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MICROFLUIDIC EXPERIMENT OF CARBONATE PARTICLES DISSOLUTION MONITORED BY SYNCHROTRON LAMINOGRAPHY

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Summary: To improve our understanding of pore scale reactive transport in porous media, microfluidic experiments were performed using Geological Laboratories on Chip. Dissolution of carbonate particles bed was monitored by synchrotron laminography. 3D registration of the resulting 3D images allows quantification of the geometry evolution.

1. INTRODUCTION

CO_2 geological Storage (CGS) is commonly accepted as one of the ways to reduce anthropogenic CO_2 emissions into the atmosphere. It consists in injecting in a deep geological formation a part of the produced CO_2 that has been captured before its emission into the atmosphere. If it is clear that the final goal is to understand CGS at large spatial and time scales, pore scale experiments are relevant to study and understand several crucial transport phenomena that, after averaging and change of scale, will control the CGS behaviour. In this context, the recently developed high-pressure microfluidic tools adapted to CGS in the CGSμLab project offer new opportunities.
This specific presentation is focused on the reactive transport (coupled fluid flows and geochemical reactions) part of the project. In this domain, if numerical models [1] and experimental data [2] are being improved, there is still a large gap: numerical models are not able to handle complex systems like real rocks at a representative scale and experimental data are generally limited due to the complexity of real 3D samples. Our GloCs (Geological Laboratories on Chip) [3, 4] are designed to allow *in situ* micro-characterizations (micro Raman, micro calorimetry, etc.) that can provide new experimental data to validate/invalidate pore scale numerical models. These data are 2D, but microfluidic devices are not 2D systems, they are rather flat-3D systems for which the effects of the third dimension must be evaluated. This is one of the objectives of this work, which aims at investigating the evolution of a 3D reconstructed packed bed of calcium carbonates micro particles within a micro channel.

2. EXPERIMENTAL METHOD

The microfluidic device used for this experiment is presented Fig.1a. The microfluidic network is patterned on a silicon wafer (<100> orientation perpendicular to the surface) by photolithography/wet etching techniques. After cutting the wafer to separate the different networks, injection holes are drilled by sandblasting. Each etched silicon wafer is then anodically bonded to a Pyrex wafer [3] to close the device, and connected to the different inlet/outlet tubing thanks to the homemade stainless steel compression connector visible Fig.1a on the left. A reactive flow experiment consists in: 1) preparing and loading the calcite particles inside the micro-channel, 2) establishing a steady state flow of a solution in equilibrium with the reactive mineral, 3) injecting a calibrated pulse of a reactive solution keeping the overall fluid flow constant thanks to a by-pass device, 4) waiting until a new steady state flow of the non-reactive solution is reached and 5) stopping the flow and closing the system for scanning. Steps 3 to 5 are repeated for each dissolution step. A carbonate particles packing is visible Fig.1a (in white). It is obvious that the horizontal to vertical dimensions ratio is not speaking in favour of classical tomography.
Acquisitions were performed on the laminography setup at the ID19 beamline of the European Synchrotron

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Radiation Facility (ESRF) in Grenoble, France [5]. For each scan, 3600 projections of 2560x2160 pixels were acquired (50 ms exposure time) over 360°. The energy was set to 25 keV, the angle between the sample rotation axis and the beam direction was equal to 62.5 °, and, with the selected optic, the pixel size was 0.649 μm.

3. RESULTS

After reconstruction by filtered back-projection [6], volumes of 2560x2560x300 voxels were obtained. Because the reactive flow experiments were not performed in situ, an accurate registration of the 3D images was necessary to characterize the evolution of the carbonate particles bed. Two properties of the reconstructed images make these registrations delicate: phase contrast, and lack of information about the upper and lower limits (Fig.1c). The procedure we finally applied is the following: Statistically extract the parallel upper and lower planes limiting the network using the intensity changes along the "vertical" direction; Rotate the image to put these planes parallel to (X,Y); Detect the lateral limits using the phase contrast fringes and imposing some geometrical constraints; Use the analytical expressions of these limits to register two consecutive images by a translation and a Z axis rotation. As seen Fig.1c, this procedure allows quantification of the geometry evolution with a fraction of voxel precision.

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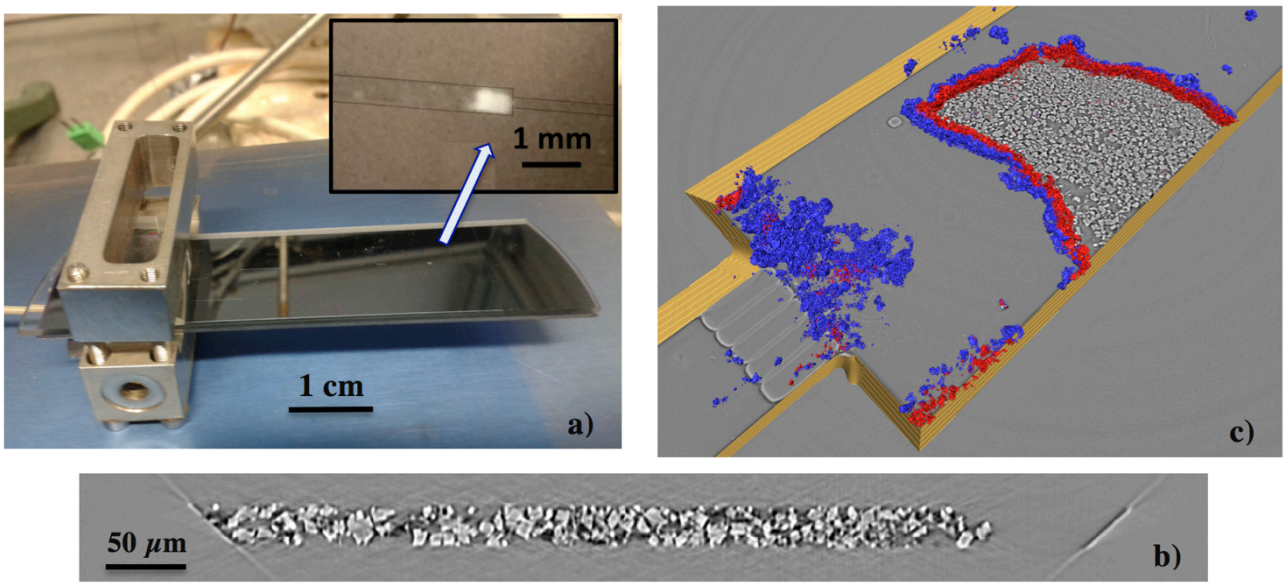

Figure 1: (a) Microfluidic device as used in this study. The carbonate particles bed is visible in white. (b) Vertical section through the 3D reconstruction showing the carbonate particles and the lateral limits of the channel. (c) Horizontal section through the 3D image at stage 20 (grey levels), 3D rendering of the differences between stages 20 and 19 (red) and stages 19 and 18 (blue), 3D rendering of the lateral limits of the microfluidic device (yellow).