Estimating Radionuclide Release from a Uranium Deposit Through Uranium-Series Systematics in Carbonates and Opal

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The Nopal I uranium deposit at Pena Blanca, Chihuahua, Mexico, has been studied as a natural analog to the proposed geologic repository for high-level nuclear waste at Yucca Mountain, Nevada, USA. The deposit has strong geologic, hydrologic, and geochemical analogies to Yucca Mountain. Uraninite, a spent nuclear fuel analog, was deposited at around 8 Ma (Pearcy et al., 1994) and has been almost completely oxidized to a series of uranyl oxide hydrates and silicates. Some late-stage uranyl minerals formed at 3 Ma (Pickett and Murphy, 1999a). The oxidized uranium has been partially redistributed by aqueous transport around the deposit into fractures filled with iron oxyhydroxide, carbonate, and silica phases (Prikryl et al., 1997; Pickett and Murphy, 1999a). Thus, Nopal I may provide insights on radionuclide release and migration at the proposed Yucca Mountain site.

Caliche and opal in and around the deposit record mobilization and deposition of uranium and may provide information on rates and episodicity of release from the deposit. Visually pure opals covering uranium mineralization have uranium as high as 6900 ppm. Fracture-filling, carbonate-rich caliches contain up to 320 ppm uranium, demonstrating near-surface mobilization. Caliche uranium contents are highest down-slope from the deposit, but reach over 100 ppm in other directions. Crystalline calcite occurs in several forms around the deposit, representing various generations. Calcite ages are uncertain relative to uranium mineralization, and field relationships suggest some may predate the deposit. Nevertheless, their uranium contents are relatively high for calcite, ranging up to 34 ppm. Partitioning of uranium into calcite can be described by a D value--the ratio of U/Ca in calcite to that in water. Using a typical Nopal I crystalline calcite uranium concentration of 10 ppm and an approximate Nopal I perched water uranium concentration of 10^{-8} molar (Pickett and Murphy, 1999b) yields a D of 0.4, consistent with experimental partitioning data (Nugent and Reeder, in preparation). Higher uranium concentrations in caliches--which clearly post-date the deposit--may be attributable to higher water concentrations and/or a fraction of uranium not in calcite. Regardless of mechanisms, uranium-rich caliches reflect mobilization from the deposit under low-temperature oxidizing conditions likely to occur over long time periods at Yucca Mountain.

Uranium-thorium ages of U-rich caliche and opal testify that release has been recent at 54 ka, and suggest episodic release. Uranium concentrations in the caliche and opal, combined with isotopic age data and estimates of material mass, can be used to calculate rates of redeposition of mobilized uranium. These estimates provide a lower bound to rates of episodic release from the uranium ore body. Using the approach of Murphy and Pearcy (1992), Murphy and Codell (1999) estimated an upper bound to the mass of uranium removed from the Nopal I deposit [U(r), moles] based on a water infiltration rate estimated from meteoric precipitation rates, uranium concentration in water from solubility limits, and duration time of removal from ages of oxidized uranium minerals. The amount of uranium trapped in carbonates and opals [U(T)] can be compared with U(r) to help evaluate the reasonableness of each estimate. The estimated rate of uranium redeposition can be compared to the long-term estimate [U(r)/time]. Another long-term rate estimate--greater than U(r)/time--was considered by Murphy and Codell (1999) using the total mass of uranium oxidized. This rate was used in a U.S. Nuclear Regulatory Commission (NRC) performance assessment model for Yucca Mountain; the calculated dose from repository radionuclide release was lower than those using laboratory-derived release parameters from spent nuclear fuel tests. If release is episodic, short-term rates may differ from these long-term averages.

U.S. Department of Energy plans include study of natural analogs for building confidence in dissolution models for predicting spent fuel degradation rates at Yucca Mountain. It is in this context that release rates from Nopal I may prove useful. Radionuclide release rate is a function of both the degradation rate and the flux of water. Rates from episodic release and transport at Nopal I may further test the assumptions used to predict radionuclide release from Yucca Mountain.

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