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Understanding Methane Hydrate Behavior Using X-ray Computed Tomography

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Introduction

To understand hydrate behavior, either in a pure phase or in a porous medium, we measure its properties and responses to changes in conditions such as pressure, temperature, and chemistry. Natural hydrates occur at low temperatures and high pressures (substantially outside the range of ambient conditions), making observation difficult. Some investigators have successfully used pressure vessels with windows, but visual observations are limited to the outer appearance of the sample.

We use x-ray computed tomography (CT) scanning as a visualization technique to observe hydrate related laboratory processes occurring within the entire volume of laboratory samples. This activity is a component of our NETL-funded project in which we are determining (through history matching of laboratory data) parameters and processes of critical importance to the feasibility of gas production from hydrates.

In our tests, it is critical to understand what is occurring at each location within a sample to avoid misinterpreting local behavior for bulk behavior. Our work involves hydrate samples on multiple spatial scales. In the laboratory, we use core-scale and smaller samples to gather data that can be extended to the field scale. We are also interested in processes at the pore scale, which may ultimately control the ability to produce natural gas from a hydrate-bearing reservoir.

In CT scanning, x-rays are passed through the sample from many different directions, and detectors record the intensity of the x-rays passing through relative to the directions. Materials within the sample absorb x-rays roughly in proportion to their density and to a lesser extent their atomic number, thus denser and higher-atomic-number materials absorb more x-rays than less dense, lighter materials. The density image is calculated from the x-ray intensities. X-ray imaging is performed without altering the sample, making it ideal for tracking both temporal and spatial changes.

Our visualization studies employ a variety of tools. We use a modified medical CT scanner (Figure 1), several lab-built CT scanners (including one featured in Fire in the Ice, Fall 2003), and perform microtomography using the bright soft x-
rays at the Advanced Light Source (ALS) at Berkeley Lab. These instruments offer a variety of capabilities and resolutions - from rapid scanning and image computation of ~30 cm-scale samples at a resolution of ~ 250 microns, to very high resolution (~3 microns or less) scanning of cm-scale samples at the ALS, to portable equipment for scanning core-scale samples at field sites. All these tools provide an understanding of processes in different sample types and multiple scales.

![Figure 1](image_url)  
Figure 1. Arvind Gupta (Colorado School of Mines), Liviu Tomutsa, and Tim Kneafsey (LBNL) setting up an experiment on the modified medical CT scanner

The results of several of the experiments in which we have used CT scanning are presented below.

**Experiment 1: Tracking the Dissociation Front: Thermal Dissociation**  
**Collaborators: Laura Stern and Steve Kirby (USGS)**

In this test, we observed and measured the dissociation within a 2.5 cm diameter sample containing two regions with different mixtures of pure methane hydrate and silica sand. The success of this study hinged on our ability to use CT to distinguish between hydrate and the ice produced upon dissociation. Figure 2 shows a photo and an initial CT scan of the sample. We allowed the sample to
warm from the top and scanned while continuously measuring temperature and the volume of gas produced. To identify the changes in the sample, we looked at the differences in x-ray attenuation from the initial condition (Figure 3). As the hydrate dissociates from the top, the mass is reduced by the loss of methane, showing a decrease in attenuation (blue and purple colors). From these images, we can track the dissociation front and correlate it with temperature and volume data.

**Figure 2.** Photo of the sample (courtesy of Laura Stern, USGS) and x-ray CT image of the methane hydrate/sand sample
Figure 3. Attenuation changes as the dissociation front (at arrow) progresses downward in the sample

Experiment 2: Methane Hydrate Dissociation: Portable X-ray CT System

In Experiment 2, we placed cold, stable methane hydrate, ice, and silica sand in a 7.6 cm inner diameter aluminum vessel and allowed ambient heat to warm the vessel and cause dissociation. This experiment was performed to test the Berkeley Lab’s portable CT system, which is able to scan as much as a 10 cm length of core at a time with about 200 micron resolution. Figure 4 shows the initial CT scan (baseline) and the changes that occur over time. In the baseline, sand is the densest material and is the darkest in the image. The ice is the magenta object in the lower right quadrant. The hydrate chunks are the irregularly shaped yellow-orange objects. Over time, as dissociation occurs the
hydrate becomes less dense (more purple in color), while the sand and ice maintain a constant density.

**Figure 4.** Baseline CT image (top left) and changes in density over time (others) for a 7.6 cm inner diameter vessel containing sand (darkest in the baseline), hydrate (irregular yellow-orange shapes in the baseline), and ice (large magenta shape in the lower right quadrant). The density of the sand and ice remain constant, whereas the hydrate becomes less dense (darker colors) on account of dissociation. The scale bar does not apply to the baseline image, but only to the density changes.

**Experiment 3. Hydrate Formation and Dissociation in a Core-Scale Partially Saturated Sand**

**Collaborators:** Chuck Taylor (NETL) and Arvind Gupta (Colorado School of Mines)

In this test, we formed methane hydrate in wet sand (not fully water saturated) in a 7.6 cm inner diameter by 25 cm long aluminum vessel (Figure 5). We built this vessel to perform hydrate studies using CT scanning, and it includes multiple thermocouple ports allowing us to collect temperature data at four locations and a fluid jacket for temperature control. After forming the hydrate
(Figure 6b.), we dissociated some of it by heating from the outside (Figure 6c.), formed hydrate again (Figure 6d.), and dissociated it by depressurization and heating (Figure 6e.). The resulting density changes are indicative of the physical/chemical and hydrological processes that occurred during the formation and dissociation of the hydrate. With the CT scanning (Figure 6), we can see water saturation changes, the locations where hydrate forms and dissociates, and even gas pressure changes. Thermal properties and dissociation kinetics parameters are being determined by history matching using the TOUGHFx/HYDRATE code developed at Berkeley Lab.

Figure 5. Core-scale pressure vessel in the water jacket for temperature control. Thermocouples extend from the left side.

Figure 6. Initial scan at one location (a.) (Note: thermocouple location is shown by the white circles) and subsequent changes at that location caused by hydrate formation (b.), thermal dissociation (c.), second hydrate formation (d.), and dissociation by depressurization and heating (e.)
Conclusions

CT scanning is a valuable tool in hydrate research, one that can be used to help understand processes occurring in hydrate formation and dissociation in the laboratory, as well in characterizing natural cores. CT scanning, in conjunction with pressure and temperature measurements, is a powerful system for understanding hydrates, as well as providing data for numerical model extension and validation.