Lattice Boltzmann pore-level modelling of dissolution-precipitation processes in porous media

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The porosity and mineralogical evolution of the technical barriers and their respective interfaces, plays a key role in the performance assessment of future deep geological repositories for radioactive waste. Geochemically induced changes of the porosity and changes of local transport properties at interfaces are most likely to occur at the contact of geochemically different barrier materials, e.g. clay host rock or concrete. Such localized changes can influence the intermediate and long term evolution of the entire barriers, and directly affects for example re-saturation times, corrosion rates, or the gas pressure build up within the repository. Experimentally exploring the processes with real interface materials is demanding due to the very complex chemistry and the extremely slow mineralogical evolution caused by limited transport of reactants in the barrier materials. In order to assess and model the effect of mineral dissolution-precipitation on porosity evolution, the laboratory experiment described in Ref. [1] has been performed considering a simplified chemistry and fast transport in a porous environment. Moreover, the selected barite-celestite system is of high relevance for retention of radium in geochemical systems and technical installations. Dissolution of strontium sulfate (celestite) and precipitation of barium sulfate (barite) alter the pore space in a non-linear way. The continuum scale reactive transport modelling of the experiment showed that it is impossible to reproduce the experimental results without detailed knowledge of on-going processes at the pore scale. The microscopic structure examination identified at least two important mechanisms for this specific experiment: a) homogeneous nucleation which results in nanocrystaline barite, and b) heterogeneous nucleation, which results in barite epitaxial rim formation on celestite crystals which prevented further celestite dissolution.

A Lattice Boltzmann model was built to enhance understanding of the underlying microscopic processes. The model successfully reproduces one of the main observed mechanisms of barite precipitation, namely the epitaxial rim formation. In Figure xx.1, a typical microscopic image of samples originating from the reactive zone of the in-house experiment of Ref. [1] is shown. The part of the image that was used as input geometry for the computations has been colored-coded and shows large celestite crystals in blue, the epitaxial barite layer in green, nanocrystalline barite in grey and liquid filled pore space in black. Simulations were conducted in this microscopic 2D geometry neglecting the presence of nanocrystalline barite and placing a supersaturated aqueous solution of Ba^{2+} and SO_4^{2-} between the celestite crystals. On the right side of Figure xx.1, the evolution of the reactive simulation is presented from top to bottom for a total reaction time of 20 hours. The barite layer formation (green), changes the pore space in a way that affects the permeability and the effective diffusivity of the domain of interest [2].

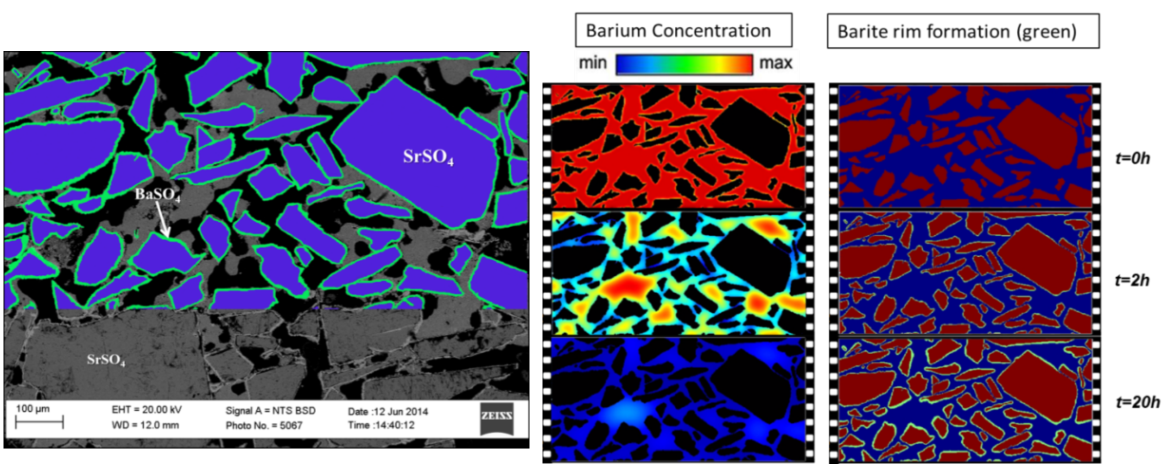

Figure xx.1: ***[Left]:*** *Scanning Electron Microscopy, Back Scattering method. Large celestite crystals (Blue color) are passivated by the formation of barite rims (Green color). Nano-crystalline Barite is in between. Black is open porous where aqueous solution existed by the end of the reactive transport simulation. Original setup contained pure celestite phases.* ***[Right]:*** *Lattice Boltzmann numerical simulation of the rim-formation mechanism and the precipitation evolution. Barium concentration drops from initial values (red) to equilibrium values (blue). Barite layer is colored green at the rightmost set of plots.*

References:

[1] Poonoosamy, J., Kosakowski, G., Van Loon, L., Mäder, U. (2015). Journal of Contaminant Hydrology, 177-178, 1–17.

[2] Prasianakis N.I., Poonoosamy J., Gimmi T., Kosakowski G., Churakov S., (2015). 6th international conference of Clays in natural and engineered barriers for radioactive waste confinement, Brussels, Belgium, March 23-26, 2015