In Situ Mechanical Properties of Shallow Gas Hydrate Deposits in the Deep Seabed

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Abstract Natural gas hydrates (or methane hydrates) could become a major energy source but could also exacerbate global warming, because as the climate warms, hydrate deposits deep under the oceans or in permafrost may release methane into the atmosphere. There are many shallow deposits of gas hydrates in fine-grained muddy sediments on the seafloor. However, the mechanical properties of these sediments have not yet been investigated because of the engineering challenges in coring and testing at in situ temperatures and pressures. Here we present the first uniaxial and triaxial strength and stiffness measurements of pure massive natural gas hydrates and muddy sediments containing hydrate nodules obtained by pressure coring. As a result, we were able to observe the hydrate undergoing a catastrophic brittle failure. Its strength and deformation moduli were 3 and 300 MPa, respectively. Muddy sediments containing hydrate nodules had the same strength as that of hydrate-free sediments.

1. Introduction

Natural gas hydrates (or methane hydrates) are solid clathrate compounds of gas and water molecules (Sloan, 2003). Gas hydrates are stable at relatively low temperatures and high pressures (i.e., −80 °C at atmospheric pressure and 10 °C at 7 MPa) but dissociate into mostly methane and water when heated or depressurized. They may represent a larger source of hydrocarbons than the entire world’s oil, natural gas, and coal deposits combined. As such, they could become a major energy source but also a major threat to the climate, exacerbating global warming through the release of methane which leads to positive feedback on climate warming in the glacial cycle (Beaudoin et al., 2014; Paull et al., 2007; Skarke et al., 2014). Several numerical analyses have been conducted to simulate hydrate instability due to climate change (Elliott et al., 2010, 2011; Reagan & Moridis, 2007, 2008, 2009), and they suggested that shallow hydrate-bearing deposits can be unstable due to seafloor temperature increase and release significant quantities of methane from hydrate.

There is opposing research which indicates that methane release and transport into the atmosphere may be net greenhouse gas sinks due to methane and carbon dioxide exchange across the sea-air interface (Pohlman et al., 2017). Currently, there is no conclusive proof that hydrate-derived methane is reaching the atmosphere (Ruppel & Kessler, 2017).

Moreover, hydrates can trigger large-scale underwater landslides due to climate change (Paull et al., 2007; Elger et al., 2018), while hydrate dissociation in the course of gas production by human activity may not significantly affect slope stability (Moridis et al., 2018; Rutqvist et al., 2010; Rutqvist & Moridis, 2012). Additionally, the release of gas bubbles is hypothesized to be responsible for the sinking of ships (Deming, 2004). However, the in situ mechanical properties have not been investigated because of the difficulty in testing and maintaining the low temperatures and high pressures at which such subsurface hydrates exist.

Generally, two types of natural gas hydrates are found. The first is hydrate-bearing sediments that consist of sand with hydrate-filled pores. Research on the potential for extracting natural gas from hydrate-bearing sandy sediments has progressed thanks to the advances in oil and gas industry technology. Gas has successfully been produced from gas hydrates in tests by pore fluid depressurization or thermal stimulation from a production well, which is drilled into a hydrate-bearing reservoir in permafrost or offshore (Hancock, Collett, et al., 2005; Hancock, Dallimore, et al., 2005, Anderson et al., 2011, Boswell et al., 2017, Yamamoto et al., 2017; Li et al., 2018; Yamamoto et al., 2019). Further, mechanical properties of gas hydrates under high pressure and low temperature conditions have been studied by both experimental and numerical analyses (Boswell et al., 2017; Yamamoto et al., 2017; Li et al., 2018; Yamamoto et al., 2019). The second type is hydrate nodules that can form under the seafloor. Hydrate nodules can be found in natural gas deposits that consist of hydrate/nodules and hydrate-free sediments. Mechanical properties of these sediments have not yet been investigated because of the engineering challenges in coring and testing at in situ temperatures and pressures.

Here we present the first uniaxial and triaxial strength and stiffness measurements of pure massive natural gas hydrates and muddy sediments containing hydrate nodules obtained by pressure coring. As a result, we were able to observe the hydrate undergoing a catastrophic brittle failure. Its strength and deformation moduli were 3 and 300 MPa, respectively. Muddy sediments containing hydrate nodules had the same strength as that of hydrate-free sediments.
hydrate-bearing sediments were demonstrated by using synthetic methane hydrates (Clayton et al., 2005; Ebinuma et al., 2005; Hyodo et al., 2005; Masui et al., 2005). The latest coring technology allowed us to recover natural hydrate-bearing sediments, and we investigated their petrophysical properties to predict future gas production (Jin et al., 2016; Konno et al., 2017; Priest et al., 2015; Santamarina et al., 2012, 2015; Yoneda et al., 2015).

The second type of natural gas hydrates is massive gas hydrate deposits in fine-grained muddy sediments. The hydrate exists as chunks, nodules, veins, and lenses, and fractures. In previous studies, these types of gas hydrate deposits were not attractive targets for gas production using oilfield technology, because this method is not ideal due to both the potential size limit and the likelihood for significant disturbance of sensitive sea-floor ecosystems (Boswell & Collett, 2006). A special class of gas hydrate occurrences is massive gas hydrate mounds that lie exposed on the seafloor and extend to unknown depths (Boswell & Collett, 2006).

According to the latest survey by the Japanese government, the Sea of Japan contains 1,742 gas chimney structures (Agency for Natural Resources and Energy, Ministry of Economy, Trade and Industry, 2016) in which hydrates are expected to be distributed (Matsumoto, 2017). Using three different methods (i.e., logging while drilling, controlled source electromagnetic sampling, and core sampling), the total amount of hydrates in the Umitaka Spur (with an area of 200 × 250 m and a depth of 120 mbsf [meters below the seafloor]) is estimated to be equivalent to 600 Mm³ of natural gas. This corresponds to a mound composed of around 60% hydrate by total volume fraction. Since the density of hydrates is 0.91 g/cm³, which is less than that of water, the mound is expected to have a large buoyancy and is considered to be mechanically metastable. An assessment of the mechanical stability of the gas hydrate mound is important for our understanding of methane releases from the seafloor to the sea or atmosphere in the global carbon cycle on the earth. In addition, especially if human activities change the shape of the mound by mining or drilling for energy resources, shaved or hollow out hydrate mounds will become a potential geohazard and it may have an effect on seafloor ecosystem. However, research on the mechanical properties of massive gas hydrate deposits in fine-grained muddy sediments is very limited, even using synthetic samples. Moreover, there is no information about mechanical properties of natural samples. There have been studies on the compression strength of methane hydrates using synthetic samples. Powder ice was used to form a hydrate in a triaxial testing apparatus, and high-pressure methane gas was then injected into it. The compression strength was obtained under a confining pressure of several to tens of megapascals (Durham et al., 2003; Hyodo et al., 2002; Nabeshima et al., 2005; Stern et al., 1996, 1998, 2000). According to these studies, hydrates depend on the compression strain rate, confining pressure, and temperature. Methane hydrates are about 20 times stronger than ice (Durham et al., 2003). These studies were conducted at a relatively higher confining pressure and lower temperature than those of the deep-seabed environment. To assess the impact of hydrate veins on the mechanical properties of fine-grained muddy sediments, triaxial compression tests were conducted using vertical cylindrical tetrahydrofuran hydrate veins (Smith et al., 2018). They reported that increasing the hydrate vein diameter significantly increases strength and stiffness.

Here we present the first strength and stiffness data for massive natural gas hydrates (MNGHs) collected from a shallow gas hydrate deposit using pressure coring and analysis technology. During the coring process in the Sea of Japan and transport to the testing equipment, high-pressure and low-temperature conditions were maintained to be within the hydrate stability boundary.

2. Sampling and Testing Procedure

Gas hydrate cores were taken from the seabed deep under the Sea of Japan by Meiji University under the program for shallow methane hydrate research sponsored by the Ministry of Economy, Trade and Industry of Japan. The drilling vessel Poseidon-1, operated by Fukada Salvage and Marine Works, was used during the PS15 drilling expedition, which was undertaken from August to November 2015. A hybrid pressure coring system (Kubo et al., 2014) was used, and the recovered cores were cut on the ship by a pressure core analysis and transfer system (Schulteiss et al., 2009) and stored in a special chamber. Samples were then brought from Niigata to Sapporo and kept at the National Institute of Advanced Industrial Science and Technology in Hokkaido under a pressure of 10 MPa at a temperature of 4 °C until the test. Pressure and temperature were kept within the methane hydrate stability boundary during the entire process of coring, transferring from drill site to laboratory, and loading into the testing apparatus.
The MNGH mound from where the hydrate samples were recovered is shown in Figure 1a (Umitaka Spur in the Sea of Japan). There was an acoustically blank gas chimney structure in the mound, likely because of the large acoustic reflections from the MNGHs at shallow depths below the seafloor (Freire et al., 2011). Coring was conducted at an underwater depth of around 900 m, and samples were retrieved from the core sampler and placed in the onboard storage chamber by the pressure core analysis and transfer system. We obtained core samples from four locations (Figure 1a; Borehole names are J23R, J14R, J07RB, and J13R), which were categorized as massive hydrates and nodule hydrates formed in muddy sediments (Figure 1b and Table 1). Massive hydrates were recovered from 14.2 and 14.3 mbsf and muddy sediments with hydrate nodules were taken from 11.8, 29.1, and 64.4 mbsf (Table 1). The geometry of hydrates in the core samples are shown in Figure 1c and supporting information Movies S1–S5. Hydrate was segregated by image processing. Massive hydrates have some rind of mud, but inclusions like mud or carbonate were not observed from cross-sectional X-ray CT (Table 1. and supporting information Movies S6–S10). The temperature of the seafloor of this region is approximately 0 °C, and the temperature gradient is approximately 0.1 K/m, which is a relatively high value considering the upstream methane flux (Machiyama et al., 2009).

The samples were transported from the drilling vessel to an onshore laboratory in a reefer container under pressure control. X-ray computed tomography scans of some of the collected samples were taken using an X-ray transparent pressure chamber (Jin et al., 2016; Yoneda et al., 2015) (Figure 1b). The massive hydrate appeared to be homogeneous and no cracks were evident in the scans (Figure 1b; J23R). The samples were then removed from the pressure chamber and subsampled for further testing under pressure. They were then loaded into a transparent acrylic cell triaxial testing system (Yoneda et al., 2015) (see Supporting Text S1 and Figure S1), and a uniaxial compression test was carried out at a water pressure of 10 MPa and a temperature of 4 °C, similar to the conditions at the bottom of the ocean. Each sample was sandwiched between a top cap and a pedestal in the transparent acrylic cell triaxial testing cell, which was filled with distilled water. Hydraulic pressure was applied isotropically, and the sample was then compressed in the vertical direction. The photographs taken of the core in the acrylic pressure cell show that it is composed of a translucent crystalline material, although there was drilling mud attached to its surface (Figure 1b). Nodule hydrate sediments contain hydrate crystals distributed in mud, which range in size from several millimeters to several centimeters. From the outside, the samples look like mud.

The load (P) acting on a specimen, which does not depend on the temperature and pressure of the surroundings, was measured using a load cell. The compression speed was specified according to the specimen height as the strain rate, which is 0.1%/min and 0.01%/min (Table 1). The axial stress q (MPa)

\[ q = \frac{P}{A \times 10} = \frac{P}{A_0(1 + \varepsilon_r/100) \times 10} = \frac{P}{A_0(1 - \varepsilon_a/100) \times 10} \tag{1} \]

at each strain level was calculated using the obtained load (P) [kN] and the cross-sectional area \( A \). The cross-sectional area is \( A = A_0 \varepsilon_n \), where the initial cross-sectional area of the sample is \( A_0 \) [cm\(^2\)], and the lateral strain is \( \varepsilon_n \) [%]. Here we assume the volumetric change during the compression equals zero, so the axial (or vertical) strain is \( \varepsilon_a = - \varepsilon_r \) [%]. An exterior view of a specimen during testing can be found in supporting information Movie S11, which runs at a real-time speed. A triaxial test of the nodule hydrate muddy sediments was conducted using sealing sleeves to apply in situ effective stress (Yoneda et al., 2013, 2015, 2017). The consolidation process was conducted until the pore pressure drop reached a consolidation degree of more than 98%. The total gas volume of each core sample was measured using a wet gas flowmeter through a gas/water trap by depressurization.

### 3. Results and Discussion

The stress-strain curves obtained by applying uniaxial compression to MNGH samples are shown in Figure 2a (see supporting information Movie S1 for an exterior view during compression and Table 1 for the testing conditions and sample index properties). The MNGH samples were compressed at two different strain rates: 1.0%/min and 0.1%/min. They exhibited an elastic behavior up to approximately 1% axial strain. The maximum deformation (Young's) modulus in this elastic region was ~300 MPa for both samples, and the peak stress values of the two samples were also similar, at 3 MPa. The nodule hydrate sediments were compressed under undrained triaxial conditions, which simulate the in situ stress state corresponding to the overburden and drainage conditions (Figure 2b). Strength increased with increasing sampling depth below the seafloor.
Figure 1. Sample information. (a) Locations of the drilling sites for PS15 (red) and previous wells (Kataoka & Matsumoto, 2012) (yellow) as well as the acoustic tomography of the Umitaka Spur. The map was drawn using the Generic Mapping Tools (Wessel & Luis, 2017). (b) X-ray computed tomography images and photographs through an acrylic pressure cell under a water pressure of 10 MPa at a temperature of 4 °C. (c) Hydrate geometry in the sample. Hydrate in X-ray CT images were applied thresholds and constructed 3-D view by ImageJ (Rasband, 1997–2012). Supporting information Movies S6–S10 show 360° rotation.
However, the strength of the nodule hydrate sediments was less than 1/10 that of the massive hydrate because of the unconsolidated muddy sediment, which generates excess pore pressure during compression. Both MNGH and nodule hydrate sediments were depressurized, and the total gas volume was obtained (Table 1). The dissociation behavior of MNGHs is shown in supporting information Figure S2 and Movie S12. The hydrate volume fraction of MNGHs (sample from J23R) was approximately 93% which is calculated based on diameter $D = 5.08$ cm (coring bit size). Precise diameter of the sample cannot be measured from X-ray CT images because of very small density gap between hydrate and surround water. In addition, fine-grained mud rind blocks photographic measurement. Assuming that the actual sample diameter is 1 mm smaller than the calculated value, the hydrate saturation falls down from 100% to 96%. Continuous observation of cross-sectional X-ray CT images confirm that there are no clear inclusions like mud, carbonate, or gas (supporting information Movies S1–S5). At this moment, there are still possibility of water existence in the massive hydrate, but it should be less than a few percent. We may conclude that the total volume fraction of MNGH sample in this study is close to 100%. And the volume fraction of nodules hydrate in total volume (sample from J14R, J07RB, and J13R) is approximately 20% which was calculated based on a diameter $D = 5.36$ cm. Another 80% consists of fine-grained muddy sediments, which has a mean particle size of approximately 5 μm. A visual observation of Translucent sample (see supporting information Movie S11) may identify the MNGH sample as a polycrystalline hydrate, although the transparency observed in a single hydrate crystal cannot be confirmed using this method. The strength of the massive gas hydrate is compared to that of polycrystalline ice in Figure 3a. The strength of ice depends on the strain rate; this dependency is particularly apparent in the ductile failure mode that arises during a compression test (Gold, 1977; Kvenvolden & Lorenson, 2013) and is relatively small in tensile failure mode (Petrovic, 2003; Haynes, 1978). For our samples, no clear differences in strength were observed at different strain rates. We believe that this is due to the brittle failure mode and because tensile stress has more influence compared to shear stress in the samples. Polycrystalline ice shows brittle failure below $-50 \, ^\circ\text{C}$; above this temperature, it undergoes ductile failure. The maximum stress decreases as the ice approaches its melting point. We see that the strength of MNGHs is almost equal to

### Table 1: Core Samples

| Sampling Site | Symbol | Unit | J23R(a) | J23R(b) | J14R | J07RB | J13R |
|---------------|--------|------|---------|---------|------|-------|------|
| Sample Name   |        |      | Massive | Massive | Nodule | Nodule | Nodule |
| X-Ray CT Image (Supporting Information Movies S1–S5) | | | | | | | |
| Cross Section (Diameter = 33.6 mm) | | | | | | | |
| Dark Gray: Hydrate | | | | | | | |
| Light Gray: Fine-Grained Sediment | | | | | | | |
| Depth Below Seafloor (m) | 14.2 | 14.3 | 11.8 | 29.1 | 64.4 |
| Averaged Horizontal P Wave Velocity ($V_p$) (m/s) | 3,010 | 2,990 | 1,560 | 1,550 | 1,470 |
| Vertical P Wave Velocity ($V_p$) (m/s) | 3,350 | 3,500 | 1,880 | 1,850 | 1,860 |
| Vertical S Wave Velocity ($V_s$) (m/s) | 1,730 | 1,610 | 580 | 1,110 | 710 |
| Sample Height (cm) | 7.53 | 7.46 | 9.98 | 9.14 | 7.08 |
| Core Diameter ($D$) (cm) | 5.08 | 5.08 | 5.36 | 5.36 | 5.36 |
| Test Condition | Uniaxial | Uniaxial | Triaxial | Triaxial | Triaxial |
| Confining Pressure (MPa) | 10 | 10 | 10.05 | 10.15 | 10.22 |
| Effective Stress | 0 | 0 | 0.05 | 0.15 | 0.22 |
| Compression Strain Rate | 1.0 | 0.1 | 0.05 | 0.05 | 0.05 |
| Maximum Deviator Stress ($q_{\text{max}}$) (MPa) | 3.03 | 3.24 | 0.0585 | 0.119 | 0.186 |
| Maximum Deformation Modulus ($E$) (MPa) | 250 | 287 | 3.07 | 21.8 | 27.6 |
| Total Gas Volume (0 °C, 1 atm) ($V_{\text{gas}}$) (cm$^3$) | 23,100 | 22,600 | 4,480 | 5,580 | 5,050 |
| Hydrate Total Volume Fraction ($\phi_h$) (%) | 93.4 | 92.4 | 14 | 19 | 19 |
| Porosity Without Hydrate ($\phi_s$) (%) | — | — | 73.5 | 73.4 | 67.7 |
| Saturated Soil Density Without Hydrate ($\rho_{\text{sat}}$) (g/cm$^3$) | — | — | 1.409 | 1.410 | 1.536 |
| Grain Density of Sediment ($\rho_g$) (g/cm$^3$) | — | — | 2.54 | 2.54 | 2.66 |
| Mean Particle Size ($D_{50}$) (μm) | — | — | 4.2 | 5.4 | 5.1 |

$^a$Coring bit size. $^b$Inner diameter of core liner.

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that of ice at \(-10\ °C\). The hydrate phase boundary at 10 MPa is approximately 13 °C, which means the temperature depression is \(13 - 4 = -9\ °C\). The similar temperature depression of ice and hydrate may indicate that they have similar strength. The maximum deformation modulus of ice is almost inversely proportional to the temperature (Figure 3b). The maximum deformation modulus of MNGHs is approximately \(1/5\) that of ice at \(-10\ °C\). Thus, MNGHs are more easily deformed than ice, even though they show brittleness and are prone to catastrophic failure.

To determine the type of methane hydrate in the MNGH sample, we compared our results with previously published values with wide ranges of crystalline grain size. The Young modulus of a single methane hydrate crystal (monocrystalline hydrate) was measured to be 8.52 GPa experimentally (Shimizu et al., 2002) and estimated to be 7.68–9.71 GPa according to a molecular dynamics (MD) simulation (Wu et al., 2015). The results of MD simulations suggest that the maximum compression strength of monocrystalline and polycrystalline methane hydrates is approximately 750 and 240 MPa, respectively, for a crystal grain size of 20 nm (Cao, Wu, Zang, Fang, & Ning, 2018; Wu et al., 2015). Both the Young modulus and the strength of the MNGH are lower than these values. The MD simulation suggests that the Young modulus and strength decrease as the grain size increases (Wu et al., 2015), and this crystal grain-size dependency has been confirmed in ice (Currier & Schulson, 1982) and other materials such as steel (Hall, 1951; Petch, 1953). According to these studies, the failure strength is in inverse proportion to the grain size of crystal. Because of the failure theory posed by Griffith (1921), it is known that all existing materials contain small cracks, which fracture at a small force of \(1/100\) to \(1/1000\) of the theoretical strength because these cracks cause stress concentration. It has been confirmed with ice that such small cracks will be generated when stress is applied (Gold, 1972), leading to the failure of the material. The cracks that cause failure are scratches on the material surface, and there are also cases of concentration and movement of dislocations in the structure which are crystallographic defects (Taylor, 1934, Webb & Hayes, 1967). In the case of hydrates in nature, it is inferred that narrowing in the crystal grain boundaries during the crystal growth process and the fine particles left behind are the causes of failure at a low stress level. We summarized a relationship between maximum stress and crystalline grain size for methane hydrate and ice from both experimental and numerical results in Figure 3c. Extrapolation the result of using the Hall-Petch empirical equation (Currier & Schulson, 1982) for ice shows good correlation with the MD simulation result (Cao, Wu, Zang, Fang, Peng, Li, Vlugt, & Ning, 2018). The crystalline grain size of massive gas hydrates may be estimated by considering the Hall-Petch empirical equation using the results of this study maximum stress 3.02–3.24 MPa under 4 °C. Therefore, we conclude that MNGHs are polycrystalline gas hydrates with a relatively large grain size on the order of tens to hundreds of microns.

The relationship between the strength and depth of nodular gas hydrate sediments is shown in Figure 3d. The strength of the nodule hydrate sediments obtained at depths of 11.8, 29.1, and 64.4 m in this study
correspond to the results of a previous study on hydrate-free sediments, where an increase in strength was observed at depths down to 30 m below the seafloor (Kataoka & Matsumoto, 2012). The strength of hydrate-free sediments obtained via piston coring may be underestimated because of the expansion of the volume of dissolved gases. The pressure core result, which varies only slightly, is at the upper limit of the strength. Our results suggest that the nodule hydrates that occupy 20% of the total volume provide no contribution to the strength of the sediment. The mechanical behavior is governed by the host muddy sediments, which occupy 80% of the volume of the sample. Lei and Santamarina (2019) performed numerical simulations to investigate the influence of hydrate lens on sample strength. They suggest that when a hydrate lies on the potential shear band of the sample, the strength increases by hydrate

Figure 3. Comparison of strength and stiffness of MNGHs and ice. (a) Maximum stress (or strength) as a function of temperature for grains of size 0.9 ± 0.4 mm (Arakawa & Maeno, 1997). (b) Maximum deformation modulus as a function of temperature (Arakawa & Maeno, 1997). (c) Maximum stress as a function of crystalline grain size. (d) Relationship between shear strength ($q_{max}/2$) and depth below the seafloor in vane shear tests for samples from wells 3296, 3299, 3301, 3304, and 3307 in the same area (Kataoka & Matsumoto, 2012).
existence. Smith et al. (2018) conducted triaxial compression with cylindrical tetrahydrofuran-hydrate veins and reported that increasing hydrate vein diameter significantly increases strength and stiffness but there is no significant difference if the hydrate area ratio $A_{ vein}/A_{ specimen} < 0.014$. Hydrate nodules in this study are similar to a cube and relatively small for the sample size, which may not disturb shear banding. Therefore, the hydrate likely has no contribution to the sample strength.

4. Conclusions

In summary, pressure-core-based uniaxial and triaxial compression tests were carried out to investigate the mechanical properties of shallow gas hydrate deposits. Natural core samples were recovered from the deep seabed under the Sea of Japan. The hydrates were kept at a pressure and temperature necessary for stability during the entire processes of coring and transfer from the drill site to the laboratory. MNGHs showed brittle failure under uniaxial compression, with strength and deformation moduli of 3 and 300 MPa, respectively, making it approximately 1/200 weaker than a single crystal. This strength is equivalent to that of polycrystalline ice under atmospheric pressure at $-10 ^{\circ}C$. The deformation modulus was 1/5 that of polycrystalline ice. Muddy sediments containing hydrate nodules had the same strength as that of hydrate-free sediments recovered from the same regions. Centimeter-sized nodules of gas hydrates may not affect the strength of the sediment because 80% of the volume pertains to the muddy host sediments, which govern their mechanical behavior.

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