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The Influence of Vertical Cylindrical Tetrahydrofuran Hydrate Veins on Fine-Grained Soil Behaviour

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9 ABSTRACT

10 Over the last 10 years, marine gas hydrate drilling expeditions utilizing advances in pressure coring
11 techniques and imaging have routinely encountered gas hydrates residing in fine-grained sediments. The
12 hydrate typically occurs as fracture-filling, near vertical, veins that displace the sediment, potentially
13 leading to increased sediment strength that may prevent normal consolidation of the sediment thus leading
14 to underconsolidation. Destabilization of this hydrate, through climate change or human activity on the
15 seafloor, may cause dramatic loss of strength of the sediment and pose a significant geohazard. To assess
16 the impact of hydrate veins on sediment behaviour, this paper reports on a series of consolidated (CU) and
17 unconsolidated undrained (UU) triaxial tests carried out on fine-grained soil specimens hosting
18 simplified, vertical, cylindrical THF hydrate veins of varying diameters. The results show that increasing
19 hydrate vein diameter significantly increases strength and stiffness, including the development of post
20 peak strain softening. The mode of failure of the hydrate veins influenced the specimen strength, but did
21 not affect the specimen stiffness. Hydrate dissolution during CU tests prevented quantitative comparison
22 with UU tests. However, CU test results on the soil specimens containing the largest hydrate vein suggest
23 that increasing lateral confining stresses increase the sediment strength.

24

25 **Key Words:** undrained shear strength, soil stiffness, THF hydrate, hydrate veins, triaxial testing.

1. INTRODUCTION

Methane gas hydrates are naturally occurring, ice-like compounds which exist under low temperature and high pressure conditions and are readily found within sediments along marine continental margins and onshore beneath permafrost (Kvenvolden and Lorenson 2001). Gas hydrate within these sediments sequester significant volumes of methane gas and therefore have attracted global interest due to their potential as an unconventional energy resource (Boswell and Collett 2011), their potential role in climate change (Kvenvolden 1988; Dickens et al. 1997; Ruppel 2011) and potential impact as a geotechnical hazard (Maslin et al. 1998; Rothwell et al. 1998; Rutqvist and Moridis 2009; Grozic 2010; Kwon et al. 2010). The presence of stiff, ice-like gas hydrate within the sediment matrix can increase the strength and stiffness of the sediment, which can be lost if the environmental conditions are no longer conducive to hydrate stability, leading to hydrate dissociation. In addition, hydrate dissociation involves the release of free gas into the pore space which can reduce the effective stress on the sediment (Nixon and Grozic 2007), thus exacerbating any reduction in strength; this may be more acute in fine-grained sediments due to their reduced ability to dissipate excess pore fluid (Kayen and Lee 1991). Processes that will bring about hydrate dissociation such as future gas hydrate production or ongoing climate change may pose a significant risk to submarine slope stability.

Assessing the potential impact of hydrate dissociation on slope instability requires a detailed understanding of the geomechanical properties of hydrate-bearing sediments. Historically, studies on the geomechanical properties of natural hydrate-bearing sediments have been hindered by the degradation of in situ properties due to hydrate dissociation during sample recovery from the seafloor. Significant advances in pressure coring, storage and transfer techniques (Pettigrew et al. 1992; Schultheiss et al. 2006) alongside the development of integrated analysis tools (Schultheiss et al. 2008; Yun et al. 2006; Priest et al. 2015; Santamarina et al. 2012) have enabled more accurate assessments of hydrate saturations (Dickens et al. 2000), hydrate morphology (Holland et al. 2008) and the physical properties of natural hydrate-bearing sediments (Yun et al. 2010; Priest et al. 2015; Santamarina et al. 2015; Yoneda et al. 2015). However, sampling disturbance associated with recovering hydrate-bearing sediments (Dai and

Santamarina 2014) and the limited number of samples available for testing gives rise to a paucity of reliable and representative test results, thus preventing a detailed understanding of the in-situ geomechanical properties of natural hydrate bearing sediments.

To overcome these constraints, experiments involving the formation and testing of laboratory synthesized gas hydrate-bearing specimens have been undertaken. Results have confirmed an increase in sediment stiffness and strength relative to gas hydrate-free sediments (Masui et al. 2005; Priest et al. 2005, 2009; Ghiassian and Grozic 2013; Clayton et al. 2011; Miyazaki et al. 2011; Hyodo et al. 2013) with the magnitude of increase dependent on degree of gas hydrate saturation, grain mineralogy, applied stresses and formation method adopted (Waite et al. 2009). The tests highlighted above were conducted on coarse-grained specimens, because the low solubility of methane gas coupled with the low permeability of fine-grained sediment introduce significant challenges in the controlled formation of gas hydrate within these soils. To form hydrate-bearing fine-grained soil specimens, analogues of gas hydrate such as tetrahydrofuran (THF) hydrate have been used (Yun et al. 2007; Lee et al. 2010), and within these specimens the presence of hydrate was observed to increase the strength and stiffness of the sediment.

In the aforementioned experiments, THF hydrate was likely formed homogeneously disseminated within the pore space of the soil, similar to that observed in nature for coarse-grained sediments (Dallimore et al. 1999; Winters et al. 2011). However, within fine-grained sediments, gas hydrate has been observed exhibiting grain-displacing, fracture-filling, complex subvertical vein structures (Rees et al. 2011). Tests on sediment samples recovered from this location showed that the hydrate veins have significantly higher strength than the sediment matrix in which they formed (Yun et al. 2010) and the undrained shear strength of the sediment matrix was significantly lower than what could be expected at the burial depths at which the samples were recovered (Winters 2011; Priest et al. 2014). This suggests that the hydrate veins may prevent normal consolidation of fine-grained sediment leading to high water contents (Collett et al. 2008), high initial void ratios and high sediment compressibility (Lee et al. 2013). Given that a significant portion of gas hydrates are hosted within fine-grained sediments (Klauda and Sandler 2005), they may pose the greatest risk as a geohazard if dissociation occurs.

To gain a better understanding of the geomechanical impact of heterogeneous, grain-displacing hydrate that is observed within natural fine-grained sediment, a series of triaxial compression tests were conducted on hydrate-bearing soil specimens. This paper discusses the procedures adopted in forming simplified vertical, cylindrical THF hydrate veins of varying sizes within fine-grained soil specimens and the results of undrained shear tests carried out on these specimens to investigate their geomechanical behaviour.

2. FORMATION OF VERTICAL CYLINDRICAL TETRAHYDROFURAN HYDRATE VEINS IN FINE-GRAINED SOIL

Natural gas hydrate veins exhibit complex morphology within fine-grained sediment making them difficult to replicate and subsequently analyze due to potential non-symmetric stress-strain behaviour. Therefore, a simplified formation procedure was developed to form vertical cylinders of THF hydrate in a void created within the centre of pre-consolidated clayey silt specimens. The veins were aligned with the principal stress direction to approximate natural near-vertical structures. Cylindrical veins were formed due to the difficulty of creating thin, planar veins typically observed in nature, while also creating a specimen that would respond axisymmetrically ($\epsilon_2 = \epsilon_3 = \epsilon_r$) to horizontal confining stress (σ_3) applied in a triaxial test. Hydrate veins in natural cores have been observed to range from sub-millimetre size to several millimetres in aperture, with average hydrate saturations of 20% to 30% (Rees et al. 2011). Therefore, four hydrate vein sizes were selected to investigate their influence on soil behaviour: 6.35 mm (0.25"), 12.7 mm (0.50"), 19.05 mm (0.75") and 25.4 mm (1") diameter, corresponding to hydrate vein saturations ranging from 2 to 26%.

2.1 Synthetic Hydrate

Tetrahydrofuran (THF) was used as a hydrate former for this study due to its practical advantages over methane gas: THF is fully miscible in water, has similar mechanical properties to methane hydrate (Lee et al. 2007), and when mixed with water at an appropriate molar ratio allows for rapid, homogeneous

hydrate formation at atmospheric pressure and temperatures below $\sim 4^{\circ}\text{C}$. The molar ratio at which THF and water form only hydrate is 1:17 respectively, however THF has a significantly higher vapour pressure than water that can lead to the vaporisation of THF during the formation process and result in incomplete hydrate formation (Zeng et al. 2006). Through extensive preliminary tests, the creation of a rapid and repeatable THF hydrate formation procedure was achieved by mixing THF and pre-cooled water ($\sim 2^{\circ}\text{C}$) at a molar ratio of 1:15 and then cooling the well-stirred mixture to -20°C .

2.2 Soil Specimen Preparation

The soil used in this study was prepared at a mixture by weight of 35% kaolin and 65% silica silt to resemble the grain-size distribution of formerly hydrate-bearing fine-grained sediment (Figure 1). The dry soil was mixed with distilled, de-aired water to form a slurry with an initial water content of 55%, and then preconsolidated in a specially constructed consolidation cell which applied a unidirectional vertical consolidation stress of 100 kPa. Specimens were taken by subsampling the preconsolidated soil using a 70 mm internal diameter cylindrical sampling tube and were then placed on a dummy pedestal and enclosed in a rubber membrane secured by a split mold. Cylindrical holes were drilled vertically through the centre of the specimen to correspond to a chosen hydrate vein diameter and the soil was subsequently cooled to 2°C to allow for rapid hydrate formation. Material properties of the prepared soil are highlighted in Table 1 along with those for natural fine-grained sediment associated with hydrate deposits.

2.3 Hydrate Vein Formation Procedure

Two vein formation procedures were adopted due to constraints associated with forming the THF veins. The 6.35 mm diameter vein was formed by placing 1:15 THF-water liquid mixture within the vein void, covering the specimen and cooling to -20°C . A small soil plug was tamped into the bottom of the vein void to prevent leakage of the THF-water mixture during hydrate formation. The formation of the hydrate

vein occurred within 30 minutes, with minimal freezing of the host soil. For larger diameter veins, this in situ formation method resulted in partial freezing of the soil's porewater, leading to ice lensing that may result in thaw-consolidation effects within the soil (Nixon and Morgenstern 1973).

To overcome the soil freezing issues, hydrate cylinders were formed independently and subsequently transferred into the vein void. This was done by forming cylindrical aluminium foil molds of the required vein diameter and height, which were then filled with 1:15 THF-water mixture, covered, and placed in a freezer to initiate hydrate vein formation. Once formed, hydrate cylinders were quickly unwrapped and carefully inserted into the vein void. THF hydrate formed using this method was observed to contain subhorizontal, planar macroscopic structural features along which THF hydrate dissociation appeared to initiate, as shown in Figure 2. Due to the high slenderness ratio of the 6.35 mm diameter vein, this procedure led to the fracturing of the small vein, and so could not be adopted.

Once the vein was either formed in situ or inserted into the soil, a second soil plug was tamped on top of the vein to seal it within the specimen. The specimen was then stored between 0 and 2°C prior to geomechanical testing.

3. GEOMECHANICAL TESTIGN OF HYDRATE-BEARING SOIL SPECIMENS

3.1 Test Apparatus and Specimen Mounting

A specially adapted double walled, computer-controlled triaxial system was used for testing of the hydrate-bearing specimens. The apparatus featured a load frame with an external load cell for measuring the vertical load applied by a convex loading piston. Axial strain was measured by a linear variable displacement transducer on the load ram. Cell and back pressure were applied using computer servo-controlled hydraulic pumps. An electronic pore pressure transducer in the base plate was used to measure pore pressure. Refrigerated circulator systems were used to maintain the temperature of the cell fluid and the apparatus base plate at approximately 2°C; within the THF hydrate stability field. An insulation jacket

was installed around the cell to maintain the temperature with a thermocouple installed in the cell to monitor the temperature of the cell fluid surrounding the soil specimen.

Prior to specimen mounting, the drainage lines, base pedestal and top cap were saturated with de-aired water in accordance with ASTM Standard D4767. The prepared specimen was removed from the refrigerator, saturated filter papers were placed on its top and bottom, and radial drains were applied around specimens to aid reconsolidation. The specimen was mounted, and the triaxial cell was assembled quickly and filled with cooled cell fluid such that no hydrate dissociation occurred during this process.

3.2 Triaxial Test Procedures

The testing program consisted of consolidated undrained (CU) and unconsolidated undrained (UU) triaxial compression tests on the specimens. For the CU tests, the specimens were first consolidated at a cell pressure of 500 kPa and back pressure of 400 kPa to an effective isotropic confining stress of 100 kPa. The consolidation stage was terminated when 95% of the excess pore pressure was dissipated, as per ASTM Standard D4767. The drainage lines were then closed, and the specimen was sheared at a strain rate of 0.05%/min. For the UU tests a cell pressure of 200 kPa was applied to the specimens prior to shearing at an axial strain range of 0.3%/min, as suggested by ASTM Standard D2850. Specimens were sheared until either an observed peak in axial load was observed or 15% axial strain was reached. For CU tests axial load, axial displacement and pore pressure were recorded throughout the shearing stage, while for UU tests axial load and displacement were recorded. After shearing, each specimen was cut open to expose and photograph the hydrate vein. The hydrate vein was then removed and its weight, and the moisture content of the specimen were determined.

4. RESULTS AND DISCUSSION

In this section, the results obtained from CU and UU tests conducted on fine-grained specimens with cylindrical hydrate veins are presented and discussed before comparing changes in strength and stiffness as a function of hydrate saturation (also defined in terms of an area ratio) for the two different tests.

4.1 CU Tests

The behaviour observed during CU compression tests on soil specimens containing THF hydrate veins of different diameters are shown in Figures 3a-c and summarized in Table 2. Figure 3a highlights the change in deviatoric stress ($q = \sigma_1 - \sigma_3$) versus axial strain (ϵ_a) showing that the inclusion of the 25.4 mm diameter vein gives rise to a large increase in q , reaching a peak value of ~ 660 kPa at an ϵ_a of $\sim 4.5\%$ (~ 4.6 times the peak deviatoric stress measured for the specimen without hydrate) before significant post-peak strain softening indicating brittle failure. In contrast, the 19.05 mm diameter vein leads to a 1.8 times increase in deviatoric stress with no appreciable strain softening, while the 6.35 mm and 12.7 mm diameter vein-bearing specimens show a slight reduction in peak deviatoric stress compared to the specimen with no hydrate.

Figure 3b presents the change in pore pressure coefficient ($A = \Delta u/q$, where Δu is the change in pore pressure) versus ϵ_a showing a significant reduction in the post-peak value of A for the 19.05 mm and 25.4 mm vein when compared to the non-hydrate-bearing specimen. This coupled with the lower A value at failure would indicate the axial stress is predominantly carried by the hydrate vein. A slightly greater A -value response is seen in the 6.35 mm and 12.7 mm veins when compared to the non-hydrate-bearing specimen. In considering Figure 3c, which highlights the stress paths followed by the specimens in q - p' stress space ($p' = \sigma'_1 + \sigma'_3$), it can be seen that the rapid reduction in A corresponds to the soil being able to withstand stresses that exceed its critical state due to the support of the hydrate vein (for the 19.05 mm and 25.4 mm vein). For the specimens with 0, 6.35 mm and 12.7 mm diameter veins, the increase in pore pressure relative to the increase in q leads to specimen failure as it reaches the critical state line.

Figure 4 shows a number of the tested specimens cut open after shearing, highlighting the significant loss of hydrate within the vein void. Measurement of the hydrate veins indicated that 79%, 76% and 78% of the THF hydrate vein remained by weight for the 12.7 mm, 19.05 mm and 25.4 mm vein respectively, while the 6.35 mm vein had disappeared. This is likely due to the time-dependent

dissolution of THF hydrate, driven by the concentration gradient between the THF and free water in the pore space. In addition to the loss of hydrate, contrasting hydrate failure modes could be observed. The 25.4 mm diameter vein appears to have fractured subhorizontally, subsequently leading to specimen rotation about the fracture, while the 19.05 mm diameter vein appears to have fractured diagonally leading to shear plane development through the rupture and surrounding soil. This may explain the large difference in peak deviatoric stress and post-peak behaviour between these two specimens, even though the relative loss of hydrate was comparable. The 12.7 mm vein was significantly fragmented and the 6.35 mm vein had disappeared, thus neither provided structural support and indeed may have weakened the specimen due to the creation of a fluid filled void within the specimen. To account for the change in vein geometry due to the hydrate dissolution seen in the CU test specimens, the final vein volume (shown in Table 2) was calculated by assuming that the vein remained cylindrical during dissolution, and its average diameter was determined by measuring the vein at three locations after shear.

4.2 UU Tests

To reduce THF hydrate vein dissolution, the testing time was minimized by carrying out unconsolidated undrained (UU) triaxial compression tests, which reduced the time the THF hydrate vein was in contact with the specimen porewater from around 30 hours (CU testing time) to approximately 30 minutes. Post-shear analysis of the hydrate vein suggested that no appreciable dissolution of the hydrate had occurred, preserving the overall dimensions of the veins (Figure 5).

A summary of the results from the UU compression tests are shown in Table 3, with Figure 6 showing q vs. ϵ_a results. Generally, an increase in hydrate vein diameter is seen to give rise to an increase in peak deviatoric stress, an increase in post-peak strain softening along with an increase in the initial gradient (stiffness) of the q vs. ϵ_a curves. The mode of failure of the hydrate veins (shown in Figure 5) has a significant influence on the peak deviatoric stress, with results for the 25.4 mm specimens showing that subhorizontal fractures lead to higher deviatoric stresses compared to diagonal fractures. This is similar to

the CU test results, and is likely due to the geometry of the vein fractures and the post-rupture behaviour of the specimen. Diagonal fractures allow for shear plane development through this zone of weakness allowing vein segments to translate past one another, while subhorizontal fractures lead to specimen rotation around the point of weakness with increasing axial strain.

4.3 Quantifying the Hydrate Veins

The impact of hydrate on soil behaviour is typically considered in terms of hydrate saturation (S_h) defined as the ratio between the volume of hydrate within the voids (V_h) and the volume of voids within the soil (V_v) shown in Eq. 1.

$$(1) \quad S_h = \frac{V_h}{V_v} \times 100\%$$

In these tests, where the hydrate is contained entirely in the vein and not disseminated within the pore space, the total volume of voids space will include the volume of voids within the soil ($V_{v(\text{Soil})}$) and the volume of the hydrate vein (V_h) such that

$$(2) \quad S_h = \frac{V_h}{V_h + V_{v(\text{Soil})}} \times 100\%$$

Areal relationships have been employed previously in defining the contribution of competent cylindrical bodies to a soil's behaviour, for example stone columns (Barksdale and Bachus 1983; Priebe 1995). Therefore, given the hydrate veins and specimens have constant diameter and height, and the specimens void ratio is known, hydrate saturation can be more simply defined by an area ratio (A_r) that relates the cross-sectional area of the vein (A_{vein}) to the specimen area (A_{Specimen}):

$$(3) \quad A_r = \frac{A_{\text{vein}}}{A_{\text{specimen}}}$$

This value ranges from 0 in hydrate-free sediment to 1 if the specimen is entirely composed of hydrate. The hydrate saturation and the area ratio can be related through:

$$(4) \quad A_r = \frac{n}{\frac{100\%}{S_h} - (1-n)}$$

where n is porosity. This relationship is approximate since it assumes that the soil and hydrate height are equal, when the small plugs of soil placed at the bottom and top of the vein reduced its height relative to the specimen height by $\sim 5\%$.

4.4 Undrained shear strength

The undrained shear strength (C_u) of a specimen is typically defined as half of the deviatoric stress at failure, $C_u = 0.5q_f$, where failure is determined by the maximum deviatoric stress recorded during a test. A number of relationships have been developed that relate C_u to effective vertical preconsolidation stress (σ'_v) for normally consolidated clays, such as that given in Equation 5 (Skempton 1957) in terms of plasticity index (PI), and Equation 6 (Mesri 1989):

$$(5) \quad \frac{C_u}{\sigma'_v} = 0.11 + 0.0037(PI)$$

$$(6) \quad \frac{C_u}{\sigma'_v} = 0.22$$

For the UU tests, where the fine-grained specimens were preconsolidated to $\sigma'_v = 100$ kPa, the values predicted by Eq. 5 and 6 ($C_u = 17$ kPa and 22 kPa respectively) are close to that measured for the non-hydrate bearing specimen ($C_u = 18.5$ kPa).

Generally, the presence of hydrate veins is seen to give rise to a linear increase in C_u for increasing vein diameter when plotted versus area ratio, although the 6.35 mm vein can be seen to have little effect on C_u . C_u derived from the UU compression tests are plotted versus A_r in Figure 7. Applying a linear line of best fit to the specimens containing 12.7 mm, 19.05 mm and 25.4 mm veins that were fractured horizontally, with the intercept set to pass through the origin, gives a slope of 1350 kPa. Extrapolation of this line to $A_r = 1$ (hydrate vein diameter = specimen diameter) would suggest a deviatoric stress at failure (q_h) of 2700 kPa for a THF vein, which closely matches the average shear stress (2680 kPa) obtained from the shearing of standalone THF hydrate veins (Wu 2016). Using the extrapolated shear stress at failure from this testing ($q_h = 2700$ kPa), a 6.35 mm vein would reach failure under a deviatoric load of $Q_f = 0.09$ kN, which is lower than the load at failure for the non-hydrate bearing sediment (0.14 kN). This

suggests that the hydrate vein increases the specimen strength only once the vein strength exceeds the strength of the soil ($C_{u(\text{Soil})}$). Therefore, the threshold when the undrained shear strength of a hydrate-bearing soil transitions from a soil-dominated behaviour to a hydrate vein-dominated behaviour ($A_{r(\text{thresh})}$) defined as the intercept of the two straight-line relationships, given C_u of 18.5 kPa for the soil, would occur when $A_r \sim 0.014$ as shown in Figure 7. This relationship can be generalised as:

$$(7) \quad \text{If } A_r \leq \frac{C_{u(\text{soil})}}{0.5q_h} \quad , \quad C_u(A_r) = C_{u(\text{soil})}$$

$$(8) \quad \text{If } A_r > \frac{C_{u(\text{soil})}}{0.5q_h} \quad , \quad C_u(A_r) = 0.5q_h A_r$$

Equations 7-8 can be rewritten in terms of S_h using Eq. 4:

$$(9) \quad \text{If } \frac{S_h}{100\%} \leq \frac{1}{n \frac{0.5q_h}{C_{u(\text{Soil})}} + (1-n)} \quad , \quad C_u = C_{u(\text{Soil})}$$

$$(10) \quad \text{If } \frac{S_h}{100\%} > \frac{1}{n \frac{0.5q_h}{C_{u(\text{Soil})}} + (1-n)} \quad , \quad C_u = \frac{0.5q_h n}{\frac{100\%}{S_h} - (1-n)}$$

The relationship suggested may be a simplification of the material behaviour due to the small dataset, as the transition between the two different behaviours may not be so abrupt. Differences in behaviour between specimens hosting the 6.35 mm vein and 12.7 mm vein may also be due to the difference in hydrate vein formation method, leading to a difference in material characteristics. In addition, the observed behaviour may be related to the strength of the soil matrix surrounding the hydrate vein (the lateral resistance provided by the soil to the hydrate vein). Figure 8 compares the deviatoric stress versus axial strain measured in both CU and UU tests on soil specimens containing 25.4 mm diameter hydrate veins that ruptured subhorizontally. It can be seen that the peak deviatoric stress obtained in the CU test was significantly higher than that from UU tests. All soil specimens were preconsolidated one-dimensionally under an effective vertical stress of 100 kPa producing a lateral stress around 38 kPa as predicted from K_0 tests carried out on the soil material (Smith 2016). However, in the case of the CU tests, the soil is further isotropically consolidated under a radial stress of 100 kPa, which would be transmitted directly to the hydrate vein and appears to increase the load the hydrate vein can carry before

failure occurs. Therefore, Eq. 7-10 may apply only to hydrate-bearing soils under low lateral effective confining stress.

4.5 Stiffness

Soil stiffness is typically determined by considering the gradient of the line joining the origin on the stress-strain curve to the point corresponding to half of the peak deviatoric stress (E_{50}), which helps overcome some of the issues associated with seating/bedding errors. Due to the non-linearity of the stress-strain response of soils, stiffness usually reduces with increasing strain. However, it can be seen in Figure 6, that some specimens exhibited low stiffness values initially before increasing (e.g. at ~2% axial strain for the 12.7 mm vein-bearing specimen), which may be related to the soil plugs placed at the bottom and top of the hydrate vein. Therefore, in our tests the undrained stiffness, E_u , of the specimens was determined either by considering the E_{50} value or the maximum gradient of the stress-strain curve before peak stress (e.g. the stiffness of the UU test on the 12.7 mm vein-bearing specimen was determined between 2.5% and 3.7% axial strain). Figure 9 shows the calculated values of E_u plotted against A_r for all UU tests and for the CU tests for specimens containing 0, 19.05 mm and 25.4 mm veins. In the case of the CU test data, the A_r values have been corrected to account for the dissolution of hydrate. In general, an increase in E_u is observed with increasing hydrate vein diameter for both sets of data. The vein failure mode was seen to have no effect on the undrained stiffness.

The stiffness determined in UU tests is not immediately increased by the presence of a small vein (6.35 mm vein), while at a certain threshold value it approximately follows a straight-line relationship in E_u vs. A_r space, as seen with the undrained shear strength. As with the undrained shear strength, a straight-line relationship with an intercept of zero implies that above the threshold value the hydrate vein stiffness dominates specimen behaviour. In a similar manner to that adopted for C_u , an A_r threshold value where stiffness becomes hydrate vein dependent can be predicted by extrapolating a linear line of best fit applied to the UU test data for specimens containing 12.7 mm, 19.05 mm and 25.4 mm veins (shown in Figure 9). Adopting this method gives an $A_r \sim 0.02$, slightly higher than that predicted for C_u . The

gradient of the line, which represents the stiffness of the THF hydrate vein, E_h , was found to be 185 MPa somewhat less than 251 MPa that was measured for standalone THF veins (Wu 2016). The lower stiffness coupled with the higher A_r suggests that the soil plugs, used to seal the vein in the specimen, may have a greater influence on initial stiffness than on strength. The relationships between E_u and A_r can be generalized by assuming that below the threshold area ratio, E_u is controlled by the soil stiffness ($E_{u(soil)}$) and above the threshold it is controlled by the stiffness of the hydrate vein (E_h) as

$$(11) \quad \text{If } A_r \leq \frac{E_{u(soil)}}{E_h} \quad , \quad E_u(A_r) = E_{u(soil)}$$

$$(12) \quad \text{If } A_r > \frac{E_{u(soil)}}{E_h} \quad , \quad E_u(A_r) = E_h A_r$$

As with C_u , Equations 11-12 can be rewritten in terms of S_h using Eq. 4:

$$(13) \quad \text{If } \frac{S_h}{100\%} \leq \frac{E_{u(soil)}}{E_h n + E_{u(soil)}(1-n)} \quad , \quad E_u = E_{u(soil)}$$

$$(14) \quad \text{If } \frac{S_h}{100\%} > \frac{E_{u(soil)}}{E_h n + E_{u(soil)}(1-n)} \quad , \quad E_u = \frac{E_h n}{\frac{100\%}{S_h} - (1-n)}$$

The relationship presented above does not consider the reduction in stiffness that may result from the soil plugs. In addition, it is applied only to UU test results, since undrained stiffness values derived from CU test results are higher for all specimens with and without veins. As highlighted previously, this is likely due to the isotropic consolidation to 100kPa in CU tests, increasing the lateral confining stress on the vein thereby increasing specimen stability during loading. However, the lack of CU test data on specimens with smaller veins and at different effective stresses precludes the development of a more comprehensive relationship. As previously mentioned, differences in behaviour between specimens hosting the 6.35 mm vein and 12.7 mm vein may be due to the difference in hydrate vein formation method, leading to a difference in material characteristics.

4.6 Potential impact on natural sediments

The results from this study show that increasing hydrate vein size leads to an increase in the strength and stiffness of the soil in which they are hosted. THF hydrate veins were created within specimens up to an

area ratio of 0.13 that equates to 26% hydrate vein saturation, which is representative of that observed in fine-grained fracture-hosted hydrate deposits (Rees et al. 2011).

Our tests were conducted on soil specimens that were subject to one-dimensional consolidation under a vertical effective stress of 100 kPa, which is representative of marine sediments at a burial depth of approximately 20 metres. Therefore, our results are applicable to near surface hydrate-bearing sediments. As the growth of hydrate veins within the sediment would lead to an increase in the sediment C_u and E_u , Equations 7-14 can be used to predict the change in strength of near-surface sediments as a function of area ratio/hydrate saturation. The incorporation of the developed relationships into geomechanical reservoir-scale numerical codes and slope stability programs would allow the impact of hydrate formation on the geomechanical response of these sediments to be assessed. Further work exploring the impact of effective stress on the strength of hydrate-bearing sediments, and the inclusion of more complex vein geometry, is required to understand the impact of hydrate veins on the behavior of more deeply buried, natural hydrate-bearing sediments.

5. CONCLUSIONS

A laboratory testing program was carried out to investigate the influence of hydrate veins on the behaviour of fine-grained sediments. Soil specimens composed of silt-sized silica (65% by weight) and kaolin clay (35%) with simplified cylindrical THF hydrate veins of different sizes located centrally within the specimens, were subjected to CU and UU triaxial compression tests. Four different hydrate vein diameters were considered (6.35 mm, 12.7 mm, 19.05 mm and 25.4 mm) representing hydrate saturations from 2% to 26%. The 6.35 mm hydrate vein was formed by placing THF/water mixture within a pre-drilled hole in the specimen and then forming the hydrate, while the larger veins were formed separately and subsequently inserted into pre-drilled holes.

Initially, CU compression tests were carried out on soil specimens with and without hydrate veins. Significant increases in stiffness and strength were observed for specimens with the large diameter veins (19.05 mm and 25.4 mm) compared to the non-hydrate bearing specimen. In contrast, the smaller

diameter veins (6.35 mm and 12.7 mm) led to a slight reduction in strength and stiffness. Inspection of the hydrate veins after shearing showed that a reduction in hydrate volume, possibly due to dissolution of the hydrate into the soil porewater, had occurred with 76% and 78% of the 19.05 mm and 25.4 mm THF hydrate veins remaining by weight respectively. The 12.7 mm vein had completely fragmented with 79% remaining by weight, while the 6.35 mm vein had disappeared.

To reduce the potential for hydrate dissolution, UU tests were carried out, which reduced the time the hydrate vein was in contact with the porewater of the soil from 30 hours to 30 mins. The results confirmed that hydrate vein diameter increases soil specimen strength and stiffness, and led to simple relationships describing changes in C_u and E_u with hydrate vein saturation/area ratio for soil specimens that were formed by one-dimensional consolidation under a vertical effective stress of 100 kPa. The mode of failure of the vein was seen to influence the peak stress at failure and the post-rupture behaviour of the specimen due to the geometry of the vein fractures, with subhorizontal fracturing leading to the largest increase in strength. In contrast, the stiffness of the soil specimen was unaffected by the mode of failure of the vein.

Although the dissolution of hydrate veins in CU tests prevented direct comparison with the UU tests, the results suggest that increasing lateral confining stress also increases the specimen strength and stiffness. Further CU testing at higher effective confining stresses, where THF hydrate dissolution is prevented is required to determine the exact relation between effective confining stress and specimen strength/stiffness.

Knowledge gaps still exist related to the effect of gas hydrate veins on fine-grained soil behaviour, making it important to carry out further investigations into the geomechanical behaviour of this potential energy source, climate change driver and marine geohazard.

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Figure Captions:

Figure 1: Grain size distribution curve of the prepared fine-grained soil compared to formerly gas-hydrate-bearing soil recovered from the KG Basin (Clayton et al. 2008) and the Gulf of Mexico (Winters 2011)

Figure 2: Image of outlined a) THF hydrate vein and b) same vein during dissociation at room temperature ($\sim 20^{\circ}\text{C}$).

Figure 3: Results from CU tests on soil specimens with different vein diameters. Increasing vein diameter (for 19.05 mm and 25.4 mm veins) leads to (a) an increase in peak strength and stiffness, (b) a decrease in A_f and (c) the soil exceeding its critical state, along with a significant influence on post-peak behaviour. The 6.35 mm and 12.7 mm veins appear to have minimal impact on observed behaviour. The failure modes for each hydrate vein are indicated.

Figure 4: Photographs of soil specimens cut open after CU testing highlighting different failure modes of THF hydrate veins, with (a) the 12.7 mm diameter vein fragmented, (b) the 19.05 mm diameter vein ruptured diagonally, and (c) the 25.4 mm diameter vein ruptured subhorizontally.

Figure 5: Photographs of specimens cut open after UU testing with hydrate veins outlined of (a) 6.35 mm, (b) 12.7 mm, (c) 19.05 mm and (d) 25.4 mm diameter ruptured subhorizontally, and (e) the 25.4 mm diameter vein ruptured diagonally.

Figure 6: Stress-strain response from UU tests on hydrate vein bearing specimens showing an increase in strength and stiffness with increasing hydrate vein diameter. The failure modes for each hydrate vein are indicated.

Figure 7: Changes in undrained shear strength with hydrate vein area ratio. Empirical lines of best-fit showing two distinct strength behaviours, namely soil controlled (flat slope) and hydrate vein controlled

(positive slope) strength behaviour. Extrapolation of the two lines (dashed line) highlights a transition threshold for vein diameter between the two strength behaviours.

Figure 8: Differences in peak strength obtained from CU and UU tests on specimens with 25.4 mm diameter veins highlighting the impact of vein failure mode.

Figure 9: Changes in undrained stiffness from UU (diamond) and CU (square) tests with hydrate vein area ratio. Empirical lines of best-fit applied to the UU tests results suggest two distinct stiffness behaviours, namely soil controlled (flat slope) and hydrate vein controlled (positive slope) stiffness behaviour. Extrapolation of the two lines (dashed line) suggests a transition threshold (circle) for vein diameter between the two stiffness behaviours.

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Table 1: Soil characteristics of different natural hydrate-bearing sediment and laboratory mixed soil

Characteristics (%)	Experimental Soil	Krishna-Godavari Basin ¹	Ulleung Basin ²	Gulf of Mexico ¹
Average Sand	1.4	5	0	1
Average Silt	63.6	55	80	22
Average Clay	35	40	20	77
Liquid limit range	34	70-98	12-129	N/A
Plastic limit range	18	33-49	17-88	N/A

¹Winters (2011)²Lee et al. (2011)

Table 2: Experimental Results from Consolidated Undrained (CU) Compression Tests

Initial Hydrate Vein Diameter (mm/in)	Final Area Ratio after Dissolution, A_r	Final Hydrate Vein Saturation after Dissolution, S_h (%)	Void Ratio of Soil, e_{soil}	Axial Strain at Failure, ϵ_{af} (%)	Peak Deviatoric Stress, q_f (kPa)	Pore Pressure Parameter at failure, (A_f)	Undrained Stiffness, E_{50u} or E_{secu} (MPa)	Vein Failure Mode
0/0	0	0	0.54	12.0	136	0.34	6.2	N/A
6.35/0.25	0	0	0.53	12.0	109	0.57	9.6	Dissolution
12.7/0.50	0.027	7.7	0.52	8.9	134	0.33	7.0	Fragmented
19.05/0.75	0.060	15.1	0.57	6.3	245	0.17	15.4	Diagonal Rupture with Shear Band
25.4/1	0.105	23.8	0.60	4.5	609	0.08	24.6	Subhorizontal Rupture

Table 3: Experimental Results from Unconsolidated Undrained (UU) Compression Tests

Hydrate Vein Diameter (mm/in)	Area Ratio, A_r	Hydrate Vein Saturation, S_h (%)	Void Ratio of Soil, e_{soil}	Axial Strain at Failure, ϵ_{af} (%)	Peak Deviatoric Stress, q_f (kPa)	Undrained Shear Strength, C_u (kPa)	Undrained Stiffness, E_{50u} or E_{secu} (MPa)	Vein Failure Mode
0/0	0	0	0.67	12.0	37	18.5	3.6	N/A
6.35/0.25	0.008	2.0	0.70	14.2	33	16.5	3.2	Fragmented
12.7/0.50	0.033	7.8	0.68	4.6	105	52.5	3.9	Subhorizontal Rupture
19.05/0.75	0.074	15.4	0.74	2.3	183	91.5	11.2	Subhorizontal Rupture
25.4/1	0.132	26.0	0.74	1.5	360	180	25.1	Subhorizontal Rupture
25.4/1	0.132	26.0	0.74	1.2	235	116.5	24.9	Diagonal Rupture with Shear Band