# **OBSERVED GAS HYDRATE MORPHOLOGIES IN MARINE SEDIMENTS**

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#### ABSTRACT

Small-scale morphology of gas hydrate is important for understanding the formation of gas hydrate deposits, for estimating the concentrations of gas hydrate from geophysical data, and for predicting their response to climate change or commercial production. The recent use of borehole pressure coring tools has allowed marine gas-hydrate-bearing sediments to be recovered with centimeter to sub-millimeter gas hydrate structures preserved in their in situ condition. Once these sediment samples are recovered at in situ temperature and pressure, nondestructive analyses, including gamma density, P-wave velocity, and X-ray imaging, are used to examine the character of the gas hydrate relative to the structure of the surrounding sediment. Gas hydrate morphology from pressure core data is summarized from the recent national gas hydrate expeditions of India, China, and Korea, as well as from Ocean Drilling Program Leg 204, Integrated Ocean Drilling Program Expedition 311, and the Gulf of Mexico Chevron-Texaco Joint Industry Project. The most striking result is the variability of gas hydrate morphology in clay, ranging from complex vein structures to an invisible pore-filling matrix. Both of these morphologies have been observed in clay sediments at gas hydrate saturations equivalent to 30-40% of pore volume. A clear knowledge of detailed gas hydrate morphology will provide important data to help determine the mechanisms of gas hydrate deposit formation and also provide crucial data for modeling the kinetics of deposit dissociation, from both natural and artificial causes. The morphology also has large effects on sedimentary physical properties, from seismic velocities on a large scale to borehole electrical resistivities on a smaller scale, and gas hydrate morphology will therefore impact estimation of gas hydrate saturation from geophysical data. The detailed morphology of gas hydrate is an essential component for a full understanding of the past, present, and future of any gas hydrate environment.

Keywords: gas hydrate, pressure core, dissociation, formation, veins, disseminated, fractures, production

#### **INTRODUCTION**

Gas hydrate morphology describes the relationship between gas hydrate and the surrounding marine sediments. The morphology of gas hydrate determines the basic physical properties of the sediment-hydrate matrix. It provides clues to the formation of gas hydrate deposits, and the nature of the disruption that will occur on dissociation. Many remote techniques for gas hydrate detection and quantification are highly dependent on the hydrate morphology.

Little attention has been paid to hydrate morphology until now because previous methods

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of hydrate collection preserved only the grossest morphologies: lumps and nodules of hydrate. Models of gas hydrate formation & dissociation, as well as the models underlying remote gas hydrate quantification, generally assume that gas hydrate is distributed finely and uniformly within sediments. Recent advances in pressure coring and pressure core analysis have allowed collection of samples with intact gas hydrate-sediment morphologies, which show that gas hydrate often takes on complex forms which will require new approaches to both conceptualizing and modeling gas hydrate dynamics.

## **METHODS**

Morphology of gas hydrate was examined in HYACINTH pressure cores from recent seagoing expeditions. Visual and infrared observations from non-pressurized (conventional) cores were also used.

## Expeditions

Gas hydrate expeditions that have collected HYACINTH pressure cores include Ocean Drilling Program (ODP) Leg 204 to Hydrate Ridge, Oregon Margin [1]; the Chevron/DOE (US Department of Energy) Naturally-Occurring Hydrates JIP (Joint Industry Project), Gulf of Mexico (GOM-JIP; [2]); Integrated Ocean Drilling Program (IODP) Expedition 311, Cascadia Margin [3]; the Indian National Gas Hydrate Program Expedition 1 (NGHP1; [4]), Bay of Bengal; the Chinese Guangzhou Marine Geological Survey Expedition 1 (GMGS1; [5]), South China Sea; and the Korean Ulleung Basin Gas Hydrate Expedition 1, East Sea (UBGH1; [6]).

## **Pressure Core Analysis**

All pressure cores examined were collected with the HYACINTH pressure corers [7, 8]; and analyzed in the MSCL-P (Multi Sensor Core Logger-Pressure) which is now a component part of the integrated PCATS (Pressurized Core Analysis and Transfer System, [8]). The MSCL-P was used to collect routine nondestructive data on HYACINTH-compatible pressure cores, including gamma density, P-wave velocity, and X-ray images. Gamma density was measured using a 137-Cs source and NaI detector. Ultrasonic Pwave velocity (Vp) was measured using two 500 kHz acoustic transducers mounted inside the pressure chamber, perpendicular to the core and the gamma ray beam. X-ray images were obtained through an aluminum pressure chamber using a linear X-ray device consisting of a lead-shielded microfocal X-ray source and phosphor image

intensifier; the intrinsic spatial resolution of the images is approximately  $150 \ \mu m$ .

X-ray computed tomographic (CT) analysis was completed on selected cores from two of the most recent expeditions using equipment in local hospitals. Cores from NGHP1 were analyzed in Singapore on a Philips Mx8000 (slice resolution 0.7 mm), and cores from UBGH1 were analyzed in Daejeon, Korea, on a Siemens SOMATOM Sensation (slice resolution 0.6 mm).

Natural gas mass balance determined from the depressurization of pressure cores ([9-11], refined in [3, 4]) was used to estimate the saturation of gas hydrate, by quantifying the total amount of hydrate-forming gases present in the cores. Mass balance from pressure cores is the only way to quantify the total concentration of natural gas in a core, from which the gas hydrate saturation of a sediment can then be calculated assuming dissolved gas, free gas, and gas hydrate phases are in thermodynamic equilibrium. Gases released from pressure cores during depressurization experiments were collected and their composition measured. The total amount of gas contained in a pressure core was then compared to the pore volume of the core (measured in the MSCL-P from the X-ray images and gamma density profiles), and gases that were present at a level beyond saturation at in situ conditions were assumed to exist as a gas hydrate or free gas phase.

## **GAS HYDRATE MORPHOLOGIES**

Gas hydrate morphologies come in two basic types: pore-filling and grain-displacing. Porefilling morphologies of gas hydrate replace pore fluid between grains of sediment; this gas hydrate may or may not cement grains together. Graindisplacing gas hydrate does not occupy pore volume between grains and instead forces grains apart, forming veins, layers, and lenses of pure gas hydrate. Grain-displacing hydrate may cover a vast range of sizes, from thin veins of possibly only a few microns thick to nodules tens of centimeters or even meters in diameter.

"Grain-displacing" and "pore-filling" are not equivalent to the terms "massive" and "disseminated" which are often used to describe gas hydrate. These terms are used on core which has already undergone gas hydrate dissociation, where "massive" gas hydrate is still visible, and "disseminated" gas hydrate is invisible to the naked eye, and may have already completely dissociated. "Disseminated" gas hydrate is generally identified as cold regions (negative thermal anomalies) in a core using infrared thermal imaging [12]; once dissociated, thin veins and layers would be indistinguishable from porefilling hydrate and classed as "disseminated."

The details from the pressure core MSCL-P and Xray CT data were used to determine the geometry of gas hydrate in relation to the sediment structure. Grain-displacing gas hydrate structures, such as veins and lenses, are visible in density profiles and X-ray images as low-density structures and, if they are large enough or oriented properly, visible in the P-wave velocity data as high velocities. Porefilling gas hydrate is not likely to be evident in density and X-ray data, as gas hydrate has a similar density to pore fluid, but can produce anomalously high P-wave velocities if the sediment structure is made more rigid by gas hydrate cementation.

#### Grain-displacing marine gas hydrate

Gas hydrate has been observed in grain-displacing forms in all of these recent gas hydrate expeditions, with the exception of Expedition GMGS1 to the South China Sea [5]. Gas hydrate was found in structures ranging from thin, wispy veins (Figure 1) to large, photogenic multicentimeter chunks.

Four expeditions have drilled and cored locations that can be categorized as "focused, high-flux" sites (after [13]), which may have had active or relic seafloor venting: ODP Leg 204 [1], IODP Exp. 311 [3], NGHP1 [4], and UBGH1 [6]. While conventional cores from each of these locations recovered layers and chunks of "massive" hydrate, pressure cores from the two most recent of these expeditions (NGHP1 & UBGH1) showed that the habit of gas hydrate within these sediments was much more complex than had been previously imagined (Figures 2, 3). Though these cores were predominately silty clay, X-rays of cores show these veins cross-cutting sedimentary layers, sometimes in groups of subparallel veins.



Figure 1. X-ray image of thin, subvertical vein (light vertical stripe on left) in Core NGHP1-12A-9Y from the Krishna-Godavari Basin. This core, 57 mm in diameter, contained only 2% gas hydrate calculated as a percent of pore volume. Dense structures (e.g., sulfide) are dark; less dense structures (e.g., gas hydrate) are light.

Figure 2 (left). X-ray image of Core NGHP1-10B-8Y, collected in the Krishna-Godavari Basin [4]. Core is 57 mm in diameter and 90 cm long; core image is to scale. Dense structures (e.g., carbonate) are dark; less dense structures (e.g., gas hydrate) are light.



Figure 3. Cross-sections of X-ray CT data from Core NGHP1-10B-8Y (Figure 2), collected in the Krishna-Godavari Basin [4]. Core is 57 mm in diameter. Dense structures (e.g., carbonate) are dark; less dense structures (e.g., gas hydrate) are light.

While there were prodigious quantities of veins at these "focused, high-flux" sites, other locations that were closer to the "diffuse, low-flux" endmember [13] also showed thin veins of gas hydrate similar to that in Figure 1. These veins were primarily subvertical in nature and they occurred in clays.

#### Pore-filling marine gas hydrate

Most pore-filling marine gas hydrate has been observed in coarse-grained sedimentary layers in a fine-grained matrix. These silty or sandy layers are most often associate with the bases of mass flow deposits. Cores from IODP Exp. 311 [3], NGHP1 [4], and UBGH1 [6] all show pore-filling gas hydrate in coarse-grained layers. Most of these observations have been made on nonpressurized cores, and the layers have been identified by the use of infrared thermal imaging [12]. In these coarse-grained layers, the gas hydrate filling the pore spaces is often visible to the naked eye once the core is opened. Instances of such layers in pressure cores are rare but they do occur; one is shown in Figure 4.



Figure 4. X-ray image of gas-hydratebearing sand layer in Core NGHP1-15A-11Y from the Krishna-Godavari Basin [4]. Dense structures are dark (e.g., carbonate layers, shear pin in upper left corner); less dense structures are light (e.g., gas hydrate). Core is 57 mm in diameter. Arrow shows gas-hydrate-bearing sand; the pore-filling hydrate does not change the density appreciably but its presence was confirmed by porewater freshening analysis [4].

Pore-filling gas hydrate in fine-grained sediment (clay) has only been positively identified at three sites sampled during GMGS1 [5], situated within the same area. This gas hydrate deposit, in the South China Sea, contained 20-40% gas hydrate as a percent of pore volume, measured by methane mass balance analysis. Analysis of X-rays and gamma density logs showed no decrease in density relative to surrounding sediments; however, the acoustic (P-wave) velocities were over 2000 m/ sec, indicating possible cementation of clay particles.

There are many instances of cores with lower gas hydrate saturations where the gas hydrate morphology could not be constrained. Sometimes this was due to lack of data: non-pressure cores, early pressure cores with incomplete nondestructive data sets or no X-ray CT. However, low saturations of pore-filling gas hydrate that do not cement grains (and hence do not elevate P-wave velocity) are difficult to distinguish from the thinnest gas hydrate veins, which may be only a fraction of a millimeter thick. Full resistivity imaging of the core surface under pressure could provide a method of distinguishing these two morphologies.

#### IMPACT OF HYDRATE MORPHOLOGY

The abundance of gas hydrate veins and layers in marine sediments has major implications for the study of gas hydrate deposits. Qualitative and quantitative models for the formation, evolution, and dissociation of gas hydrate deposits, as well as quantitative models underlying remote geophysical measurement of gas hydrate, must explicitly state the geometry of the gas hydrate in relation to the sediment. Often the assumption is that gas hydrate is pore-filling, uniform, and isotropic. The many observations now made from pressure core analysis suggest that this is not the normal case, even at sites with relatively low overall gas hydrate saturations (< 5% by pore volume).

The largest impact of the discovery of common grain-displacing gas hydrate morphology is on the quantification of gas hydrate using remote geophysical techniques. Calculation of gas hydrate saturation from downhole electrical resistivity data currently relies on Archie's relation [14], which assumes an isotropic, porous medium where conductive (seawater) and resistive (gas hydrate) materials occupy the pore volume. For gas hydrate with pore-filling morphology, this assumption is valid. For gas hydrate with graindisplacing morphology, it is invalid, and Archie's relation, even if ground-truthed and calibrated, is unlikely to yield satisfactory results. It is certainly misleading to use gas hydrate saturations based on an Archie-type analysis when it is known that gas hydrates are predominately of the grain displacing vein type. Quantification of gas hydrate from seismic data has similar problems; many current questions surrounding the cementation or otherwise of sediment grains in pore-filling gas hydrate become irrelevant when veins and layers are the predominant morphologies in the sediment.

The predominance of subvertical vein/fracture morphologies has implications for the formation of gas hydrate in sediments. Rather than slow advection or slower diffusion, the veins are more likely evidence of hydrate deposit formation by pneumatic or hydraulic fracture of the clay sediment. Observations of the nature of multiple "sets" of very thin veins at different angles [4] give the distinct impression that the fracturing activity is episodic and may come in bursts at different times. Conversely, gas (and possibly also the fluid) released by hydrate dissociation could migrate through similar fractures out of the formation, creating a very different type of sediment disturbance and releasing much more gas directly to the seafloor than previously imagined. Examination of gas hydrate morphology will give us insight to some of the dynamics of gas hydrate deposits, extending our understanding both forwards and backwards in time.

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