Can a macroscopic modeling approach properly describe transport in porous media during mineral precipitation induced clogging?

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The understanding of permeability and tortuosity evolution due to porosity changes is of paramount importance for the description of various hydrogeological processes, such as fluid circulation in acid mine drainage remediation or hydrothermal systems, management of aquifer recharge, efficiency of permeable reactive barrier and the long-term geochemical evolution of host rock considered for high-level nuclear waste repositories. The description of clogging phenomena in a reactive transport code is challenging, particularly over the long time periods (1000 to 1,000,000 years) that are of interest in most safety assessment cases. In addition to the validation of numerical scheme, experiments to validate the implemented laws used for the coupling of geochemistry (e.g. dissolution-precipitation reactions) to the transport parameters (induced porosity changes) and overall fluid flow processes (e.g. Carman-Kozeny relationship or Archie's law) are required.
The so-called Archie's law (Archie, 1942) is widely used in reactive transport models (Navarre-Sitchler et al., 2009) and describes the relationship between the effective diffusivity (D_e, in m^2s^{-1})of a dissolved species in a porous media and the diffusion coefficient of that same species in pure water (D_w, in m^2s^{-1}). The D_e/D_w ratio is a function of the tortuosity of the material (τ) and its constrictivity (δ) and therefore on the accessible porosity of the material (ε_a). As both the tortuosity and the constrictivity are difficult to measure experimentally, their ratio is often described as the geometrical factor, G. The relationship can be written as follows:

$$\frac{D_e}{D_w} = \frac{\delta}{\tau} = \frac{1}{G} = \varepsilon_a^n$$

where m is a called the cementation exponent. Though this empirical power law can accurately describe simple porous natural systems such as poorly compacted sands and sandstones, its applicability to more complex, low porosity and heterogeneous systems is questionable (Navarre-Sitchler et al., 2009)

In order to tackle this issue and to check the domain of validity of Archie's law, X-Ray transparent diffusion column experiments have been performed. Different porous media (clay, sand) have been used. In the present paper the focus is given on poorly compacted sand. Reduction of the pore space was induced by diffusion controlled precipitation of a simple salt, celestite ($SrSO_4$). A multi-scale and multi-technique approach has been used in order to characterize the transport properties before, during and after the reduction of the porosity as well as the amount of material that has precipitated. In this manner, (i) conservative tracer experiments (HTO) before and during the formation of a precipitation front, (ii) computed micro-tomography (µ-CT) coupled to pore-morphology modeling before and after the formation of a precipitation front, and (iii) post mortem analysis of the precipitate (dissolution, SEM-EDX) have been performed.

The conducted experiments are reproducible and after approximately 15 days the measured flux of through diffusing tracer HTO was constant and seemed to indicate that a steady state was reached, and thatthe pore space within the precipitation front was not fully clogged. Indeed, it was found that the reduced pore space in the precipitation zone remained fully connected (no isolated pores) under the resolution of CT (~6 µm), allowing for the migration of the tracer. The porosity and transport parameters prior to clogging estimated by the first 2 methods were consistent, as was the mass of precipitate estimated by the last two methods.

However, the reactive transport model built upon the experimental results underestimated the remaining connected porosity in the precipitation zone and overestimated the amount of precipitate. The results presented demonstrate that 1D continuum models might fail in the long-term prediction of transport in geochemically disturbed systems and argue for more complex 2D pore-scale reactive transport and/or a modified diffusivity/porosity relationship.

References

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