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MORPHOLOGY OF METHANE HYDRATE HOST SEDIMENTS

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Introduction

Results from simulated experiments in several laboratories show that host sediments influence hydrate formation in accord with known heterogeneity of host sediments at sites of gas hydrate occurrence (1). For example, in Mackenzie Delta, NWT Canada (Mallik 2L-38 well), coarser-grained units (pore-filling model) are found whereas in the Gulf of Mexico, the found hydrate samples do not appear to be lithologically controlled. We have initiated a systematic study of sediments, initially focusing on samples from various depths at a specific site, to establish a correlation with hydrate occurrence (or variations thereof) to establish differences in their microstructure, porosity, and other associated properties. The synchrotron computed microtomography (CMT) set-up at the X-27A tomography beam line at the National Synchrotron Light Source (NSLS), Brookhaven National Laboratory was used as a tool to study sediments from Blake Ridge at three sub bottom depths of 0.2, 50, and 667 meters. Results from the tomographic analysis of the deepest sample (667 m) are presented here to illustrate how tomography can be used to obtain new insights into the structures of methane hydrate host sediments. The investigation shows the internal grain/pore space resolution in the microstructure and a 3-D visualization of the connecting pathways obtained following data segmentation into pore space and grains within the sediment sample. The analysis gives the sample porosity, specific surface area, mean particle size, and tortuosity, as well. An earlier report on the experimental program has been given by Mahajan et al. (2).

Experimental

Sediment Sample. The sediment sample was obtained during Ocean Drilling Program Leg 164 on the Blake Ridge at a latitude of 31° 48.210' N and a longitude of 75° 31.343' W, at water depth of 2278.5 m and a depth below the mud line of 666.7 m. The sample was stored in the original container and refrigerated at 4° C prior to the experiment. For the CMT analysis, a small portion of the material was taken and from the container and used to fill a cylindrical polyethylene tube

that had an inside diameter of 4.7 mm and a wall thickness of 0.8 mm to a height of about 2 cm. Only a small portion, about 1.2 mm in height, was examined during the experiment.

Experimental Apparatus. The CMT experiment was performed at the Brookhaven National Synchrotron Light Source facility (3). X-radiation from a bending magnet at an energy of 14.89 Kev illuminated a sample area about 7 mm in width and about 2 mm high. The x-rays passing through the sample were detected using a YAG scintillator 0.50 mm in thickness. Light from the scintillator emitted in the beam direction was reflected at 90° through a magnifying lens onto a charge-coupled device camera with a size of 1335 x 1017 pixels. A series of exposures was made at angles from 0° to 180° in steps of 0.18°. Exposures were also made of the white field response to use in normalization of the attenuation values calculated from each exposure and of the dark field response to correct for background. The data was also corrected for overexposed pixels, the images were then used to construct a 2-dimensional (2-d) map of the attenuation coefficients for each pixel in a matrix that corresponded to the number of pixels in the horizontal size of the stored CCD pictures. A 3-dimensional (3-d) volume can then be assembled by stacking the 2-d sections into a 3-d matrix. The pixel size for the data reported here was 0.0067 mm. Jones et al. have given an overview of the experimental applications of synchrotron CMT (4) and Dowd et al. (5,6) have discussed more details of the experimental apparatus and further examples of experimental results.

Results and Discussion

The visualization of the 3-d volume obtained from the tomographic investigation of the sediments is shown in Figure 1. This is a partial volume cut from the full data set. The pixel size is 0.0067 mm so that the volume shown is 0.67 mm x 0.67 mm x 1.34 mm. An expanded view of an even smaller portion of the data is given in Figure 2. Similar views are shown for another data set in Figures 3 and 4. Only the higher values of the measured attenuation coefficients are shown. The complexity of the sediment morphology is clearly shown in these figures. The potential use of CMT for investigating questions related to the effects of sediment properties on methane hydrate formation on a grain-size scale is obvious.

The data can be analyzed in a more quantitative way. We used the 3dma software developed by Lindquist et al. (7) to segment the data into solid and pore space working on a histogram that shows the distribution of the measured attenuation coefficients. The histogram can be used to make rough determination of the distribution of different minerals in the sediments. The 3dma software then uses the histogram to make a pixel-by-pixel assessment to solids or pores based on several different algorithms. The result of the segmentation is used in several ways. These include calculations of the sediment porosity and the 2-d correlation function. The 2-d correlation function in itself yields estimates of the porosity, specific surface area, mean particle size, and permeability as discussed by Berryman and Blair (8), Berryman (9), and Blair et al. (10). More complex 3dma analyses can be done to give

the non-intersecting pathways through the volume (tortuosity) pore volumes, permeability and throat sizes.

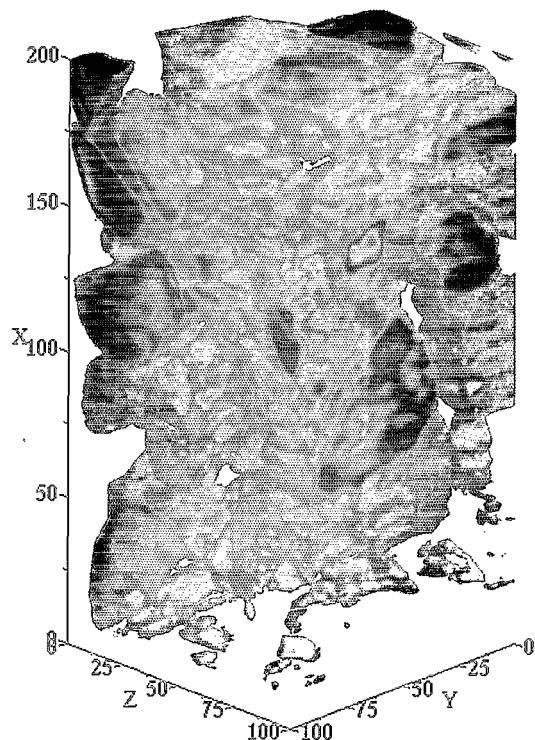


Figure 1. This figure is a partial view of the tomographic volume measured for the Blake Ridge sediment. It shows the morphology of the sediment on a micrometer scale. The pixel size is 0.0067 mm.

The values that we measured for the porosity of the sample using CMT are in good agreement with the bulk measurement found using gravimetric methods. The value of the tortuosity found for the sample using the 3dma program was 1.2. This low value is expected based on the very high porosity of the material. We did not try to apply the 3dma methods for throat and pore sizes since these concepts are not well defined for a very porous material. The estimated value for the specific surface area was 1700 m^{-1} and the particle-size estimate was very roughly about 0.100 mm. More extensive analyses can be found in the papers by Mahajan et al. (2) and by Jones et al. (11).

The results discussed here show that application of CMT to methane host sediments can give a unique picture of the interaction between the sediments and methane hydrates contained therein. It will be able to determine if the particles are incorporated in the sediments without changing the porosity or whether the sediment particles are extruded from the hydrates. Differences in the 2-d correlation functions will also be useful in considering that question and also in searching for evidence of cementation between the particles.

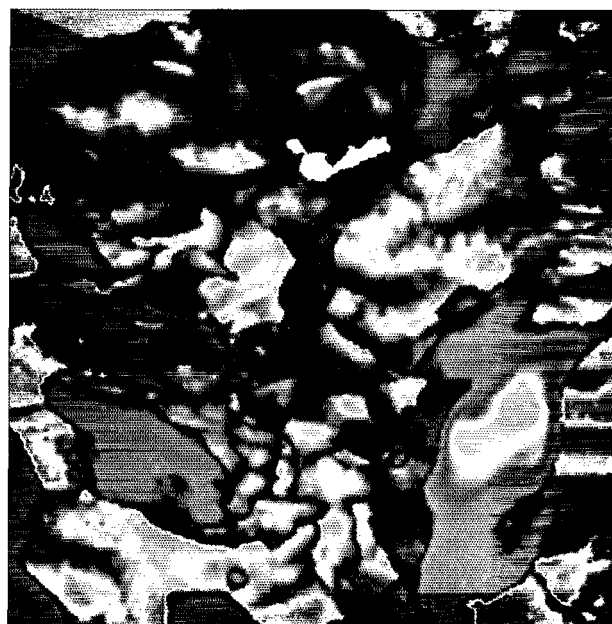


Figure 2. A small portion of the tomographic volume displayed in Figure 1 is shown to further emphasize the complex morphology of the sediments.



Figure 3. A volume representation of another measurement of the sediment sample is given that shows only the voxels with the highest values of attenuation coefficients. The visualization emphasizes the complex relationship between large and small sediment particles.



Figure 4. Magnified view of the larger volume that is given in Figure 3.

Conclusions

The data presented here demonstrates an innovative application of CMT to the study of methane hydrate host sediments that complements data from other methods. In future studies, the approach can be used to study both the evolution of methane hydrates formed in laboratory reactors and samples taken from specimens obtained from ocean sediments or in soils. Special sample cells that can provide regulated temperatures and pressures will need to be designed for these measurements, but the highly penetrating nature of the x-ray beams will make it feasible to implement the needed hardware to do experiments under realistic conditions.

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