New frontiers in experimental geoscience: X-ray microcomputed tomography and fluid flow

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INTRODUCTION
The flow of multiple fluids through the subsurface governs systems such as the flow of water in aquifers, the extraction of oil and gas reserves and the injection of carbon dioxide underground for the purposes of carbon capture and storage. In this article we review some of the recent advances in experimental techniques used to look at these processes, focusing on a single problem, that of capillary trapping for the purposes of carbon capture and storage.

X-ray microcomputed tomography (µCT) has developed over the last 25 years to be the primary method for the non-invasive imaging of rock cores for both experimental and modelling purposes [1-3]. One great advantage of this technique, recognised from the very earliest works on microtomography [4], is that, as it is non-invasive, it is possible to examine “contained systems under conditions of temperature, pressure and environment representative of process conditions”. This is a very attractive advantage for the examination of both single- and multiphase fluid flow through rocks as both the pore structure and fluid properties can be affected by the external conditions [5]. In multiphase flow this is particularly important, as the most important macroscopic parameters for the control of multiphase flow are fundamentally rooted in pore-scale thermo-physical properties, such as contact angle and interfacial tension, which are in turn a strong function of system conditions such as temperature, pressure and salinity, especially for the carbon dioxide–brine system [6-8]. Until recently, however, examining systems under these process conditions has remained a challenge.

Our recent work [9-11] has established a method for the imaging of multiple fluids at conditions representative of subsurface flow while maintaining chemical equilibrium between the fluids and the rock.

MICRO-CT TECHNIQUES
In µCT the sample is rotated relative to a static X-ray source and detector, and X-ray transmission images (radiographs) are collected at multiple discreet angular steps. The set of radiographs is then converted into tomography data using a filtered back-projection algorithm. In traditional laboratory micro-CT architectures resolution has been simply a function of the source spot size and source–sample–detector geometry. This is because the systems relied solely on the geometric magnification of the cone-beam X-rays. This architecture severely limited the size of the equipment and, critically, the size of sample which could be put into the X-ray beam. High resolutions required extremely small samples which could be placed extremely close to the X-ray source.

Modern systems, such as the Versa XRM-500 used in this study (Zeiss X-Ray Microscopy, Pleasanton, CA, USA), incorporate additional optical magnification in the visible spectrum using microscope objectives after the scintillator plate. This allows for high resolutions (at the level of 1 µm) whilst retaining sample sizes of several mm and source-sample and sample-detector spacings of a few cm. This allows enough space for the incorporation of high-pressure high-temperature equipment, allowing for the in-situ imaging of multiple fluid phases at conditions representative of subsurface flow.

The challenge when designing such equipment is how to maintain such extreme conditions in a vessel small enough to fit into the scanner and which is sufficiently X-ray transparent to allow for effective imaging. The solution to this lies in the central section of our core-holder, which is constructed of M55 carbon fibre. This is capable of maximum working pressures of 50 MPa with a vessel wall thickness of just 1 mm, making it virtually transparent to X-rays. The equipment can take cores ranging in size from 4 to 6.5 mm in diameter.

This coreholder is then attached using flexible flow lines to high-pressure flow systems which could be placed extremely close to the X-ray source.

FIGURE 1
Experimental apparatus. (A) The pumps used to control flow and the siting of the flow cell within the µCT enclosure. (B) Detail of the flow cell and heating apparatus and the siting of the core assembly. (C) Detail of the core assembly showing a triple wrap of aluminium around the core to prevent diffusive exchange across the Viton sleeve. (Modified from [11]).
syringe pumps (Teledyne Isco, Lincoln, NE, USA), which maintain and control the flow throughout the system. When examining reactive systems (such as the CO₂–brine–carbonate system) it may be necessary to equilibrate the different phases (solid and liquid) together prior to fluid injection. This is achieved by mixing the different phases together using an entrainment stirrer in a heated reactor (Parr Instruments, IL, USA).

The experimental apparatus is shown in Figure 1.

APPLICATIONS IN CARBON CAPTURE AND STORAGE – CAPILLARY TRAPPING
Carbon capture and storage (CCS) is a process where CO₂ is captured from large point sources and stored in the porous rock formations in the subsurface, principally saline aquifers. Once injected into the subsurface the CO₂ can be immobilised by four principal mechanisms, stratigraphic trapping, solubility trapping, mineral trapping and residual trapping, where the CO₂ is held by surface forces as tiny droplets in the pore space of the rock. This process can occur either naturally, by the migration of the CO₂ plume, or can be induced by the injection of chase brines [12]. The injection of chase brines is simulated in our apparatus by the injection of brines which have been equilibrated both with CO₂ and with the rock into which the CO₂ was injected. This represents conditions of geo-chemical equilibrium present in an aquifer far from the injection site.

Experiments were conducted on five different rock types (two sandstones: Bentheimer and Doddington, and three carbonates: Ketton, Estaillades and Mount Gambier) in order to examine how trapping behaviour varied across pore geometries and rock chemistries (a more detailed description of this study can be found in [11]).

Pore geometry was characterised by extracting network models from the pore-space of dry scans of each rock type by the use of a maximal ball algorithm [13]. In a network model the pore space is represented by an array of perfect shapes, typically triangular prisms. This not only greatly simplifies the flow behaviour for modelling purposes, but also provides descriptive metrics of the pore space, such as the Pore Volume Weighted Connectivity, which are used to compare and understand differences in behaviour between different rock types.

Tomographies were acquired after both drainage (CO₂ injection) and imbibition (brine injection), and reconstructed using the proprietary XMRReconstructor software from a set of 400 radiographs acquired at source acceleration voltages of 80 kV and exposure times of 1.5 seconds. The reconstructed tomography consisted of a volume of roughly 1000³ voxels. The voxel size (6.6 μm) was chosen such that the entire core was in the field of view. For each experiment five individual tomographies were acquired at multiple points along each core and composite volumes were created by stitching together these overlapping sections. This was then cropped to create a cuboidal volume of around 900 x 900 x 3300 voxels. Experiments were repeated for each rock type five times to examine imbibition and once to examine drainage.

After acquisition the images were filtered using a non-local means edge preserving filter (see Buades et al. [14, 15]). The images were then segmented into two phases, with the supercritical (sc) CO₂ being treated as one phase and the brine and rock treated as the other phase. The segmentation of images containing partial saturations of multiple fluids is significantly more difficult than the segmentation of dry images [16], so simple greyscale segmentation was insufficient. Instead a seeded watershed algorithm was used, where the seed was generated by the use of a 2D histogram [17].

This segmented image was then analysed in 3D in order to identify and measure each of the disconnected ganglia. This process is shown in Figure 2.

TRAPPING BEHAVIOUR AND CONNECTIVITY
The saturation (the proportion of the pore-space which is occupied by CO₂) before and after brine injection in each case is shown in Table 1. Large variations in initial and residual saturation are seen as both porosity and the ratio between micro- and macroporosity varies widely across the samples. The overall trapping behaviour can be better characterised by the trapping efficiency, or the ratio of the residual saturation to the initial saturation. This shows very little variation across the different rock types. The ganglia at residual saturation are shown for two example rock types, Mt Gambier and Doddington, in Figure 3.

As well as looking at the bulk trapping behaviour we can look at the distribution of the sizes of the residual ganglia. This is important as it is possible to make predictions of these distributions from the fundamental description of multi-phase flow in porous media, percolation theory. Specifically, the number n of ganglia of a size s should scale as $n \sim s^{-t}$, where t is the Fisher exponent [18]. Its value represents the ratio of large ganglia to small ganglia formed in this system. More small ganglia increases storage security as small ganglia are less easy to mobilise by viscous or gravitational forces and also present a relatively larger surface area for dissolution and reaction. The Fisher exponent should have a value of around 2.189 for cubic lattices in 3D.
[19]. An obvious way to find the value of the Fisher exponent for a set of residual ganglia is to plot the binned quantity, as defined by Dias and Wilkinson [20]

\[ N_s = \sum_{s=2}^{L-1} n_s \]  

(1)

This is shown for the set of rock types in Figure 4.

Our results show power law-like behaviour for large ganglia, but show that small ganglia are significantly underrepresented. This means that the exact nature of local pore filling during the formation of the residual ganglia may not be strictly percolation-like, as co-operative pore filling or piston-like displacement becomes more or less important compared to snap-off [21]. Large ganglia do, however, appear to show approximately power-law behaviour. Fitted Fisher exponents are reported in Table 1 and plotted in Figure 5 against the pore volume weighted connectivity.

**CONCLUSIONS**

These results provide important insights for researchers and policy makers alike, as it shows that for the first time the fundamentals of multiphase flow can be experimentally probed at realistic subsurface flow conditions. The results of the example application discussed
in this article are useful not only as it shows that capillary trapping is a viable trapping mechanism in a wide range of different rock types but also because it shows the link between rock pore structure and trapping behaviour. Capillary trapping for carbon capture and storage, however, represents only the first of many low-hanging fruit that these techniques can address. Imaging and system technology is sufficiently advanced that many of the previously inaccessible properties which control the flow of multiple phases in porous media, such as capillary pressure and contact angle, are now within grasp.

What’s more, recent advances in extremely bright synchrotron light sources have reduced acquisition times by orders of magnitude, allowing for the imaging of the displacement of fluids in real time [22]. This will allow us to look at the processes behind the fluid flow in rocks at conditions representative of subsurface flow with an unprecedented level of detail and precision.


7. Li, X. et al. Interfacial tension of brines + CO$_2$ (0.864 NaCl + 0.136 KCl) at temperatures between 298 and 448 K, pressures between 2 and 50 MPa, and total molalities of 1 to 5 mol/kg. Journal of Chemical and Engineering Data 57(4):1078-1088, 2012.


