Biogeochemical alteration of uranic particles under flowing conditions: A micro/nanofocus study

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Decades of nuclear energy production and atomic weapons development has led to numerous authorised and accidental releases of radioactive material to the environment. Current impact assessments take little account of the fact that many radionuclides are dispersed in particulate form, and lack of knowledge concerning the evolution of particle-bound radionuclides makes prediction of their impact difficult. Here, uranic micro-particles (fuel proxy (UO_2), refined U ore (UO_3), and surrogate yellowcake (U_3O_8)) have been emplaced in flowing sediment / groundwater column systems, using sediments representative of the UK's Sellafield site. Two contrasting groundwater chemistries were employed to alter the particles under oxic and anoxic (Fe(III) to SO_4^{2-} reducing) conditions. The columns were then sacrificed after ~6 and ~12 months and, using the micro- and nano-focus spectroscopy beamlines at the Swiss Light Source (X05-LA) and the Diamond Light Source (I18, I14), elemental/energy mapping and targeted U L_{III}-edge XAS measurements were used to document the variable dissolution rates of the contrasting U particles, and follow the subsequent migration of U throughout the sediment, eventually precipitating secondary U phases (under both oxic and anoxic conditions). The results demonstrate the complexity of U association with the sediment/organic fractions at the micro-scale, whilst targeted μEXAFS and XANES energy mapping across regions of interest reveal intermediate and end product U speciation. This fundamental knowledge can be used to expand models of U biogeochemistry at contaminated nuclear sites and inform nuclear incident response.