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Characterization of reactive flow-induced evolution of carbonate rocks using digital core analysis - part 2: Calculation of the evolution of percolation and transport properties

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A B S T R A C T

Percolation of reactive fluids in carbonate rocks affects the rock microstructure and hence changes the rock macroscopic properties. In Part 1 paper, we examined the voxel-wise evolution of microstructure of the rock in terms of mineral dissolution/detachment, mineral deposition, and unchanged regions. In the present work, we investigate the relationships between changes in two characteristic transport properties, i.e. permeability and electrical conductivity and two critical parameters of the pore phase, i.e. the fraction of the pore space connecting the inlet and outlet faces of the core sample and the critical pore-throat diameter. We calculate the aforementioned properties on the images of the sample, wherein a homogeneous modification of pore structure occurred in order to ensure the representativeness of the calculated transport properties at the core scale. From images, the evolution of pore connectivity and the potential role of micropores on the connectivity are quantified. It is found that the changing permeability and electrical conductivity distributions along the core length are generally in good agreement with the longitudinal evolution of macro-connected macroporosity and the critical pore-throat diameter. We incorporate microporosity into critical length and permeability calculations and show how microporosity locally plays a role in permeability. It is shown that the Katz-Thompson model reasonably predicts the post-alteration permeability in terms of pre-alteration simulated parameters. This suggests that the evolution of permeability and electrical conductivity of the studied complex carbonate core are controlled by the changes in the macro-connected macroporosity as well as the smallest pore-throats between the connected macropores.

1. Introduction

A wide variety of geochemical processes including geological storage of CO_2, contaminant transport in groundwater resources and acidizing of petroleum reservoirs involve reactive transport in the subsurface. In such systems, chemical change is driven by the interactions between migrating fluids and solid phases. The evolution of these complex systems involves a variety of processes including mass transport and heterogeneous chemical reactions occurring at the disordered pore-solid surface (Lichtner et al., 1996). The geometry and connectivity of the pore space may evolve due to the interactions between reactive fluids and mineral solid. This may, in turn, induce changes in physical properties of porous rocks. Among a wide variety of natural porous systems, carbonate sediments are of great importance since they comprise > 50% of the world's petroleum reservoirs and almost all deep saline aquifers (Arns et al., 2005b; Gouze and Luquot, 2011). In petroleum industry, porosity, permeability and their distributions play a major role in the productive capacity of the reservoir, predicting reservoir performance, and determining well production rates. It is well known that reactive flow plays an important role during the life cycle of a reservoir, since dissolution/deposition and fines mobilization and clogging have a direct influence on porosity, permeability and their distributions, and hence the recovery from the reservoir. The characterization of the complex interplay between the pore microgeometry, connectivity, and transport properties of carbonate rocks is an intriguing but extremely challenging research field. This is because most carbonate rocks exhibit complex pore structure comprising a variety of pore types with irregular spatial distributions and a wide spectrum of pore sizes from a few nanometres to several millimetres (Cantrell and Hagerty, 1999). The broad nature of most carbonate rock structures has limited accurate determination of the relation between rock microstructure, pore geometry and flow properties which is of vital

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importance in modelling of reactive flow and transport in carbonate formations.

Over the past two decades, digital core analysis or more widely as digital rock physics (DRP) is increasingly being used to compute the petrophysical and transport properties of porous materials via tomographically recorded 3D media (Spanne et al., 1994; Arns et al., 2003; Sakellariou et al., 2004; Arns et al., 2005a; Youssef et al., 2007; Knackstedt et al., 2009; Derzhi et al., 2010; Grader et al., 2010; Knackstedt et al., 2011; Amabeoku et al., 2013; Sungkorn et al., 2014; Guibert et al., 2015; Alyafei et al., 2016; Sun et al., 2016). In this technique, high resolution images of the material specimen are acquired at micro-metre scale and processed to create accurate 3D digital data. The material properties are then computed by advanced numerical techniques at the pore scale.

A number of pore-scale imaging techniques such as optical light microscopy (Ehrlich et al., 1984), scanning electron microscopy (SEM) (Bekri et al., 2000), transmission electron microscopy (TEM) (Timur et al., 1971), atomic force microscopy (AFM) (Krekeler et al., 2005) and 2D nuclear magnetic resonance (NMR) spectroscopy (Kessler et al., 1988) have been used to describe pore and grain at extremely high resolution, down to a few nanometres. These methods, however, can only provide 2D representations of complex materials. Recent progresses in 3D imaging methods, such as conventional X-ray computed tomography (CT) (Ketcham and Carlson, 2001), X-ray micro-computed tomography (μ-CT) (Sakellariou et al., 2003), neutron tomography (De Beer and Radebe, 2012), electron tomography (Weyland et al., 2001) a variety of ultrasonic methods (Shen et al., 2012), photoacoustic tomography (PAT) (Cai et al., 2013), magnetic resonance imaging (MRI) (Sarkar et al., 1992), focused ion beam scanning electron microscopy (FIB-SEM) (Kubis et al., 2004) and laser scanning conformal microscopy (Fredrich et al., 1995) make it possible to directly visualize and analyse the 3D microstructure of porous materials from low to high resolution.

Particularly, X-ray μ-CT has proved itself as a distinctive method for non-destructive imaging of complex material microstructure in recent years (Coles et al., 1994; Spanne et al., 1994; Lindquist, 2002; Arns et al., 2005b; Sheppard et al., 2006; Knackstedt et al., 2007; Youssef et al., 2007; Knackstedt et al., 2011; Georgiadis et al., 2013; Al-Yaseri et al., 2015; Oughanem et al., 2015; Shah et al., 2015; Peche et al., 2016). The major advantage of μ-CT technique over conventional medical CT is to create high-resolution images with large field of view (FOV) which allows one to compute macroscopic properties, representative of the main core plug. The capability of imaging and characterizing pore structure in 3D, coupled with the ability to numerical computation of macroscopic properties directly on image data allow a systematic study of the role of pore structure and connectivity on transport properties and their alteration during a variety of processes, particularly reactive flow-induced structural evolution. A number of comprehensive review articles have been published detailing theoretical and technical aspects of X-ray μ-CT technique and the relevant applications in digital rock physics (Denison et al., 1997; Ketcham and Carlson, 2001; Stock, 2008; Cnudde and Boone, 2013; Wildenschild and Sheppard, 2013).

Regarding carbonate rocks, X-ray μ-CT has proven to be a valuable tool for characterization of their complex structure at high resolution (Arns et al., 2005b; Knackstedt et al., 2006a; Ghous et al., 2007; Knackstedt et al., 2007; Youssef et al., 2007; Sok et al., 2010; Garing et al., 2014; Hebert et al., 2014; Saenger et al., 2016). Recent progress has extended the application of μ-CT to characterize the impact of reactive flow on physical properties of carbonate rocks. Most studies have focused on porosity, permeability and their distribution as well as the closely related microstructural descriptors such as pore connectivity and pore size distribution. Bernard (2005) examined the structural modifications induced by reactive percolation of CO_2-saturated water within a natural limestone sample. He computed the sample permeability along the core axis at different stages of the reactive percolation using a solution of the closure problem associated with the volume averaging of Stokes equations and compared with the experimental results obtained by Noiriel et al. (2005). Even though, the employed method yielded results in the same order-of-magnitude range as the permeability values measured experimentally, it is computationally demanding. Cai et al. (2009) studied changes in pore structure of a special natural sediment subject to a simulated caustic tank leachate. From the acquired images, they quantified changes in coordination numbers, connectivity and tortuosity in pre- and post-alteration states. Since their results are limited to a small sub-dataset, the REV requirements for the reactive flow effects are doubtful. The evolution of porosity, permeability and elastic properties during carbon dioxide flooding of a limestone sample was studied by Nur et al. (2011) using high resolution CT-scanning before and after the CO_2 injection. They showed that the permeability changes did not conform to the porosity changes due to displacing of fine particles. Yet, they did not give any evidence for fines migration. Hao et al. (2013) studied numerically CO_2-induced dissolution of carbonate cores of varying heterogeneity to evaluate the evolution of pore space connectivity and permeability in various wormhole-like dissolution regimes. From images, they determined spatial and temporal development of wormhole within the samples after the reactive-flow experiments and examined the impacts of heterogeneity on the initiation and development of dissolution front. They also employed a simple power-law equation to relate local permeability change to porosity and discussed the power law exponent for different carbonate rocks affected by heterogeneous dissolution. Qajar et al. (2013) have used high resolution images for quantification of porosity changes in terms of pore growth/clogging and fines migration during mineral dissolution. They found an initial decrease in permeability due to particle mobilization and clogging at early stage of the dissolution experiment. The changes in pore structure, absolute and relative permeability resulting from carbonate precipitation during CO_2 storage were numerically studied by Jiang and Tsuji (2014). They used 3D images of a Berea sandstone core at a resolution of 3.2 μm in numerical simulation of reactive transport. They concluded that pore clogging in the nonwetting phase has the major influence on relative permeability reduction. The heterogeneous dissolution of a carbonate core plug induced by percolation of CO_2-rich fluids was studied by Vialle et al. (2014). They used X-ray CT imaging and showed that the experimental porosity-permeability relationship is in consistent with the dissolution regime revealed by the CT images. Luquot et al. (2014) examined the evolution of various types of porosity and transport properties during CO_2-rich brine injection into four oolitic limestones. They concluded from the μ-CT pre- and post-dissolution images that the initial disconnected macropore phase became connected for one of the samples while for other samples the macroporosity remained connected by the micropore phase after the dissolution experiments. They also calculated the effective diffusion coefficient on the segmented images by solving the Laplace equation. It was found that the effective diffusion coefficient increases after the dissolution experiments. In a similar work, Garing et al. (2015) studied anti-correlated porosity-permeability changes in the course of CO_2-rich deionized water injection. A clear particle detachment/dissolution, displacement and deposition were observed on the micro-tomographic images by comparing the images in pre- and post-dissolution states. They also proposed simple phenomenological models to address the impact of the driving mechanisms on the porosity-permeability correlations. In a recent paper, Luquot et al. (2016) investigated changes in total and effective porosity, pore connectivity, pore size distribution and diffusion coefficient induced by reactive flow-through experiment in a limestone sample using an acidic solution other than CO_2-rich brine. They compared the evolution of the aforementioned parameters computed by X-ray μ-CT images and laboratory experiments and obtained good agreement between them. The authors argued that despite high dependence of image-based computed parameters on the voxel size particularly for fluid-rock interface, the μ-CT technique yields similar results for the micro- and macro-scale rock properties as the laboratory experiment in much shorter time and in a