



## Review article

## Thermal instability of gas hydrate bearing sediments: Some issues

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## ABSTRACT

The overall stability of marine strata holding gas hydrates is dependent on their shear strength characteristics. These characteristics, in turn, are dependent on thermal flux that is imposed for dissociation of the hydrates for the safe and efficient extraction of methane gas from the hydrate bearing sediments. Due to the imposition of thermal flux on these sediments, their fabric structure and pore space hydrate saturation changes, which impacts the overall stability of the sea bed. Estimating stability conditions in such a 'multiphase and dynamic system' necessitates collection of undisturbed samples without compromising their in-situ thermodynamic conditions. This is a daunting task given the huge cost of procuring samples and the challenge of maintaining an undisturbed sample with in-situ thermodynamic conditions till it is brought to the laboratory. Synthesizing hydrate bearing sample sediments in laboratory for conducting studies to identify heat migration mechanisms and thermal property measurements and linking them to the shear strength characteristics provides an affordable solution to this problem. With this in view, a critical review of the available literature, dealing with laboratory synthesis of hydrate bearing sediments, their thermal and strength characteristics, the coupled phenomenon of heat and fluid migration, and its impact on the overall stability of marine sediments, has been conducted and presented in this paper. This will facilitate understanding the factors governing and the mechanism of heat transfer in a multiphase system, the changes in the system brought about by the hydrate dissociation front, and the overall impact on the stability of seabed.

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## 1. Introduction

Gas hydrates are cages of water molecules in solidified form, holding gas molecules (methane in more than 95% cases) within the interstitial spaces of the crystal lattice, with no chemical bonding between the gas and water molecules (Kvenvolden, 1993).

Rich deposits of natural gas hydrates have been discovered in permafrost regions and within the continental margins the world over, which are estimated to hold more than half of the world's organic carbon (Kvenvolden and McMenamin, 1980; Grevemeyer et al., 2000; Zillmer et al., 2005; Hester et al., 2007; Cook et al., 2008; Jiang et al., 2008; Pecher et al., 2008; Long et al., 2009; Dewangan et al., 2010; Lee et al., 2013). As such, gas hydrates

may prove to be a lucrative alternative to meet the world's energy demands well into the foreseeable future. Studies conducted into the exploration, existence, and properties of natural gas hydrates show that they exist within the pores of marine sediments where the thermo-mechanical conditions (high pressure and low temperature) are conducive for their stable existence and adequate methane gas is available in solution to promote hydrate formation (Sloan and Koh, 2007; Gabitto and Tsouris, 2010). Gas hydrates, on being brought out from their stability zone, dissociate to release methane gas and water, and this property is targeted for exploitation (mostly by raising temperature) for the production of gas (Waite et al., 2009).

Currently, a significant research focus on gas hydrates is directed towards understanding the temperature-pressure regime (the combination of temperature and pressure conditions necessary for the stable existence of gas hydrates) of the hydrate bearing sediments, and its implications on their stability (Helgerud et al., 1999; Waite et al., 2004; Clayton et al., 2005; Lee et al., 2007; Jung and Santamarina, 2012; Jiang et al., 2014). The seabed stability is

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directly related to the strength and deformation characteristics of marine sediments, such as stiffness, shear strength parameters (cohesion and friction angle), and settlement parameters. The dissociation of hydrates, brought about by supplying heat energy, is an energy intensive process which in turn is dependent on the heat migration mechanisms, and thermal characteristics of the sediment matrix (Cortes et al., 2009; Cheng et al., 2011). However, the hydrate saturation of the pore space changes as the hydrate dissociation occurs in the soil mass and progresses through it (Xu and Ruppel, 1999; Jayasinghe and Grozic, 2013). This brings about changes in the arrangement of grains (or sediment fabric), the contact stresses between the grains, the pore space shape and dimensions, and the arrangement and contact of hydrate crystals with the sediment grains. Such a dynamic environment will have a major impact on heat migration, fluid migration, strength and stiffness characteristics, and settlement behaviour.

Proper understanding of heat migration and the changes brought about by dissociation of hydrates within the sediment matrix necessitates the collection of undisturbed samples from sub-sea strata – a task involving huge investments in time and money. To overcome this, researchers have resorted to laboratory synthesis of gas hydrates within various sediments. Various methods of laboratory synthesis of hydrates in sediments have been developed, each leading to a different growth pattern within the pore space of sediments. These different growth patterns have implications for the various characteristics of hydrate bearing sediments such as seismic wave velocities, thermal characteristics (such as conductivity, diffusivity, specific heat, and enthalpy of reaction etc.) (Handa, 1986; Waite et al., 2002, 2004, 2007; Huang and Fan, 2004, 2005; Kumar et al., 2004; Turner et al., 2005; Zhang et al., 2014), and strength and deformation characteristics (such as elastic modulus, cohesion, and friction angle) (Soga et al., 2006; Masui et al., 2008; Wu and Grozic, 2008; Grozic and Ghiassian, 2010; Zhang et al., 2012; Lu et al., 2013).

In light of the above discussion, this paper takes a critical look at the available literature dealing with the laboratory synthesis of hydrate bearing sediments, the thermal and strength characteristics determination carried out in laboratory on such sediments, the influence of the synthesis methods on the thermal and strength properties, the coupled phenomenon of heat and fluid migration in a dynamic and multiphase system, and the overall impact on the stability of sea bed. This will facilitate understanding the factors governing and the mechanism of heat transfer in a multiphase system, the changes in the system brought about by the hydrate dissociation front, and the overall impact on the stability of seabed, thus providing a better understanding of the issues and challenges facing the production of gas from hydrate bearing sediments.

## 2. Properties of gas hydrates

### 2.1. Composition of gas hydrates

Hydrates are a class of substances called clathrates (Kvenvolden, 1993). Clathrates are solids comprised of a crystal lattice of molecules of one kind (called host molecules) which entrap molecules of another kind (called guest molecules) within the intermolecular cavities of the lattice (See Fig. 1). The guest molecules provide structural support to the cavities of the crystal lattice, adding to its stability (Sloan and Koh, 2007). The bonding between host and guest molecules is principally physical attraction between adjacent molecules, and is different from chemical or hydrogen bonding. The term 'hydrate' indicates that the host are water molecules. The water in hydrates is in solidified form, and thus, the hydrates are somewhat similar in nature and physical properties, to ice (Waite et al., 2009).

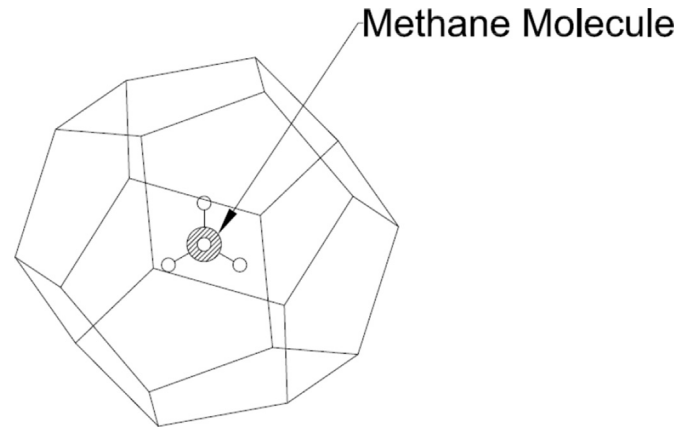


Fig. 1. Molecular structure of methane hydrate. Each node of a pentagonal face represents a water molecule.

The guest molecules are usually from a suite of gases such as halogens, noble gases, hydrogen sulphide ( $H_2S$ ), carbon dioxide ( $CO_2$ ), and low molecular weight hydrocarbons such as methane, ethane etc. (Kvenvolden, 1993). The type of guest molecules depends on the crystal structure of the lattice (i.e., the intermolecular space available). Most (more than 95%) naturally occurring gas hydrates are methane hydrates (i.e., the guest molecules are methane) (Kvenvolden, 1993; Waite et al., 2009). It is to be noted here that not all the intermolecular lattice spaces are occupied. Most naturally occurring hydrates have 70–90 % lattice spaces occupied with methane gas. The rest of the interstitial lattice spaces are usually blank. For the hydrates to have stable existence at the locations at which they are found (continental margins, permafrost offshore and onshore), at least 30–40% of the lattice spaces should have guest molecules (Kvenvolden, 1993; Waite et al., 2009).

One volume of natural gas hydrate usually accommodates about 150–180 volumes of gas, when brought to standard temperature and pressure (STP) (Kvenvolden, 1993). This has implications not only for the amount of natural gas that can be obtained from a hydrate deposit, but also for how this gas may be transported from the continental margins in deep sea. Typically, one volume of methane, in liquefied form, yields only 2 volumes if methane gas is at STP (Kvenvolden, 1993). Hydrates, thus, offer an economic way of transportation of the gas from the offshore production well to the nearest port/shore.

It should be noted here that naturally occurring gas hydrates are different from artificial gas hydrates that are formed in gas pipelines. Artificial gas hydrates are formed when there is presence of moisture, in a closed pipeline where a mixture of hydrocarbons flows under high pressure (Waite et al., 2002, 2009). These inevitably end up plugging the pipelines and wells used for conveying gases.

### 2.2. Existence of hydrates in nature

Gas hydrates have been found to exist at a depth of up to 700 m beneath the sea floor, where the thermo-mechanical (temperature and pressure) conditions are compatible for their stable existence (typically pressures exceeding 140 atm and temperature between 4 and 6 °C as in Krishna-Godavari Basin (K-G Basin), off the Eastern Coast of India, or pressures exceeding 200 atm and temperature between 10 and 17 °C as in Andaman Deep Sea, east of South Andaman Island, India) (Kumar et al., 2008; Ramana et al., 2008; Lee et al., 2011). The zone of sediments in which the gas hydrates are known to exist, is called 'hydrate stability zone' (HSZ) and the

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