STORAGE OF PRESSURIZED CARBON DIOXIDE IN COAL OBSERVED USING X-RAY TOMOGRAPHY

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Introduction

The sequestration of CO_2 in coal seams has been proposed as a mitigation strategy for climate change. To maximize sorption potential it is essential that the heterogeneity of the coal seam be represented in the computational models used to predict the complex flow and sorption within the seam. It is well known that the porosity and sorption of gases are maceral, and hence lithotype dependent[1]. The presence of lithotypes and mineral layers in the seam will influence the fluid-flow, sorption capacity, and sorption rate. As potential sequestration sites are deeply buried, un-mineable coal seams realistic conditions will involve considerable lithostatic and hydrostatic pressure. These may be important because of the tendency of some coals to swell when carbon dioxide is absorbed[2]. Unfortunately, this is not easily reproduced in the traditional sorption experiments such as thermogravametric analysis. An additional problem is that the coal samples are often pulverized and hence the heterogeneity of the lithotypes is lost in the average values obtained. This is undesirable if we wish to predict sequestration potentials and realistic CO_2 behavior within a particular seam.

Objective

The objective in this study was to characterize the uptake diversity of CO_2 into a coal core under simulated in situ conditions in the coal seam. Of particular importance is the heterogeneity of the sorption, and its subsequent influence on fluid-flow into the coal seam. Here we demonstrate the application of Computerized Tomography (CT) technology to sequestration for the first time.

Approach

To approach this objective, we have subjected coal samples to CO_2 pressure while confining them under moderate overburden stress. X-ray CT images were taken and quantitatively interpreted to map the heterogeneous uptake of the gas as a function of time and applied gas pressure. The change in CT numbers was followed for the coal core, confined in X-ray transparent pressure vessels which applying in situ stress at room temperature, upon CO_2 sorption.

Technology

X-ray CT provides three-dimensional mapping of X-ray absorption of a sample. Differences in X-ray attenuation (related to the atomic composition and bulk density) are determined non-destructively. Quantitative maps of X-ray absorption are produced using sophisticated computer signal processing to combine many "slices" of the sample. By placing the resulting two-dimensional cross sections (slices) together in 3-dimensional space, the image of the physical form of the subject can be viewed and manipulated. Simple visual images of these data can be used to observe macro structures in the coal including fractures and cleats, mineral distribution, and differences in matrix density. Using appropriate calibrations the data can be interpreted quantitatively to map density, porosity and fluid saturation. Both a medical scanner and a high-resolution material scanner were utilized in this study. The high-resolution instrument utilizes cone-X-ray tomography to scan a volume rather than single "slices", resulting in greater pixel resolution and an order of magnitude decrease in the "slice" thickness equivalent.

In addition to simple characterization of the cleat, pore structure, and lithotypes present, the technique can be used to follow the absorption and desorption processes. Coal lithotypes (macroscopically distinguishable in coal seams from their thickness, friability, and luster) and macerals (optically recognizable organic components of coal under the microscope) have different pore volumes and pore size distributions [3-5]. Due to the density differences in coal, for most of the rank range, the lithotype layers can be identified. Thus, CT-derived lithotype maps can be identified quantitatively. Since introduction of CO_2 changes the bulk density and composition of the coal, the quantity and distribution of the sorbent can be followed as a function of time and pressure conditions. In the following, we demonstrate the potential of CT technology for use as an enhanced formation characterization tool for following CO_2 sequestration within coal and its influence on the physical structure.

Methods

Coal cores were obtained from the Pennsylvanian Department of Environmental Protection and from the Penn State Coal Sample and Database Library. The experiment reported here was performed on a coal core section from the Fruitland coal seam in the San Juan basin, Colorado. This seam is currently the site of a large-scale CO_2 sequestration effort. This location is well known for coalbed methane production and has been well studied. The coal core is from a high volatile A bituminous part of the seam.

The core was first tested under as-received conditions (water wet) and subsequently evacuated. The measurements were performed on cores confined in an X-ray-transparent pressure vessel designed to simulate in situ stress and temperature conditions. The cylindrical coal cores were sheathed in a rubber jacket and housed within the hydrostatic cell, which is capable of applying differential confining stresses to 30 MPa. Confining load is applied through the rubber sleeve. The cell schematic is shown in Figure 1.

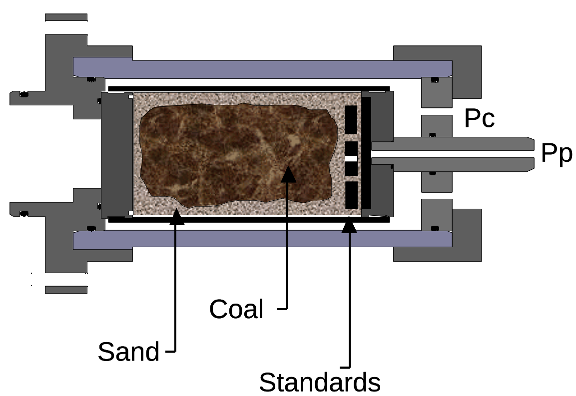

Figure 1. Pressurized Cell Schematic

Uptake of CO_2 was determined from the change in CT numbers (a normalized measure of X-ray absorption) over time for specific locations within "slices" of the coal core. The CT numbers were calibrated against density standards to allow quantitative interpretation of CO_2 sorption. As the density was determined, and the slice thickness is known, the volume of the coal can be calculated. A hole drilled in the one of the density standards served as a location where the CT number of the injected CO_2 could be determined under the conditions of pressure and temperature used. This permitted the uptake to be measured on a mass-per-mass basis. The kinetics of uptake was also followed over a 6,000-hour period.

Results

A digital radiograph (equivalent to a simple transmission X-ray) of the pressurized vessel and coal core are shown in Figure 2. The bedding plane of the coal core is clearly visible, and is parallel to the core diameter. Different density bands (lithotypes) are evident within the core. Density calibration standards, sand, water, and the rubber gasket, used to apply pressure to the core, are also shown. Two-millimeter-thick tomographic scan slices were taken at intervals of one centimeter along the core's axis. An axial reconstruction formed by combining many of these slices is shown in Figure 3. The black lines signify the "slice" locations for subsequent figures. The red colors indicate higher density regions whose CT numbers are indicative of mineral matter (Figure3).

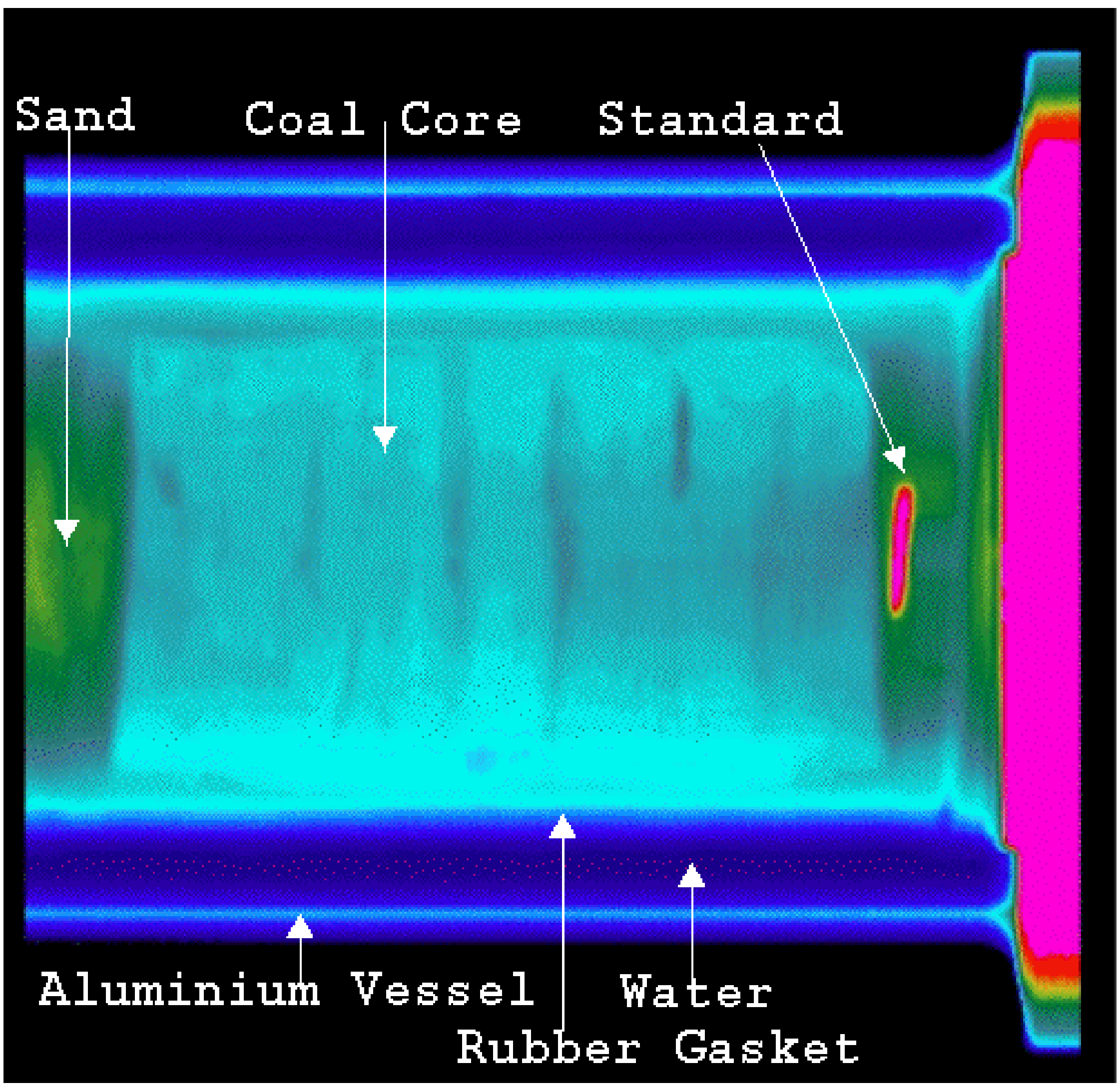

Figure 2. A Digital Radiograph of the Fruitland Coal Core within the X-ray Transparent Vessel. Brighter colors indicate a higher CT attenuation (a higher density)

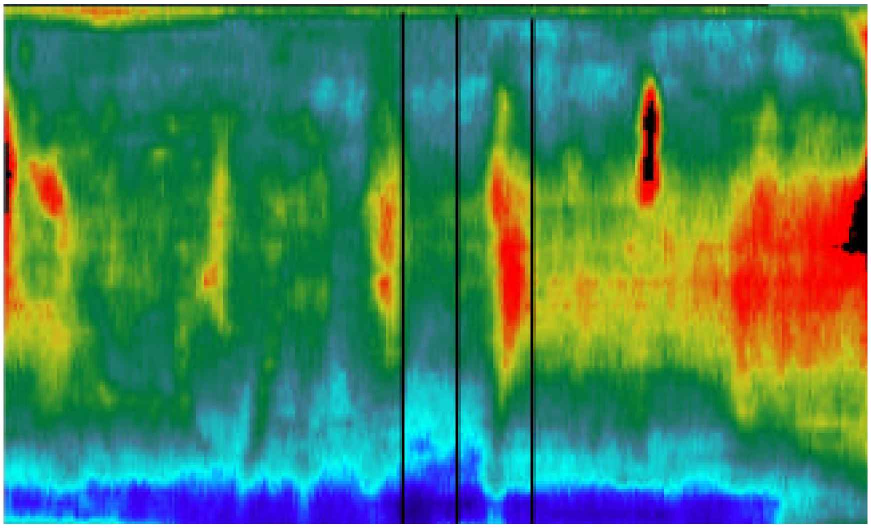

Figure 3. Reconstruction of the Fruitland Coal Core. Black Lines Signify "Slice" Locations Numbers 8, 9, & 10 from left to right. Slice Thickness is 2 mm.

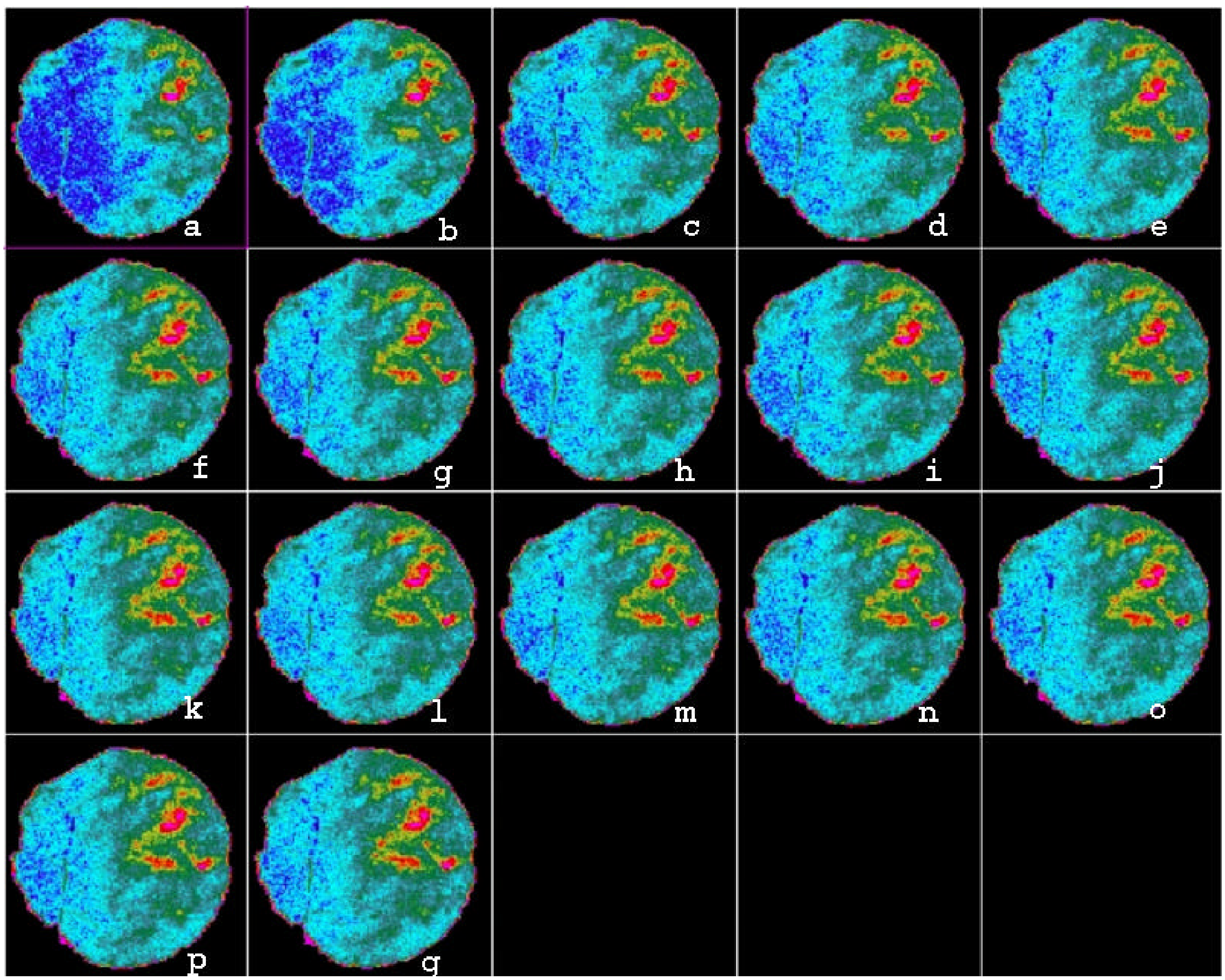

Figure 4. Time Sequence of Slice 8. Changes in image sequence are due to the sorption of Carbon Dioxide.

A time sequence of an individual tomographic slice (slice 8) is shown in Figure 4 as a series of images advancing in time from left to right. Color changes indicate the change in density due to the sorption of CO_2. The lower density region in the left-hand side and the high-density region in the middle right-hand of the slice uptakes the majority of the CO_2 for this slice. The heterogeneous lithography of the slice is the likely cause of the differences in the sorption behavior[6]. The higher density region of this slice contains a mineral matter band that may indicate a porous region of the coal. A linear relationship between pore volume and ash has been determined for a bituminous coal[3].

Observing the CT number change in the hole of the density standards on pressurized CO_2 exposure, it is possible to quantify the uptake of CO_2 in selected regions as presented in Figure 5. The uptake follows a similar pathway for each of the areas selected (two from slice 7 and one from slice 10). However, the storage capacity is different. Area 1 uptakes more CO_2, by a factor of 1.6, than area 2 (Figures 5 & 6). This is to be expected, as lithotypes are known to have different sorption capacities. Typically the bright vitrain bands have the highest capacity but this is not always the case. The influence of the specific maceral groups and the diversity across the rank range makes the absolute assignment of the higher sorption lithotypes difficult without microscopic identification and experimentation. The advantage of the technique used here is that sorption capacities can be measured on "large" samples providing the overall uptake *and* the uptake diversity. The regions used for the figure are shown for "slices" 7 & 10 in Figure 6.

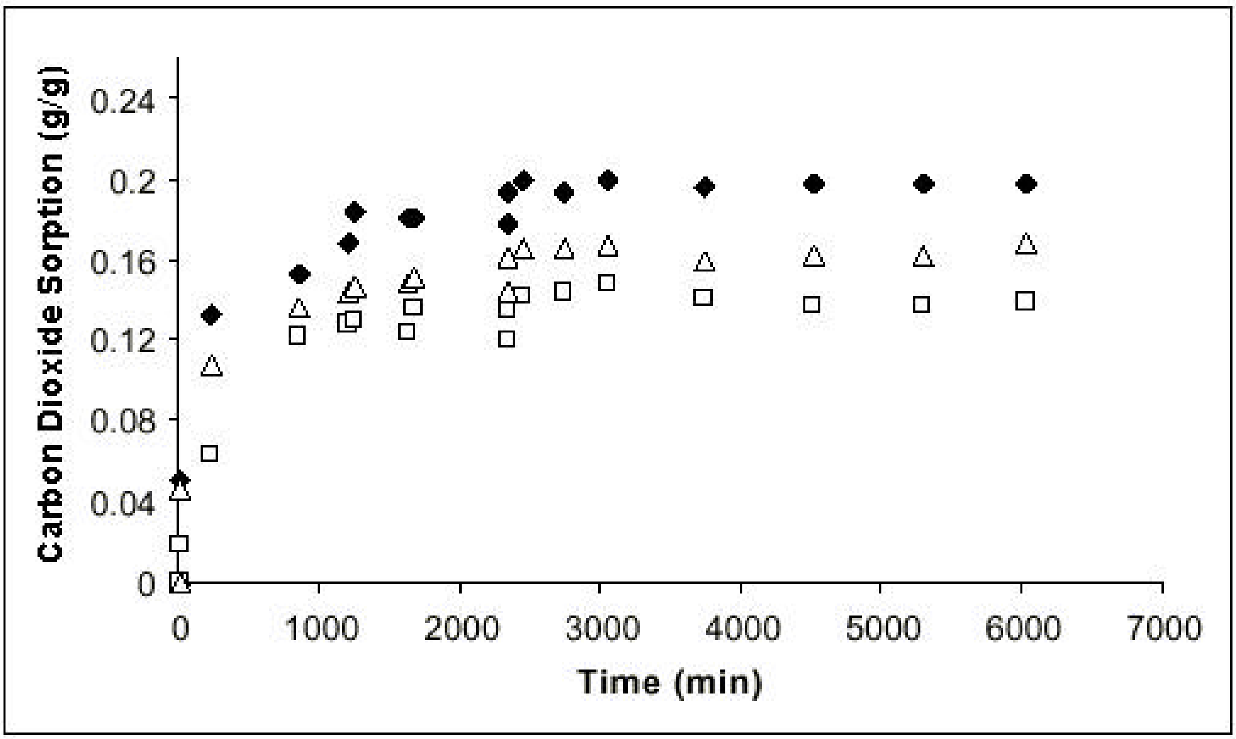

Figure 5. Sorption of Carbon Dioxide in Fruitland Coal vs. Time for Two Regions of "Slice" 7 and One from "Slice" 10. The Highest and Lowest Uptake Capacities are for Regions 1 and 2 Respectively, Shown in Figure 6. The Middle Region is from an intermediate density area from "Slice" 10. Carbon dioxide pressure increased from 300 psi to 500 psi after 2,200 minutes.

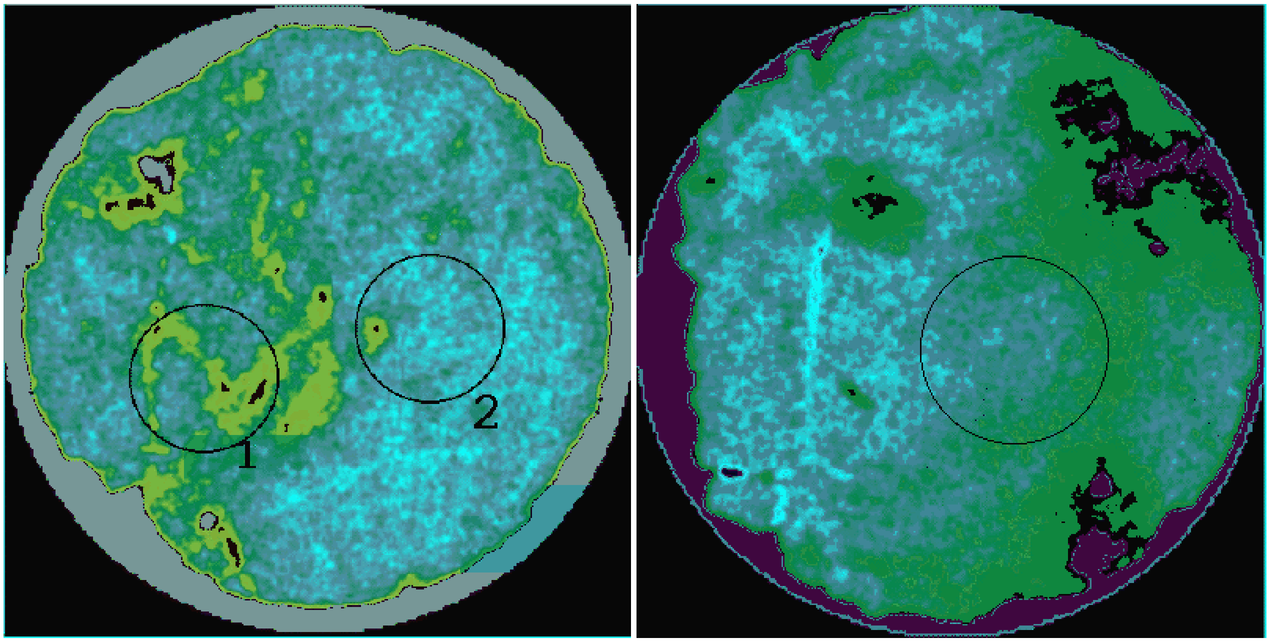

Figure 6. "Slices" 7 and 10 of the Fruitland Coal Core. Marked Regions are used in Calculating Figure 5.

Time sequence plots of selected areas (Figures 5 & 6) within a "slice" are used to calculate the region specific uptake. Selecting multiple areas that represent the lithotype diversity (in multiple slices) will permit the sequestration to be followed with respect to the individual coalbed lithography. This data can be collected following the completion of the sequestration using traditional, but destructive, microscopy.

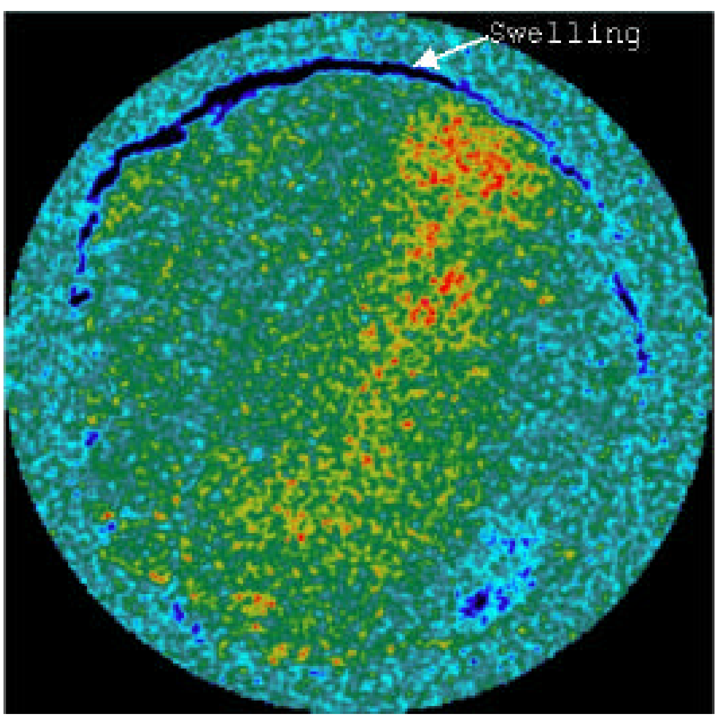

Figure 7. Subtracted Image Showing Expansion of Fruitland Coal "Slice" 10 After 100 Hours of Exposure to Carbon Dioxide.

Also of interest is the change in the physical structure of the coal during sequestration. In the coalbed the removal of moisture, a good swelling agent, and perhaps methane may cause shrinkage[7]. Swelling is known to occur upon CO_2 sorption. Image subtraction of the original unexposed image of slice 10 from that of slice 10 after exposure produces Figure 7. The dark blue crescent ring at the top of the figure is indicative of the coal swelling, displacing the sand. The sand has a higher CT number, so that the image subtraction produces an area of negative numbers where the coal has displaced the sand. Note that the swelling is not uniform. From pixel counting, length measurements indicated the coal has swelled by 2.4%

of its original diameter, at this location, despite the confining pressure. A similar expansion was determined for the Pittsburgh seam coal core (not shown). The red-to-yellow areas of Figure 7 represent positive CT numbers, indicating that central location of this slice has preferentially sorbed the CO_2 as indicated by a CT number increase. This heterogeneity in CO_s uptake again represents different absorption by different lithotypes. Increasing the CO_2 pressure from 300 to 500 psi did not significantly increase the uptake (Figure 5). Increasing the CO_2 pressure to 1,400 psi has been shown to more than double the capacity of CO_2 in a wet Fruitland coal in comparison to the lower pressures (at 115 °F)[8]. This result was attributed to multilayer adsorption. Initial attempt to follow the sorption of CO_2 with CT on the wet Fruitland coal core failed to produce significant uptake on exposure to CO_2. The moisture was presumably blocking the pores. Removal of water was necessary prior to uptake. Flow-through experiments however will be a better simulation of the system.

High resolution images are shown in Figure 8. The advantage of the higher resolution is the slice thickness equivalent is smaller by an order of magnitude. This allows for an improved accuracy for the core description and behavior. The mineral banding and different density regions are clearly shown.

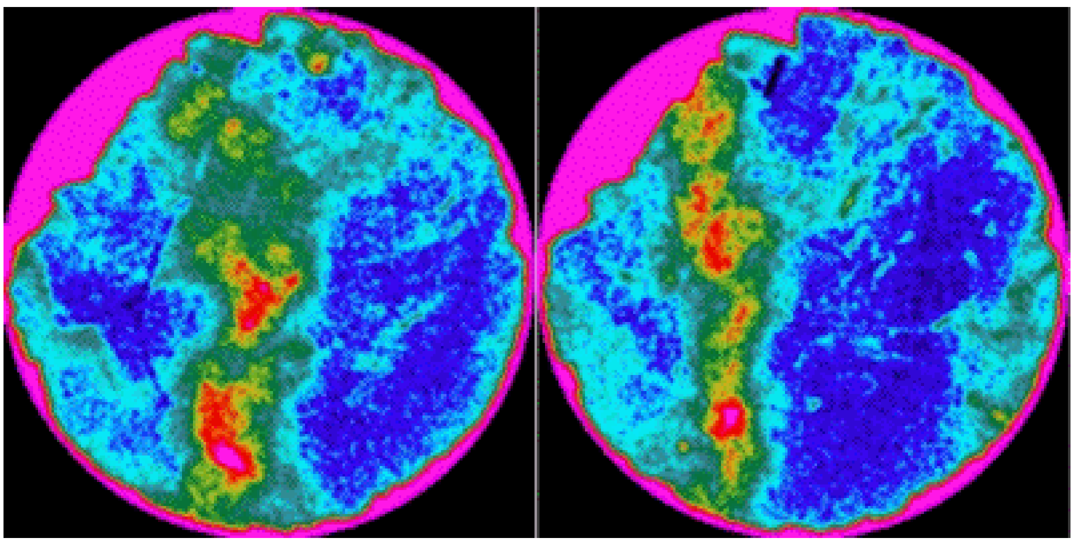

Figure 8. High Resolution Images of Two Locations 1mm Apart, Slice Thickness Equivalent is 150 µm.

Benefits

This enhanced characterization of coal and its sorption and uptake capacity allows improved modeling for the optimized storage of CO_2 within a coal seam. Such approaches are necessary if the sequestration site is to maximize uptake while still being economically viable. The CT technique has many benefits over traditional techniques. The most important is a quantitative image sequence from which the diversity of sorption behavior and uptake kinetics can be followed.

Future Activities

Linking the observed behavior to specific lithography's within the core will aid in determining sequestration potential and variability. The core will be dissected and the lithography determined under the microscope. We also wish to continue this work on smaller cores but at a much higher resolution (20 μm) utilizing a high energy micro-focus industrial scanner. We also plan to follow flow-through experiments to determine the competitive sorption / replacement of methane on recently extracted gassy coal samples.

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