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Experimental Study of Methane Hydrate Dissociation and Gas Production Behaviors under Depressurization

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***Abstract*—The objective of this study is to evaluate into detail on the effect of sensible heat in methane hydrate cores for gas production behavior using depressurization method under various initial temperature conditions. Firstly, methane hydrate was synthesized under low temperature and high pressure conditions in the laboratory. Methane hydrate cores were made by using methane hydrate and sand. Dissociation experiment using those cores was conducted by using depressurization method. Then, pressure, temperature, gas production rate and cumulative gas production were measured during the experiment. In the experiment, gas production rate of low porosity core with T_{ini} of 2.5 ℃ was higher than that of low porosity core with T_{ini} of 1.5 ℃ in the first 10 minutes. That implies sensible heat used for methane hydrate dissociation in the low porosity core with T_{ini} of 2.5 ℃ was higher than that in the other core due to initial temperature deference between these low porosity cores. Therefore, it is important to increase initial temperature of methane hydrate reservoir for enhancement of gas production rate under depressurization.**

***Index Terms*—energy, methane hydrate, depressurization, dissociation, gas production, sensible heat**

I. INTRODUCTION

In Japan, methane hydrate, which is regarded as new energy resource, exists beneath seafloor under low temperature and high pressure conditions. The analysis of basic physical properties and commercial utilization for methane hydrate has been developed for decades [1]. Meanwhile, it is necessary to extract methane gas from methane hydrate reservoir for using it as energy resource. Depressurization and thermal stimulation as basic techniques to produce methane gas from methane hydrate layer were proposed [1], [2].

Furthermore, proposals to utilize methane hydrate by using these production methods were suggested by many researchers [3], [4]. For example, a power generation system utilizing methane hydrate with low carbon dioxide emission was proposed by Maruyama *et al* [4]. The conceptual diagram is shown in Fig. 1. In that system, carbon dioxide resulting from combustion is isolated from the atmosphere by dissolving carbon dioxide with compressed seawater, which is heated by exhaust heat of the system. Moreover, hot sea water with carbon dioxide is injected into methane hydrate reservoir for gas production and carbon dioxide storage. Meanwhile, it is required to produce methane from methane hydrate layer stably for practical use of that system. In order to achieve that goal, several studies have been focused on methane

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hydrate dissociation in methane hydrate reservoir [5]-[8]. For example, in order to extract methane from methane hydrate layer efficiently by using thermal stimulation, Lacaille *et al*. [8] evaluated heat and mass transport phenomena during methane hydrate dissociation by using methane hydrate mimicking sample. However, in that power generation system, depressurization method has to be only used to dissociate methane hydrate in early stage of gas production because hot sea water cannot be produced. Hence, there is a need to study efficient depressurization method for enhancement of gas production.

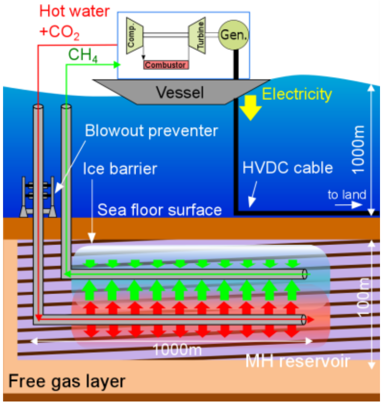

Figure 1. Conceptual diagram of the power generation system [4].

In depressurization method, sensible heat is important to enhance gas production because methane hydrate dissociation is endothermic reaction. Therefore, it is necessary to evaluate the influence of sensible heat for gas production. Youslf *et al*. [9] investigated methane hydrate dissociation in core samples experimentally and theoretically. Zhou *et al*. [10] observed ice formation in methane hydrate core during methane hydrate dissociation using depressurization method. Konno *et al*. [11] concluded gas production rate under depressurization of high-permeability methane hydrate cores is probably limited by heat transfer from ambient. Oyama *et al*. [12] determined core sensible heat is rapidly consumed by methane hydrate dissociation and this heat is not supplied to dissociate all bearing methane hydrate during depressurization. X. Yang *et al*. [13] found mass transfer resistance and thermal buffering due to ice formation have an important effect on gas production rate during methane hydrate dissociation in porous sediment by depressurization method. Konno *et al*. [14] concluded gas production from methane hydrate cores induced by depressurization can be accelerated by heat supply from ice formation in the first 50 minutes. Konno *et al*. [15] determined the latent heat of ice formation is efficiently used for hydrate dissociation using depressurization method. Zhao *et al*. [16] determined that methane hydrate reformation and ice formation always occur due to endothermic reaction of methane hydrate dissociation and insufficient heat transfer from ambient under depressurization. Chen *et al*. [17] found ice formation during the endothermic reaction of the methane hydrate dissociation could possibly affect the methane gas production process numerically.

However, these studies have not analyzed into detail on the effect of sensible heat for gas production under various initial temperatures. It is expected increment of initial temperature in methane hydrate layer may lead to increase of sensible heat for methane hydrate dissociation under depressurization. Hence, it is necessary to evaluate the influence of sensible heat for gas production under various initial temperature conditions.

In the present study, methane hydrate was synthesized in laboratory. Then, dissociation experiment was conducted by using cores under various initial temperature conditions. Methane hydrate dissociation and gas production behavior were evaluated in pure methane hydrate core and low porosity cores. Additionally, the effect of sensible heat for gas production from low porosity cores was analyzed in this study.

II. EXPERIMENTAL APPARATUS AND PROCEDURE

A. *Methane Hydrate Synthesis Apparatus*

Methane hydrate forms under low temperature and high pressure conditions. Hence, methane hydrate synthesis apparatus with pressure resistance was constructed in this study. Schematic of methane hydrate synthesis apparatus in this study is shown in Fig. 2

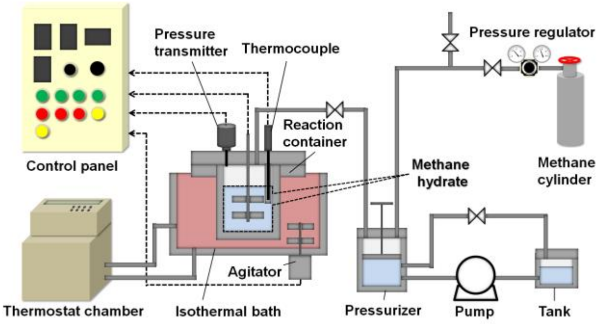

Figure 2. Schematic of methane hydrate synthesis apparatus.

This synthesis apparatus is composed of a reaction container, an agitator, an isothermal bath, a thermostat chamber, a pressurizer, a thermocouple, a pressure transmitter and a control panel. The volume of the reaction container is 980 ml. It is known that methane hydrate formation is enhanced by increasing interface area between methane and water. Therefore, the agitator is installed in the container in order to synthesize methane hydrate efficiently. The isothermal bath is covered with thermal insulations and filled with antifreeze liquid. The temperature of the antifreeze liquid was controlled by the thermostat chamber with an accuracy of ±0.03 K. The pressurizer is used for increasing pressure in this chamber. Temperature and pressure in the reaction container were measured by the thermocouple and the pressure transmitter with the maximum errors of ±1.0 K and ±0.1 MPa, respectively. All data were displayed on the control panel.

B. *Methane Hydrate Synthesis Procedure*

Methane hydrate was synthesized according to the following steps:

- The temperature in the isothermal bath was set to 4 ℃.
- Deionized water was injected into the reaction container, of which the mass was approximately 0.3 kg.
- The reaction container was purged with nitrogen twice and then evacuated for approximately 30 seconds to ensure the absence of air from it.
- The pressure was pressurized up to 7 MPa gradually by injecting methane into the reaction container and using pressurizer.
- The reaction container was pressurized up to 7 MPa again by injecting methane using the pressurizer after pressure in that container became less than 5 MPa.
- The reaction container was removed from the isothermal bath after pressure in that container became stable.
- The reaction container was cooled less than approximately -80 ℃ by using liquid nitrogen.
- Pressure in the reaction container was decreased to atmospheric pressure.

C. *Experimental Apparatus for Depressurization*

Schematic of experimental apparatus for depressurization is shown in Fig. 3.

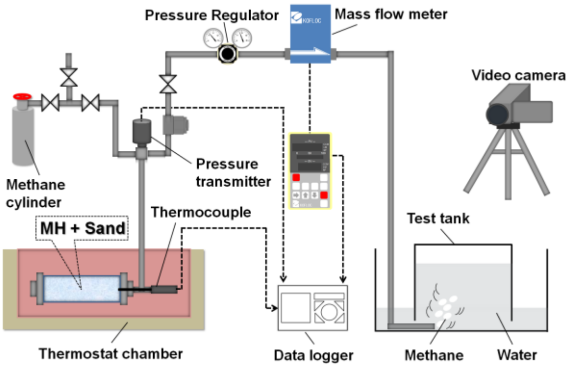

Figure 3. Schematic of experimental apparatus for depressurization.

This experimental apparatus is composed of a cylindrical case, a thermostat chamber, a pressure regulator, a mass flow meter, a thermocouple, a pressure transmitter, a video camera and a data logger. The volume of the cylindrical case is approximately 52.8 ml, which is capable of withstanding pressure of 15 MPa. The thermostat chamber is filled with antifreeze liquid. The temperature of the antifreeze liquid was controlled by this thermostat chamber with an accuracy of ±0.2 K. The pressure in the cylindrical case was controlled by the pressure regulator. The gas production rate and the cumulative gas production were measured by the mass flow meter with a full scale of 1.00 SL/min (Standard liter per min) and an accuracy of ± 0.1 %. Temperature and pressure in the cylindrical case were measured by the thermocouple and the pressure transmitter with accuracies of ±1.0 K and ±0.35 MPa, respectively. A behavior of the gas production was recorded by the video camera in this experiment. All data were recorded every 0.1 s by the data logger.

D. *Depressurization Procedure*

Depressurization experiment using methane hydrate cores was conducted. Toyoura sand was used to simulate porous sediments in order to adjust porosity of methane hydrate cores. Grain diameter distribution of the sand particle was in the range of 0.1–1.0 mm [18]. Depressurization experiment using methane hydrate cores was conducted according to the following steps:

- The cylindrical case and sand were cooled less than approximately -80 ℃ by using liquid nitrogen.
- Synthesized methane hydrate and sand were mixed manually and the cylindrical case was filled with that mixture manually.
- Pressure in the cylindrical case was pressurized to approximately 3.5 MPa by injecting methane gas into that case.
- The cylindrical case was installed in the thermostat chamber of which temperature settings was each production temperature.
- The cylindrical case was left for approximately 2 hours in the thermostat chamber after temperature in that case became the production temperature designed for each experimental run.
- After temperature and pressure in the cylindrical case became constant during approximately 2 hours, the pressure regulator was adjusted to the production pressure designed for each experimental run.

After the pressure regulator was adjusted, pressure in the cylindrical case decreased drastically. Temperature in the thermostat chamber was maintained at production temperature throughout this experiment. The temperature and pressure in the cylindrical case, gas production rate and cumulative gas production were recorded by the data logger.

III. RESULT AND DISCUSSION

A. *Methene Hydrate Synthesis*

Figure 4. Methane hydrate synthesized in this experiment. (Left: Methane hydrate, Right: Combustion of methane hydrate).

Methane hydrate synthesized in this experiment is shown in Fig. 4. The time variation of temperature and pressure during the methane hydrate synthesis process is shown in Fig. 5. It can be seen that temperature increased dramatically just after pressure increased in Fig. 5, because methane gas was injected into the reaction container drastically. Afterward, temperature increment was observed while pressure decreased gradually. That result shows exothermic reaction of methane hydrate formation occurred during the pressure reduction.

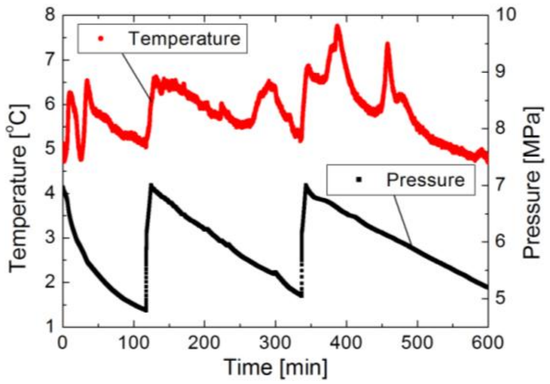

Figure 5. Time variation of temperature and pressure during methane hydrate synthesis process.

B. *Depressurization Experiment*

Values of initial temperature, initial pressure, mass saturation of synthesized methane hydrate, mass of methane hydrate, minimum temperature and duration of 0 ℃ condition in all cores are shown in Table I.

TABLE I. THE SUMMARY OF DEPRESSURIZATION EXPERIMENT

Core	Pure methane hydrate core	Low porosity core	
Initial *T* [℃]	1.6	2.5	1.5
Initial *p* [MPa]	3.5	3.9	3.5
Mass of MH [g]	38.7	13.2	5.4
Minimum *T* [℃]	-1.6	-1.3	-1.0
0 ℃ duration [min]	80	15	10

The mass saturation of methane hydrate in all cores was calculated from experimental result of depressurization by using (1) and expressed as follows:

$$S_{MH} = \frac{m_{MH}}{m_{MH} + m_w + m_{CH_4} + m_s} \quad (1)$$

where S_{MH} [-] is the mass saturation of methane hydrate, m_{MH} [kg] is the mass of methane hydrate, m_w [kg] is the mass of water, m_{CH4} [kg] is the mass of methane gas in the pore space and m_s [kg] is the mass of sand.

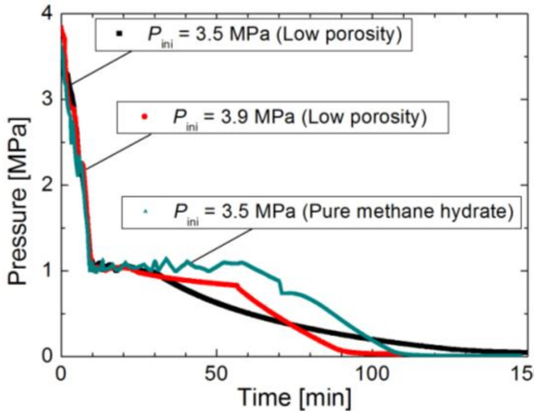

Figure 6. Time variation of pressure during depressurization process in each core.

Time variation of pressure during depressurization process in each core is shown in Fig. 6. It can be seen in Fig. 6 that pressure in all cores decreased to 1 MPa in the first 10 minutes. Also it is shown in Fig. 6 that pressure in all cores decreased to atmospheric pressure after pressure in all cores was maintained at approximately 1.0 MPa during constant time. Especially, 1.0 MPa duration of pure methane hydrate core was longer than that of low porosity cores. That result is because volume of generated methane from pure methane hydrate core was bigger than that from low porosity cores.

Time variation of temperature during methane hydrate depressurization process in each core is shown in Fig. 7. In the first 7 minutes, it can be seen that temperature decreased drastically in pure methane hydrate core and low porosity cores in Fig. 7. That temperature reduction in the cores is caused by endothermic reaction of methane hydrate dissociation. Especially, temperature reduction in the low porosity core with T_{ini} of 2.5 ℃ was higher than that in the other cores. That result is because sensible heat used for methane hydrate dissociation was higher than that in the other cores. Then, just after the temperature reduction, temperature increased to approximately 0 ℃ in all the cores. That increment indicates ice formation occurred in all the cores.

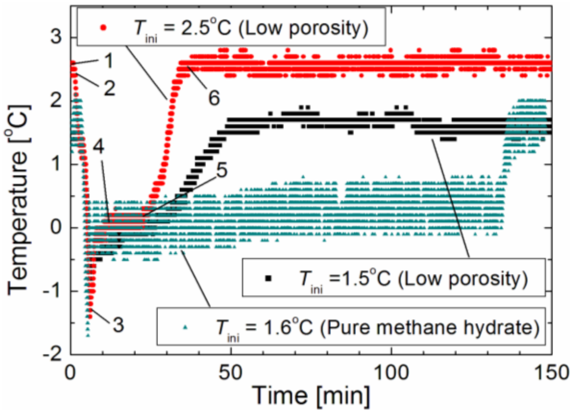

Figure 7. Time variation of temperature during depressurization process in each core.

After temperature became approximately 0 ℃, this condition was maintained in all the cores. That implies heat from boundary of the cores was mainly used for ice melting. Especially, duration of 0 ℃ condition in pure methane hydrate core was longer than that in the low porosity cores. That reason is the effect of ice formation in pure methane hydrate core was bigger than that in the low porosity cores. After ice melting finished, temperature increased to each initial temperature in all the cores. That is because heat from boundary of the cores was used for temperature increment in the cores.

C. *Discussion on Detailed Disssociation Process*

Temperature change with respect to pressure change in the low porosity core with T_{ini} of 2.5 ℃ is shown in Fig. 8. Phase equilibrium curve of methane hydrate proposed by Kamath [19] is shown in Fig. 8.

That result shows that the process of gas production from methane hydrate dissociation can be divided in several stages. Also these stages are shown in Fig. 7. In the first stage (1-2 in Fig. 8), pressure decreased without temperature change. In that process, the core was depressurized and free gas in the porous media was liberated. However, methane hydrate in the core remained stable. In the second stage (2-3), methane hydrate in the core dissociated along the equilibrium curve. Temperature reduction in this stage was caused by endothermic reaction of methane hydrate dissociation. Then, sensible heat of the core was mainly used for methane hydrate dissociation in this process. In the third stage (3-4), temperature increased to approximately 0 ℃ quickly. In that process, temperature increment implies ice formation occurred in the core. In the fourth stage (4-5), temperature remained approximately 0 ℃ with pressure reduction. That result indicates heat from boundary of the core was used for ice melting in the core. Then, gas production nearly finished in that stage because methane hydrate almost dissociated. In the fifth stage (5-6), temperature increased to initial temperature of the core drastically. That result shows heat from boundary of the core mainly was used for temperature increment of the core after ice melting finished. That trend of temperature change with respect to pressure change during depressurization was observed in the other cores.

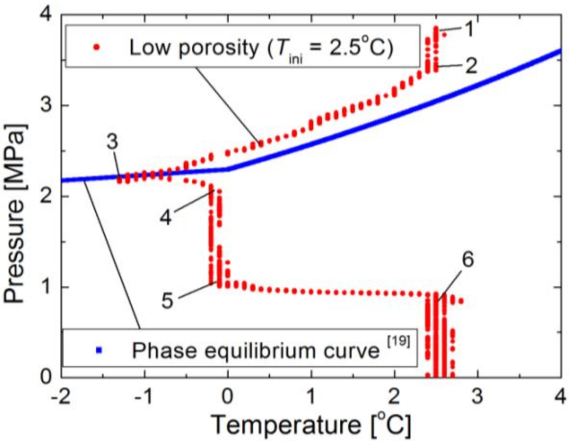

Figure 8. Temperature change with respect to pressure change in low porosity core of which initial temperature was 2.5 ℃.

D. Gas Production Analysis

Cumulative gas production in pure methane hydrate core and low porosity cores is shown in Fig. 9. It can be seen in Fig. 9 that gas production rate for pure methane hydrate core with T_{ini} of 1.6 ℃ was quick in the first 10 minutes. That result indicated sensible heat and latent heat of ice formation in the core were used for methane hydrate dissociation. Then, it is shown in Fig. 9 that gas production rate in pure methane hydrate core decreased gradually after the first 10 minutes. That is because heat supply from boundary of the core was mainly used for ice melting.

It can be seen in Fig. 9 that cumulative gas production in low porosity cores increased drastically in the first 10 minutes. That result is because pressure propagated quickly in those cores during depressurization and sensible heat and latent heat of ice formation in the cores were used for methane hydrate dissociation. Then, it is shown in Fig. 9 that gas production in low porosity cores nearly finished after the first 10 minutes because methane hydrate almost finished dissociating in the first 10 minutes.

However, gas production rate in low porosity core with T_{ini} of 2.5 ℃ was higher than that in low porosity core with T_{ini} of 1.5 ℃ in the first 10 minutes. That implies sensible heat used for methane hydrate dissociation in the low porosity core with T_{ini} of 2.5 ℃ was higher than that in the other core due to initial temperature deference between those low porosity cores.

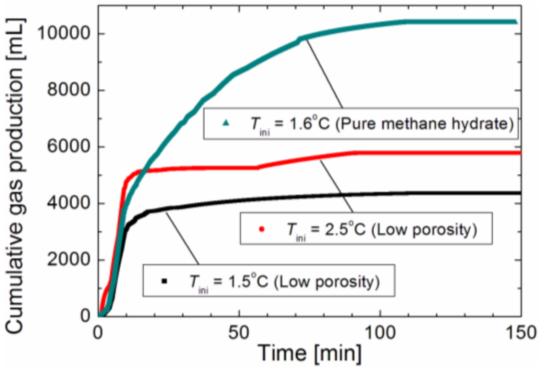

Figure 9. Comparison of cumulative gas production with respect to time in each.

From those results, it can be concluded that sensible heat of core is dominant factor for enhancement of gas production from methane hydrate core in first stage of depressurization. Therefore, it is important to increase initial temperature of methane hydrate reservoir for enhancement of gas production rate during depressurization.

IV. CONCLUSIONS

In this study, the effect of sensible heat for gas production behavior during methane hydrate dissociation using depressurization method was evaluated under various initial temperature conditions. Dissociation experiment was conducted using depressurization method. Methane hydrate dissociation and gas production behavior were evaluated in pure methane hydrate core and low porosity cores. The effect of sensible heat for gas production behaviors in low porosity cores was analyzed. The following conclusions can be drawn:

- Methane hydrate was synthesized under low temperature and high pressure conditions in laboratory.
- The process of gas production from methane hydrate dissociation can be divided into several stages. In the first stage (1-2 in Fig. 8), pressure decreased without temperature change. Then, methane hydrate in the cores dissociated along the equilibrium curve (2-3). In the third stage (3-4), temperature increased to approximately 0 ℃ dramatically. Then, temperature remained approximately 0 ℃ with pressure reduction (4-5). In the fifth stage (5-6), temperature increased drastically to initial temperature of the core.
- Gas production rate in low porosity core with T_{ini} of 2.5 ℃ was higher than that in low porosity core

with T_{ini} of 1.5 ℃ in the first 10 minutes. That implies sensible heat used for methane hydrate dissociation in the low porosity core with T_{ini} of 2.5 ℃ was higher than that in the other core due to initial temperature deference between those low porosity cores.

- It is important to increase initial temperature of methane hydrate reservoir for enhancement of gas production rate during depressurization.

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He received many prestigious honors and awards for his continuous outstanding contributions to thermal science and engineering, including the Award for Young Engineers (1989), Medal for Outstanding Paper (1999) and (2013), Thermal Engineering Division, Award for Contribution (2003), Award for Achievements (2012) from the Japan Society of Mechanical Engineers (JSME), Award for Scientific Contribution (1998), Award for Technical Contribution (2002) from the Heat Transfer Society of Japan, Scientific Research Video Award (2015) form Elsevier, and the Medal of Honor (Medal with Purple Ribbon) from the Japanese Government, etc.

Dr. Maruyama has published more than 200 academic papers and contributed to more than 10 books. The textbooks edited by him, the “Thermodynamics” and “Heat Transfer”, have become the best seller among many Japanese engineering textbooks.