Visualization of the flow pattern in methane hydrate reservoir model

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Abstract
The objective of this study is to visualize the flow pattern in methane hydrate (MH) reservoir model under atmospheric pressure condition. A method to mimic a real MH reservoir was introduced into the present research to visualize the multiphase flow pattern in porous media under thermal fluid injection. First, porous media mimicking real MH reservoir were prepared in a visualization cell with dual horizontal wells, which were composed of glass beads, ice of sodium bicarbonate (NaHCO₃) aqueous solution and ethanol (C₂H₅OH). Thereafter, hydraulic fracturing by injecting C₂H₅OH aqueous solution was executed to generate flow path that increases permeability between the injection well and production well. The flow pattern in the reservoir model with the flow path was then visualized during the injection of hydrochloric acid (HCl) aqueous solution. The dominant factors governing the multiphase flow in fractured porous MH mimicking reservoir were evaluated. It was found that the flow path formation with high permeability by hydraulic fracturing and permeability increment by ice melting are of critical importance for the reservoir mimicking system. In addition, it is found that the liquid phase flow may also be affected by the formation of gas phase inside the porous media that mimic the dissociation process in the real MH reservoir.

Keywords: Methane hydrate, Visualization, Hydraulic fracturing, Flow pattern, Permeability change

1. Introduction
1.1 Methane hydrate

Methane hydrate (MH) is crystalline compound in which methane molecules are hosted by water molecules cage (Sloan and Koh, 2008). MH exists stably under low-temperature and high-pressure conditions such as permafrost and seabed. In Japan, vast amounts of MH exist in pore spaces of sandy sediments under the seafloor such as Nankai trough. Hence, MH must be dissociated into methane (CH₄) and extracted from the reservoir to utilize it as a novel energy resource. Basic techniques for the production of methane from MH reservoir, such as depressurization method, thermal method, and inhibitor injection method have been developed (Kurihara, 2011; MH21 Research Consortium, 2018). In the depressurization method, the MH reservoir pressure is reduced by using a pump, which allows MH dissociation to be induced. Additionally, at present, the depressurization method is regarded as a promising production method, and a considerable number of studies have been conducted thus far regarding this method (Konno, et al., 2012, 2014; Chen et al., 2016, 2017; Yamada, et al., 2017; Chen, et al., 2018). In the thermal method, the temperature in the MH layer is increased by heat conduction or hot water injection, and MH dissociation is thus promoted. In the inhibitor
injection method, the phase equilibrium of MH is shifted to the high-pressure and low-temperature side by injecting the inhibitor, such as salt or alcohol.

1.2 Thermal flooding method

In these production methods, the thermal method is divided into the thermal stimulation method and the thermal flooding method. In the thermal stimulation method, the temperature in the region around the bottom of the production well is increased by heat conduction. Additionally, the rate of change of temperature in the MH layer is considerably slow because the MH reservoir is heated by heat conduction from the production well. However, in the thermal flooding method, the MH reservoir temperature is heated by injecting thermal fluid such as hot water. Furthermore, in the thermal flooding method, MH dissociation is promoted by expanding the flowage region of hot water in the MH reservoir. Therefore, the thermal flooding method is regarded as the better production method than the thermal stimulation method from the view-point of heat transfer characteristic in the MH reservoir. Moreover, power generation systems using the thermal flooding method to utilize MH as an energy resource have been proposed (Maruyama, et al., 2012; Sasaki, et al., 2014; Chen, et al., 2017). Maruyama et al. (2012) proposed a power generation system with low carbon dioxide (CO\textsubscript{2}) emission as shown in Fig.1 (Maruyama, et al., 2012). In that system, CO\textsubscript{2} resulting from combustion in a turbine is isolated from the atmosphere by dissolving it into seawater, which is heated by an exhaust heat of the power generation. Hot seawater with CO\textsubscript{2} is injected into the MH layer as the thermal flooding. Additionally, the produced methane is extracted from the MH reservoir and utilized for power generation. Sasaki et al. (2014) suggested a power generation system using the thermal flooding method with several dual horizontal wells (Sasaki, et al., 2014). In this system, a large number of dual horizontal wells is used on the thermal flooding. Furthermore, Chen et al. (2017) proposed an offshore power generation system with carbon capture and storage (CCS) for utilizing oceanic MH (Chen, et al., 2017). In this system, warming the MH reservoir and hydraulic fracturing are executed by hot seawater injection. However, there are challenges to the thermal flooding method in terms of practical uses of MH. For example, it is difficult to expand the flowage region of hot water in the MH reservoir where there is low permeability condition. In other words, the heat supply required for MH dissociation is affected by low permeability. Hence, it is important to clearly understand the flow pattern of multiphase flows in porous media with low permeability and consider a way to improve the low permeability condition.

To resolve these problems, research on the practical use of the thermal flooding method have been conducted. Generally, it is impossible to directly observe the flow pattern in the MH reservoir while the thermal flooding method is conducted. Moreover, tight pressure chambers, such as stainless steel, must be used to treat MH experimentally, in which it is difficult to visualize the inner condition. Because of these challenges, Sasaki et al. (2009) devised a method...
to simulate the MH reservoir under atmospheric pressure condition (Sasaki, et al., 2009; Ono, et al., 2009). They visualized the flow pattern in the MH reservoir model with dual horizontal wells while hot water injection was executed in the laboratory experiment. In their research, it was concluded that the gas production rate was proportional to the enlargement speed of the flowage region of the multiphase flow in the MH reservoir model. Furthermore, Sasaki et al. (2014a, 2014b) conducted a visualization experiment with respect to the flow pattern in the MH reservoir model using the same devised method (Sasaki, et al., 2014a, 2014b). In this study, multiphase flow was observed clearly and it was found that the enlargement of the liquid flowage region was affected by the formation of a gas flowage region.

However, dominant factors governing the enlargement of the flowage region of multiphase flow in porous media with low permeability during thermal flooding were not evaluated in those previous studies. Sasaki et al. (2009) referred to only the relationship between the gas production rate and the enlargement speed of the flowage region in the MH reservoir model (Sasaki, et al., 2009; Ono, et al., 2009). Sasaki et al. (2014b) evaluated the flowage characteristic of multiphase flow in porous media with high permeability, in which the pore spaces were filled with only liquid phase (Sasaki, et al., 2014b). Therefore, it is necessary to quantitatively evaluate the dominant factors governing multiphase flow in porous media with low permeability during thermal flooding to improve the challenges for the thermal flooding method.

The objective of this study is to visualize the flow pattern in the MH reservoir model during thermal flooding and evaluate dominant factors governing multiphase flow in porous media with low permeability. A visualization cell with dual horizontal wells was constructed, and porous media mimicking MH reservoir were prepared in the visualization cell. Thereafter, hydraulic fracturing was executed in the reservoir to generate a high permeability path between the injection well and production well. The flow pattern in the MH reservoir model with the flow path was then visualized during the thermal flooding, and the dominant factors governing multiphase flow in porous media with low permeability were evaluated.

2. Principle of the MH reservoir model

In previous studies, a method to mimic real MH reservoir under atmospheric pressure condition was proposed to visualize the flow pattern in porous media during thermal flooding. For example, Sasaki et al. (2009) devised a method to mimic a MH reservoir with the thermal flooding under atmospheric pressure condition (Sasaki, et al., 2009). In the method, MH dissociation, which is phase change phenomena, is mimicked by the chemical reaction between hydrochloric acid (HCl) aqueous solution and ice of NaHCO₃ aqueous solution, as shown in Fig. 2.

As shown in Fig. 2, hot water injection in the thermal flooding method and MH in the pore spaces of sandy sediments are mimicked by HCl aqueous solution and ice of NaHCO₃ aqueous solution, respectively. Additionally, a change of permeability and gas generation attributed to MH dissociation are mimicked by ice melting and gas generation due to the chemical reaction, as shown in equations (1) and (2), respectively.
\[ \text{CH}_4 \cdot 5.75\text{H}_2\text{O} = \text{CH}_4(g) + 5.75\text{H}_2\text{O} - 56.9\text{kJ/mol} \quad (1) \]

\[ \text{NaHCO}_3 + \text{HCl} = \text{NaCl} + \text{H}_2\text{O} + \text{CO}_2(g) + 56.5\text{kJ/mol} \quad (2) \]

Furthermore, an endothermic reaction of MH dissociation is mimicked by melting heat of an ice of NaHCO$_3$ aqueous solution, although the chemical reaction is an exothermic reaction. Details of the mimicking method is summarized in Table 1. Moreover, Lacaille et al. (2017) proposed a way to control the permeability of the MH reservoir model by adding C$_2$H$_5$OH to NaHCO$_3$ aqueous solution (Lacaille, et al., 2017). In this way, the permeability of the reservoir can be controlled by partially forming an ice of NaHCO$_3$ aqueous solution with a liquid phase of C$_2$H$_5$OH in the pore spaces of the porous media. Both methods were introduced into the present research to prepare the MH reservoir model, of which permeability is controlled.

**Table 1 Details of MH reservoir model of the thermal flooding method (Ono, et al., 2009).**

| Methodology | Thermal flooding method | This study |
|-------------|-------------------------|------------|
| Environment | High pressure           | Atmospheric pressure |
| Porous media | Sand layer              | Glass beads layer |
| Thermal fluid | Hot water              | HCl aqueous solution |
| Extraction target | MH                  | Ice of NaHCO$_3$ aqueous solution |
| Phase change phenomena and Permeability change | MH dissociation | Ice melting + Chemical reaction |
| Gas generation | CH$_4$              | CO$_2$ |
| Heat consumption by phase change | -4.27×10$^5$ J/kg | -3.34×10$^5$ J/kg |
| Remarks | –                      | Chemical reaction is an exothermic reaction |

**3. Preparation of the MH reservoir model**

A visualization cell with dual horizontal wells was constructed as shown Figs. 3 and 4, which is 340 mm in height, 360 mm in width, and 64 mm in thickness.

![Figure 3](image-url)  
**Fig. 3** Photograph and schematic of the MH reservoir model in a visualization cell. ((a): Photograph of front view of the cell, (b): Schematic of side view of the cell).
The cell was composed of polycarbonate plates, an aluminum frame, an aluminum plate, high-tension bolts, sealing rubbers, thermocouples, an injection well, and a production well. The aluminum frame was installed between polycarbonate plates to prevent injected HCl aqueous solution from leaking on the interface between the polycarbonate plate and the surface of the MH mimicking sample. Moreover, the high-tension bolts were used in the cell to prevent leakage from the cell. As shown in Fig. 3, four thermocouples were installed in the center of the aluminum plate at 30 mm, 105 mm, 180 mm and 230 mm from the bottom of the sample space. Additionally, the maximum measurement errors of the thermocouples were ±1.0 K. The injection and production wells were installed at center of the aluminum plate at 80 mm and 130 mm from the bottom of the sample space, respectively. The distance between both wells was 50 mm, as shown in Fig. 3. Moreover, the outer diameter of both wells was 9.5 mm. The sample space in the cell was 260 mm in height, 280 mm in width, and 10 mm in thickness, which could maintain two-dimensional flowage of multiphase flow in the MH reservoir model. The MH reservoir model was formed in the sample space of the visualization cell. The experimental procedure for forming the reservoir is as follows:

1. Glass beads were packed tightly in the space, of which the average diameter of the distribution was 177–250 μm.
2. Mixed aqueous solution was prepared, which was composed of distilled water, NaHCO₃, and C₂H₅OH.
3. The pore spaces of the glass beads layer were filled with the mixed aqueous solution.
4. The visualization cell was cooled at −50°C for approximately 2 days and the MH reservoir model was formed, as shown in Fig. 3.

In this experiment, three samples were prepared in order to evaluate effect of porosity of glass beads layer on hydraulic fracturing behavior in the MH reservoir model. The mass of glass beads was 1.39 kg, 1.38 kg and 1.37 kg in Runs 1–3, respectively. The apparent porosity of glass beads layer in the sample space with the both well was 0.234, 0.243 and 0.245 in Runs 1-3, respectively. The mass of mixed aqueous solution was 3.46×10⁻¹ kg, 3.65×10⁻¹ kg and 3.27×10⁻¹ kg in Runs 1–3, respectively. In Runs 1–3, the mass fractions of C₂H₅OH and NaHCO₃ were 10 wt% to NaHCO₃ aqueous solution and 5 wt% to distilled water. Additionally, the permeability of the MH reservoir model in this experiment was approximately 0.80 D, which was estimated from experimental results of the previous research by Lacaille et al (Lacaille, et al., 2017).

4. Hydraulic fracturing experiment in the MH reservoir model
4.1 Experimental system and procedure

Hydraulic fracturing experiment was executed before visualization experiment of the flow pattern was conducted to generate flow path that increase permeability of the MH reservoir model. The visualization cell was installed into experimental system for visualization after the MH reservoir model was formed, as shown in Fig. 5. The experimental system was composed of an isothermal bath, a thermostat chamber, an agitator, a pump, pressure transmitters,
thermocouples, a balance, a mass flow controller, a data logger, and a video camera. The isothermal bath was covered with thermal insulation materials and filled with antifreeze liquid, whose temperature was controlled at −10°C by the thermostat chamber with an accuracy of ±0.03 K and the agitator. The pump was used to inject fluid into the MH reservoir model. The temperature and pressure in the injection well and tank were measured by the thermocouples and the pressure transmitters with accuracies of ±1.0 K and ±0.35 MPa, respectively. The mass flow rate of fluid captured from the production well was measured using the balance. All data were recorded every 0.1 s and stored in the data logger. The flow pattern in the MH reservoir model was recorded by the video camera. The hydraulic fracturing experiment was conducted according to the following steps:

1. A valve between the MH reservoir model and a tank was closed.
2. C$_2$H$_5$OH aqueous solution was injected into the tank using the pump.
3. The valve was opened after pressure in the tank increased to approximately 1 MPa.
4. Injection of C$_2$H$_5$OH aqueous solution from the injection well to the MH reservoir model was started by using the pump.
5. Pressure in the injection well and the flow pattern in the MH reservoir model during injection of C$_2$H$_5$OH aqueous solution were measured and recorded.

In this experiment, C$_2$H$_5$OH aqueous solution was used as fluid for hydraulic fracturing. The mass fraction of C$_2$H$_5$OH was 30 wt% to the aqueous solution and its temperature was controlled at −10°C by the thermostat chambers, as shown in Fig. 5. Red ink was added to the aqueous solution to observe the flow pattern in the recording by the video camera. Additionally, the mass flow rate of C$_2$H$_5$OH aqueous solution was set to 2.5×10$^{-5}$ kg/s in the pump.

4.2 Results and discussion

The time evolution of the zoomed flow pattern in the MH reservoir model and time variation of pressure in the injection well during hydraulic fracturing are shown in Figs. 6 and 7, respectively. Especially, the zoomed visualization results when the pressure decreased to approximately 0.5 MPa was shown in middle time pictures of Fig. 6. It can be seen in Fig. 6 that flowage of C$_2$H$_5$OH aqueous solution occurred around the injection well in Runs 1–3 until 20 min after the injection was started. As shown in Fig. 7, pressure in the injection well increased drastically in Run 1 after the injection was started. This pressure increment indicates that the injected C$_2$H$_5$OH aqueous solution was stagnant in the injection well because permeability of the MH reservoir model was quite low, as shown in step 1 of Fig. 8. Thereafter, the pressure in the injection well increased or decreased in Run 1 as time advanced. This result implies that hydraulic fracturing for ice in the reservoir was executed because of the pressure gradient between both wells, and the flow path with high permeability was formed, as shown in step 2 of Fig. 8. Other groups have reported that a similar tendency of
the pressure change occurred when hydraulic fracturing was executed for unconsolidated porous media (Bohloli and de Pater, 2006; Ito, et al., 2008; Ito, et al., 2011). The pressure in injection well then decreased drastically to approximately 0.5 MPa in Run 1. This shows that the injected fluid from the injection well reached the production well, as shown in step 3 of Fig. 8. As shown in Fig. 7, although each tendency of the pressure change in Runs 2 and 3 was similar to that in Run 1 during the fluid injecting, the respective maximum pressure in the injection well in Runs 2 and 3 was lower than that in Run 1. Additionally, the respective duration at which the pressure in the injection well decreased to 0.5 MPa in Runs 2 and 3 was shorter than the duration it took in Run 1. These results are attributed to differences in the mass of glass beads in the MH reservoir model. In other words, these may be caused by differences in the porosities of the glass beads layers.

Therefore, it was found that the flow path with high permeability was formed between the injection well and production well by hydraulic fracturing. Moreover, it is implied from the experimental results that maximum pressure and duration at which the fluid reached the production well for hydraulic fracturing depends on the porosity of the glass beads layers.

![Fig. 6 Time evolution of zoomed visualization results during hydraulic fracturing in Runs 1–3.](image-url)
5. Visualization of the flow pattern in the MH reservoir model
5.1 Experimental system and procedure

The flow pattern in the MH reservoir model with the flow path was visualized when HCl aqueous solution was injected. The experimental system used in this experiment was the same as that used in the hydraulic fracturing experiment, as shown in Fig. 5. The gas production rate of CO$_2$ captured from the production well was measured by the mass flow controller with a full scale of 8.3×10$^{-6}$ m$^3$/s and an accuracy of ±0.1%. The experimental procedure for the flow pattern visualization is as follows:

1. The valve between the MH reservoir model and the tank was closed.
2. HCl aqueous solution was injected into the tank by using the pump.
3. The valve was opened after pressure in the tank increased to approximately 1 MPa.
4. Injection of HCl aqueous solution from the injection well to the MH reservoir model was started using the pump.
5. The flow pattern in the MH reservoir model during the injection of HCl aqueous solution was recorded.
6. The temperature in the MH reservoir model during the fluid injection and the gas production rate of CO$_2$ captured from production well were measured, respectively.

Fig. 7 Time variation of pressure in injection well during fluid injection in Runs 1–3.

Fig. 8 Schematics of process of hydraulic fracturing in MH mimicking reservoir.
In this experiment, HCl aqueous solution was used in thermal flooding, which was composed of distilled water, HCl and C\textsubscript{2}H\textsubscript{5}OH. The mass fractions of HCl and C\textsubscript{2}H\textsubscript{5}OH were 3 wt\% and 30 wt\% to distilled water, respectively. The temperature of the HCl aqueous solution was controlled at \( -10^\circ \text{C} \) by the thermostat chambers, as shown in Fig. 5. Furthermore, blue ink was added to the aqueous solution to observe the flow pattern in the recording by the video camera. Each mass flow rate of HCl aqueous solution in Runs 1–3 was set to \( 3.2\times10^{-4} \) kg/s, \( 1.6\times10^{-4} \) kg/s and \( 2.5\times10^{-5} \) kg/s in the pump, respectively.

### 5.2 Results and discussion

The time evolution of the flow pattern in the MH reservoir model is shown in Fig. 9. The time variation of the gas production of CO\textsubscript{2} captured from the production well in only Run 1 during the injection of the HCl aqueous solution is shown in Fig. 10 because measurement of the gas production rate of CO\textsubscript{2} was failed in runs 2 and 3. The position of the thermocouple between the both wells is shown in Fig. 9. It can be seen in Fig. 9 that flowage of HCl aqueous solution occurred between the injection well and production well in Runs 1–3 until 30 min after the fluid injection was started. CO\textsubscript{2} production from the production well was measured in Run 1 just after the fluid injection was started, as shown in Fig. 10. Additionally, the temperature between the both wells increased in Runs 1–3 after the fluid injection was started, as shown in Fig. 11. These results were caused by heat generation attributed to the chemical reaction between HCl and NaHCO\textsubscript{3} as shown in equation (2), and indicate that the injected HCl aqueous solution flowed selectively in the formed flow path with high permeability between both wells. However, the duration in Run 3 at which the flowage can be observed was longer than the durations it took in Runs 1 and 2. It is implied from the results that the duration at which the flowage occurred in the MH reservoir model depended on the mass flow rate of the HCl aqueous solution.

![Fig. 9 Time evolution of the flow pattern in the MH reservoir model during injection of HCl aqueous solution in Runs 1–3.](image-url)
It can thus be observed from Fig. 9 that the flowage region of the multiphase flow in the reservoir expanded gradually from the flow path between both wells in Runs 1–3. Moreover, the region around both wells were colored clearly in Runs 1–3 after the visualization experiment, as shown in Fig. 12. Especially, although the mass flow rate of the HCl aqueous solution was set to various value in Runs 1–3, the flowage region expansion in the MH reservoir model in Runs 1–3 exhibited a similar tendency. These results are attributed not to the mass flow rate of the HCl aqueous solution but rather to the permeability increment on the fringe region of the flowage by ice melting, which was induced by an exotherm of the chemical reaction between HCl and NaHCO$_3$. Furthermore, it is assumed that the flowage region enlargement of the HCl aqueous solution may be affected by the formation of CO$_2$ flowage on the fringe region of the liquid phase. In previous research, it was reported that similar phenomena occurred when multiphase flow formed in porous media (Sasaki, et al., 2014).

Hence, it can be deduced from the experimental results that the dominant factors governing multiphase flow in the MH reservoir model are flow path formation with high permeability by hydraulic fracturing and permeability increment by ice melting. In addition, it can be seen that the liquid phase flow may also be affected by the formation of the gas phase inside the porous media that mimic the dissociation process in the real MH reservoir.

![Fig. 10](image1.png)

**Fig. 10** Time variation of gas production of CO$_2$ captured from production well in Run 1.

![Fig. 11](image2.png)

**Fig. 11** Time variation of temperature between the both wells during injection of HCl aqueous solution in Runs 1–3.
6. Concluding remarks

In this study, the flow pattern by injection of the HCl aqueous solution in the MH reservoir model with the hydraulic fracturing was visualized. Additionally, the dominant factors governing multiphase flow in the reservoir were evaluated from the experimental results. The following conclusions can be drawn:

1. The flow path with high permeability was formed between the injection well and production well by hydraulic fracturing, which was induced by pressure gradient between both wells.
2. It implies that maximum pressure and duration at which the fluid reached the production well on hydraulic fracturing depend on porosity of glass beads layers.
3. It can be deduced that the dominant factors governing multiphase flow in the MH reservoir model are flow path formation with high permeability by hydraulic fracturing and permeability increment by ice melting.
4. The liquid phase flow may be affected by the formation of a gas phase inside the porous media that mimic the dissociation process in the real MH reservoir.

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