Physicochemical controls on Uranium(VI) migration at the intermediate scale

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Prediction of uranium migration within the context of long-term stewardship of contaminated sites requires models that can incorporate temporal and spatial heterogeneities at the field scale. However, most transport studies are completed at the bench scale, and often simplify either the physical or chemical heterogeneity in order to determine fundamnetal migration behavior. Thus there is an informational gap in how fundamental behaviors are exhibited at the field scale. In this presentation we report on experiments at the intermediate scale (~2m); the express goal of these experiments is to elucidate methodologies to 'up-scale' reactive transport models from the bench to the field.

An intermediate scale tank (2.44m x 0.61m x 7.6 cm) has been constructed; a macroscopically heterogeneous packing has been completed using the <0.250mm and 0.250-2mm size fractions of uranium contaminated sediment from the Naturita Uranium Mill Tailings Remedial Action (UMTRA) site. The size fractions were generally packed in layers, with an added 'block' of <0.250mm fraction present at the upgradient end to further perturb the flow field. Kinetic hindrances to uranium desorption were examined through the use of stop flow events. Once the tank exhibited tailing behavior, the composition of the influent was altered (CO_2 concentration increased from atmospheric to 2%) to determine the effects of temporal chemical gradients.

Effluent uranium concentrations started at 12.3 µM, and declined to between 2-3 µM within 30 days (0.5 pore volumes). This corresponds to a uranium flux out of the tank of 0.0756 µmols U/(kg sediment * L water) after four days of flow, and 0.0299 µmols U/(kg sediment * L water) after 35 days of flow. Uranium distribution within the tank was found to vary with pH, alkalinity, dissolved calcium, rate of U release from the different particle size categories, and the nature of the heterogeneity distribution.

Rhenium isotopes in natural systems

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We investigate variations of $^{187}Re/^{185}Re$ in various natural systems. Samples are analyzed by MC-ICP-MS; instrumental mass fractionation is corrected for using a combination of standard-bracketing and external W-correction according to an exponential law. External standard reproducibility is 0.04‰ (2SD). Data are reported as $\delta^{187}Re$, relative to NIST SRM 989 [1].

Total range of natural isotopic variation is approximately 2‰. Isotopic variation is observed in every substrate type observed to date, including sulphide minerals, meteorites, organic-rich sediments, liquid samples, and industrial Re. In sulphides, we see values of 0.75‰ and 0.12‰ for Archaean and Proterozoic MoS_2 while a modern ReS_2 has a value of -0.22‰. Among meteorites, Allende has a $\delta^{187}Re$ of -0.50‰ while that of Canyon Diablo Troilite is -0.21‰. Organic-rich sediments show a range of about 0.7‰. Dissolved samples analyzed thus far include seawater (-1.25‰) and acid pit mine water (-0.13‰). Industrially available Re exhibits a total isotopic range of ~0.3‰.

Data will be presented on Re isotope variations in the modern marine environment (seawater, reducing marine sediments, oxide crusts) for comparison with the better established Mo, and U isotopic systems [2, 3]. Solid and dissolved phase samples will be used to describe the effects of weathering on the Re system. Case studies will include a naturally weathering black shale profile, as well as an anthropogenically influenced mine site.

[1] Gramlich *et al.* (1973) *J. Res. NBS* **77A**, 691-698. [2] Barling *et al.* (2001) *EPSL* **193**, 447-457. [3] Weyer *et al.* (2007) Goldschmidt Conference Abstracts 2007, *Geochim. Cosmochim. Acta* **71**, A1105.