

Consolidation of surface speciations by a combined spectroscopic and modeling approach

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A reliable risk assessment of a deep geological waste disposal site for spent nuclear fuel refers to thermodynamic data bases which must contain resilient data sets. This information can be obtained from Surface Complexation Modeling (SCM) by fitting experimental batch sorption data, which is in turn consolidated by molecular information of relevant, prevailing species at mineral surfaces derived from spectroscopic investigations.

A combined approach of optical spectroscopic techniques, such as vibrational and luminescence spectroscopy, classical batch sorption studies and SCM was applied for the study of the surface speciation of actinide- and selenium-oxyanions on mineral phases. In this study, spectroscopic findings of the surface speciation of uranium(VI) and selenium(IV) and (VI) on different mineral oxide phases are presented. These phases serve as models for complex naturally occurring minerals in a host rock of a nuclear waste repository.

For the ternary sorption system U(VI)/phosphate/SiO₂, the formation of two binary uranyl surface species and of a precipitate was derived from spectroscopic findings which was sufficient to satisfactorily fit the respective batch results by SCM. For the sorption of selenium(IV) or selenium(VI) on alumina phases, a single predominant inner-sphere selenite and outer-sphere selenate surface species were identified by vibrational spectroscopy. With respect to the bidentate binding mode observed for both oxyanionic surface species, SCM provided excellent fitting results of the batch sorption data.

The results of this study demonstrated that the combined approach of *in situ* spectroscopy and batch sorption studies contributes to an improved performance of future assessments for the migration of radionuclides and fission products in the environment of a repository site.