STRENGTH AND DISSOCIATION PROPERTY OF METHANE HYDRATE BEARING SAND

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ABSTRACT
A series of triaxial tests has been carried out on the mechanical properties and dissociation characteristics of sands containing methane hydrate using an innovative high pressure apparatus which has been developed to reproduce the in-situ conditions expected during proposed methane extraction methods. It was found that the strength of MH sand increased with MH saturation due to particle bonding. Dissociation by heating caused large axial strains for samples with an initial shear stress and total collapse for samples consolidated in the metastable zone. In the case of dissociation by de-pressurisation, axial strains were generated by increasing effective stress until a stable equilibrium was reached. However re-pressurisation led to the collapse in the metastable zone.

Keywords: methane hydrate-bearing sand, triaxial compression test, strength, stiffness, stability

INTRODUCTION
Methane hydrate (MH) is a solid compound in which a large amount of methane is trapped within a crystalline structure of water, forming a solid similar to ice. It is known to exist in a stable condition under certain temperature and pressure conditions. Its existence has been confirmed in permafrost layers and in deep ocean floors[1]. Although Japan has no permafrost zones, it is believed that MH exists in the seafloor, and development is underway for MH to be a future energy resource, replacing oil and coal [2, 3]. Recently, the existence of a large-scale MH natural gas reservoir has been investigated in the Nankai Trough [4, 5]. Worldwide, MH is believed to exist in various forms, such as massive structures within muddy layers or at the surface of deep seabeds, or embedded within the pores of

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sandy layers[6]. The seabed in the Nankai Trough consists of alternating layers of sand and clay, and it has been confirmed that the MH-enriched zone is buried within the voids of the sand layer [7].

Currently, the method proposed for abstracting MH in the Nankai Trough is by drilling a shaft into the MH-rich layer, and heating, depressurizing, or inserting hydrate inhibitors, causing the MH to become dissociated into methane and water after which the gas could be collected [8]. Using these methods, the solid MH existing in the pores within the soil is transformed into gas for collection; in the process, complex physical events, such as changes in the soil structure and thermal conductivity, pore fluid and gas migration, and other complicated phenomena need to be considered. It is predicted that a combination of such phenomena could cause consolidation and shear deformation of the ground due to changes in the effective stress and decrease in soil particle strength. Therefore, it is important to investigate the mechanical properties of MH-bearing sediments, for safe and economical exploitation.

**TESTING EQUIPMENT AND SAMPLE PREPARATION**

**Triaxial Testing Apparatus**

Figure 1 shows the temperature-controlled high pressure triaxial testing apparatus which was developed such that the back pressure and confining pressure could be controlled under various temperature and high pressure conditions in order to examine the mechanical behaviour of MH-bearing sand specimens under deep seabed

![Experimental apparatus](image-url)
stress and temperature conditions. The maximum permissible load was 200kN. To remove the influence of piston friction, a cylindrical-shaped loading cell that was not affected by temperature and pressure was set up in the cell as shown in Figure 1(c). Cell pressure could be increased up to 30MPa. To reproduce back pressure associated with this high pressure condition, a syringe pump was installed as shown in Figure 1(d). By using incompressible solution in the cylinder, the measurement of volume change of the specimen was enabled by calculating the amount of penetration of the piston in the cylinder. To measure the volume change of partially saturated soil during dissociation, an inner cell was installed, with a mechanism similar to the syringe pump for back pressure. Temperature was controlled by a system which circulated the cell fluid from a low temperature water tank set up outside allowing the temperature to be adjusted from -35°C to +50°C.

**Specimen Preparation**

Based on visual observation of the undisturbed core samples obtained from the Nankai Trough [9], it is believed that MH in-situ is buried within the pores between grains of the sand. Based on this, MH-bearing sand was artificially produced using the grain size distribution curve of the undisturbed core sample [7] as shown in Figure 2, and Toyoura sand was chosen as the host material. First, the amount of water for the target MH saturation was mixed with sand whose volume corresponded to a target density. The moist soil was placed in 15 layers in a mould measuring 30 mm in diameter and 60 mm high, with each layer compacted by a tamper 40 times. In order for the specimen to stand by itself, the mould containing the sand was placed in a freezer. The frozen specimen was then removed from the mould and placed on the pedestal, and the membrane was installed as shown in Figure 1(e). Because the specimens in the tests were subjected to low temperature and high pressure, rubber membranes conventionally used in triaxial tests were avoided; instead, silicon-type membranes were used because of their flexibility under low temperature/high pressure conditions and butyl rubber was used in long term tests, such as MH dissociation because silicone is to some degree permeable to methane gas. The inner cell set up is shown in Figure 1(f).

**Generation of MH and Experimental Procedure**

After forming the specimen, it was subjected to a series of processes under specific temperatures and pressures, as depicted in Figure 3. First of all, the frozen specimen (a) was thawed to room temperature inside the triaxial cell (b). Then, the back pressure was gradually increased to 4MPa while methane was injected into the specimen (c) by filling the pores of the specimen with methane. At this time, the gas pressure was increased over a period of time so that the specimen's moisture content would not become non-uniform as a result of the pressurized injection. Next, the temperature in the triaxial cell was lowered to 1°C where the MH was stable, and the specimen environment was kept under constant temperature and pressure conditions for 24 hours. By keeping the gas pressure constant in the connection between the specimen and the syringe pump and by observing the amount of gas flowing at various times, the transformation of water within the pores into hydrate was judged to be complete if there was no marked change in the amount of gas, as indicated in the figure. Note that the plots in the figure show some irregularities because in inducing the gas to flow into the specimen to promote the generation of MH, the volume of gas in the specimen was increased and decreased slightly while adjusting the upper and lower syringe pumps.
After the hydrate was generated, water under constant pressure was allowed to infiltrate the specimen. Then, the pore water pressure was applied (e) and the temperature was adjusted to the prescribed test condition (f). While keeping the pressure constant, consolidation was carried out until the specified effective stress was reached and shearing was conducted with a strain rate of 0.1%/min. After shearing, the temperature in the specimen was increased and MH dissociated; the amount of gas was measured using the gas mass flow meter shown in Figure 1(b)-(i). The amount of gas measured was then converted into MH saturation, (assuming the density of MH was 0.912 mg/m³).

TRIAXIAL COMPRESSION TESTS

Testing Condition

In the experiments, the specimens were subjected to different levels of effective confining pressure (1, 3, 5MPa), back pressure (5, 10, 15MPa) and temperature (1, 5, 10°C). For all tests, a shearing rate of 0.1%/min was adopted. Tests were performed on two sets of specimens, porosity n=40% (relative density Dr=90%) and n=45% (Dr=40%), with various MH saturation ratios. The maximum deviator stress indicated in the table refers to the peak stress obtained from the deviator stress-axial strain relation; however, if peak was not observed, the deviator stress corresponding to 15% axial strain was used.
Testing Results

Figure 4 shows the deviator stress, axial strain and volumetric strain relations for isotropically consolidated $n=40\%$ specimen ($Dr=90\%$) with different MH saturation and subjected to effective confining pressure $\sigma_c'=5\text{MPa}$. From the figure, it is observed that the specimens show compressive volume change and strain hardening behaviour at this level of confining pressure, notwithstanding the high relative density. Moreover, a marked increase in the initial stiffness and strength is observed as the MH saturation of MH-bearing specimen increases. The volumetric strain changed from compressive to dilative, and for specimens with $S_{MH}=50\%$, significant dilative behaviour was observed. This is believed to be due to the hardening action induced by MH on the sand particles. Although axial strain $\varepsilon_a=15\%$ was exceeded in most tests, high residual strengths were observed in all cases, as shown in Figures 4. In order to examine what extent of axial strain would affect the residual strength, tests were performed on a pure sand specimen and one with MH saturation $S_{MH}=53.1\%$ for axial strains up to $\varepsilon_a=50\%$, and the results are shown in Figure 5. From the figure, the peak strength for the MH-bearing sand took place near $\varepsilon_a=12\%$ after which strain hardening started to occur at $\varepsilon_a=15\%$ and, although not shown in the figure, the deviator stress and volumetric strain remained constant up to about $\varepsilon_a=42\%$, indicating steady state conditions.

On the other hand, although the pure sand specimen demonstrated a similar steady state condition, significant difference in residual strength can be observed even at $\varepsilon_a=50\%$. To explain this, reference is made to previous research conducted on the hardened soil structure of sand to which cement was added [10]. Based on observation of the internal structure of the specimen with 4% and 7% cement, the authors postulated that when peak strength was mobilized, a mass of particle with strength corresponding to the hardened strength was generated at the shear surface; consequently at the residual state, a mass of the same strength as the particle was destroyed. Thus, they reported that the residual strength of the cemented sand was the same as the uncemented specimen. For the present test on MH-bearing sand with $S_{MH}<30\%$, a similar phenomenon can be assumed to occur. On the other hand, the higher residual strength observed in specimens with high $S_{MH}$ is postulated to be due the contribution of MH flakes which were peeled off from sand particle surfaces and were buried within the pores of the sand during shearing, resulting in increased residual strength.

Tests under different effective confining pressures ($\sigma_c'=1\text{MPa}$, $3\text{MPa}$, $5\text{MPa}$) but with constant back pressure, temperature and MH saturation were performed to examine the influence of the effective confining pressure on the shear behavior of MH-bearing specimens. The test results for each confining pressure are shown in Figure 6 together with the results for pure sand. For an effective confining pressure $\sigma_c'=1\text{MPa}$, the deviator stress-axial strain behaviour showed a strain softening tendency after peak, while the volumetric strain was compressive at first changing to dilative. This tendency changed with
increasing effective confining pressure, i.e., the response changed gradually to a strain hardening and compressive tendency. When $\sigma_c = 5\text{MPa}$, a clear peak was not observed and the volumetric strain became compressive.

Figure 8 considers the effect of back pressure on the shear behaviour by shearing specimens with similar effective confining pressure, temperature and MH saturation but different back pressure. From the figure, it is seen that the initial stiffness increases markedly while the peak strength remains higher when the back pressure was doubled. The results of tests to examine the effect of temperature on the shear response are shown in Figure 9. Although the temperatures considered were not much different at 1°C and 10°C, large differences in response can be seen with higher stiffness and higher peak strength observed for the specimen at lower temperature. Since deep seabed temperatures would be above zero, tests were performed at above zero temperatures and confirmed the temperature and pressure dependency even for these conditions.

**Cementation of MH-bearing Sediment**

The relation between maximum deviator stress of the MH-bearing sand and MH saturation obtained from triaxial compression tests considering all experimental conditions listed in Table 2 are plotted in Figures 10(a), (b) and (c). Porosity and effective confining pressure are major factors affecting the shear behavior of sand. Figure 10 presents the maximum deviator stress plotted against MH saturation for MH bearing sand with various porosities and confining pressures. Figures 10(a), (b) and (c) show the results for effective confining stresses, $\sigma_c = 5\text{MPa}$, 3MPa and 1MPa, respectively. Furthermore, similar testing results from Miyazaki et al.[11] are also plotted on Figures 10(b) and (c). Although the effect of temperature and back pressure has been discussed above, their influence is relatively small when compared to that of confining pressure. It is observed from the figures that the maximum deviator stress increases with increase in MH saturation as well as with increase in confining pressure. When the three density states are compared, the increase in strength due to MH appears to be more significant for dense specimens. Generally, it is known that the shear behavior of sands is dependent on effective stress with stress-strain curves changing from strain softening to strain hardening with increasing effective confining pressure. Moreover, loose sands have a higher tendency for strain softening behaviour compared to dense. It is therefore difficult to isolate the effects of the MH cementation. Figures 11 to 13 show the deviator stress differences relative to the host sand for different MH saturation, effective confining stress and relative porosity.
density. The stress differences for different MH saturations at the same confining pressure of 3MPa and porosity around 40% are summarized in Figure 11. The peak deviator stress difference as a result of MH cementation was apparent in each case at 1%~2% strain regardless of the MH saturation. The strength decreased almost linearly with strain after the peak was reached. In Figure 12 the relations between the deviator stress difference and axial strain for constant porosity \(n=40\%\) and MH saturation \(S_{MH}=50\%\) but varying effective confining stress \(\sigma_c' = 1, 3 \text{ and } 5\text{MPa}\) are depicted. The stress increment increased with increasing effective confining pressure and the maximum deviator stress difference again occurred in the vicinity of 1%~2% axial strain regardless of effective confining pressure. It should be noted that there is a frictional contribution to the strength increase rather than purely being due cementation.

To examine the influence of soil density the relation for the deviator stress differences for porosities of \(n=40\%\) (loose) and 45\% (dense) are shown in Figure 13 for a constant effective confining stress \(\sigma_c' = 3\text{MPa}\) and low and high MH saturations. In this case the major influence was the MH saturation although there was a more rapid decay of strength for the loose specimens. The maximum deviator stress difference occurs at an axial strain of 1%~2% regardless of the porosity. It can also be seen that the decrease in the deviator stress difference after the peak is as marked for loose sand as it is for dense.

The strain level at which the peak increment in strength occurred due to MH cementation was 1%~2% regardless of the effective confining stress, porosity and MH saturation. For the MH-bearing sand the behaviour changes from compressive to dilative between 1% and 2% axial strain. Thus, it is clear that the MH cementation effect is at a maximum when the soil particles begin to roll over each other. The relation between the deviator stress difference at an axial strain of 1.5% and MH saturation is shown in Figure 14. Regression lines for each effective confining pressure are drawn on the figure. It is observed that the strength increases due to MH cementation increases exponentially with MH saturation. The regression lines also show that the deviator stress difference increases with increasing effective confining pressure.
MH DISSOCIATION TESTS

Testing Condition and Stress Path of Dissociation Tests

After consolidation at an effective stress equivalent to in-situ conditions, MH dissociation was performed by either increasing the temperature or decreasing the pressure. The experimental conditions and the corresponding experimental results are summarized in Table 2. Figure 16 shows the stress paths followed for three different cases. In Case 1 there was no initial shear stress and the temperature was increased to simulate thermal recovery or the mean principal effective stress was increased from a to a’ simulate the depressurization method. In the case of depressurization, the pore water pressure was allowed to increase again after dissociation to represent post-production equilibrium conditions being restored. In Case 2 an initial shear stress was applied before using thermal recovery or depressurization. Following this the temperature was allowed to increase at b or the pore water pressure was decreased from b to b’, and from b’ back to b after the dissociation of all the MH. Finally in Case 3 the stress path was taken into the metastable zone between the failure envelopes for pure sand and MH-bearing sand. The purpose of this test was to study the effect of dissociation and consequent loss of particle bonding in the zone where failure could occur for an unbonded sand.

Thermal Recovery Method

The temperature was increased to cause dissociation by circulating heated oil through a double walled annulus surrounding the inner cell. Figure 17 shows the deviator stress, axial strain and volumetric strain relations for isotropically consolidated n=40% specimens (Dr=90%) for Case1, Case2 and Case3 subjected to an initial effective confining pressure $\sigma_c = 5$ MPa. From the figure, it is observed for Case1 that no deformation was occurring during the dissociation of MH. Next, in case2, the axial strain increased to $\varepsilon_a = 9\%$, however, failure conditions were not reached. Finally, in case3, it was observed that the sediment failed due to dissociation of MH. That is, the sediment will collapse when the methane hydrate dissociates in the state between the failure envelopes for pure sand and MH-bearing sand. Figure 18 shows the variation of temperature, MH saturation, and volumetric and axial strains with elapsed time. For stress state (a) there was only a small change in axial strain related to the small volumetric compression. For stress state (b) there was a marked increase in axial strain reaching about 10% as the MH dissociated. Total collapse did not occur as the soil was still below the failure envelope. In the case of stress state (c) the strain initially increased until the start of MH dissociation when a rapid collapse occurred as the failure envelope moved towards that for the uncemented sand.

The thermal method required the input of heating energy. During this process deformation of the soil occurred under high initial shear stresses indicating that should this method be used for methane production high levels of deformation could occur.
De-pressurisation Method

Figure 19 shows the variation of back pressure, MH saturation, volumetric and axial strains during the de-pressurization and re-pressurization stages. In each case there was an initial compressive volumetric strain because of the increase in effective stress. Further volumetric strain occurred as the MH dissipated and bonds dissolved. Dissociation occurred when the back pressure dropped to 4.3MPa. Dissociation continued for approximately 2 to 3 hours from initiation, with axial strains still increasing up to 3 hours after the start of the test. After 5 hours the pore water pressure was ramped back up to 10MPa over one hour to represent a restoration of post-dissociation equilibrium conditions being restored in-situ. In Cases 1 and 2 there was an elastic recovery of axial and volumetric strains due to a decrease in effective stress. However in Case 3 collapses occurred as the now non-bonded soil moved outside the failure envelope.

Figure 20 shows the stress ratio $\eta (q/p')$, axial strain and volumetric strain relations during the tests. It can be seen that the axial strain increased dramatically when the stress strain curve reached the strength of pure sand as the water pressure recovered.

In contrast to the thermal method no deformations were observed despite the increase in effective stresses. After dissociation during pore
water pressure recovery large deformations may occur in the metastable zone. In the Nankai trough the geology consists of turbidite alternating layers of sands and clays. Where the sands containing the methane hydrate are sandwiched between impermeable clay layers the depressurization method would be most effective.

CONCLUSIONS
Tests have been carried out on the mechanical and dissociation properties of sands containing MH using an innovative high pressure apparatus which allows control of back pressure and temperature. The following conclusions were drawn:

1) A double walled high pressure triaxial testing equipment with wide ranging temperature control was developed for studying the dissociation and strength characteristics of methane hydrate bearing soils.

2) Artificial samples containing methane hydrate were successful produced by controlling stress and temperature conditions.

3) Volume change of partially saturated dissociated samples was measured by monitoring fluid volume changes of the inner cell.

4) The strength of MH sand increased with MH saturation due to particle bonding.

5) The peak strength increased with increasing effective confining pressure indicating that there is a frictional contribution to the strength increase rather than purely being due to cementation.

6) The maximum deviator stress difference occurred in the vicinity of 1%~2% axial strain regardless of effective confining pressure. Thereafter the strength decreased linearly with increasing axial strain.

7) The strength of MH sand increased with increasing back pressure and decreasing temperature.

8) Dissociation by heating caused large axial strains for samples with an initial shear stress and total collapse for samples consolidated in the metastable zone.

9) In the case of dissociation by depressurisation, axial strains were generated by increasing effective stress until a stable equilibrium was reached. However repressurisation led to the collapse of Case 3 in the metastable zone.

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REFERENCES

Fig. 20. Stress strain behavior during MH dissociation using de-pressurisation and water pressure


