



Radionuclide retardation onto mineral surfaces: From descriptive approaches to mechanistic process understanding

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Sorption onto mineral surfaces is an important retardation phenomenon for radionuclides both in the near and the far field of potential nuclear waste repositories. Empirical distribution coefficients or simple models such as sorption isotherms (e.g. according to Langmuir or Freundlich) can nicely describe experimental sorption batch experiments. However, their predictive capability for environmental conditions beyond the specific lab set-up is severely hampered. They neither take into consideration the competitive effect of basic processes such as ion exchange, surface complexation, surface precipitation or incorporation nor do they reflect any realistic chemical speciation. The talk illustrates current state-of-the-art approaches to design useful sorption experiments, to identify the most relevant processes, to characterize the species involved in the reactions on the surfaces, to derive respective physico-chemical models, to parameterize them in a consistent way, and to consequently establish quasi-thermodynamic databases ready-to-use for reactive transport modelling. As an examples, the sorption of uranium(VI) onto quartz SiO_2 , gibbsite $\gamma\text{-Al(OH)}_3$ and goethite $\alpha\text{-FeOOH}$ is discussed, being representative model systems as well as having practical implications in the nuclear waste disposal as secondary phases. Techniques applied for the unequivocal determination of the relevant surface species are a combination of infrared (ATR FT-IR), fluorescence (TRLFS) and X-ray absorption (EXAFS) spectroscopies, supported by thermodynamic modelling. A consistent aqueous chemistry is provided by the most recent volumes of the NEA Thermochemical Database. The parameter sets derived are currently implemented into the Thermodynamic Reference Database THEREDA (www.thereda.de).