Precipitation processes in porous media and consequences on permeability

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The understanding of porosity evolution and its impact on the transport of fluids and solutes in porous media is important, for example, with respect to contaminant transport in the subsurface, CO_2-sequestration, or the performance assessment of geological disposal facilities for radioactive waste. In particular in the field of nuclear waste disposal, an in-depth understanding of reactive transport phenomena and process couplings from the nano-scale to the macro-scale is a prerequisite for a realistic description for the long-term evolution of a repository system.

To enhance the predictive capabilities of reactive transport models and to gain fundamental insight into the couplings between advective transport and kinetically controlled dissolution/precipitation reactions causing porosity and permeability changes, we conducted a series of column experiments. Experiments were carried out in plug flow columns containing celestine ($SrSO_4$) grains as model porous media. Barium chloride solutions of different concentrations were injected into the columns, resulting in a replacement of $SrSO_4$ by $BaSO_4$ or ($Ba,SrSO_4$) accompanied by an increase in the volume of solids, and in consequence a porosity and permeability decrease. In some cases, similar volume and porosity changes resulted in significant differences in the measured permeability of the porous media. Microstructural investigation suggested that supersaturation greatly influences the nature and location of the $BaSO_4$ precipitates and that shape and surface roughness of the precipitates exert a pronounced effect on the flow field and the permeability of the system.

The experiments were modelled using the reactive transport code OpenGeosys-GEMS, giving a satisfactory match of the simulated effluent concentrations and bulk mineral transformations to the experimental data. However, using the Kozeny-Carmen equation, the simulated permeability changes showed significant discrepancies to the experimental results. The outcome of this study highlights the need for more sophisticated constitutive equations for a realistic description of porosity/permeability relationships in reactive transport models.