A study of solute diffusion through unsaturated argillite micro-fractures network

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Introduction

In the context of radioactive waste management, deep geological repository in indurated argillaceous media is considered as a potential solution. Radionuclides transport through argillaceous media is then of concern and in particular via diffusion processes in pore water. The Excavated Damaged Zone (EDZ) surrounding the storage cells and the galleries of the repository presents a complex network of micro- and macro-fractures where radionuclides transport may be enhanced with respect to the diffusion in intact host rock. Especially the micro-fractures are supposed to form a connected pathway and thus the diffusion of solutes in this network is of high interest in the context of the storage safety assessment.

During the transient period after the excavation of the repository, the fractures undergo several desaturation/resaturation cycles resulting from desaturation process during the excavation phase or from hydrogen flow where hydrogen is produced from corrosion of the waste metallic canisters after repository closure. Their state can vary from fully saturated with water to almost complete desaturation depending on their location, aperture and on the time in the history of the repository. The saturation state of porous media has an important impact on the diffusion processes as it modifies the connectivity and the tortuosity of residual pore water. Thus in this work, we will focus on radionuclide diffusion through unsaturated micro-fractures network.

In order to simulate diffusion process inside micro-fractures geometry, we chose to use a Lattice Boltzmann Model (LBM) that allows to i) easily take into account the fractures geometry as available from X-ray tomographic images, ii) simulate water-gas distribution inside the fracture for different saturation level, and iii) simulate diffusion inside the resulting connected water pathway.

Opalinus clay µ-tomography samples

The micro-fractures used in this work originated from the Opalinus Clay of the Mont Terri Rock laboratory. The fractures were chosen among the available and percolating fracture geometries obtained from X-ray µ-tomography of clay samples collected inside the EDZ surrounding the borehole of the Ventilation Experiment II [1] with a voxel size of 0.7 $(\mu m)^3$. From the initial total dimension of about 1000^3 voxels smaller subsamples were extracted to study the dependence of the effective diffusion coefficient.

Lattice Boltzmann calculations

The LBM we used in this work is based on a Two-Relaxation-Time (TRT) collision operator. Water-gas distribution was simulated using the LBM described in [2] which follow the Shen-Chen approach through a particular source term. At initial time, a density was imposed inside the void space of the fracture. The presence of the wetting walls induces spontaneous phase separation leading to a low-density phase (gas) and a higher density phase (water). Based on the calculated liquid-gas distributions inside the pore space, a basic thresholding algorithm allowed to extract the connected liquid phase. For the diffusion computations, we considered a non-volatile tracer and only took into account diffusion in the liquid phase. We conducted diffusion simulations for different saturation in each micro-fracture following the procedure described in [3] and [4]. Computations were performed starting from an initial Dirac imposed concentration on the symmetry line of each fracture. The mean concentration was calculated as a function of distance to the central line and then fitted with the Gaussian curve. Effective diffusion coefficient was deduced as a function of fracture saturation.

References

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