Impact of microbial metabolism on radionuclide solubility in natural and engineered environments

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Microbial metabolism can have a controlling influence on the solubility of actinides and fission products in engineered and natural environments. In the 'far field' surrounding a nuclear repository, microbial processes can immobilise redox active radionuclides via respiratory processes that either change directly the oxidation state of the element, or produce new biogenic phases for enhanced sorption. In the 'near field' of the repository, the direct and indirect impacts of microbial metabolism are less well characterised but have the potential to have a significant impact on wasteform evolution and radionuclide mobility, and must be incorporated into the safety case of the repository.

Recent work on the redox cycling of U, Np, Pu and Tc will be discussed, including both reduction and oxidation reactions and their impact on soluble and insoluble radionuclide inventories. The roles of proteins, secreted electron shuttles and other microbial products will be discussed alongside additional controls coupled to bulk element cycles e.g. the production of new mineral phases or significant changes in the geochemical environment such as pH. Studies from a range of contrasting natural and engineered systems will highlight how microbial communities can respond to the radioactive inventory and the extreme (radio)chemistry of some disposed wasteforms, and ultimately control the biogeochemical fate of key radioactive elements.

Laser ablation with the NEPTUNE *Plus* MC-ICP-MS

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The Thermo Scientific NEPTUNE *Plus* with Jet Interface offers unparalleled MC-ICP-MS sensitivity for all elements across the mass range. An ion yield of > 3 % has previously been reported for uranium solutions [1]. Sensitivity enhancements have been achieved through a combination of a dry high-capacity (up to 130 m^3/h) interface pump, X-cone, Jet cone, and desolvating nebuliser system.

The Thermo Scientific NEPTUNE *Plus* with Jet Interface was coupled with a Photon Machines excimer laser ablation system. This system features a short pulse width (4ns) 193 nm excimer laser and the HELEX 2 volume sample cell. The 193nm wavelength has been shown to reduce the particle size distribution of the aerosol produced by the laser ablation process [2] and this in turn has been shown to help minimize the effects of fractionation by ensuring that particles are in a size range so as to avoid incomplete vaporization and ionization in the plasma [3].

The sensitivity of the Jet Interface for small spot size (< 10 μm diameter) LA-MC-ICP-MS was trialled for natural uraninite and for NBS U-030 certified reference material. The minor uranium isotopes, ^{234}U and ^{236}U, were measured simultaneously on discrete dynode SEMs, with RPQ abundance sensitivity filters, whilst ^{238}U and ^{235}U were measured on Faraday cups with 10^{11} Ω amplifiers. This 'nuclear forensic' application was previously demonstrated with an older generation of MC-ICP-MS instrument, which required larger ablation volumes due to lower analyte sensitivity [4].

Hafnium isotope ratios were measured from zircon reference material 91500 using Faraday cups with 10^{11} Ω amplifiers. Figures of merit are given for precision from decreasing ablation spot sizes (i.e. improved spatial resolution).

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