X-RAY MICROTOMOGRAPHIC IMAGING AND ANALYSIS FOR BASIC RESEARCH

J. H. Dunsmuir, S. Bennett, L. Fareria, A. Mingino, M. Sansone
ExxonMobil Research and Engineering Company
Corporate Strategic Research
Annandale, New Jersey 08801

ABSTRACT
For research facilities with access to synchrotron x-ray sources, X-ray absorption microtomography (XMT) has evolved from an experimental imaging method to a specialized, if not yet routine, microscopy for imaging the 3D distribution of linear attenuation coefficients and, in some cases, elemental concentration with micron spatial resolution[1]. Recent advances in source and detector design have produced conventional x-ray source instruments with comparable spatial resolution but with lower throughput and without element specific imaging[2]. Both classes of instrument produce 3D images for analysis. In this paper, we discuss an integrated approach for the implementation of analytical XMT to support basic research into the structure-property relationships of a variety of materials. The essential components include instrumentation for collecting quantitative 3D images, a 3D image processing environment to address questions as to the quantity, composition, geometry and relationships among the features in one or more images, and visualization to provide insight and communicate results. We give examples of image analysis of resolved and unresolved pore spaces of sandstones.

INTRODUCTION
The motivations driving the development of XMT have been diverse. These range from fundamental studies of image contrast formation that exploit the unique properties of new x-ray sources, to the development of detector technologies, and, ultimately, to the application XMT to address scientific and engineering questions. In its current state of development, XMT requires significant resources and is not yet a tool for casual inspection. The main contribution of the XMT technique lies in its ability to measure globally the 3D structures of the specimen that relate to externally observed mechanical, hydrological, electrical or acoustic properties. Frequently, inspection of the 3D image using visualization provides sufficient qualitative understanding. The presence, approximate size, and distribution of components can be quickly estimated and compared by a skilled interpreter. In complex structures, the visualized relationships are less clear and quantitative image analysis is needed in order to extract meaningful structure-property relationships. Two dimensional image processing methods do not take full advantage of the 3D nature of the data and may actually give misleading results due to sampling errors. We have developed 3D image processing software to analyze geological, catalyst, composite material and other samples scanned by XMT. Although the functions were initially developed to meet our immediate research needs, they were later modified to be more general. They are 3D by default and handle 2D images as special cases. Scalar and image arithmetic allow limited importation of external relationships. With acquisition, image processing, modeling and visualization tools, XMT has provided valuable guidance for composite and catalyst design and contributed physical insights that guide external measurements such as NMR well logging[12]. The key elements in analytical XMT are the acquisition of high quality images, measurement of the relevant properties of the image and the...
This document was presented at the Denver X-ray Conference (DXC) on Applications of X-ray Analysis.

Sponsored by the International Centre for Diffraction Data (ICDD).

This document is provided by ICDD in cooperation with the authors and presenters of the DXC for the express purpose of educating the scientific community.

All copyrights for the document are retained by ICDD.

Usage is restricted for the purposes of education and scientific research.

DXC Website  ICDD Website
communication of those results either as parameters, distributions, derived images or series of images.

IMAGE ACQUISITION

Three XMT instruments are used in our laboratory. Two are located at beamline X2B at Brookhaven National Laboratory and are operated remotely from our home institution. The third instrument uses a conventional diffraction x-ray source and is located in our laboratory. All instruments are set up for parallel beam illumination and use a polished single crystal CsI(Tl) scintillator to convert x-rays to visible light and an optical relay to a CCD camera for projection acquisition. The first X2B instrument is used for high dynamic range measurements. A Si<111> single crystal monochromator provides tunable narrow bandpass x-ray illumination in the range of 10 - 37keV. Projections are recorded using 1370x1340 16bit CCD camera with a readout rate of about 0.75 frames per second. This scanner is used for absorption edge crossing experiments and precise measurements of linear attenuation. The second X2B scanner is optimized for fast data acquisition. A 22A or 44A W/B4C multilayer monochromator provides broadband high-flux illumination from 8 - 25keV. Projections are recorded using a 1392x1040 12bit CCD with a maximum readout rate of about 13 frames per second. The fast system is used when 12 bit digitization provides sufficient signal to noise to adequately image the specimen contrast. A 1392x1392x1040 3D volume can be acquired in about 10 minutes. Both X2B scanners use high numerical aperture microscope objectives to couple the scintillator to the CCD and have a spatial resolution approaching 1 micron under optimum illumination conditions. The conventional-source scanner is primarily used for larger or more dense specimens. It is similar in design but operates at lower resolution due to penumbral broadening and low flux. Flat field copy lenses are used for optical coupling in the conventional instrument. Detective quantum efficiency ranges from about 0.3 using a 20x NA = 0.4 objective to 0.01 for low resolution imaging and is mostly determined by the numerical aperture of the relay lens. As configured, all of our XMT systems are CCD shot noise limited.

Beam hardening from the conventional source is corrected using a polynomial fit to a calibration step wedge. In all systems, ring artifacts are suppressed using both acquisition protocols and post processing. A correlated sampling method is used during acquisition[3]. Two projections are acquired, the second with the specimen position slightly displaced. Signals that move the displacement distance are assigned to the specimen and those that do not are classified as systematic errors and removed. The sorting algorithm is sufficiently fast to allow asynchronous acquisition with the high dynamic range instrument. Detector defects that are separated by the displacement distance are removed after acquisition by high-pass filtering a sinogram column sum and subtracting the result from each row of the sinogram. While very effective in removing rings associated with systematic errors, this approach can introduce rings adjacent to high contrast features in the reconstructed slice.

Where appropriate, beam shaping is used to adjust the intensity profile of the incident beam such that the transmitted intensity profile across most specimen projections is significantly flattened. This permits acquisition of projections with nearly constant signal to noise across the field of view. This approach greatly reduces ring artifacts near the center of rotation where S/N is usually poor compared to the perimeter. The horizontal beam profile is shaped using an
aluminum plate with shallow V-grooves machined lengthwise into both surfaces such that the grooves nearly touch at the center. The plate is mounted on a rotation table with the axis of rotation aligned with the grooves and centered vertically in the incident beam. The horizontal intensity profile is modulated by rotating the plate.

The axis of rotation is aligned perpendicular to the x-ray beam and parallel to the CCD pixel columns using a difference method. Sub-pixel alignment is obtained by subtracting from a projection taken at 0 degrees specimen rotation a projection taken at 180 degrees that has been digitally flipped about a pixel column boundary, usually at the center of the CCD. The position, sign and magnitude of the difference image indicate the amount of misalignment of the axis of rotation both in position and rotation with respect to the selected pixel boundary. This method uses the photometric accuracy of the CCD rather than feature position to measure alignment error and, for high contrast specimens, alignments to less than 0.1 pixel are readily achievable.

**EXPERIMENTAL METHODS**

The instruments described here detect x-ray absorption. Other "bright field" methods such as phase contrast and dark field methods such as fluorescence tomography are not supported. Edge enhanced "pseudo-phase" absorption images may be obtained by displacing the specimen a few centimeters from the scintillator screen. CT scans across the absorption edges of multiple elements may be automated using acquisition scripts. Additional methods usually involve altering the specimen or specimen environment in-situ between scans using special equipment. For example, fluid flooding is commonly used to detect both resolved and unresolved porosity. Ex-situ specimen alterations usually require computationally expensive 3D image realignment.

**IMAGE RECONSTRUCTION**

Direct Fourier Inversion is used for reconstruction[1]. This method in inherently quantitative, offers substantial inherent speed advantages when reconstructing parallel beam data and is readily parallelizable. Although local reconstruction methods, where portions of the specimen rotate outside the field of view of the detector, are not supported, the global algorithm is applied to local data using profile extension methods. The reconstructed linear attenuation coefficients will be relative rather than absolute. Sinogram preprocessing modules for beam hardening, sum rule, ring reduction, axis position and profile extension are included in the reconstruction module.

**3D IMAGE PROCESSING**

As in many microscopies, image inspection alone is sometimes sufficient and a 2D view of reconstructed slices may be adequate. 3D visualization provides an intuitive view of the structure when needed. Questions regarding the spatial relationships between the properties of the various components or structures and their relationship to external observables require a quantitative approach. Image processing tools that allow the experimenter to probe the three-dimensional structure are essential to extract the maximum benefit from microtomographic images. Rapid progress is occurring in 3D image processing [4], but essential tools are often unavailable or incompatible. During the course of investigating several classes of material, we
have developed a proprietary software package, 3DToolKit or 3dtk, for image processing. In many cases the modules in 3dtk are simple extensions to 3D of well known 2D algorithms. In a few cases the extensions are less straightforward. 3dtk currently contains 30 modules for image vector and scalar arithmetic, filtering, sampling, statistics, drawing and connectivity. Additional modules are based on FFTW[5] for Fourier methods, 3dma[6] for medial axis transform and segmentation, regfloat3d[7,8] for 3D image registration, and Numerical Recipes[9] for numerical analysis. 3dtk contains only scalar and image arithmetic to apply external relationships. For example, there are no flow simulation modules since the properties of the fluid are extrinsic to the image. These operations are best performed separately in a suitable computing environment to which microtomographic images are exported. Although separating modeling and image processing is not always optimal, it results in a robust toolkit that can be applied equally to all samples. The toolkit is written in ANSI C and builds readily on most platforms. In our examples, the user interface to the various toolkit functions has been implemented to run as extensions to a commercial 2D image processing program [10]. The commercial program supports image stacks and scripting so that the user can easily automate complex processes. In some instances, where consistency and frequency of use warrant, higher level C wrappers are used to bundle the lower level processes into a single callable function. The examples below illustrate the use of these functions.

EXAMPLE STUDIES

Random porous media are among the most common sample types for XMT studies in our laboratory. Samples of reservoir siliciclastics and carbonates make up the bulk of these specimens. The measurement of fluid saturations and the calculation of NMR T2 relaxation in the fast diffusion limit using image processing have been discussed previously[11,12,16]. Here we present experimental methods and image processing techniques used to characterize the resolved and unresolved pore spaces in rock specimens. In the first example we simulate mercury injection capillary pressure (MICP) in the resolved pore spaces of different porosity Fontainebleau sandstones. In an MICP experiment the volume of non-wetting mercury forced into the pore structure is recorded as a function of injection pressure. The pressure required to force a non-wetting liquid through a circular cross section is given by the Washburn[13] equation

$$P_{Hg} = \frac{2\sigma_{Hg/air}(-\cos(\theta))}{r_pore}$$

where $P$ is the capillary entry pressure, $\sigma$ is the mercury surface tension, $\theta$ is the mercury-air-grain contact angle and $r$ is the pore throat radius.

For a selected contact angle and interfacial surface tension the pressure dependence of the invaded volume is converted to a radius distribution. This radius distribution is frequently described as a pore size distribution assuming a bundle of cylindrical capillaries model. The measurement is subject to artifacts when a large pore volume is only accessible through small pores. This large pore volume will be counted incorrectly with the small pore radii.
Pore space geometry was obtained from XMT images of six Fontainebleau sandstones from 7 to 22% porosity by applying a gray-level threshold to segment the image into pore and grain. Simulations of MICP in pore spaces have been reported previously\cite{14,15,16} and typically rely on skeletonization to extract pore radii. We simulate non-wetting MICP using 3dtk components. We examine the connectivity of exact Euclidean distances, calculated using a 3D extension of Danielsson's algorithm\cite{17}, as a function of distance using a gray level flooding algorithm\cite{4}. Euclidean spheres are drawn at flooded radii and the flooded volume recorded. A 2D representation of the image processing steps of the algorithm are shown in figure 1 with the calls to significant 3dtk functions shown in italics.

Figure 1. 2D example of processing steps in simulation of MICP. a) original binarized image of a percolating random network. b) \textit{ExactEuclidean} distances to nearest boundary. c) \textit{FloodFill} connected regions $R_{\text{flood}} \leq R \leq R_{\text{max}}$. d) Remove unflooded regions. e) Restore flooded distances. f) \textit{DrawSpheres} at restored radii. g) Flooded region showing flooded (white) and unflooded (gray) pore spaces.

Figure 2b shows a plot of flood radius vs. volume similar to that obtained indirectly by MICP. The breakthrough radii, where the digital flood first reaches the opposite side of the sample, are also shown. Surprisingly, the 15% porosity specimens have a larger critical radius than the 22% porosity specimens. A sudden jump in volume at $R_{\text{flood}} = 4.1$ pixels in the 15% porosity specimen is a radius step that exceeds a critical throat diameter and is similar to those observed in MICP experiments. We note that the simulation tracks only geometry and topology. Effects due to fluid mass, grain surface properties not captured by XMT are ignored. In addition, even when using exact distances, the digital nature of the data results in discrete distances for small throats. A throat consisting of a single voxel and its nearest touching neighbors can have distances of 1, $\sqrt{2}$ and $\sqrt{3}$. The simulation floods smoothly at large pore throats but jumps suddenly as the resolution limit is reached. The 7% porosity sample was not imaged with sufficient resolution for successful simulation. The effective mercury interfacial radii restored by the simulation are not necessarily spherical since they can be drawn by overlapping spheres of radii $R \geq R_{\text{flood}}$. The simulation accounts for back-filling of smaller features along the sides of rough pores that are lost using skeletonization algorithms. Figure 3 shows the shape of the flooded pore space at the breakthrough for the 11% porosity specimen. We can compare the pore size distribution obtained by simulating MICP to that obtained by the local measurement of the surface to volume ratio. \textit{PoreSizeDistribution} wraps several 3dtk functions and randomly selects a large number of cubic sub regions with edge dimension that is just bigger than a typical pore and counts the number of surface and pore voxels. The counts are converted to an effective spherical pore size using $R = 3V/S$. This method does not account for the non-spherical shape of most pores but remains an easily calculated metric. This method does not require the identification of throats
and is insensitive to pore connectivity. The results of both measurements are shown in figure 2. The V/S measurement indicates a greater number of large pores in the 15% compared to the 22% specimens consistent with the breakthrough radii determined by MICP simulation.

More typically, the pore spaces in sandstone contain significant amounts of clay minerals and microporous grains that have been altered by geologic processes. Unlike Fontainebleau, these sandstones have no well-connected macroscopic pore channels. The mineral micropore connectivity plays an important role in fluid capacity and transport but is below the resolution limit of optically coupled XMT. We would like an XMT method that detects the presence of microporosity and an image processing method that allows us to assess the importance of microporous materials on fluid conductance. Two methods are available to image microporosity. In the first, the specimen is imaged at a fixed x-ray energy before and after fluid saturation. The difference image, normalized by the bulk fluid attenuation, provides the fractional volume of fluid in each voxel. The second, a synchrotron method, images the specimen at two energies that cross the absorption edge of a fluid containing an appropriate atom. This permits difference imaging without sample alteration. Normalizing the difference image by the bulk fluid jump ratio provides the voxel fractional fluid volume. For this study, we examined three sandstone samples with similar porosities but different permeabilities that

Figure 2. The pore size distributions measured from the digital volume using the V/S (a) and MICP (b) methods. The V/S distribution measures pore radii consistent with those observed by visualization. MICP radii are significantly smaller and correspond to the volume accessible at a throat radius. The longer critical length observed in the MICP of the 15% porosity samples is consistent with the large diameter pores observed in the V/S data.

Figure 3. Flooded region for $R_{\text{flood}} = 1.72$ pixels. The flood crosses the 3D volume at $R_{\text{flood}} = 1.67$. The pore image has been omitted for clarity.
contain essentially no resolvable pore space. The samples were imaged at fixed energy before and after spontaneous imbibition of 1-bromo-hexadecane. The fluid rapidly wet the specimens when contacted with a pendant droplet. The specimen position was not disturbed during the imbibition and re-registration was not required. Figure 4 shows a 2D slice of sample C dry and a difference image after imbibition.

![Figure 4](image)

Figure 4. Sample C imaged at 18keV and 2 micron per pixel sampling. In the dry image at left, carbonate cements are white and quartz grains are gray. Lower gray levels may be feldspars or clay. The difference image at right is normalized between 0 and 50% porosity. Several microporous regions correlate to regions classified as mineral in the dry image.

Figure 5 shows a 3D rendering of the microporous network. The porosity distribution is obtained by weighting the histogram with the volume fraction for each gray level and is shown in

![Figure 5](image)

Figure 5. Volume rendering of microporosity image obtained subtracting dry image from bromohexadecane flood. It is difficult to assess connectivity by inspection.

![Figure 6](image)

Figure 6. Porosity distribution obtained by weighting histogram of 3D porosity image. Sample B has more ~40% porosity pores than A or C. Sample C has more ~15% porosity pores than A. Sample A porosity is at low volume fraction.
figure 6. It is clear that sample B contains a large number of ~40% microporosity pores compared to A and C and that sample C contains slightly more voxels at ~15% porosity than sample A. The porosity distributions are integrated to obtain the total porosity shown as CT in the figure and are in good agreement with MICP experiment. Unlike total porosity, the trend in pore size seems to correlate well with permeability. The connectivity of the high porosity regions can be quantified using the same gray-scale flooding module used in the MICP simulation. It is important to note that each voxel in the fractional volume image contains only porosity information. At the voxel level, the micropore wettability, pore size and shape distributions, connectivity and permeability are unknown. By gray-scale flooding, we assume that the same linear relationship exists between porosity and permeability for all mineral phases. This may not be true for microporosity created by different chemical or physical processes. The results of the stepped flood are not directly comparable to MICP experiment, but do provide valuable information about the connectivity of microporous regions. Quantitative relationships between microporosity and gray level based on external observation can be introduced as additional data conditioning steps. These include estimates of conductance based on micropore size and shape obtained from electron microscopy and the variation in micropore size and shape[18] for the different mineral phases that can be segmented in the XMT image. These additional considerations import external data and are modeling steps for the purpose of this discussion.

Plots of the flooded volume for each porosity step are shown in figure 7. The samples show significant differences in flood behavior, most notably, the gray level at which the flood crosses the entire volume correlates well with measured permeability. This indicates that the assumption of similar porosity-permeability behavior for different mineralogy is reasonable for these specimens. The last step in the analysis is to determine if the connected porosity pathways correlate with any of the mineral phases present. 2DHistogram shows the probability of finding a voxel of intensity X in image A associated with a voxel of intensity Y in image B. Using this tool we identify the association between the opacity range associated with a mineral in the dry image with a porosity range from the difference image. Figure 8 shows a 2D histogram for the dry and porosity 3D images of sample C. The porosity scale is horizontal from 0% on the left to 100% on the right. The vertical scale is mineral linear attenuation.
increasing from top to bottom. The bright region at the left side shows the expected strong correlation between quartz grain density and zero porosity. The finger rising left to right identifies voxels with increasing porosity and decreasing mineral density relative to quartz grains. The region selected by the white box in the 2D histogram corresponds to the range of feldspar linear attenuation and non-zero porosity. The 2D histogram also points backward into the data to produce a 3D image of those voxels that lie within the selected region and satisfy a logical criterion. In this example we segment the data at breakthrough into only two components. The first is low density minerals with overlapping attenuation such as clays, partially dissolved quartz grains, or chert. The second is a high attenuation phase such as microporous feldspars, i.e. voxels that are of high opacity in the dry image but are also porous. The porosity histograms of the clay and mineral phases shown in figure 9 indicate that the bulk of the fluid lies in the low density phase. Examination of the connectivity of the feldspar and low density components using FloodFill shows that the mineral component is not connected without the low density phase and that the low density phase remains connected if the feldspar is removed. Figure 10 shows the flooded microporosity at breakthrough segmented into feldspars and low density minerals. The feldspars do not form a connected flow path.

CONCLUSIONS

Analytical x-ray microtomography provides a means to relate observable physical properties of materials to their structure. The essential components of an analytical XMT system include instrumentation to acquire high quality volume images, three dimensional image processing tools that probe the volume image and visualization to examine both raw data and the results of image processing operations. A simulation of MICP using a simple image processing toolset reproduces the observable features of the experiment probes the complex pore space. The combination of difference imaging and image analysis of microporous sandstone shows that a linear scaling of microporosity to permeability and an examination of the connectivity of the microporous voxels compares well with measured permeability. Examination of the location of microporous feldspars and low density minerals...
with respect to the maximum porosity path shows that the feldspar contribution to capacity and flow is minimal in this sample. The low density minerals play a dominant role in the permeability of this specimen. Valuable additional information relating lithology to hydrology may be obtained if an experimental method can be devised to allow segmentation of the low density minerals by rock type.

**ACKNOWLEDGEMENTS**

Use of the National Synchrotron Light Source, Brookhaven National Laboratory, was supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, under Contract No. DE-AC02-98CH10886.

**REFERENCES**

[10] IPLab, Scanalytics corp 8550 Lee Highway, Suite 400, Fairfax, VA 22031-1515