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## POROSITY IMPAIRMENT INDUCED BY DIFFUSION OF REACTIVE FLUIDS IN POROUS MATERIALS: EXPERIMENT APPROACH AND MODELING

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Disposal in geological clay formations is one of the solutions envisaged for managing the fate of high / intermediate level as well as long-lived nuclear wastes. The long-term evolution of these repositories is expected to be largely governed by a.o. geochemical processes that can irreversibly modify the containment properties of the materials used in the multi-barrier system. For instance, mineral dissolution and precipitation can significantly change their transport properties by enlarging or clogging the pore space. However, addressing the feedback of porosity changes in the long-term simulations coupling chemistry and diffusive transport is still an issue, especially because of the lack of dedicated experiments required for calibrating the numerical models. The objective of this study is to assess the ability of numerical codes (two reactive transport tools, HYTEC and CRUNCH) to reproduce experimental results obtained from reactive diffusion experiments carried out through porous media with an increasing complexity (from glass frit, to sandstone, chalk, and argillite). For that purpose, a large data set was acquired from experiments for which inert and reactive tracers diffuse through the porous media while precipitation and/or dissolution reactions take place. The solution chemistry and the tracer fluxes were monitored by regular sampling into the reservoirs. At the end of the experiments, complementary experimental techniques such as SEM-EDS or gamma-autoradiography were used, allowing the quantification of the mineralogy/porosity changes within the aged materials.

The results showed that the tracer diffusion into most of the reacting porous media was affected by precipitation, the impact intensity being roughly related to the pore size distribution and the precipitate type. For instance, the cells with chalk in which barite was expected to precipitate, displayed a clear impact of the clogging process from the experiment start, with a continued decrease of the HTO flux that became 40 times lower than the flux measured in the sound chalk sample (Figure 1). Conversely, the clogging effect on HTO fluxes induced by gypsum precipitation was observed (i) much later (i.e. 70 days after the beginning of the experiment) and (ii) with a weaker intensity, the HTO flux decreasing by a factor of 3 compared to the sound chalk sample. These results suggest that the clogging efficiency would be more related to the nature of the mineral than the amount of precipitated matter.

Finally, most of the simulations performed with CRUNCH and HYTEC were, at this stage, unable to properly reproduce the experimental results, especially those obtained in the diffusion/precipitation barite experiments (Figures 1 and 2). This issue will be discussed regarding the kinetic laws considered in the two codes and the empirical formulation of Archie's law used to relate the diffusion coefficient with the porosity change.

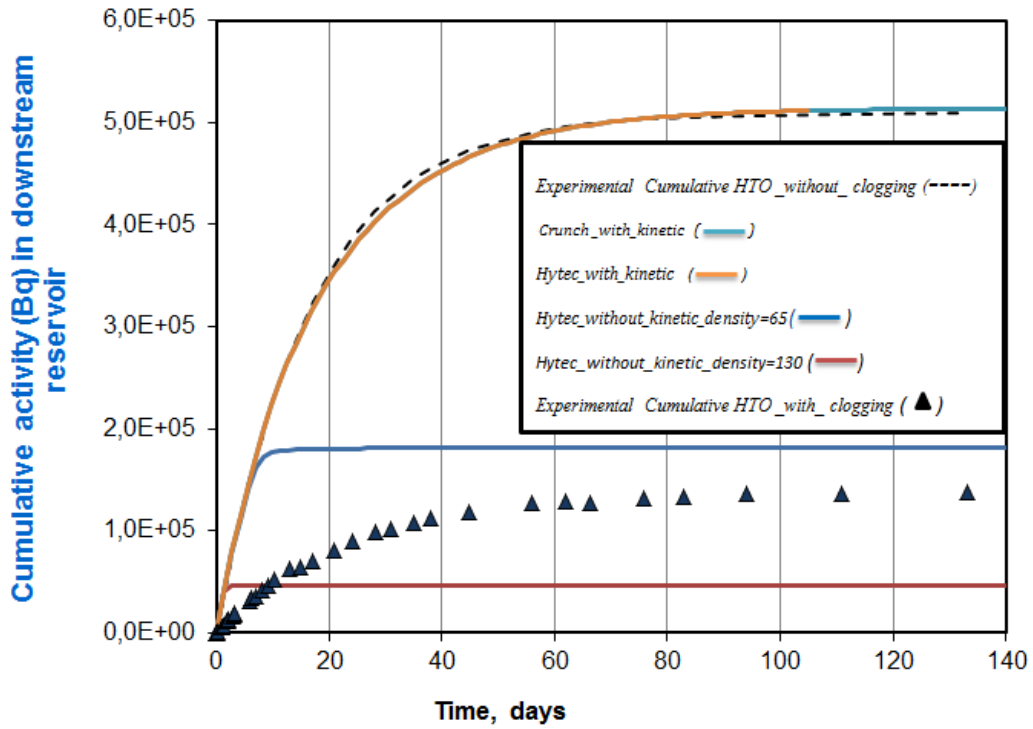


Figure 1: Cumulative activity in downstream reservoir in the chalk/barite experiment

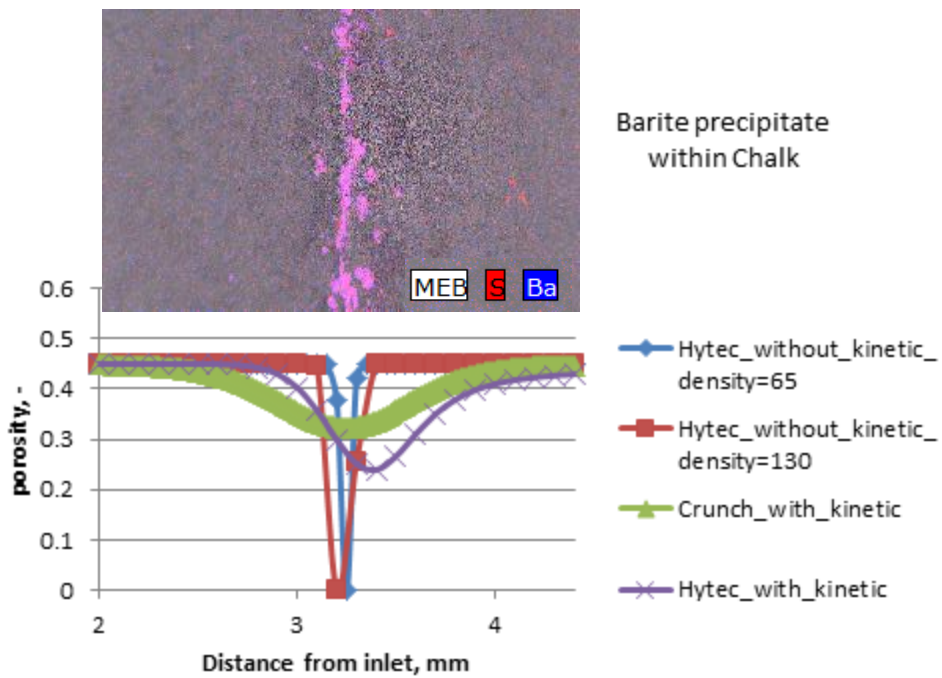


Figure 2: Comparison between the barite precipitate location in the experiment and the calculated porosity in reactive transport simulations