Use of Computed Microtomography to Visualize and Quantify Pore-scale Transport Processes

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Introduction

Diffusion in low-porosity rocks is a potentially important process in the evaluation of deep geologic waste repositories. At certain sites in Japan and Europe where repositories in crystalline rocks are being studied for nuclear waste storage, physical retardation is thought to be an important factor in delaying potential radionuclide releases. Presently there is little direct observation of diffusion and retardation in crystalline rocks, since they are studied mostly by field tracer tests and bulk laboratory measurements.

Imaging tools can greatly enhance the understanding of the physical controls of diffusion. Two-dimensional x-ray imaging has been used to study diffusion in dolomite and fractured crystalline rocks [1, 2]. However, this technique does not have a high enough resolution to be effective on materials with porosities lower than approximately 5% [3]. Use of synchrotron-based computed microtomography (CMT) allows for the high-resolution, 3-D imaging necessary to characterize small-scale heterogeneities.

The purpose of this study to examine whether CMT would be an effective technique for running diffusion experiments on low-porosity rocks. We examine using cesium chloride (CsCl) as a potential tracer.

Methods and Materials

Four rock samples from three different sites were selected for imaging (Table 1). Cores with 9-mm diameters were extracted from larger samples. The samples were pressure saturated with 400 g/L of CsCl solution and sealed in shrink-wrapped plastic and wax. Capillary tubes filled with known concentrations of CsCl were also brought and imaged. Concentrations in the tubes ranged from 8 to 400 g of Cs/L.

CMT images were obtained on the samples at beamline BM-13D, GSECARS-CAT. Images were obtained at 36.05 and 35.95 keV. The voxel size for each sample was approximately 20 µm on a side.

Samples were imaged at two different energies, above and below the absorption edge of cesium. The absorption edge is the energy required to knock an electron from an outer shell of the atom. At the energy just below the absorption edge, cesium is much less absorbing to x-rays than at energies immediately above the absorption edge. We subtracted the data arrays of the images obtained above the absorption edge from those taken below the absorption edge. If regions in the sample contain no cesium, then the voxel values above and below the edge should be close to the same value and result in a zero intensity in the difference image. In contrast, if a sample region contains cesium, then the voxel values in the difference image should be greater than zero and appear as bright areas in the image. The difference image therefore enhances the visibility of the cesium in the sample.

TABLE 1. Rock core sample summary.

Rock type	Location collected
Diorite	Äspö Underground Rock Laboratory, Sweden
Fine-grained granite	Äspö Underground Rock Laboratory, Sweden
Granodiorite	Kamaishi Mine, Japan
Shear zone	Grimsel Underground Rock Laboratory, Switzerland

Results

Figure 1 shows color-enhanced difference images of the four cores that were saturated with CsCl. Each figure shows a horizontal slice of one of the four cores listed in Table 1. The difference image was generated by subtracting the data array of the image collected above the cesium absorption edge from that of the image taken below the cesium absorption edge. Areas in red show regions containing cesium. Figure 2 presents a comparison of the expected theoretical linear absorption coefficient with the approximate range of those measured in the capillary tubes filled with known concentrations of cesium.

Discussion

The image differencing technique proved to be effective in showing areas where cesium had permeated the cores (Fig. 1). In the case of the shear zone material (Fig. 1D), it is clear that the cesium is in the pore space. However, because cesium can sorb onto clay minerals, there is some question as to whether the cesium that can

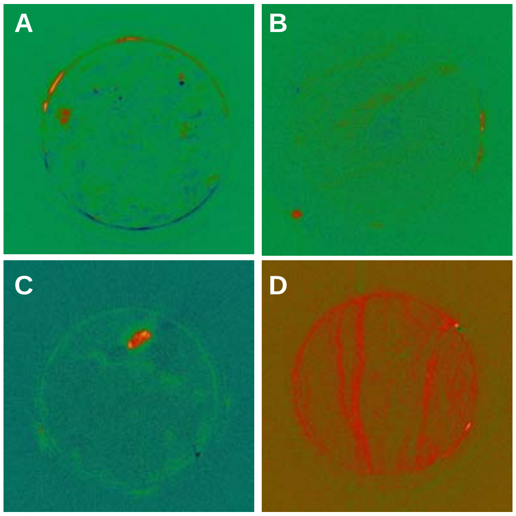

FIG. 1. Slices of CMT difference image of horizontal slice through 4 different 9-mm cores. A – diorite, B – fine-grained granite, C – granodiorite, and D – sheer zone.

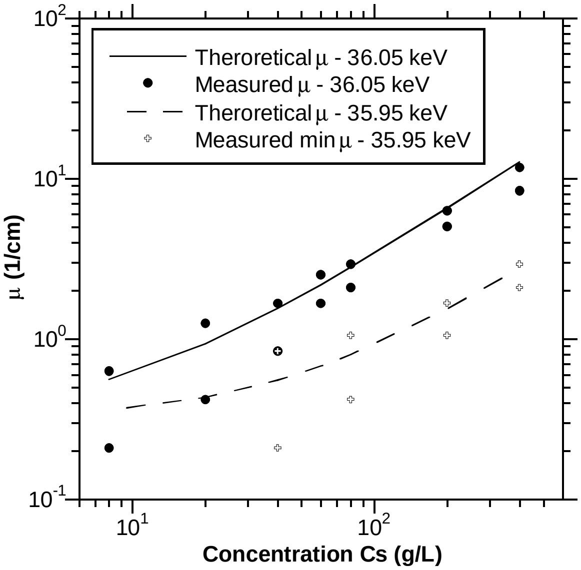

FIG. 2. Comparison of theoretical linear coefficient values (μ) to the approximate ranges of those measured in capillary tubes filled with known concentrations of cesium.

be seen in the other cores is in pore space or not. For example, the region with cesium shown in the granodiorite (Fig. 1C) is most likely a mineral that has been altered into clay. Because cesium is seen in the interior of the four cores, we do know that there is accessible pore space that leads to the regions with cesium. As a next step, we will image cores saturated with potassium iodide (KI). Iodide should not sorb in the same way the cesium has, so identification of areas with iodide will clearly show pore space. However, the absorption edge of iodine is lower that that of cesium (33.2 keV). This means that smaller diameter cores will have to be used in order to be able to get enough x-ray transmission through the cores.

The comparison of measured linear absorption coefficients to theoretical ones shows that the cesium concentrations could be inferred from measured linear absorption coefficient values (Fig. 2). However, there is enough noise in the data that only large differences in concentrations could be observed. The noise appears to increase with decreasing cesium concentrations. We are currently looking into using measurements of these calibration standards to reduce the noise in the data. It should be noted that relatively large concentrations of cesium are needed in order for it to be detected (greater than approximately 10 g/L).

This work has shown that there is promise in using CMT to conduct diffusion experiments with visualization. With some noise reduction corrections, we believe that quantitative results could be obtained. Another consideration is that the process that is being studied (e.g., diffusion) must be slow enough that significant changes will not occur during the time it takes to collect the data needed to reconstruct each image.

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References

[1] V. C. Tidwell, L. C. Meigs, T. Christian-Frear, and C. M. Boney, J. Contam. Hydrol. **42**(2-4), 285-302 (2000).
[2] S. J. Altman, M. Uchida, V. C. Tidwell, C. M. Boney, and B. P. Chambers, SAND2001-1842 (Sandia National Laboratories, Albuquerque, NM, 2001).
[3] S. J. Altman, M. Uchida, V. C. Tidwell, C. M. Boney, and B. P. Chambers, J. Contam. Hydrol. (in press).