



Assessing long-term microbial impact on mineralogical trapping of uranium from deep groundwater at Äspö Hard Rock Laboratory, Sweden

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Natural uranium (U) in deep groundwater has been extensively studied in connection to the search for suitable locations for final disposal of spent nuclear fuel (SNF).[1] The U removal process depends on environmental and geochemical conditions and is often associated with fractionation of the main 'stable' isotopes, ²³⁸U and ²³⁵U ($\delta^{238}\text{U}$). The latter thus serves as an important tracer for redox specific scenarios, in local to global temporal and spatial scales. In this contribution, one specific borehole drilled at 415 m depth into Paleoproterozoic granitoid rock at the Äspö Hard Rock Laboratory (HRL), Sweden, is investigated after 17-year experiment period (1995-2012).[2] The HRL is built and operated by the Swedish Nuclear Fuel and Waste Management Co. and serves as a full-scale tunnel as a test-facility for the actual SNF repository to be built at Forsmark. We show how various micro-analytical techniques, modelling methods, and isotope analyses can be utilized to reveal U speciation and removal pathways, associated redox changes and related U isotope fractionations in the deep aquifer and during U mineralogical trapping. Spectroscopic techniques reveal that calcite precipitated on the borehole equipment contains intermittent highly elevated U, occurring as U(IV), and thus serves as a sink for U. Thermodynamic modelling shows that aqueous Fe(II) is the main driving force for the reduction of U(VI) in the borehole water, alongside sulfides formed from bacterial sulfate reduction. The bacteria-driven degradation of technical polymer constituents present in the borehole equipment is central to processes forming the sulfides and carbonates that facilitate reduction of U(VI) and subsequent immobilisation of U(IV) into the calcite. We use $\delta^{238}\text{U}$ to show that U undergoes several redox events in granitic rock aquifers, involving mineralogical and microbial pathways. The recorded $\delta^{238}\text{U}$ provides evidence for reductive removal of U from fracture water, presumably taking place along redox fronts in the fracture network. The obtained data on U(IV) removal by calcite provide important insights for the assessment of the geochemical behaviour of U and other redox-sensitive species in deep anoxic aquifers, that is relevant for trace metal mobility and long-term storage of SNF and nuclear waste.

References

[1]. J. Suksi et al., (2021), *Chemical Geology* 584: 120551.

[2]. H. Drake et al., (2018), *Environmental Science & Technology* 52(2): 493-502.