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Chemical dissolution of carbonate rocks: A micro-CT study

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The evolution of rock microstructure is a result of complex interactions between the rock surface and reactive fluids and depends on the thermodynamic conditions, the rock and fluid composition and the flow regime [1]. At the pore-scale, the dissolution mechanism consists of mass transport of the reactant by diffusion and advection to the solid surface, chemical reaction at the fluid-mineral interface, and the product mass transport away from the surface [2]. Natural heterogeneity of the porous rocks for the case of rapid dissolution of the mineral by the reactant fluid results in localization of the flow path and the formation of highly conductive channels called wormholes [3]. Several studies have revealed a variety of dissolution patterns from face dissolution to uniform dissolution depending on the injection rate and fluid/mineral properties [4-6].

A number of imaging techniques such as Wood's metal casting, neutron radiography and scanning electron microscopy have been used to study the dissolution patterns. Among them, X-ray micro-computed tomography (micro-CT) is a superior non-destructive imaging method which can create high-resolution images with large field of view [7-10].

In this work, two carbonate core samples were considered for the dissolution experiments. The first sample was an oolitic limestone with multi-modal pore size distribution, while the second one was a wackestone-packstone carbonate with a bimodal pore size. Dissolution experiments were conducted in core plugs of 7 mm diameter using ethylenediaminetetraacetic acid (EDTA) solution at pH 12 as the reactive fluid. The experiments were performed at ambient conditions and were terminated after significant changes occurred in the cores as indicated by the pressure transducers. Details of the experimental procedure were described in [11].

The pre- and post-dissolution dry samples were imaged using a high resolution micro-CT scanner at the Australian National University (ANU) [12] with at least 8.5×8.5×8.5 mm field of view and resolutions of less than 5 μm. The post-dissolution image was superposed to the pre-dissolution image using a 3D registration technique developed by Latham, et al. [13]. In the micro-CT image, the grey-scale values of the pixels correspond to the density and composition of the material imaged. Hence, in monomineralic rock, such as the samples studied in this paper, micro-CT images are 3D porosity maps.

Figures 1 and 2 show examples of registered slices of the oolitic sample from top and side views, respectively. Visual observations of the images clearly illustrate that the reactive fluid locally increased the pore diameters across most parts of the sample and hence led to a quasi-uniform dissolution pattern. This pattern belongs to local non-equilibrium dissolution category [2] in which there is no sharp interface between the reactive fluid zone and the porous medium.

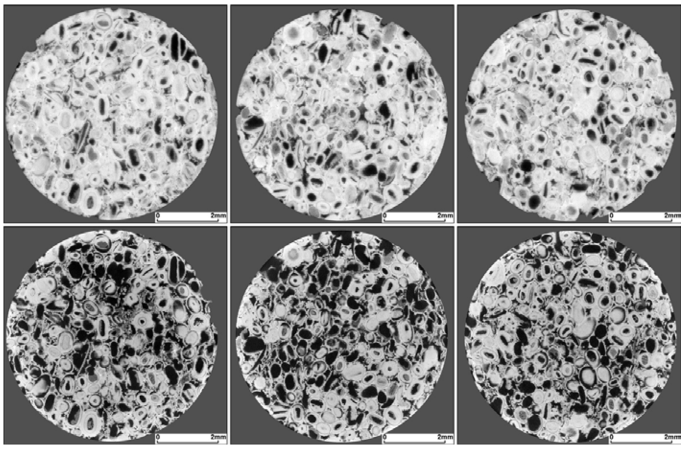

Figure 1. (Top) pre-dissolution micro-CT slices of the oolitic sample; (Bottom) the corresponding registered post-dissolution images. The slices are at different distances to the front (injection face) of the samples: (left) 6.42 mm, (middle) 9.50 mm, and (right) 11.55 mm.

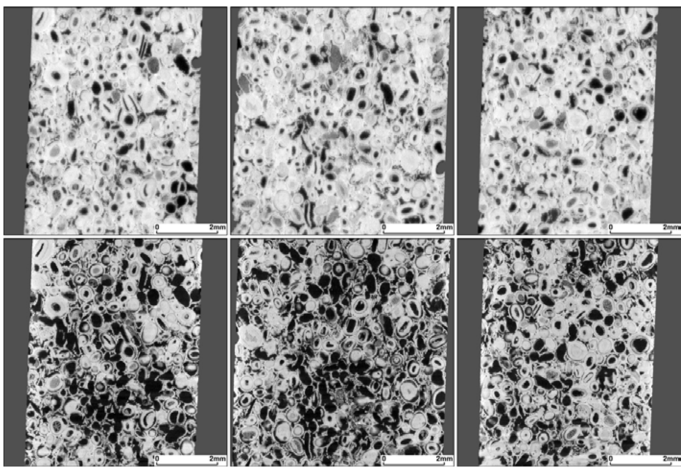

Figure 2. Side-view registered slices of the oolitic sample in (top) pre-dissolution and (bottom) post-dissolution states, at different positions.

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Figures 3 and 4 illustrate examples of registered slices of the wackestone-packstone sample from top and side views, respectively. In contrary to the observation on the oolitic sample, visual observations of the images of the second sample reveal a different dissolution pattern which is called conical wormhole dissolution pattern. In this pattern, the reactive fluid was consumed over small parts of the mineral surface area leading to the formation of a few highly conductive flow channels, i.e. wormholes. The conical wormhole corresponds to the local-equilibrium dissolution category [2] in which there is a sharp interface between the fluid zone and the medium as indicated in Figures 3 and 4.

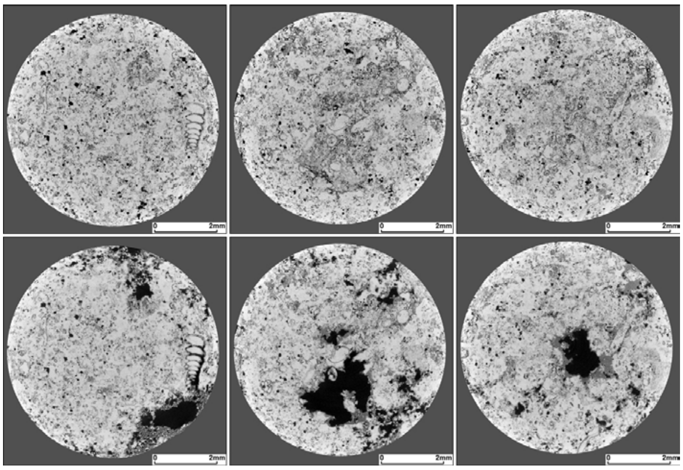

Figure 3. (Top) pre-dissolution micro-CT slices of the wackestone-packstone sample; (Bottom) the corresponding registered post-dissolution images. The slices are at different distances to the injection face of the samples: (left) 5.84 mm, (middle) 7.98 mm, and (right) 10.11 mm.

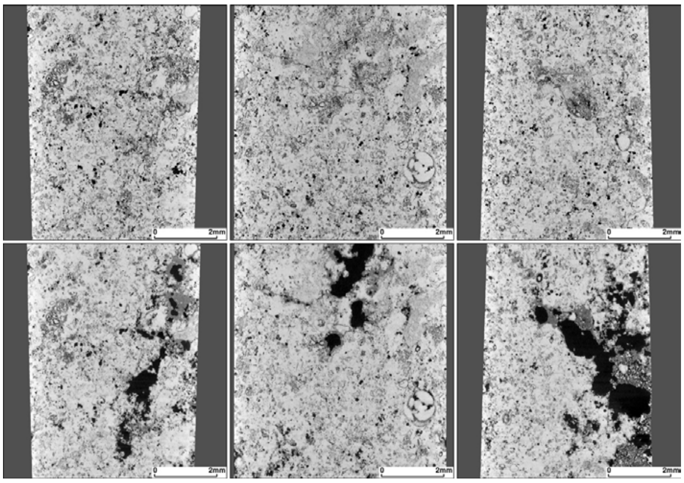

Figure 4. Side-view registered slices of the wackestone-packstone sample in (top) pre-dissolution and (bottom) post-dissolution states, at different positions.

As found in this work, the micro-CT technique can provide high resolution images of successively disturbed specimens. This, in turn provides an important basis to calculate the evolution of rock micro- and macro-scale properties using advanced numerical techniques at the pore scale. In addition, micro-CT is a useful method to study pore scale reactive displacement mechanisms and their effects on the evolution of rock properties.

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