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## COMPARISON OF METHANE GAS HYDRATE FORMATION ON PHYSICAL PROPERTIES OF FINE- AND COARSE-GRAINED SEDIMENTS

William J. Winters<sup>1</sup>\*, William F. Waite<sup>1</sup>, Ingo A. Pecher<sup>2</sup>, David H. Mason<sup>1</sup>, Deborah R. Hutchinson<sup>1</sup>, Lauren Y. Gilbert<sup>1</sup>, Richard A. Birchwood<sup>3</sup>, Sheila Noeth<sup>4</sup>, and Emrys Jones<sup>5</sup>

<sup>1</sup>U.S. Geological Survey, 384 Woods Hole Road, Woods Hole, MA 02543
 \*Corresponding author: Tel: 508-457-2358; bwinters@usgs.gov

 <sup>2</sup>Institute of Geological & Nuclear Science, 69 Gracefield Road, Lower Hutt, New Zealand
 <sup>3</sup>Schlumberger, Av. Rio Caura, Torre Humboldt, Piso 8, Prados del Este, Caracas, Venezuela
 <sup>4</sup>Schlumberger, 1325 South Dairy Ashford Road, Houston, TX 77077
 <sup>5</sup>ChevronTexaco, 2811 Hayes Road, Houston, TX 77082-6696

Field, laboratory, and theoretical analyses of methane hydrate are in general agreement that the presence of methane hydrate in natural sediments changes the physical properties of the sediment. It is much less clear how and why these changes occur. Among the discrepancies and controversies that exist are if and how cementation occurs, what role grain size plays, temporal relations of formation, whether water saturation is significant, and whether it matters if hydrate forms from a dissolved phase (as inferred in the natural environment) or from a free-gas phase (as conducted in most laboratory experiments). To answer some of those questions, the USGS has embarked on a research program to integrate field, laboratory, and modeling results by measuring physical properties of sediments containing natural- and laboratory-formed gas hydrate.

Results from field experiments show that the properties of host material influence the type and quantity of hydrates formed. In the Mackenzie Delta, NWT Canada (Mallik 2L-38 well), hydrate occurs in coarser-grained units. However, in the Gulf of Mexico, where near-surface sediments are generally fine grained, hydrates recovered in giant piston cores did not appear to be lithologically controlled. Acoustic measurements performed on Mallik sediment in the field and the laboratory are more consistent with natural hydrate occurrence filling pores, rather than by grain cementation.

In order to replicate Mallik and Gulf of Mexico field observations, controlled laboratory experiments using the Gas Hydrate And Sediment Test Laboratory Instrument (GHASTLI) have been performed on two different grain sizes (coarse grained sieved Ottawa sand and fine grained crushed quartzite clayey silt) using initially fully or partially water saturated samples. Prior to gas hydrate formation, confining stress is approximately 12 MPa and consolidation stress ( $\sigma'_c$ ) typically is 0.25 MPa for sand or as much as 5 MPa for silt. To determine the effect of consolidation, sand will be tested at higher  $\sigma'_c$  and silt at lower  $\sigma'_c$ . Hydrate is formed when methane gas is slowly pushed into a 70-mm diameter by 140-mm long cylindrical specimen and subsequently the temperature is lowered to about 6 degrees C. Preliminary analysis indicates that clayey silt has a mean grain size of 4.5±1.5  $\mu$ m and a specific surface area of 1.8±0.1 m²/g.

Sieved Ottawa sand has a nearly complete grain distribution between 250 and 500 µm, classifying it as medium-sized sand. We are in the process of determining the specific surface area of sieved Ottawa sand, but theoretically it could be as low as 0.004 m<sup>2</sup>/g.

Although the precise control of test conditions using GHASTLI provides a means for studying fundamental gas hydrate behavior during formation, engineering loading, and dissociation, laboratory results may not fully agree with field observations. For example, direct observations and modeling indicate that laboratory-formed methane gas hydrate cements Ottawa sand, whereas natural hydrate may not. This is important since cementation has a profound effect on acoustic velocity and other sediment behavior, including shear strength.

Some of the discrepancies between field and laboratory results can be explained by different formation mechanisms in situ and the lab. In the field, hydrate is typically, although not exclusively, produced from gas dissolved in the aqueous phase, whereas in the lab hydrate is formed from bubble-phase methane. Cementation in the lab may be caused by hydrate forming at locations coated with water molecules, that is, on grain surfaces and at grain contacts.

We have also found that grain size may greatly influence results. P-wave velocity (Vp) increases abruptly in clayey silt during hydrate formation in test GH090 and reaches a maximum of 1.97 km/s (Fig. 1). This is much lower than Vp obtained in GH085 conducted with Ottawa sand where Vp>3.0 km/s. The samples contained comparable amounts of gas hydrate (pore saturation ≥21% and 19%, respectively). Although this is not totally unexpected since finer-grained sediment typically has lower acoustic velocities than coarser sediment, this poses a challenge for interpreting field seismic data to estimate the volume of gas hydrate in areas prior to drilling. We are constructing models to determine if gas hydrate induced cementation occurs in silt. For test GH090 we also noticed an abrupt increase and decrease in signal amplitude during initial gas hydrate formation followed by a fairly steady increase (Fig. 2). The reason for the spike in Pwave amplitude at initial hydrate formation is unclear at this time, but it may be related to coupling between the test specimen and end caps containing the acoustic transducers. Of more interest is the similar increase in signal strength (amplitude) versus time for fine-grained (GH090) and coarse-grained sediment (GH083) (Fig. 2). Because signal strength is related to the amount of gas hydrate formed and corresponding reduction in pore-space gas, eventually the amplitude in GH083 (maximum 70% pore saturation by hydrate) surpasses that of GH090.

Field experiments rarely measure changes in pore pressure, yet these measurements are routine in the GHASTLI lab. Because pore pressures greatly influence local effective stresses, knowledge of their magnitude is crucial in predicting the potential for slope instability and other geohazards. We have documented a number of interesting pore-pressure-related phenomena during gas hydrate formation, including the partitioning of different pore pressure zones within Ottawa sand and the episodic flow of methane to regions of low pore pressure resulting in effective stress and P-wave velocity changes.

We have also observed abrupt pore pressure changes during shear in silt containing gas hydrate (Fig. 3). Although shear stress increases significantly, pore pressure (measured at the base of the specimen) does not increase until about 3% strain is reached. This can partly be explained by a reduction in permeability caused by gas hydrate formation. The shearing process opens up fluid pathways that allow communication with the external pressure transducer. Pore pressure increases and then instantly decreases at about 5% strain (Fig. 3C) and then continues to climb from a lower base-line value. This markedly affects the stress path (Fig. 3A), however, the abrupt decrease in pore pressure does not affect the shear stress versus strain behavior at that point (Fig. 3B) because the measured pressure decrease is localized.

The triaxial strength test (Fig. 3) shows that even though pore pressure varies considerably, sometimes over small strain increments, the pressure change is always positive (relative to the start of shear) for this fine-grained sample. This results from contractive behavior and is different from the dilatant behavior noticed during shear on sandy sediment (Fig. 4). Contractive behavior induces positive pore pressure response (reduces effective stress) and reduces shear strength, whereas dilatant behavior reduces pore pressure (increases effective stress) and increases strength. Exploratory drilling or other mechanisms that open pathways between pressured zones separated by gas hydrate could potentially cause marked changes in local pore pressure, even without gas hydrate dissociation. Such effects could be similar to those produced by shallow water flows.

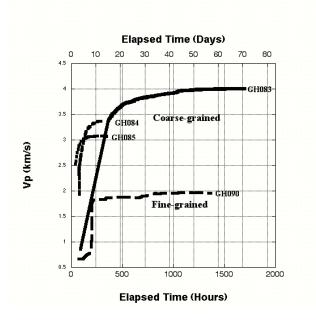


Figure 1 - P-wave velocity versus elapsed time for a clayey silt (GH090) and sieved Ottawa sand (GH083, 84, 85) samples tested using GHASTLI. Measured temperature increase indicates that gas hydrate began forming in GH090 at approximately 198.6 hours, after being in the stability field for 9 hours.

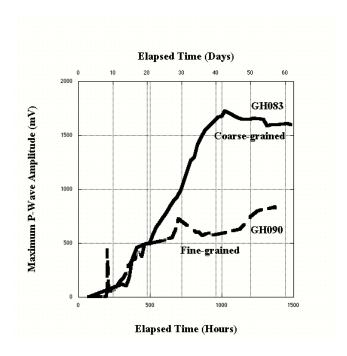


Figure 2 - Maximum P-wave amplitudes versus elapsed time for clayey silt (GH090) and sieved Ottawa sand (GH083) samples. Note the abrupt spike in amplitude for GH090 at initial gas hydrate formation (approximately 198.6 hours) followed by a steady increase for the rest of the test.

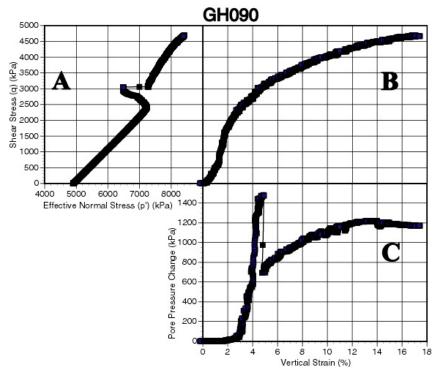


Figure 3 - Shear strength results for a clayey silt sample containing methane gas hydrate tested using GHASTLI. Individual plots are: (A) shear stress (q) versus effective normal stress (p') on a plane inclined at 45 degrees from the horizontal; (B) shear stress versus strain, and (C) change in

pore pressure versus strain. The abrupt offsets in Fig. 3A and Fig. 3C are caused by a rapid decrease in measured pore pressure at the base of the test specimen. Pore pressure increased a smaller amount at the top of the specimen and did not exhibit a similar, abrupt decrease.

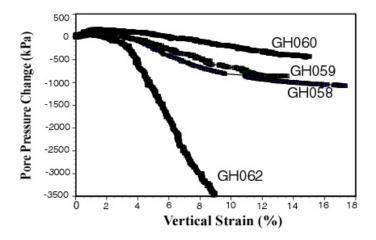


Figure 4 - Pore pressure change versus strain for natural sandy sediment from the Mallik 2L-38 well drilled in the Mackenzie Delta, NWT, Canada. Tests GH058, GH059, and GH060 did not contain gas hydrate at the time of shear, whereas GH062 did contain natural gas hydrate during the shear phase. All samples exhibit dilatant behavior as illustrated by the generation of negative (related to the beginning of shear) pore pressure. Contrast this behavior with the positive pore pressure response in Fig. 3C.