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Reactive transport modelling within a statistical framework to quantify sorption parameter uncertainties for Ni diffusion in clay

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Sensitivity analyses play an important role in understanding of complex geochemical processes and related reaction and solute transport equations when identifying most important processes and parameters [1]. It is also the driving force for designing waste disposals for radionuclides, where sorption is identified being the most important process responsible for their retention; and understanding of sorption processes, their parameters importance and accounting for their uncertainties allows quantification of long-term repository performance.

A comprehensive uncertainty and sensitivity analysis study of a detailed sorption chemistry model used for Ni reactive transport modeling in clay is presented using different statistical methods (once-at-a-time, Morris, Sobol). Migration of Ni in clay is analyzed according to uncertainties related to 25 site-specific equilibrium sorption reactions constants and clay pore water cations' concentration. The main influencing parameters have been identified, where the Fe2+ concentration shows the dominating uncertain parameter for Ni migration in clay. Three statistical methods identified the same most important parameters, but with different quantifications and related computational effort, i.e., thousands of reactive transport calculations, which were used to construct a new Ni isotherm and Kd uncertainty range, which shows lower Ni Kd values than reported in the literature for reference illite.

The presented framework can be directly transferred to other bivalent cations, as for example Co, Mn, Cu, Pb ... with similar sorption reactions [2]. Additional uncertainties for radionuclide transport could be investigated in future (spatial heterogeneity in transport, sorption, rock properties parameters etc.), which should be set in perspective to sorption parameter uncertainties investigated here. Then, it might be necessary to replace reactive transport calculations for different uncertain parameters by surrogate model calculations, which are much faster and allowing a larger set of uncertain parameters to be investigated. An example is given.

[1] Ayoub, A., Pfingsten, W., Podofillini, L. and Sansavini, G. (2020). Uncertainty and sensitivity analysis of the chemistry of cesium sorption in deep geological repositories. Applied Geochemistry 117: (doi:10.1016/j.apgeochem.2020.104607).

[2] Pfingsten, W. (2014). The influence of stable element inventory on the migration of radionuclides in the vicinity of a high level nuclear waste repository exemplified for 59Ni. Applied Geochemistry, 49, 103-115.