Lee *et al.* 2005). Previous research on the swapping method involved with bulk CH₄ hydrate and pure CO₂ yields approximately 64% CH₄, and with flue gas injection, N₂ molecules can replace CH₄ molecules in the small cavities, yielding approximately 85% CH₄ (Park *et al.* 2008). Since the deposited hydrate in nature remains within the sediment porous system, it is important to investigate the swapping behavior in sediments.

This study investigates the effect of the swapping speed on production behavior and swapping efficiency by constructing an experimental flue gas swapping system for the production of CH₄ recovery.

2. EXPERIMENT DESIGN

2.1. High pressure cell

Fig. 1 shows an experimental system and its components. High pressure cell was constructed that can operate at a pressure of 30MPa in order to simulate a gas hydrate-bearing sediment. The dimensions of specimens are 5.5cm of the diameter and 46cm of the length. Thermocouple probes were installed along the sidewall. A cooling jacket was installed to give the increased efficiency of a constant temperature.

2.2. Fluid injection unit

A syringe pump was used to inject distilled water into the specimen. Mass flow controllers were used to control the injection speed of the CH₄ and flue gas.

2.3. Temperature control unit

The temperature of high pressure cell, as well as that of the injected fluids was controlled by coolers. The flow lines, as well as the high pressure cell, were insulated with elastomeric closed-cell thermal insulation to minimize heat loss.

2.4. Gas analysis unit

A wet test meter was used in order to measure the produced gas volume. Gas chromatography was used to analyze the concentration and the composition of the produced gas during the swapping and dissociation process.

3. EXPERIMENT METHOD

3.1. Hydrate-bearing sediment specimen

A sand pack (particle size: 100 μ m, permeability: 202md, porosity: 33%) was used to replicate a gas hydrate-bearing sediment. After sand pack preparation, CH₄ gas was injected into the high pressure cell to obtain an irreducible water saturation (approximately 40%). Subsequently, the temperature of the system was lowered to 274.15K to form hydrate. After the hydrate formation and the system pressure had been stabilized, the hydrate saturation (approximately 41%) could be

calculated using Eq. (1)(Sakamoto *et al.* 2005).

$$S_{hyd} = \frac{V_{wd}}{172} \frac{\Delta P}{P_0} \frac{T_0}{T_f} \frac{1}{AL\phi} \quad (1)$$

Where, V_{wd} [m³]: volume of CH₄ gas in the pore
 ΔP [MPa]: pressure drop due to the hydrate formation
 P_0 [MPa]: pressure at standard condition
 T_0 [K]: temperature at standard conditions
 T_f [m³]: temperature used for the hydrate formation
 A [m²]: cross-sectional area of the specimen
 L [m]: length of the specimen
 Φ : porosity of the specimen.

3.2. Operating conditions for swapping production method

Once hydrate formation was completed, the flue gas was injected into the high pressure cell to initiate the swapping production. Each experiment consists of the cycles of production which includes dynamic production period and static soaking period. Cycle 1 is defined as the dynamic CH₄ swapping process by the flue gas injection at a constant swapping speed into the gas hydrate-sediment. The flue gas continues to be injected until no more CH₄ gas is being recovered. The starting point of the soaking process is set when CH₄ gas is no longer being recovered in Cycle 1. This accelerates the swapping phenomenon between the flue gas and the CH₄ molecules by allowing more reaction time. The Cycle (N+1) indicates an additional production procedure that takes place after soaking occurs N times over a certain period. In other words, Cycle 2 is the next step of the first soaking process after Cycle 1. In this study, Cycle (N+1) was conducted until Cycle 4. The production operational pressure and the temperature conditions in this study were 9.83MPa and 274.15K, respectively. These conditions are within the equilibrium condition for both CH₄ and CO₂+N₂ hydrate, and it is assumed that there is no gas production originated by hydrate dissociation within the high pressure cell.

A set of experiments have been conducted by varying swapping speed. The swapping speed was varied from 0.38 to 6.13m/day at constant system pressure and temperature in each experiment, in order to examine the change in swapping efficiency in terms of the swapping speed. The CH₄ recovery was calculated using the real time gas composition and volume data. The CH₄ recovery in Cycle (N+1) was calculated in each production process using the same method in the Cycle 1.

3.3. Hydrate dissociation and swapping efficiency analysis.

The remaining CH₄ gas volume after the swapping process should be taken accounted to calculate the swapping efficiency in Cycle 1 to Cycle (N+1). The remaining CH₄ gas volume was calculated by dissociating all the hydrates in a specimen. The actual swapping efficiency was calculated by deducting the initial free CH₄ gas volume from the total produced CH₄ gas volume. The swapping efficiency from each experiment was calculated using Eq. (2).

$$\text{Swapping Efficiency} = \frac{\text{Recovered CH}_4 \text{ Gas Volume @ Cycle N}}{\text{Total Injected CH}_4 \text{ Gas Volume}} \times 100(\%) \quad (2)$$

We converted all results as a function of the injected pore volume(PV), an dimensionless unit, in order to offset the time effect and to have consistent comparison and analysis. The definition of dimensionless unit of injected PV could be calculated using Eq. (3).

$$\text{Injected Pore Volume(PV)} = \frac{\text{Flue Gas Injection Time(T)} \times \text{Flue Gas Injection Rate(L}^3/\text{T)}_{\text{@ Operating Conditions}}}{\text{Pore Volume of Porous Media(L}^3)} \quad (3)$$

4. EXPERIMENT RESULT

4.1. Results of blank run experiment

Before conducting the swapping production experiment, a blank run experiment was performed in order to analyze the production without hydrates in the specimen. This process is used to determine the point of the complete displacement of the CH₄ gas at each flue gas injection rate. The experimental method consists of fully saturating an equal amount of CH₄ gas volume at irreducible water saturation, in accordance with the each stage. The injected PV was calculated during the injection of flue gas until a CH₄ concentration was 0%. The operating conditions (6.89MPa, 288.15K, respectively) were outside of CH₄ and CO₂+N₂ hydrate equilibrium conditions. Fig. 2(a) shows the changes in concentration during the CH₄ gas displacement process in relation to the injection rate. The difference shown at the initial displacement, according to the respective injection rate, converged at one point of the PV, suggesting equal amounts of CH₄ gas have been recovered as initially injected. Fig. 2(b) depicts the CH₄ recovery, which was converted to a cumulative value using the CH₄ concentration data in Fig. 2(a) and the volume of recovered gas. According to the results of the blank run experiment, about 2.5~2.9PV of the flue gas needs to be injected at the operating conditions for this study in order to recover all the CH₄ gas from the specimen.

4.2. Results of swapping production experiment

The CH₄ concentrations in the produced gas as a function of the produced gas volume are shown for each swapping speed in Fig. 3(a). More than 11PV of flue gas on average was injected until no more CH₄ gas is produced for swapping production while about 3PV of flue gas injection were needed during blank run experiment. These phenomena results from the reduced permeability induced by the hydrates in the pore throats and by the retarded rate of the swapping reaction in the porous system.

In Fig. 3(a), the CH₄ concentration variation curve to the left of the gray solid line indicates the Cycle 1, and hence pre-soaking, verifying that the final concentration converges at 0%. To the right of the gray solid line, the CH₄ concentration of the Cycle (N+1) is depicted. The peaks to the right of gray line indicate the additionally produced CH₄ by the soaking process. The soaking number(N) matches the number of peaks on the graphs, with the exception of Run#6(3.07m/day), which has no production after Cycle 3 and Run#7(6.13m/day), which has no production after Cycle 1. This confirms

that soaking process works as an additional recovery mechanism.

Fig. 3(b) shows the cumulative CH₄ recovery(both from CH₄ hydrate and free CH₄ gas). The plateau period that appears on the cumulative recovery graph in every experiment indicates the soaking process, and the difference in the final recovery indicates the swapping efficiency varied by swapping speeds.

The swapping efficiency in Fig. 4 suggests that the swapping efficiency is inversely proportional to the swapping speed due to the increase in the reaction time for swapping. Alternatively, an increase in swapping speed corresponds to a decrease in swapping efficiency, due to the faster gas flow, and thus the decrease in the reaction time allowed for swapping process. At the swapping speed of 3.07m/day and 6.13m/day, no additional CH₄ has been produced during some of the production cycles, suggesting the swapping efficiency has been decreased significantly. This also suggests the existence of a critical swapping speed for swapping production; the results of this experiment indicate a maximum critical swapping speed of between 2.30 to 3.07m/day, where no significant increase in recovery occurs with increase in swapping speed.

The quantitative values discussed here are presented in Table 1. The swapping efficiency of Run#1 (0.38m/day) shows a maximum value of approximately 40%(after deducting the free gas CH₄ volume). The swapping efficiency is significantly lower than the bulk hydrate cases(maximum value of 85%) because the relative permeability is significantly lowered by sediments and the multi-phase condition.

CONCLUSION

The effect of the swapping speed and the soaking process on the CH₄ recovery using the swapping method have been explored at a constant production operation pressure and temperature, through the simulation of a hydrate reservoir. Two variables (1) the swapping speed and (2) the soaking process play a crucial role in the additional recovery mechanism.

- (1) The decrease in the swapping speed corresponds to an increase in swapping efficiency when additional CH₄ gas is recovered during soaking process. The decrease in swapping speed corresponds to an increase in the amount of production due to an increase in the contact time, which results in an increase in the amount of swapping reaction. However, in the field, if the production well is operated with an extended soaking time, or with low swapping speed, problems can arise related to production delay and increased operation cost. Thus, an ideal critical swapping speed and the appropriate soaking time should be considered during the establishment of a quantitative production plan.
- (2) A numerical value lower than that of the swapping efficiency of the hydrate in bulk status was shown to be pertinent. This is due to the change in the flow conditions in the experimental system since the hydrate in this study is formed within the porous system of the specimen, which differs from the conditions in the bulk hydrate systems.

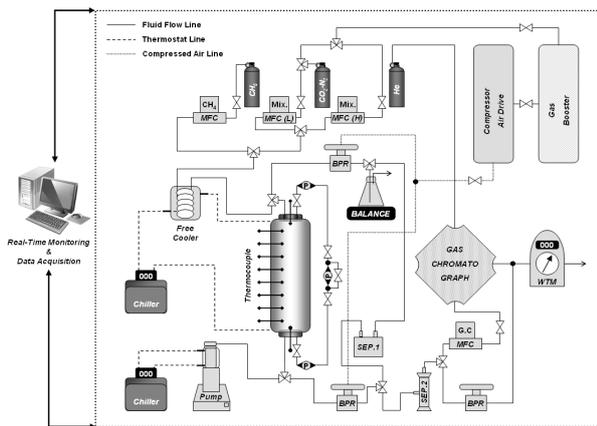


Fig. 1 Schematic diagram and pictorial view of experiment apparatus

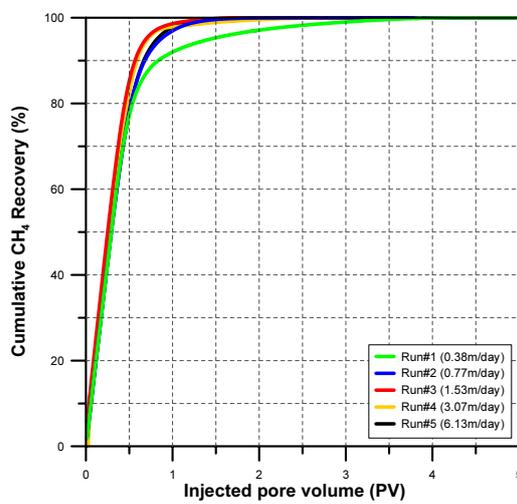
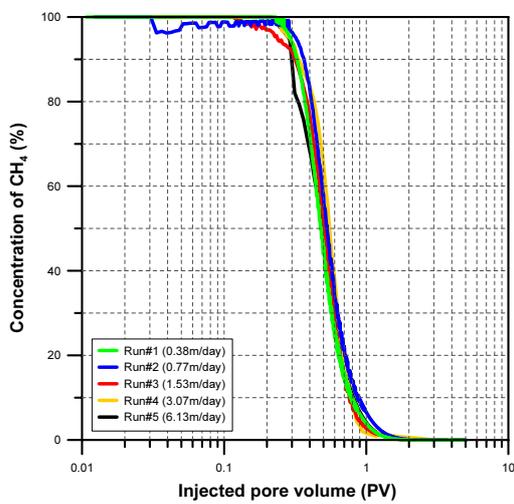


Fig. 2 Production profile of CH₄ gas for blank run:
(a) CH₄ concentration (b) Total cumulative CH₄ recovery

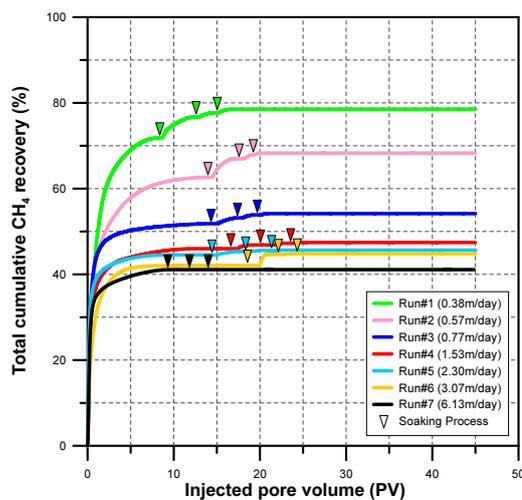
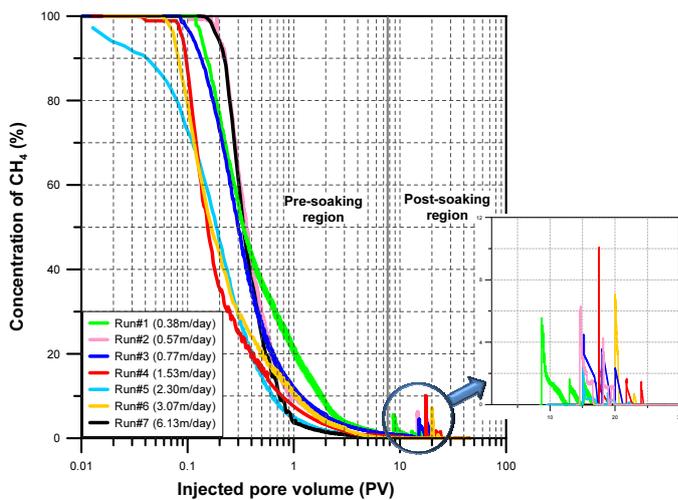


Fig. 3 Production profile of CH₄ in swapping process:
(a) CH₄ concentration, (b) Total cumulative CH₄ recovery

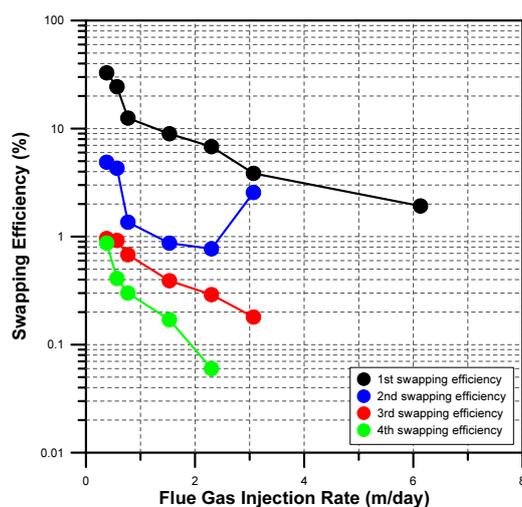


Fig. 4 Analysis of swapping efficiency

Table 1 Conditions and Results of Experiment for Swapping Process

| RUN # | Swapping Speed [m/day] | Hydrate Saturation [%] | Soaking Time [day] | Injected PV for Cycle 1 [PV] | Total Recovery [%] | Swapping Efficiency [%] | | | |
|-------|------------------------|------------------------|--------------------|------------------------------|--------------------|-------------------------|------|------|-----------------|
| | | | | | | 1st | 2nd | 3rd | 4 th |
| 1 | 0.38 | 41.50 | | 8.43 | 78.55 | 32.81 | 4.88 | 0.96 | 0.87 |
| 2 | 0.58 | 40.03 | | 14.08 | 68.29 | 24.40 | 4.29 | 0.92 | 0.41 |
| 3 | 0.77 | 41.27 | | 14.39 | 54.17 | 12.52 | 1.36 | 0.68 | 0.30 |
| 4 | 1.53 | 41.15 | 0.17 | 12.41 | 47.41 | 8.95 | 0.87 | 0.39 | 0.17 |
| 5 | 2.30 | 40.81 | | 9.49 | 45.65 | 6.80 | 0.77 | 0.29 | 0.06 |
| 6 | 3.07 | 42.39 | | 10.84 | 44.80 | 3.85 | 2.56 | 0.18 | 0 |
| 7 | 6.13 | 41.38 | | 9.67 | 41.08 | 1.92 | 0 | 0 | 0 |

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