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Research paper

# Comparison of methane mass balance and X-ray computed tomographic methods for calculation of gas hydrate content of pressure cores

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A B S T R A C T

Gas hydrate saturation was calculated for twelve pressure cores taken during National Gas Hydrate Program (NGHP) Expedition 01 in the Krishna–Godavari Basin, Bay of Bengal, at a location where thin gas hydrate veins were common. One of two methods were used to calculate gas hydrate saturation for each core: methane mass balance after depressurization and gas collection, considered the "gold standard" for measurement of gas hydrate saturation; or voxel intensity analysis (rather than binary segmentation) of X-ray computed tomographic (CT) reconstructions. Gas hydrate saturation in cores measured by methane mass balance was calculated to be 17.8%, 10.9%, 11.9%, 13.6%, 9.5%, 1.4%, and 0% by percent of core volume. Gas hydrate saturation in similar cores measured by intensity analysis of CT reconstructions was 17.2%, 9.1%, 6.7%, 7.8%, and 3.1% by percent of core volume. This quantitative CT intensity analysis contained systematic errors and therefore the calculated hydrate saturations are lower bounds. The systematic errors can be removed from the quantitative CT analysis by converting the CT intensities to real densities, though this was not possible for this study. All pressure core gas hydrate saturations were similar in magnitude to each other as well as to independent estimates of gas hydrate saturation from porewater freshening, and all variations in saturation could be explained by natural variation between samples. CT intensity (or preferably density) analysis of pressure cores showed promise for calculation of the saturation of vein hydrate in natural samples, allowing pressure cores to be used for further analyses under pressure after hydrate quantification. Theoretical examination of CT density analysis showed that this method would be unable to detect pore-filling gas hydrate; judicious examination of the results from CT density analysis versus other hydrate quantification methods on the same samples might allow quantification of pore-filling hydrate.

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## 1. Introduction

Quantification of natural gas hydrate in sedimentary formations has been attempted by a variety of techniques. Each measurement has its own spatial scale and pitfalls, and only one, mass balance from pressure cores, is considered accurate in all cases. Two remote surveying techniques, seismic (e.g., Westbrook et al., 2008) and electromagnetic (e.g., Weitemeyer et al., 2006), can detect the presence of gas hydrate in subsurface formations, and modeling of these data can provide generalized estimates of the saturation over large scales. Downhole electrical resistivity measurements can be used to estimate gas hydrate saturation due to the insulating properties of gas hydrate (Hyndman et al., 1999; Collett and Ladd, 2000; Lee and Collett, 2012). Such resistivity logging provides a profile of an entire borehole, but the calculations are highly dependent on the geometry of gas hydrate within the sediment (e.g., Cook et al., 2010). Continuous infrared imaging on individual non-pressure cores can detect the thermal anomalies caused by gas hydrate dissociation, though the correspondence between the magnitude of the anomalies and the quantity of gas hydrate often varies from core to core (Tréhu et al., 2004). Porewater salinity or chlorinity measurement to detect freshening from gas hydrate dissociation (Ussler and Paull, 2001; Malinverno et al., 2008) is a straightforward technique but has the disadvantages of being a

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point measurement and requiring a known “zero-hydrate” chlorinity at each point measured.

“Degassing” of hydrate-bearing pressure cores and measurement of natural gas quantity are used to calculate gas hydrate saturations (Dickens et al., 2000; Milkov et al., 2004; Heeschen et al., 2007). Pressure cores trap a volume of sediment along with any gas hydrate, dissolved gas, or free gas, enabling a mass-balance approach to be applied to this sealed system, and thermodynamic equilibrium is assumed in these calculations. The degassing of pressure cores is the only hydrate quantification method that can prove that gas hydrate does not exist in a sample of sediment (by showing that the porewater is undersaturated in natural gases), and is considered the “gold standard” for hydrate quantification, as it is used to ground-truth other hydrate quantification techniques (e.g., Riedel et al., 2010; Lee and Collett, 2009). One major drawback of the destructive depressurization of pressure cores is that the valuable pressure core is no longer available for other studies. If a nondestructive method of testing could be used for hydrate quantification in pressure cores, this would allow further analyses to take place on the core.

X-ray computed tomography (CT) is a good candidate for hydrate quantification via nondestructive testing. X-ray CT has been used in the field of geosciences to visualize internal structure without the need to serially section the objects (Orsi et al., 1992; Ketcham and Carlson, 2001; Cnudde and Boone, 2013). In X-ray CT, a single three-dimensional X-ray CT data set is generated from many standard two-dimensional X-ray radiographs, each of which capture an X-ray projection of the object: the X-ray attenuation of the object when irradiated at a particular angle. The X-ray attenuation of geological materials is related to the thickness of the object imaged and to the linear attenuation coefficient of the material, which is a function of the density and the elemental composition. The X-ray linear attenuation coefficients of marine sediment and gas hydrate are quite different, mainly due to the density contrast of the two materials, and this means that gas hydrate will be distinguishable from marine sediment in an X-ray CT reconstruction.

X-ray CT is already in use for hydrate quantification in experimental systems (e.g., Jin et al., 2004; Sato et al., 2005; Kneafsey et al., 2011; Hu et al., 2014). There has also been some experimentation with imaging natural samples using X-ray CT. With samples from the Mackenzie Delta in Canada, Uchida et al. (2000) examined the morphology of gas hydrate using X-ray CT, and Mikami et al. (2000) observed the samples by X-ray CT while gas hydrate dissociation was occurring. Matsumoto et al. (2000) used X-ray CT to image a sample of massive gas hydrate from Blake Ridge which had been stored under pressure, and X-ray CT was used by Abegg et al. (2007) to quantitatively analyze massive gas hydrate samples from Hydrate Ridge that had been stored in liquid nitrogen. There have also been high resolution studies of X-ray CT of gas hydrate on the pore scale using synchrotron radiation (Murshed et al., 2008; Klapp et al., 2012).

**Table 1**
Pressure cores referred to in this study.

| Core ID | Top of core (mbsf) | Length recovered (cm)[a] | Pressure at core depth (MPa) | Pressure recovered (MPa) Logged[b] | Pressure recovered (MPa) gage[c] | Method of hydrate quantification | Latitude | Longitude | Water depth (m) |
|---|---|---|---|---|---|---|---|---|---|
| NGHP-01-10B-08Y | 50.1 | 86 | 11 | 10 | 10 | X-ray CT | 15° 51.8571′ N | 81° 50.0789′ E | 1049.4 |
| NGHP-01-10B-15P | 98.2 | 100 | 11.5 | 2.3 | 1.7 | Mass balance | 15° 51.8571′ N | 81° 50.0789′ E | 1049.4 |
| NGHP-01-10B-18Y | 117.4 | 86 | 11.7 | 11.8 | 11.8 | Mass balance | 15° 51.8571′ N | 81° 50.0789′ E | 1049.4 |
| NGHP-01-10B-28P | 175.1 | 100 | 12.3 | 6 | 6 | Mass balance | 15° 51.8571′ N | 81° 50.0789′ E | 1049.4 |
| NGHP-01-10D-12E | 77.8 | 68 | 11.3 | 9.5 | 9.5 | Mass balance | 15° 51.8647′ N | 81° 50.0709′ E | 1050.4 |
| NGHP-01-10D-22E | 145.1 | 44 | 12 | –[d] | 11 | Mass balance | 15° 51.8647′ N | 81° 50.0709′ E | 1050.4 |
| NGHP-01-10D-25P | 164.4 | 100 | 12.2 | 9 | 8.7 | Mass balance | 15° 51.8647′ N | 81° 50.0709′ E | 1050.4 |
| NGHP-01-21A-02Y | 58 | 84 | 11.1 | 10.6 | 11 | X-ray CT | 15° 51.8531′ N | 81° 50.0827′ E | 1049.0 |
| NGHP-01-21A-03E | 59 | 108 | 11.1 | 10 | 10.5 | X-ray CT | 15° 51.8531′ N | 81° 50.0827′ E | 1049.0 |
| NGHP-01-21A-07E | 70 | 109 | 11.2 | 10 | 10.3 | Mass balance | 15° 51.8531′ N | 81° 50.0827′ E | 1049.0 |
| NGHP-01-21C-02E[e] | 56.5 | 110 | 11.1 | 10.4 | 10.5 | X-ray CT | 15° 51.8492′ N | 81° 50.0866′ E | 1049.0 |
| NGHP-01-21C-04E | 77 | 108 | 11.3 | 10 | 10.5 | X-ray CT | 15° 51.8492′ N | 81° 50.0866′ E | 1049.0 |

P = Pressure Core Sampler, Y = Fugro Pressure Corer, E = HYACE Rotary Corer.
[a] Length measured from X-ray and gamma density analysis, which may not match curated core length.
[b] Last pressure recorded before data logger disconnected from corer autoclave. Temperature 2–4 °C.
[c] Pressure recorded when autoclave pressure transducer connected to external gage. Pressure measured at 7 °C.
[d] Data logger did not record.
[e] Stuck in autoclave; no nondestructive test data.

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