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Microtomography-based Inter-Granular Network for the simulation of radionuclide diffusion and sorption in a granitic rock

Aitor Iraola[a], Paolo Trinchero[a,*], Mikko Voutilainen[b], Björn Gylling[c], Jan-Olof Selroos[c], Jorge Molinero[a], Urban Svensson[d], Dirk Bosbach[e], Guido Deissmann[e]

[a] AMPHOS 21 Consulting S.L., Passeig de Garcia i Faria, 49-51, 1-1, Barcelona 08019, Spain
[b] Department of Chemistry, University of Helsinki, P.O. Box 55, Helsinki 00014, Finland
[c] Swedish Nuclear Fuel and Waste Management Company, Box 250, Stockholm 101 24, Sweden
[d] Computer-aided Fluid Engineering AB, Frankes väg 3, Lyckeby 371 65, Sweden
[e] Institute for Energy and Climate Research: Nuclear Waste Management and Reactor Safety (IEK-6) and JARA-HPC, Forschungszentrum Jülich GmbH, Jülich 52425, Germany

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ABSTRACT

Field investigation studies, conducted in the context of safety analyses of deep geological repositories for nuclear waste, have pointed out that in fractured crystalline rocks sorbing radionuclides can diffuse surprisingly long distances deep into the intact rock matrix; i.e. much longer distances than those predicted by reactive transport models based on a homogeneous description of the properties of the rock matrix. Here, we focus on cesium diffusion and use detailed micro characterisation data, based on micro computed tomography, along with a grain-scale Inter-Granular Network model, to offer a plausible explanation for the anomalously long cesium penetration profiles observed in these in-situ experiments. The sparse distribution of chemically reactive grains (i.e. grains belonging to sorbing mineral phases) is shown to have a strong control on the diffusive patterns of sorbing radionuclides. The computed penetration profiles of cesium agree well with an analytical model based on two parallel diffusive pathways. This agreement, along with visual inspection of the spatial distribution of cesium concentration, indicates that for sorbing radionuclides the medium indeed behaves as a composite system, with most of the mass being retained close to the injection boundary and a non-negligible part diffusing faster along preferential diffusive pathways.

1. Introduction

In the last two decades, the description of subsurface flow and transport processes at the pore scale has received increasing attention (Molins, 2015, and references therein). These modelling efforts have gone hand-in-hand with the increasing availability of laboratory instruments, such as micro computed tomography (CT) scanners, which allow rock samples to be studied down to a resolution of about a micron (e.g. Blunt et al., 2013; Fusseis et al., 2014; Voutilainen et al., 2012).

Besides its academic value, pore scale modelling has been and is being extensively used by, or in the context of, the oil and gas industry, as a cheaper (and, often, more reliable) alternative to laboratory experiments; e.g. to derive continuum-scale properties to be used in reservoir-scale models (e.g. Øren and Bakke, 2003) or to study physical and geochemical processes that can influence oil recovery or CO_2 storage in applications related to geological carbon sequestration (e.g. Andrew et al., 2013; Molins et al., 2014; Huang et al., 2016).

Recently, increasing interest has emerged in using these micro characterisation and modelling techniques for the support of safety analyses of deep geological repositories for nuclear waste. In this context, micro-scale studies are typically applied to fractured crystalline rock formations and focused on assessing the influence of the heterogeneous rock matrix on transport and geochemical processes (Voutilainen et al., 2013; Trinchero et al., 2017a). Compared to the aforementioned applications in the oil and gas industry, one of the main differences here is that diffusion, which is retarded sometimes by sorption mechanisms, is the main (and sometimes only) transport driver in the rock matrix. Moreover, fractured crystalline rocks have usually a very low amount of pore space whose characteristic size is typically below the resolution of micro characterisation techniques. This pore space is typically found in-between grains of different mineral phases (Voutilainen et al., 2016) and this is why these types of micro-scale modelling approaches are denoted here as grain-scale models rather than pore-scale models.

* Corresponding author.
E-mail address: paolo.trinchero@amphos21.com (P. Trinchero).

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A recent diffusion experiment (LTDE-SD experiment, (Nilsson et al., 2010)) carried out in the Äspö Hard Rock Laboratory (HRL) in Sweden has shown that in fractured crystalline rocks strongly sorbing radionuclides can travel surprisingly long distances driven by molecular diffusion. These anomalous penetration profiles are being assessed by an international team of experimentalists and modellers in the framework of the SKB Task Force GWFTS (Groundwater Flow and Transport of Solutes; www.skb.se/taskforce). In this study, we use grain-scale reactive transport models at different levels of detail to give a qualitative explanation to the aforementioned experimental results.

More specifically, we apply micro-CT analyses of a crystalline rock sample to provide a description of the inter-granular porosity which is used as topological space for numerical diffusion experiments. A detailed segmentation of the different mineral phases allows chemically reactive grains (i.e. grains where sorbing tracers could potential be sorbed on) to be identified, and sorption sites to be distributed accordingly. The numerical experiments focus on assessing whether the complex porosity network combined with the sparse distribution of sorption sites have a significant impact on the cesium penetration patterns.

2. X-ray micro-CT characterisation of the rock sample

The granodiorite sample studied here was taken from the Äspö Hard Rock Laboratory (HRL) is Sweden. The sample (LTDE-SD1) with diameter of 24 mm and length of 45 mm was drilled from an exposed fracture surface, which was the object of the LTDE-SD experiment. The rock sample includes a fracture coating with thickness of about 5 mm, an alteration rim, and an underlying less altered rock matrix (Fig. 1). The fracture coating consists of calcite and quartz together with minor amounts of chalcopyrite and small crystals of barite and fluorite, and the alteration rim consists mainly of epidote and chlorite (Widestrand et al., 2010). In this work we focus on the less altered rock matrix that is fine-grained and according to previous studies (Widestrand et al., 2010) consists mainly of quartz (30–35%), plagioclase (26–33%), K-feldspar (26–32%) and biotite (3–7%). The samples studied by Widestrand et al. (2010) have shown that plagioclase is partly saussuritised and sericitised, and biotite is mostly altered to chlorite. An average porosity of (0.26 ± 0.08) % has been determined for the rock matrix and it has been shown that the majority of the porosity is located at the grain boundaries around quartz and feldspar grains, and in biotite grains (Widestrand et al., 2010).

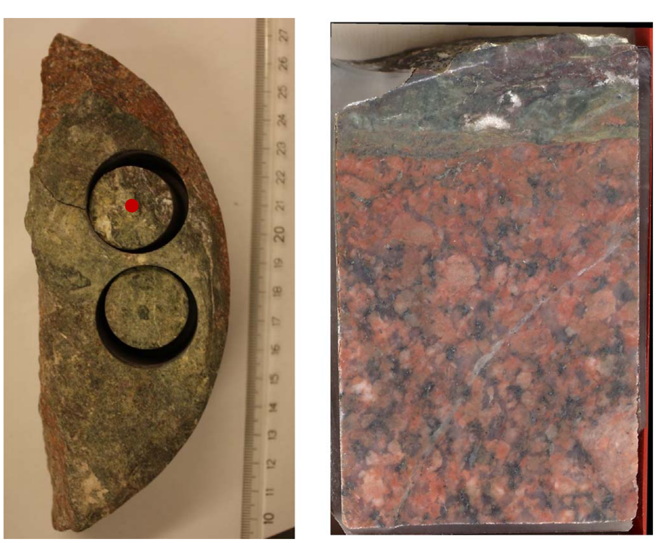

Fig. 1. (left) The LTDE-SD samples from the Äspö HRL (Sweden) and (right) cross section of sample LTDE-SD1, which is the object of this study (sample LTDE-SD1 is also indicated with a red dot in the picture to the left).

The 3D structure of the sample was determined with a SkyScan 1172 micro-CT scanner which has a conventional X-ray tube with a spot size less than 5 μm and a conical X-ray beam. The sample was scanned using a voxel size of 13.58 μm, an acceleration voltage of 100 kV and a current of 80 μA. Both aluminum and copper filters were used to treat the energy spectra. During the scan 2040 shadowgrams were taken with a rotation step of 0.10°. The reconstruction of the 3D image was done using a commercial software (NRecon Reconstruction) with beam hardening and ring artefact removal. After 3D reconstruction, 3D realization of Gaussian blur with a radius of 3 voxels was applied to reduce noise from the 3D image and to assist segmentation. Different minerals and the alteration rim were segmented from each other. First the alteration rim was segmented from the rest of the sample by detecting the interface between these two parts using the Carpet algorithm (Turpeinen et al., 2015). This was followed by segmentation of different minerals based on their grey value in the 3D image using histogram presentation. Threshold values for segmentation of each mineral were decided such that the thresholds were in between the peaks in the histograms. The rock matrix could be segmented into four mineral phases that include quartz and plagioclase, K-feldspar, mica minerals (biotite and chlorite) and dense accessory minerals. Quartz and plagioclase could not be segmented from each other as their grey values were overlapping due to their similar X-ray attenuation coefficients. Furthermore, distance transform and a watershed algorithm were applied to segment the grain structure of the minerals (Sardini et al., 2001). This was done separately for each mineral phase. The original micro-CT cross section and the images obtained after the segmentation steps are shown in Fig. 2.

The analysis described above was performed for a $1280 \times 1280 \times 1938 = 3.18 \cdot 10^9$ voxels sized representation of the sample with a resolution of 13.58 μm. The dataset was subsequentially reduced by aggregating voxels up to a resolution scale of 41.19 μm that lead to a $422 \times 422 \times 315 = 5.61 \cdot 10^7$ voxels sized representation of the sample after removal of parts including alteration rim. The resulting three-dimensional distribution of mineral grains is shown in Fig. 3. The identified mineral phases in the intact rock matrix are quartz and plagioclase, K-feldspar, biotite and accessory minerals, with an abundance of, respectively, 56.1%, 42.2%, 1.6% and 0.1% of the total matrix volume. These values were found to be in fair agreement with the mineral abundances determined previously for Äspö granodiorite (Widestrand et al., 2010).

3. Conceptual model

The model mimics a typical diffusion experiment, with a cocktail of radionuclides (in this set of calculations a conservative tracer and cesium) that is in contact with one face of the considered rock volume and diffuses into the rock matrix. Previous studies have shown that the properties of alteration rim and fracture coating vary highly within the experimental area of LTDE-SD in Äspö (Nilsson et al., 2010). Thus our analysis focuses on the intact rock and the alteration rim was left out of this study (see Fig. 3). It is worthwhile noting that in the Äspö LTDE-SD experiment, the diffusion experiment was carried out from an exposed fracture surface but also from a slim hole directly in contact with the intact rock matrix (Nilsson et al., 2010). The numerical model presented in this work is qualitatively consistent with this second experimental set-up (i.e. diffusion into the intact rock matrix).

Here, a constant concentration of the conservative tracer and cesium is applied to the bottom boundary of the sample. Both species can penetrate into the sample driven by molecular diffusion while cesium can also sorb onto biotite surfaces.

3.1. Construction of an Inter-Granular Network

A previous study on the distribution of porosity in the LTDE-SD samples carried out using C-14-PMMA autoradiography has shown that