The ternary sorption system U(VI)/ phosphate/SiO_2: a consistent surface speciation derived from a multimethodological approach

H. FOERSTENDORF[1]*, M.J. COMARMOND[2], K. HEIM[1], R. STEUDTNER[1], M. STOCKMANN[1] AND T.E. PAYNE[2]

[1]Helmholtz-Zentrum Dresden-Rossendorf e.V., Institute of Resource Ecology, P.O. Box 510119, 01314 Dresden, Germany (*correspondence: foersten@hzdr.de)

[2]Australian Nuclear Science and Technology Organisation, Locked Bag 2001, Kirrawee DC, NSW 2232, Australia

The surface speciation of the ternary system containing aqueous U(VI), phosphate and the model mineral phase SiO_2 was comprehensively investigated at a low micromolar concentration level by batch experiments, *in situ* Attenuated Total Reflection Fourier-transform Infrared (ATR FT-IR), luminescence spectroscopy, and Surface Complexation Modeling (SCM). In the absence of phosphate, two predominant U(VI) surface species were independently identified by luminescence and *in situ* IR spectroscopy. The concordance of the two species is corroborated by the shifts of the signals which were found to be of same extent in terms of energy units in the spectra of both spectroscopic techniques.

In the presence of phosphate, batch sorption studies indicate an increase in U(VI) uptake, consistent with previously reported studies. *In situ* IR spectroscopic sorption experiments strongly suggest the formation of a solid U(VI) phosphate phase as a surface precipitate on the silica phase, evidenced by characteristic bands observed in spectra after prolonged sorption and following sequential sorption of U(VI) then phosphate. Again, the results obtained from luminescence spectroscopy support these findings.

SCM provides excellent fitting results only when exclusively considering two binary uranyl surface species and formation of a solid uranyl phosphate phase as suggested from spectroscopic results. The results of this study indicate that the sorption of U(VI) on SiO_2 in the presence of inorganic phosphate initially involves binary surface-sorption species and then evolves towards surface precipitation.