



# 4D multi-scale imaging of reactive flow in carbonates: Assessing the impact of heterogeneity on dissolution regimes using streamlines at multiple length scales

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## ABSTRACT

We have experimentally investigated the impact of heterogeneity on the dissolution of two limestones, characterised by distinct degrees of flow heterogeneity at both the pore and core scales. The two rocks were reacted with reservoir-condition CO<sub>2</sub>-saturated brine at both scales and scanned dynamically during dissolution. First, 1 cm long 4 mm diameter cores were scanned during reactive flow with a 4 μm voxel size between 10 and 71 times using 4D X-ray micro-tomography (μ-CT) over the course of 90 min. Second, 3.8 cm diameter, 8 cm long cores were reacted at the same conditions inside a reservoir-condition flow apparatus and imaged using a medical-grade X-ray computed tomography scanner (XCT). Each sample was imaged ~13 times over the course of 90 min at a 250 × 250 × 500 μm resolution. These larger cores were then scanned inside a μ-CT at a 27 μm voxel size to assess the alteration pore-space heterogeneity after reaction. Both rock types exhibited channel widening at the *mm* scale and progressive high porosity pathway dissolution at the *cm* scale. In the more heterogeneous rock, dissolution was more focussed and progressed along the direction of flow. Additionally, the dissolution pathways contained a distinct microstructure captured with the μ-CT that was not visible at the resolution of the XCT, where the reactive fluid had not completely dissolved the internal pore-structure. This microstructure was further analyzed by performing a direct simulation of the flow field and streamline tracing on the image voxels.

We found that at the larger scales the interplay between flow and reaction significantly affects flow in the unreacted regions of the core. When flow is focussed in large reacted channels, this focusing is carried through to the unreacted parts of the rock where flow continues to be confined to preferential pathways after passing the reaction front. This focussing effect is greater with increasing pore space heterogeneity indicating that the representative elementary volume (REV) for dissolution is far greater than the dissolution front itself. This study of scale dependence using *in situ* 4D tomography provides insight into the mechanisms that control local reaction rates at the *mm* and *cm* scales. Furthermore, this work suggests that under these conditions at larger scales it is likely to be structural heterogeneity that dominates the pattern of dissolution and therefore the evolution of high permeability pathways.

## 1. Introduction

A major concern in the implementation of carbon capture and storage (CCS) is long-term storage security (Herzog et al., 2003; Metz et al., 2005). Carbon dioxide (CO<sub>2</sub>) injected into the subsurface will dissolve in the resident brine and form carbonic acid (Langmuir et al., 1997; Morse and Mackenzie, 1990; Peng et al., 2013). Carbonate host rocks have the potential to react with and be dissolved by CO<sub>2</sub> acidified brine (Peng et al., 2015). Dissolution of the host rock is commonly

associated with changes in fluid flow patterns and can weaken carbonate cements and damage the formation around injection wells (Birkholzer et al., 2011). Therefore, it is important to have a holistic understanding of dissolution in the brine-rock system to predict the distribution and the rate of fluid movement, as well as the amount and impact of dissolution in reservoir rocks (Daccord et al., 1993a, 1993b; Maheshwari et al., 2013).

However, the nature and rate of dissolution in carbonates is dependent on both the chemical properties of the brine (El-Maghraby

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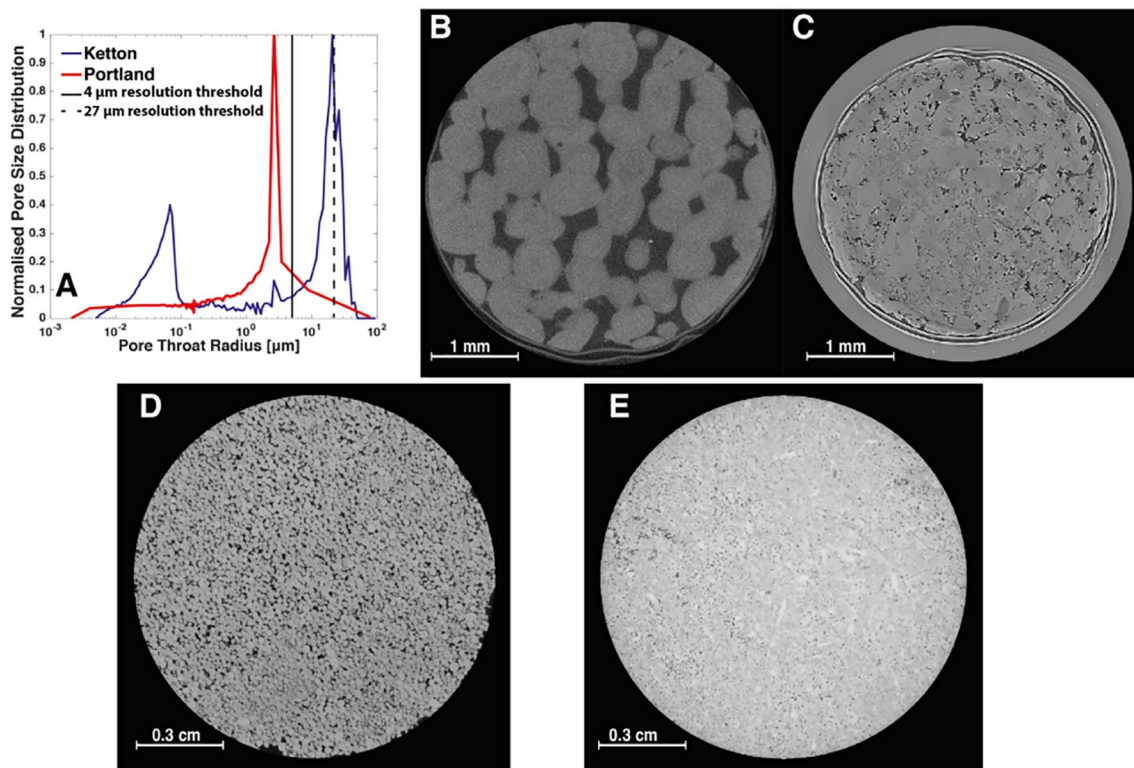


Fig. 1. The pore throat size distribution of Ketton and Portland (A) derived from mercury injection tests by Weatherford labs, UK. 2-D slices of 3D  $\mu$ -CT images of Ketton (B), Portland (C) at 4  $\mu$ m resolution, and Ketton (D), and Portland (E) at 27  $\mu$ m resolution.

et al., 2012; Fredd and Fogler, 1998; Luquot and Gouze, 2009) and the geometrical properties of the pore-space of the host rock (Li et al., 2006; Rötting et al., 2015). Carbonate dissolution rates are also strongly dependent on brine temperature and pressure (Peng et al., 2015), making it necessary to develop experimental techniques to measure complex time-dependent processes at representative reservoir conditions. All combined, physical and chemical heterogeneities have been shown to be the cause of different dissolution regimes (Fredd and Fogler, 1999, 1998) including wormholing, channelling, uniform, and face dissolution.

Reactive transport in porous media is commonly investigated using core-flooding techniques coupled with three-dimensional imaging to track the reaction at the fluid/solid boundary. One accepted imaging technique is medical-grade computed tomography scanners (XCT), which can operate at different scales. XCT scanners (Mettler Jr et al., 2000) are commonly paired with core-flooding to image rocks at the core ( $\sim$ cm) scale. Core-scale imaging is valuable because it is three-dimensional, and depending on the experimental conditions, can be temporally resolved. However, it has a maximum resolution of  $\sim$ 100  $\mu$ m and thus can only resolve the fluid/solid boundary of large features, such as fractures and vugs. The greater portion of the pore space is only resolved as an average greyscale value because several pores or parts of pores will be contained in a single voxel, thus making segmentation and analysis of local changes in connectivity and reaction rate very difficult. X-ray microtomography ( $\mu$ -CT) (Ritman, 2004) provides higher resolution, and although it is possible to image cm-scale samples, the time required for imaging increases with increasing sample size. Thus,  $\mu$ -CT is ideal for mm-scale samples when studying dynamic  $\mu$ m-scale processes on short time-scales ( $\sim$ 1 h). However, due to the small sample size  $\mu$ -CT does not always capture the full range of physical heterogeneities seen at the core scale and may not capture the representative elementary volume (REV) for heterogeneous dissolution processes.

Imaging of dissolution in limestone rock has been performed at the

core ( $\sim$ cm) scale using XCT. Ott et al. (2012) investigated dissolution using core flooding and XCT where, for several rock types and flow regimes, it was found that reaction of the dissolved  $\text{CO}_2$  with the rock matrix increased physical heterogeneity. Ellis et al. (2011) used imaging to show that exposure to  $\text{CO}_2$  progressively reduced the sealing capacity of fractured claystone caprocks. Smith et al. (2012) imaged dissolution of a dolomitic core before and after reaction with  $\text{CO}_2$  acidified brine and found dissolution rates  $17\times$  slower than in calcite. Lamy-Chappuis et al. (2014) used CT scanning to measure calcite dissolution in calcareous gritstone and found that the permeability increase with porosity did not agree with classical models (e.g. Kozeny-Carman) and that the dissolution rate was impacted largely by the acid supply. Tutolo et al. (2014) examined dolomite dissolution in the presence of  $\text{CO}_2$  saturated brine and found evidence of dissolution at low temperatures followed by exsolution and precipitation as the temperature increased. Ott and Oedai (2015) examined carbonate dissolution in the presence of two-phase flow and found large differences in dissolution regimes depending on the  $\text{CO}_2$  saturation.

There are relatively few experiments that investigate the impact of coupled dissolution and flow phenomena at the micron scale ( $\sim$  $\mu$ m) – the observation scale that is necessary for pore-scale analysis and numerical simulations of both flow and transport. Noiriél et al. (2013) used  $\mu$ -CT to measure changes in a limestone fracture at several locations during acid dissolution at ambient conditions. Additionally, Noiriél et al. (2004) reported a power law relationship between porosity and permeability during limestone dissolution by scanning the rock periodically *ex situ*. Menke et al. (2015) imaged the dissolution dynamics of a relatively homogeneous carbonate at *in situ* reservoir conditions in the advective flow regime and low brine acidity (pH 3.1) while Menke et al. (2016) used fast synchrotron tomography to image dissolution in two heterogeneous carbonates at two flow rates and identical brine conditions. They found that the porosity can either increase uniformly through time along the length of the samples, or may exhibit a spatially and temporally varying increase that is attributed to

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