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To cite this article: Dimitris Marinakis and Nikos Varotsis 2019 *IOP Conf. Ser.: Earth Environ. Sci.* **221** 012134

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Experimental Study of the Gas Hydrates Dissociation Effect on the Properties of the Host Marine Sediment

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Abstract. Gas hydrates consist of small molecules trapped in crystal lattices of water molecules in a form resembling to common ice. The hydrate forming components are abundant in subsea sediments at conditions inside the hydrate stability zone. The vast majority of natural gas hydrates is expected to be hosted at the subsea sedimentary geo-environment. Despite the fact that such hydrates are not exploitable under the current technology, they can pose an environmental risk in case of uncontrolled dissociation. In this work, the effect of gas hydrates dissociation on the mechanical properties of a muddy clayish sediment retrieved from the “Anaximander” subsea mud volcano area and on its ability to conduct flow, was studied. A marine sediment was reconstituted in vitro with in-situ retrieved mud, water and hydrocarbon gas mixtures of compositions similar to the ones measured just above the seabed and was brought at conditions favouring the formation of hydrates. Geomechanical tests were conducted on the sediment before and during the dissociation of the guest hydrates, by isobaric heating and by isothermal depressurization. Permeability was found to increase only moderately during hydrate dissociation, possibly due to the high effective stress. The consolidation of the formation was increased up to a maximum just before the hydrates start to decompose and then collapsed when the free gas was produced. The bulk sediment volume was reduced by around 15% when the liberated gas was released from the formation in sudden “bursts” which occurred sporadically. The experimental results presented in this study seem to support the possibility of subsea landslides and the hydrate gun hypothesis for such areas as a result of hydrate dissociation.

1. Introduction

Most light hydrocarbons from methane up to butane are suitable for forming hydrates in the presence of water under favourable pressure and temperature conditions. Hydrate formation can occur both at low temperatures and pressures, as well as at higher temperatures - well above 20°C - under elevated pressures such as the ones prevailing at the subsea environment. Various research groups estimate that the quantities of natural gas trapped in the form of hydrates far exceed the conventional reserves of oil and natural gas [1], [2].

Geological formations hosting hydrates are classified into four categories. Class 1 formation includes a hydrate-bearing layer with an underlying two-phase fluid zone containing mobile gas and liquid water. Class 2 deposits consist of a hydrate-bearing layer, overlaying a zone of mobile water and the Class 3 ones are composed of a hydrate-bearing layer with no underlying zone of mobile fluids. Finally, Class 4, pertains specifically to marine accumulations, and involves disperse, low- saturation hydrate deposits that lack confining geologic strata [3] and it is considered that it includes the vast majority of natural gas hydrates. Such geo-environment consists usually of clayish sediments that are formed from the erosion of the landscape terrain or from the precipitation of dissolved salts and the biodegradation of the



marine organic matter. Despite the fact that Class 4 hydrates are not exploitable under the current technology [4], they can pose an environmental risk in case of uncontrolled dissociation. Such dissociation can occur during an abnormal temperature rise, as a result of the global warming, or after the decrease of water pressure in case of geological movement of subsea formations towards the sea surface and the depletion of water from lakes and aquifers [1].

Class 4 hydrate formations, due to their occurrence and the amount of natural gas that they contain, can amplify and accelerate the effects of climate change [5]. Methane is a known greenhouse gas with an effect to global warming 20 times higher than the carbon dioxide. Furthermore, the dissociation of subsea gas hydrates contributes to the depletion of the overlaying seawater in dissolved oxygen, either through the exchange of components between the rising gas bubbles and the seawater or through the aerobic biodegradation of the dissolved methane by bacteria [6]. This oxygen depletion is a possible reason for the expansion of the subsea anoxic areas with a severe interference on the local marine biodiversity. Finally, gas hydrate dissociation can substantially deteriorate the mechanical properties (strength) of the host formation, leading to possible subsidence and subsea landslides with a snowball effect to further dissociation of GH formations [7].

The impact of hydrate dissociation on the properties of the host sediment is receiving a thorough study in natural as well as in artificially formed porous media. Hydrate dissociation and mechanical stability is also under investigation both experimentally, with tests on natural geological formations as well as theoretically. A detailed review of the recent hydrate research can be found in [8], [9].

In this communication, we have studied the effect of GH dissociation on the mechanical properties of a muddy sediment retrieved from the Anaximander MV area. The area is known to contain several active mud volcanoes that host gas hydrates in sediments very close to the seabed. The experimental results were compared with relevant studies in the area and seem to support a hydrate gun hypothesis for the specific area.

2. Description of the subsea area

Submarine mud volcanoes (MVs) are distinct geological structures in which warm fluid-rich sediments mobilized at sub-bottom depths ascend through a conduit and escape into the hydrosphere [10]. MV effluents typically originate from sediment depths of several km, and consist of fluidized mud, mud breccia and volatiles, in particular hydrocarbon gases. MVs are well known to exist in various deep-sea areas and they are considered as one of the major natural sources of methane emissions to the hydrosphere and potentially to the atmosphere. Provided that gas occurs in oversaturation within the sediments, it partitions into gas dissolved in pore water, free gas, and under appropriate conditions, into gas hydrate. Focused expulsion of mud, warm fluids and gas bubbles occurs from the MV surface in areas characterized by relatively warm near-surface sediments. With increasing distance from the MV center, the upward fluid flux diminishes and the sediment temperatures is being reduced. As a consequence, dissolved oxygen and SO_4^{2-} penetrate from the sea water deeper into the sediments, thus permitting the biodegradation of the hydrocarbons into CO_2 and water [11]. Consequently, the upper hydrate limit deepens with increasing distance from the feeder channel, while at the same time the thickness of host formation also increases with the distance from the central area of the conduit due to the lower prevailing temperatures [12].

The Anaximander sea-mountains are found in the south Mediterranean, east of Rhodes Island. Several scientific expeditions in the Anaximander area [13] revealed the existence of six active MVs. Gas hydrates were discovered at four of the area's MVs, namely the Amsterdam, Kazan, Kula and Thessaloniki ones (figure 1) [14]. The hydrates were found to be dispersed at burial depths between around 30 cm and 150 cm, where they formed aggregates with diameters ranging from 0.3 cm to 8 cm. The gas enclathrated in the hydrate is mainly of thermogenic origin, i.e. it consists of mixtures of methane with heavier hydrocarbons. Such type of hydrates are more stable at higher temperatures than the methane ones [15].

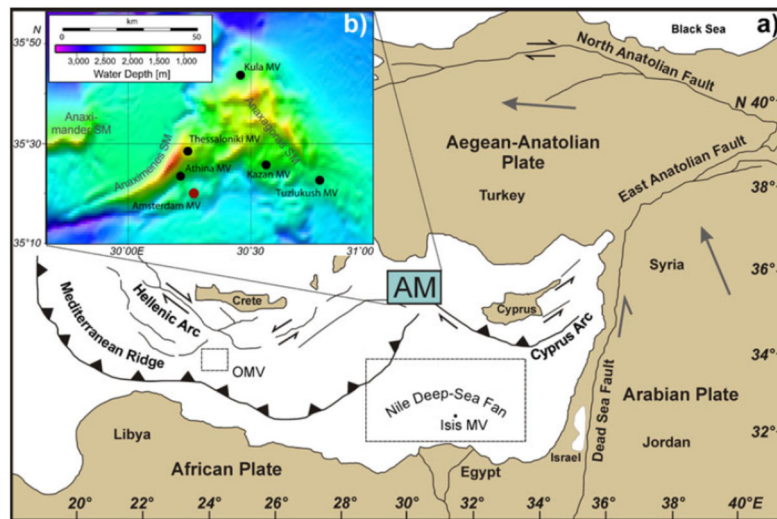


Figure 1. Location of the Anaximander area and Mud Volcanoes [15]

The most important MVs in the Anaximander area are the Amsterdam and the Thessaloniki ones. The Amsterdam MV is located in the southern part. It is the most important mud volcano in the area and possibly the largest in Eastern Mediterranean. It has been the most extensively studied MV because it is considered to host most of the gas hydrates in the area [16], while it exhibits strong similarities with respect to stromatography and mineralogy with the other ones. It appears as a flat-topped circular-shaped mound, extending over an area of about 6 km², at a water depth of 2025 m on its summit. Previous studies concluded that the formation of hydrates near the seabed can be explained either from gas dissolved in the water phase, or from the rapid cooling of hot brine and gas fluxes, which rise up through the sediment layers during geological events [17]. A detailed overview of the findings from the two most recent expeditions to the Amsterdam area can be found in [14], [15].

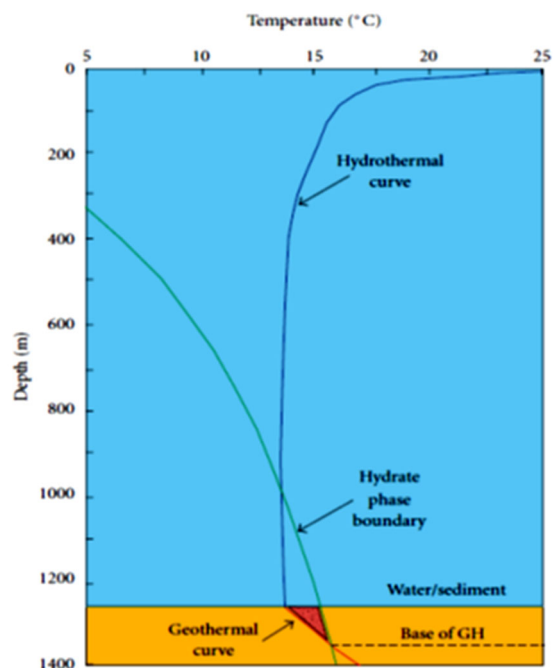


Figure 2. Hydrate stability limits at the Thessaloniki Mud Volcano [18]. The red triangle at the seafloor represents the temperature and depth limits where hydrates can be stable

Thessaloniki MV is located at water depths at which the hydrostatic pressure is marginally favorable for the stability of the hydrate deposits. It is the shallowest mud volcano in the area, lying at depths between 1315 and 1260 m, with an aerial extend of about 1.67 km² with three peaks on its summit. The sea bottom temperature (13.7°C) and the pressure (12.9 MPa) indicate that the GH present there are at the upper limit of their stability zone and are prone to dissociation at the slightest temperature increase or pressure decrease (Figure 2) [18]. In this case, an incremental change of the sea bottom temperature is enough to cause hydrate dissociation and the subsequent release of the hydrocarbon gas.

3. Materials and Methods

In order to study the behaviour of the marine host sediment containing gas hydrates, three types of tests were conducted: Isobaric heating at 20 MPa, isothermal depressurization at 12°C and isothermal compressions at a range of temperatures.

The GH host formation was reconstructed in vitro from a natural sediment sample, which was recovered by a gravity core during the ANAXIMANDER project's (EVK3-2001-000123) exploration in the area. Pretreatment of the sediment included removal of salts and mud conditioning. The sediment was flushed several times with de-ionized water, until its conductivity was reduced below 1 mS/cm. Then, the mud was dried to 80°C for 24 hours and subsequently it was softly grinded into coarse sized powder to reduce the size of the agglomerates. The mud was turned into a dry powder to ensure that the gas mixture will form hydrates of a uniform distribution. Atterberg's limits measurements were performed on the treated sediment. The liquid limit was found to be 40.3% (Cassagrande test) and the plasticity limit 15.6% (ASTM D 4318). These values indicate that the treated sediment can be classified as a lean clay.

The treated sediment was confined inside the test vessels at ambient temperature and subjected to vacuum for 1 hour. A gas mixture of molar composition 84% C1, 13% C2 and 3% C3 was then injected at 5 MPa in the pore space of the dry sediment. The gas composition was similar to the compositions of gas samples collected from near the sea bottom during the exploration cruises [15]. De-ionized water was added to increase the pore pressure up to 20 MPa. Subsequently, the temperature was reduced to 5°C, well below the hydrate formation limit, while additional de-ionized water was injected to maintain the pore pressure at 20 MPa. The system was left at constant conditions for 30 hours to reach gas hydrate equilibria. Afterwards, the temperature was set to 12°C at 20 MPa. Pore volume and gas hydrate saturation were calculated from the injected quantities of water and gas. Two experimental rigs were used in these studies: the core holder rig and the piston rig.

3.1. The core holder rig

It consists of a syringe pump, which is connected at the bottom part of two piston cells, containing gas and de-ionized water (figure 3). The pistons vessels are connected to a biaxial Hassler type core holder and provide the necessary quantities of gas and water to the host sediment. Six pressure ports along the core of the holder ensured uniform distribution of the gas and water introduced inside the host sediment during hydrate formation. In order to monitor the water permeability of the formation during the isothermal heating test, a backpressure valve (valve V2 in figure 3) was installed at the outlet of the core holder to maintain a differential fluid pressure of 2 MPa along the sediment core. The pump was also used to apply radial stress on the core holder sleeve. The volume changes on the core were indirectly measured through the pressure changes of the aforementioned chamber. More details on the setup can be found elsewhere [19].

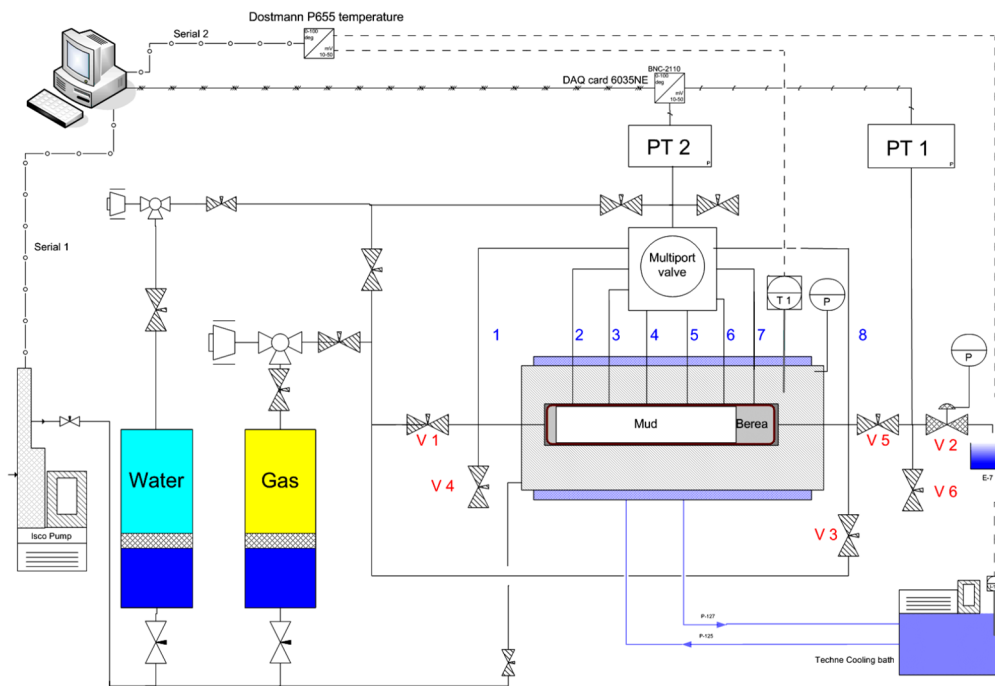


Figure 3. The core holder experimental setup

The sediment was confined in a thermally shrinking plastic tube, 1.5 in. in diameter and 5 in. long, and put inside the core holder. Two 0.5 in. long Berea sandstone taps were used at the ends of the plastic tube to confine the mud grains in place during depressurization and water/gas outflow. The tube was pre-perforated with six holes in order to achieve contact with the respective pressure taps along the core holder sleeve. The perforation holes were covered with filter paper to prevent mud grains from escaping. After the hydrate formation equilibration period, the sediment that it was at 20 MPa pore pressure, 12°C and 2 MPa radial confining stress, was left undisturbed for 3 days.

3.1.1. Isothermal depressurization. Valves V1 and V5 were closed and the core was subjected to decomposition of its hydrate content by reducing the pressure from its outlet at constant flow rate of 0.1 ml/min through valve V3 to the water piston cell, while the inlet was closed at the initial pressure. The flowrate and volume increase was controlled by the pump, while the pressures were monitored during decomposition. The pressure at the six ports along the core length was also measured regularly with the multiport valve. Gas samples were collected and analyzed with a Perkin Elmer gas-chromatograph model 8700.

A similar test was conducted for comparison, using a Berea Sandstone core inside the pores of which hydrates had also been formed and equilibrated with the same procedure as the one for the clayish sediment.

3.1.2. Isobaric Heating. Valves V1 and V5 were opened. The pressure at the inlet of the core was set to 20 MPa by injecting water from the water piston cell. At the outlet of the core, the backpressure valve V2 was set at 2 MPa lower than the pressure at the inlet. The aforementioned conditions remained constant for a period of 3 days, which was sufficient for the system to reach steady flow state and for the accurate measurement of the water flow rate through the host sediment. Subsequently, the core temperature was increased by 2°C and another 3-day long measurement cycle followed.

3.2. The piston rig

The experimental setup consists of a thermally controlled piston (figure 4). The sediment is confined at the ϵ compartment of the piston, while overburden compression pressure is applied through pressurization of the δ compartment. The fluids' pressure of the ϵ compartment corresponds to the pore pressure of the sediment and it was controlled through the auxiliary piston cylinder which contained de-ionized water. A cylindrical plug of Berea sandstone was placed at the top of compartment ϵ in contact with the sediment. A syringe pump was used to control the pressures of both the auxiliary cylinder and compartment δ .

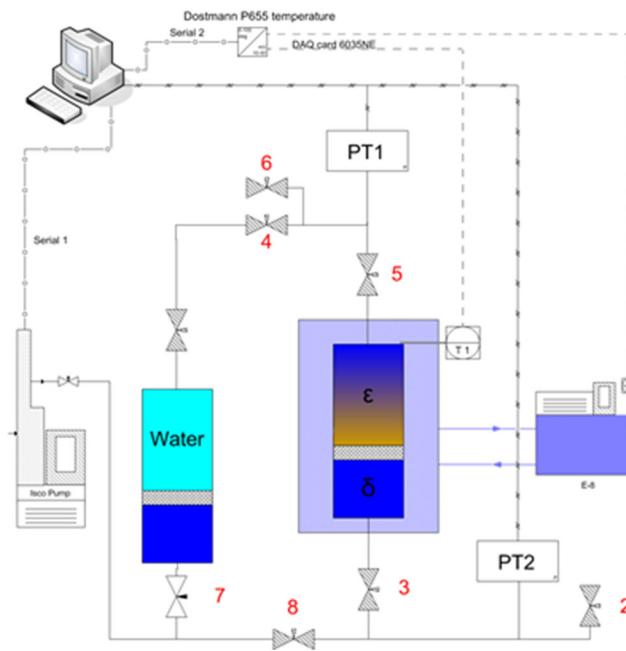


Figure 4. The piston experimental setup

3.2.1. Isobaric heating. The strain of the confined sediment was recorded over a range of compressive stresses, at constant temperature and pore pressure of 20 MPa. The measurements were repeated at temperatures between 12 and 26°C.

4. Results and discussions

4.1. Sediment depressurization at 12°C

Since the hydrates formed contain several gas components, gas was released in pressure steps at any given temperature. Sloan's CSMGem simulator was used to estimate the conditions at which the hydrates will decompose. The simulation for the gas mixture utilized revealed that at 12°C, hydrates would start to decompose at pressures below 7 MPa and the decomposition would be completed at pressures close to 3 MPa.

Figure 5 shows, with straight red lines, the experimentally measured production of the gas from the mud core (left graph) and from the Berea Sandstone (right graph). The production profile of the gas predicted by the CSMGem simulation is shown in the same graphs with dashed lines. Both the clayish sediment and the Berea core exhibited very poor fluid connectivity when the hosted hydrates were stable due to pore plugging. Even when the outlet pressure was reduced well below the incipient hydrate dissociation pressure of 7 MPa, the permeability remained very low, resulting in substantial pressure differentials across the core ends. The maximum pressure difference between inlet and outlet in the clayish sediment reached 7 MPa whereas in the Berea one it was even greater and close to 10 MPa.

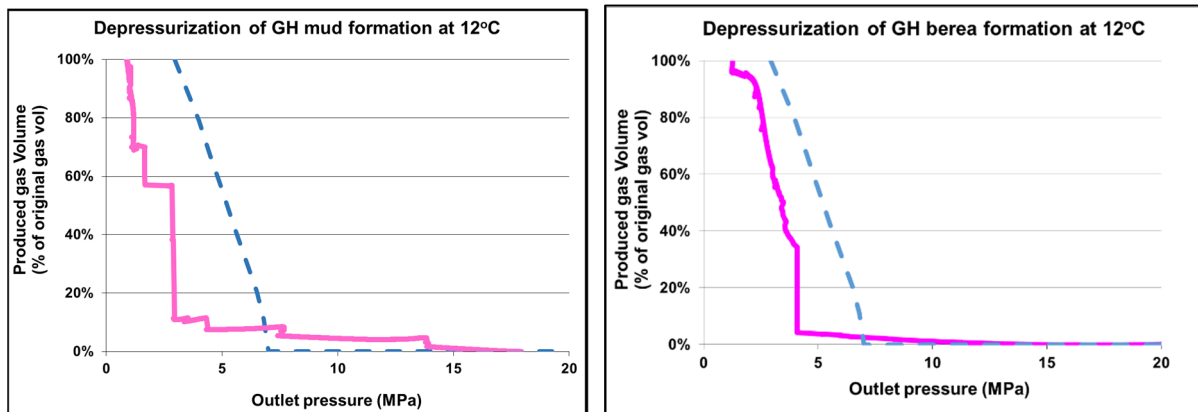


Figure 5. Production of free gas due to hydrate dissociation from the mud during depressurization (left hand side) and of the Berea Sandstone core (right hand side). The dashed line is the CSMGem simulation of hydrate dissociation and free gas production in bulk phase – no host formation.

As a result, the hydrates remained stable even at outlet pressures of less than 4 MPa, an effect which is already known as hydrate self-preservation. When pressure fell below 3 MPa in the mud sediment (4 MPa in Berea), gas hydrates were decomposed and free gas was produced at the outlet. Below that pressure, the fluid connectivity (permeability) was restored in the Berea core but not in the clayish sediment. Thus, the free gas production profile in Berea is smooth with pressure decline below 4 MPa, while in the clayish sediment it appears as step wise even at very low pressures, well below 1 MPa.

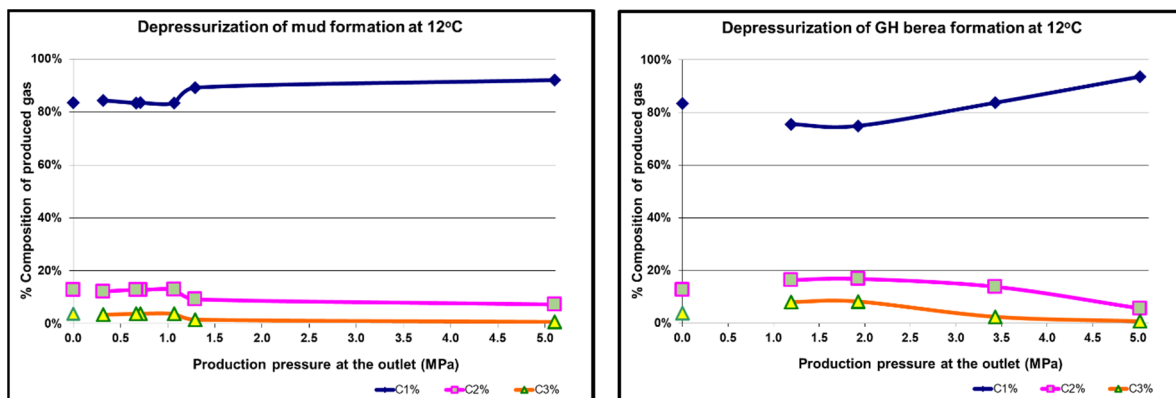


Figure 6. The compositions of the produced gas due to hydrate dissociation from the mud during depressurization (left hand side) and from the Berea Sandstone core (right hand side). At zero pressure is the feed gas composition.

In figure 6, the compositions of the gas produced from the gas hydrates in both sediments are graphically displayed. For comparison reasons, the initial feed gas composition is depicted on both graphs at 0 MPa. It is shown that the composition of the gas produced from hydrate crystals varies according to pressure. Methane tends to escape from the hydrate phase first (at relatively high pressure), while ethane and propane remain enclathrated even at pressures as low as 3 MPa. Low pore connectivity of the host core may change the production profile and the composition of the produced gas by retaining part of the initial high pore pressure and by gas back-mixing.

4.2. Core heating at 20 MPa

The sediment was confined in the core holder at 20 MPa pore pressure and at 2 MPa confining stress. The hydrate content was estimated to be close to 10% of the total sediment's volume.

The extent of hydrate dissociation as temperature increased in the clayish sediment was in agreement with the simulation runs in pure water (figure 7 left). It seems that the hydrate equilibria was rather not affected by the capillary effects of the smaller pores, so hydrates were probably located in the void space of the bigger pores, while the smaller pores remained filled with water. The very low recorded permeability values of $0.2 \mu\text{D}$ (10^{-19} m^2), which prevail due to the clayish nature of the unconsolidated grains (figure 8), do not permit the released gas to escape from the sediment and lead to a host sediment volume expansion by up to 5% (figure 7). Such formation volume expansions are significant considering the high effective stress, which is equivalent to stress values applied at formations more than a hundred meters below seafloor.

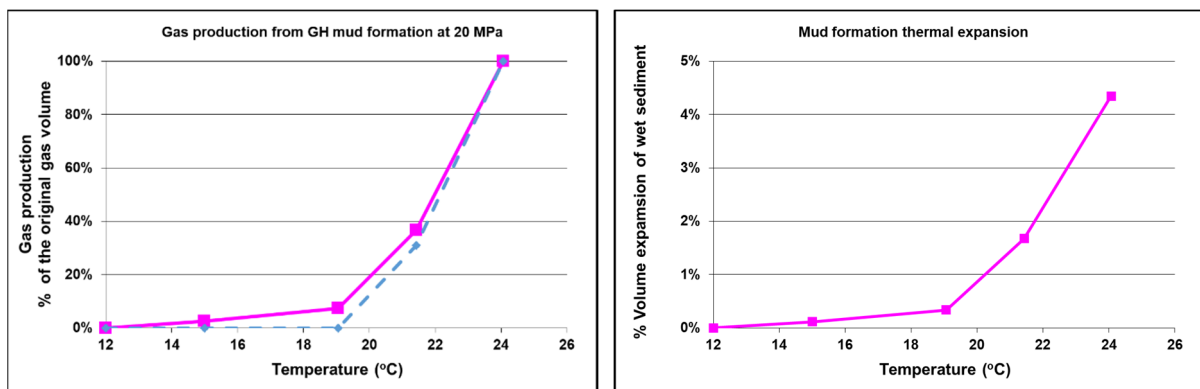


Figure 7. The production of free gas due to hydrate dissociation from the mud (left hand side) and the volume expansion of the mud (right hand side) during heating. The dashed line is the CSMGem simulation of hydrate dissociation and free gas production in the bulk phase – no host formation.

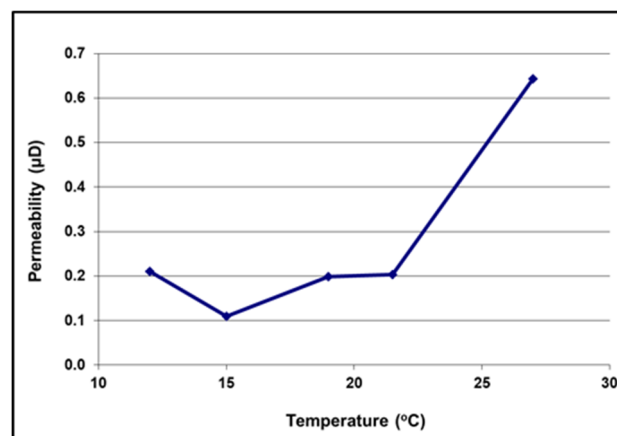


Figure 8. Mud core permeability change due to hydrate dissociation during heating [19]

The permeability of the sediment was also recorded over temperatures from 12°C, where all hydrates are stable, up to 27°C, where all the hydrates are dissociated. It was found that the formation of GH bears a rather moderate effect on the permeability of the sediment, even at high GH pore-saturation (figure 8). In literature, it is reported that hydrate dissociation in consolidated and loosely consolidated sediments results in the increase of permeability values by orders of magnitude, while in this test the increase was only threefold (figure 8). It is possible that in our case the high effective stress has reduced the effect of hydrate dissociation on the permeability values of the host sediment. This can also be explained by the selective distribution of the hydrates over the larger pores which are possibly not interconnected. In such pore configuration, permeability values are profoundly affected by the smaller

pores which interconnect the bigger pores and since the smaller pores are filled with water, their permeability is not affected by hydrate dissociation.

The results of this study indicate that in hydrate bearing clayish geological formations in the subsea environment, that lie a few tens of meters below the seabed, pore pressures can locally increase during hydrate dissociation due to heating and cause the swelling of the host formation.

4.3. Isobaric heating

The isothermal compression of the host sediment at 20 MPa pore pressure is depicted in figure 9. The strain of the confined sediment was recorded with the effective compressive stress, at constant temperature and pore pressure. The measurements were repeated at temperatures between 12 and 26°C. The host sediment had 29% pore volume with 33% saturation in gas hydrates.

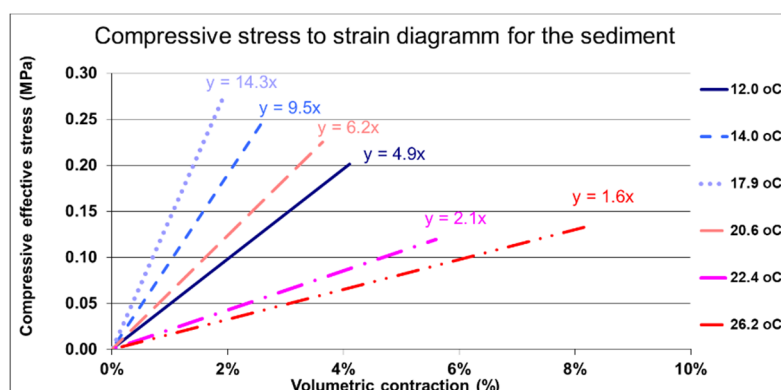


Figure 9. Host formation compressibility changes due to hydrate dissociation during heating

Gas hydrates were found to bear a profound effect on the compressive strength of the host sediment. A temperature increase above the GH stability limit (18°C), results in a sharp reduction of the sediment's stiffness, thus risking subsea landslides (figure 9).

5. Conclusions

The production profiles of the gas liberated from hydrate dissociation in the Berea and in the clayish sediment cores showed similarities. They both indicate that pressure should drop significantly –well below the incipient gas formation pressure before any gas is produced. For the permeability of both cores to be re-established, a significant percentage of the original hydrates in place needs to be dissociated. The main difference observed between the two sediments was that, below the pressure threshold of 4 MPa, the Berea core exhibits a smooth gas production profile, while the unconsolidated clayish sediment exhibits a stepwise pressure profile.

The formation of GH bears a rather moderate effect on the already very low permeability values of the sediment. This effect can be possibly appended to the high effective stresses applied which prevail in formations at few dozens of meters below the seabed.

The low permeability of the unconsolidated sediment results in the swelling of mud core during hydrate dissociation. As the gas from the dissociated hydrates cannot escape the core matrix, it increases the pore pressure.

Gas hydrates bear a profound effect on the compressive strength of the host sediment. A temperature increase above the GH stability limit, causes swelling of the host formation and sharp reduction in sediment's stiffness, thus risking a cause for subsea landslides.

Acknowledgment(s)

The authors would like to acknowledge the support from the European Community – Research Directorate, for financing the “Anaximander” project

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