



Reservoir condition imaging of reactive transport in heterogeneous carbonates using fast synchrotron tomography – Effect of initial pore structure and flow conditions



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ABSTRACT

We investigate the impact of initial pore structure and velocity field heterogeneity on the dynamics of fluid/solid reaction at high Péclet numbers (fast flow) and low Damköhler number (relatively slow reaction rates). The Diamond Lightsource Pink Beam was used to image dissolution of Estailades and Portland limestones in the presence of CO₂-saturated brine at reservoir conditions (10 MPa and 50 °C representing ~1 km aquifer depth) at two flow rates for a period of 2 h. Each sample was scanned between 51 and 94 times at 4.76- μ m resolution and the dynamic changes in porosity, permeability, and reaction rate were examined using image analysis and flow modelling.

We find 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 channel formation, a process that is distinct from wormholing, depending on initial pore structure and flow conditions. The dissolution regime was structure-dependent: Estailades with a higher porosity showed more uniform dissolution, while the lower porosity Portland experienced channel formation. The effective reaction rates were up to two orders of magnitude lower than those measured on a flat substrate with no transport limitations, indicating that the transport of reactant and product is severely hampered away from fast flow channels.

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1. Introduction

A major concern of carbon capture and storage (CCS) is long-term storage security (Herzog et al., 2003; Metz et al., 2005). Carbon dioxide, CO₂, injected into the subsurface will dissolve in the host 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 CO₂ acidified brine (Peng et al., 2015). Dissolution of the host rock may instigate unpredictable fluid flow and can weaken carbonate cements and damage injection wells (Birkholzer et al., 2011; Mohamed and Nasr-El-Din, 2012). A complete understanding of dissolution in the brine-rock system is therefore important to predict the distribution and the rate of fluid movement and the amount and impact of dissolution in the subsurface (Daccord et al., 1993a, Daccord et al., 1993b; Maheshwari et al., 2013).

However, the nature and the rate of dissolution in carbonates are dependent on both the properties of the brine (El-Maghraby et al., 2012; Fredd and Fogler, 1998; Luquot and Gouze, 2009) and 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 or measuring complex time-dependent processes at representative reservoir conditions.

Previous experiments have observed that field-scale reaction rates are typically orders of magnitude lower than experimental batch reactor measurements (Swoboda-Colberg and Drever, 1993). Mineral heterogeneity and incomplete mixing in a heterogeneous flow field are potential explanations for this phenomenon. However, it is not possible to assess the most significant factors without directly observing the evolution of the pore space during reaction. Thus, dynamic pore-scale experiments are required to provide both insights into the interplay between transport and reaction, and to validate predictive models. Traditionally, dissolution has been studied in larger (cm-scale) core floods, or in idealized two-dimensional systems (Fredd and Fogler, 1998; Marini, 2007; Ott et al., 2012). However, these experiments are unable to determine the mechanisms of dissolution within the pore space in realistic rock samples.

X-ray microtomography (μ -CT) (Berg et al., 2013; Blunt et al., 2013; Gouze and Luquot, 2011; Wildenschild and Sheppard, 2013) is an established experimental method for studying pore-scale processes in

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CCS applications. μ -CT has several benefits: it achieves high spatial resolutions of down to around 1 μm , it is non-invasive, and it provides three-dimensional images. To advance our understanding of how different transport and reaction conditions alter complex solid and pore structures, it is necessary to measure reaction-induced changes in pore-space geometry, topology and flow in subsurface rock systems at reservoir temperatures and pressures. High resolution is required to investigate pore-scale processes in detail.

This paper describes a method of studying dynamic reactive dissolution processes in rock with complex pore structures with a focus on measuring the time and spatially dependent reaction rate between a CO_2 -acidified brine and limestone rock at reservoir conditions. We observe a distinct dissolution regime, channelling, whose emergence is dependent on initial pore structure and flow conditions.

Several studies have investigated reaction in complex carbonates at the pore scale (Gharbi, 2014; Gharbi et al., 2013; Hao et al., 2013; Noiri et al., 2013; Smith et al., 2013), but due to experimental or imaging constraints they have been either limited to pre- and post-reaction images, or were not completed at representative subsurface conditions. Menke et al. (2015) performed dynamic *in situ* imaging of reaction between a CO_2 -acidified brine and Ketton limestone at the pore scale over a period of several hours and at a temperature and pressure representative of an aquifer at approximately 1 km depth. However, Ketton is a relatively homogenous carbonate rock that is easy to image in very little time (~17 min) with traditional μ -CT methods. Many carbonate rocks have complex pore structures that require more detail to accurately resolve, which can be a very time intensive process using traditional μ -CT (~hours), either with a monochromatic beam at a synchrotron source, or with bench-top x-ray scanners. Thus, a fast method of tomography is required to see dynamic reaction-induced changes in heterogeneous carbonates.

Fast synchrotron tomography has been previously used by Berg et al. (2013) to study image Haines jumps in two-phase flow, and by Andrew et al. (2015b) to study the dynamic capillary pressure changes during a reservoir condition CO_2 -brine drainage on the pore scale. We present a method that uses fast synchrotron tomography to study the effect of initial pore structure and flow conditions on the dynamics of fluid/solid reaction in two highly heterogeneous rocks at reservoir conditions. The dynamic image sequence is first analysed for porosity changes and then used as inputs into flow simulations that can elucidate changes in permeability and pore-scale velocity fields. Additionally, effective reaction rates are calculated and the reactive flow dynamics is compared in terms of the initial structure and flow conditions, demonstrating both uniform and channelled dissolution structures.

2. Methods

The photon flux and energy of the x-ray source control the image acquisition time of the sample. The use of the polychromatic beam of a synchrotron source is one method of scanning quickly (Andrew, 2015; Andrew et al., 2015b; Berg et al., 2013). This so-called 'Pink Beam' provides orders of magnitude more intense light than bench-top sources and images can therefore be taken on the tens-of-second rather than hour time scale. Using this intense x-ray flux, however, is not without its challenges. The lower energy x-rays of the Pink Beam spectrum are absorbed by the sample as heat. This can interfere with the temperature control of the *in situ* apparatus and cause CO_2 to exsolve from solution. CO_2 -saturated brine is very sensitive to both heat and pressure and therefore a small change in thermal equilibrium can significantly change the pH of the *in situ* fluid (Peng et al., 2013). Thus, careful design and control elements for the x-ray spectra must be incorporated into the beam line equipment prior to imaging. Mirrors and filters are thus used to narrow the spectrum of light to suit experimental needs and absorb unwanted x-ray spectra. Mirrors absorb the high-energy spectrum while filters absorb the lower energies. It is therefore possible to narrow the spectrum to the desired band of radiation using only these tools. The

energy of the Diamond Light Source I13-2 Pink Beam ranges from 8 to 30 keV with a maximum photon flux of 4×10^9 Ph/s. The low energy x-rays were filtered by passing the beam through 2 mm of Al and 0.1 mm of Au. A 250 μm -thick CdWO_4 scintillator with a $1.25 \times$ objective lens and a PCO EDGE camera were used in the detector assembly (Andrew et al., 2015b).

Tomographic scans were taken successively for 2 h at the centre point of the core with a 5 mm by 4 mm field of view. The dynamic scans had 1000 projections each with an exposure time of 0.04 s. Total acquisition time was ~45 s per scan with between 51 and 94 scans taken over a period of 2 h. The time between scans was controlled by the data read-off speed of the camera, which varied between experiments.

2.1. The experimental apparatus

Estailades and Portland Based carbonates were reacted using our *in situ* experimental apparatus [Fig. 1]. Reaction was imaged between calcite and unbuffered scCO_2 saturated brine in 4 mm-diameter 1.2 cm-long carbonate cores. Carbonate rocks generally have a wide

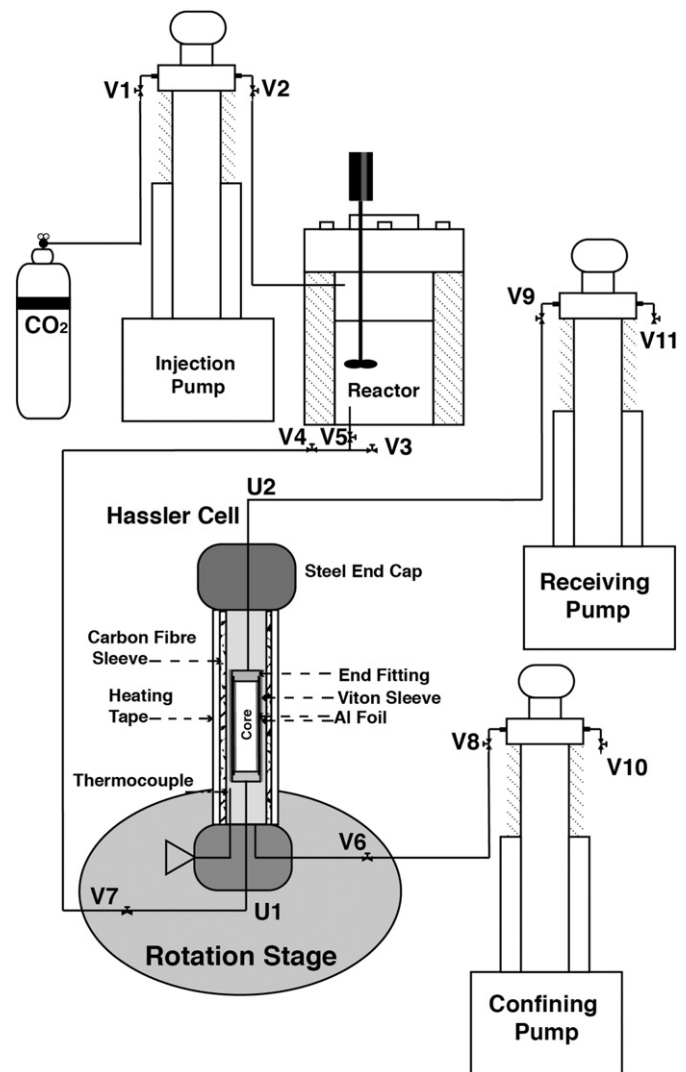


Fig. 1. The *in situ* experimental apparatus. CO_2 is pressurized by the injection pump and used to equilibrate brine in the reactor. Reactive brine is pulled through core assembly by the receiving pump. The cell is confined by deionized water in the confining pump at 12 MPa and heated using heating tape controlled by a thermocouple in the confining fluid. The experimental system is connected together using tubing and fluid flow is directed using Valves (V) and Unions (U). Modified from Menke et al. (2015).

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