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Modelling uranium retention and speciation in a chalky soil

PIERRE CRANCON[1], SIMON BAYLE[2], MARIA-ROSA BECCIA[3], PIER L. SOLARI[4] AND CHRISTOPHE DEN AUWER[3]

[1]CEA, DAM, DIF
[2]CEA, DES
[3]Université Côte d'Azur
[4]Synchrotron SOLEIL

The retention and speciation of anthropogenic uranium deposited in natural soil are investigated using an integrated and original approach combining field observations, experimental sorption isotherms and kinetics, recent developments in Extended X-ray Absorption Fine Structure (EXAFS) and Time-Resolved Laser-Induced Fluorescence Spectroscopy (TRLFS), and thermodynamic modelling using JCHESS®.

Uranium speciation along the vertical soil profile was determined by linear combination fitting of the EXAFS spectra, showing that uranium migration is strongly limited by sorption reactions.

Uranium sorption isotherms were established for calcite, chalk, and chalky soil materials, combined with EXAFS and TRLFS analyses [1, 2]. The presence of at least two uranyl tricarbonate complexes was inferred from TRLFS: the first has a liebigite-type structure and corresponds to uranyl adsorbed onto carbonate materials (calcite) at low surface loadings (< 10 mg U/kg rock), whereas the second corresponds to uranyl incorporated into calcite at intermediate to high loadings (~10 to >100 mg U/kg rock) [3, 4]. The temperature dependence of sorption kinetics was used to estimate the apparent equilibrium constant for uranyl adsorption onto carbonates [1].

The interaction of uranium with carbonates in soil materials was modelled in the presence of humic substances, highlighting the key role of both tricarbonate ions and humic substances in uranium retention and mobility in soil. This observation is of particular interest and raises the “natural organic matter paradox,” since humic substances have a versatile role in uranium migration: they are responsible for both strong uranium immobilization in soil and potential mobilization in porewater, thus enhancing uranium migration in colloidal form.

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