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A test of the effectiveness of pore scale fluid flow simulations and constitutive equations for modelling the effects of mineral dissolution on rock permeability

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ABSTRACT

Macro-scale transport properties of rocks such as permeability are bulk parameters combining the contributions of various properties only properly defined at the pore scale. Since pore-scale processes are known to modify the rock properties it is legitimate to ask if constitutive equations based on macro scale properties (e.g. porosity, permeability, formation factor, etc…) can properly describe their effect. In a previous experimental study (Lamy-Chappuis et al., 2014) we found that the effect of mineral dissolution on permeability could be much higher than predicted by such semi-empirical constitutive equations. Here we directly solve pore-scale fluid flow in high-resolution (2.5 μm) 3D models of a rock's geometry before and after mineral dissolution in order to evaluate permeability and how it is changed. This methodology is limited by the resolution of the micro-CT images used to define the rock geometry, which leads to significant overestimates of absolute permeability, but it does produce a much closer match to the change in permeability due to mineral dissolution than the constitutive equations. This is possible because the dissolution features, which enhance permeability, are large enough to be adequately resolved and produce a significant change in permeability.

1. Introduction

Prediction of the effect of pore scale modifications on permeability can be achieved with simple semi-empirical equations calibrated with experimental results, for instance the Kozeny-Carman (K-C) equations (Kozeny, 1927; Carman, 1937) have been used extensively in the past to correlate variations of permeability to those of porosity. To overcome the limitations of constitutive equations and to gain quantitative insights into the underlying mechanisms leading to changes in permeability, there has been a move towards the evaluation of transport properties by fluid flow simulation at the pore scale over the past 20 years.

Developments of pore scale simulation methods have accompanied recent progress in X-ray CT imaging of material structures (Blunt et al., 2013; Wildenschild and Sheppard, 2013). Pioneering studies focused on small models only a few pores wide obtained at resolutions of tens of microns. Initial studies were meant to serve as a proof that pore scale flow modelling in reconstructed real porous media was possible but already several authors claimed to have successfully recovered Darcy's permeability from CT images (Ferreol and Rothman, 1995; Chen and Doolen, 1998; Coles et al., 1998; Adler, 2013). As for two-phase flow simulations, they were at least qualitatively successful as the predicted general features were observed (Ferreol and Rothman, 1995; Chen and Doolen, 1998; Házi et al., 2002).

Modern CT tools can provide time series of images obtained at micron or even sub-micron resolutions, improving the quantification of properties such as reactive surface area and tortuosity (Noiriel et al., 2009), and allowing more detailed knowledge of transient rock modification. It is possible to distinguish individual constituents of the rock (Tsuchiyama et al., 2000, 2005, 2013) as well as the various fluid phases present in the pores (Gao et al., 2017). A logical continuation in this research field is therefore to study the effect of transient pore space modifications triggered by fluid-rock interactions. In this regard, many experimental studies using CT images as monitoring tools have focused on carbonate rocks dissolution (Gouze and Luquot, 2011; Gharbi et al., 2013; Noiriel et al., 2013; Al-Khulaifi et al., 2017; Menke et al., 2016), they linked variable modes of pore space dissolution (fracture dissolution, wormholing, channeling, homogeneous dissolution) with variable pore space/reactive flow properties. For instance, Nogues et al. (2013) simulated such experiments and found that the permeability could scale

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with the porosity to the power 4 to 10 depending on rock intrinsic properties (heterogeneity, connectivity and anisotropy) and on the Damkhöler and Peclet numbers of the reactive flow.

Concerning fluid flow simulations, the current production of high-resolution pore space models constitute an important step towards the quantitative interpretation of porosity-permeability relationships in rocks, but the limited computing power still forces a trade-off between resolution and model size/representativity. One way to circumvent this is to simplify the model geometry by extracting so-called "pore network" models (Dong and Blunt, 2009; Nogues et al., 2013) or to simplify fluid flow computations by adopting a pore scale-continuum approach, thereby assigning a Darcy permeability to micron-size rock subvolumes (Hao et al., 2013). Contemporary studies using direct simulation of fluid flow in accurate pore space models are scarce and deal with model sizes not larger than a few millimeters. In the latter, fluid flow is usually obtained by solving the discretized Navier Stokes equation (Adler, 2013; Bijeljic et al., 2013a, 2013b) or by using the Lattice-Boltzmann (LB) method (Ferreol and Rothman, 1995; Kang et al., 2002; Zu and He, 2013; Xie et al., 2017).

In the work presented in Lamy-Chappuis et al. (2014), experiments were conducted to study CO_2-fluid-rock interactions effects on the permeability of sandstone cores (length: 5 cm, diameter: 3.75 cm) containing 4% (in volume) of dispersed calcite grains. Due to the creation of new efficient flow paths, the effect of dissolving a relatively small amount of the rock was found to be much larger than the one calculated with semi-empirical constitutive equations. This motivated the development and testing of methods able to accurately predict changes in permeability brought by calcite dissolution. This paper presents results of direct pore-scale fluid flow simulations performed with the commercial software FLUENT using high-resolution (2.5 µm to 12 µm) 3D images of the rock previously used in experiments.

2. Micro-CT scans, mesh extraction and mesh modification

A fresh piece of Cayton Bay sandstone was extracted from a block already sampled for experimental studies and was scanned using a Scanco µCT 100 scanner housed in the Department of Mechanical Engineering, University of Leeds. X-rays were generated with a fixed X-Ray source operated at 50 kV and 160 µA, with a 0.1 mm copper filter to reduce noise and beam hardening and to increase image contrast. The volume of interest was a 5 mm long and 10 mm in diameter. This volume was reconstructed by stacking 2000 cross-sections of 5000 × 5000 pixels each so that individual volumetric pixels (voxels) would have the shape of 2.5 µm large cubes.

Each voxel has a grey scale intensity that represents the mean X-Ray attenuation coefficient of the corresponding cubic volume in the sample, itself a function of the density and the atomic number of the material. The result is a 3D image composed of 50 billion voxels. The center of the image, a $8 \times 8 \times 3.5\,mm^3$ sub-volume (Fig. 1) corresponding to about 15 billion voxels, was extracted in order to eliminate edge irregularities and imaging artifacts.

Image segmentation was performed using a trial version of the purpose-built ScanIP software obtained online at "http://www.simpleware.com/". A first segmentation was done with a flooding algorithm to select the connected porosity (i.e. the selection propagates from an initial voxel towards its neighbors if their grey scale values are within prescribed bounds). For this selection, the lower bound of the voxels grey scale value was zero and the upper bound was adjusted so that the final image porosity would correspond to the experimentally measured connected porosity (30.9%).

A second segmentation was done to isolate calcite grains by directly selecting the 4% brightest voxels, this value correspond to the calcite content of the Cayton bay sandstone. Image segmentation of calcite was facilitated by the fact that calcite is significantly brighter than the other minerals (quartz, kaolinite, microcline, albite) present in the CT images, at the exception of pyrite which is significantly brighter than calcite but

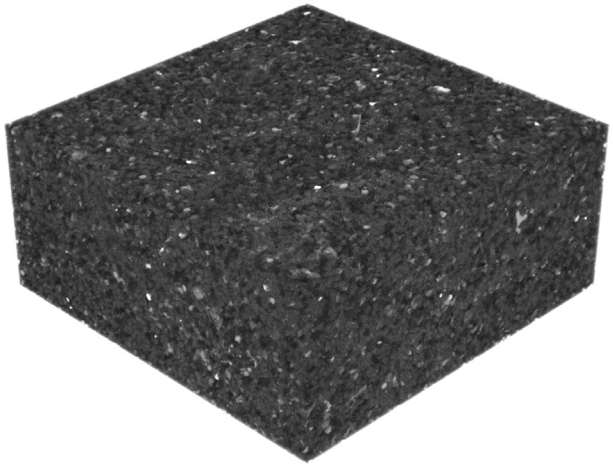

Fig. 1. 3D volume reconstruction composed of 1400 horizontal CT scans, the dimensions are $8 \times 8 \times 3.5\,mm^3$. Black zones correspond to pores, grey zones correspond to minerals of variable density/atomic number. For instance, very bright dots correspond to dense pyrite grains while very light grey zones correspond to calcite. Most of the image is composed of darker grey quartz.

is present in minute quantities. With a final visual inspection, we verified the correct segmentation of the three different phases (pores, calcite and "other solids"). Fig. 2 presents the result of the segmentation process on a small portion of the model.

Several sub-volumes were extracted from this large initial volume and were converted into finite element meshes using the finite element module of the ScanIP software: ScanFE; this generated regular meshes in which all elements were forced to be cubes of the same size. Mesh adaptation techniques and tetrahedral elements were avoided to facilitate the adaptation of the models to FV and LB methods used by collaborators. Here we report the results obtained for four sub-volumes: A1, B1, B2 and C1. The basic properties of these models are shown in Table 1.

For the models B1 and B2, a nearest neighbor resampling was used to produce ten parent models with a reduced number of voxels; from 400^3 to 350^3, 300^3, 250^3 and 200^3 voxels. The goal was to determine the trade-off between permeability prediction accuracy, model size and resolution. These ten initial models plus initial model A1 were also modified by increasing the porosity, either by numerically dissolving the calcite and reassigning it as pores, hence mimicking previous flow through core experiments, or by dilating the existing pores. For simplicity, we chose to simply remove all calcite from the model instead of simulating its dynamic dissolution. Simulating this end-member dissolution case is justified since experiments revealed that complete calcite dissolution happened very fast, not taking more than 1 h for the model sizes considered. In experiments, calcite dissolution was found to be dominantly transport limited even at the highest relevant reservoir flow rates so that dissolution dynamics (e.g. homogeneous versus heterogeneous dissolution typically studied for carbonate rocks) were not of interest here.

The structuring element size used for pore dilatation was found by trial and error so that the variation in porosity in pairs of "dilatation" and "dissolution" cases was as close as possible. The rationale was to understand the different effects on permeability when two vastly different processes modify the porosity and to make a direct comparison between the pore-scale modelling method, the Kozeny-Carman (K-C) relationship and the experimental results for the relationship between porosity and permeability. In Lamy-Chappuis et al. (2014) the K-C approach largely underestimated the permeability change in the experimental "calcite dissolution" case (see Fig. 10). Since the K-C approach assumes an idealized representation of the pore space as capillaries of a given radius and tortuosity we envisioned that it would be