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Characterization of cores from an *in-situ* recovery mined uranium deposit in Wyoming: Implications for post-mining restoration



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ABSTRACT

In-situ recovery (ISR) of uranium (U) from sandstone-type roll-front deposits is a technology that involves the injection of solutions that consist of ground water fortified with oxygen and carbonate to promote the oxidative dissolution of U, which is pumped to recovery facilities located at the surface that capture the dissolved U and recycle the treated water. The ISR process alters the geochemical conditions in the subsurface creating conditions that are more favorable to the migration of uranium and other metals associated with the uranium deposit. There is a lack of clear understanding of the impact of ISR mining on the aquifer and host rocks of the post-mined site and the fate of residual U and other metals within the mined ore zone. We performed detailed petrographic, mineralogical, and geochemical analyses of several samples taken from about 7 m of core of the formerly the ISR-mined Smith Ranch–Highland uranium deposit in Wyoming. We show that previously mined cores contain significant residual uranium (U) present as coatings on pyrite and carbonaceous fragments. Coffinite was identified in three samples. Core samples with higher organic (>1 wt.%) and clay (>6–17 wt.%) contents yielded higher ²³⁴U/²³⁸U activity ratios (1.0–1.48) than those with lower organic and clay fractions. The ISR mining was inefficient in mobilizing U from the carbonaceous materials, which retained considerable U concentrations (374–11,534 ppm). This is in contrast with the deeper part of the ore zone, which was highly depleted in U and had very low ²³⁴U/²³⁸U activity ratios. This probably is due to greater contact with the lixiviant (leaching solution) during ISR mining.

EXAFS analyses performed on grains with the highest U and Fe concentrations reveal that Fe is present in a reduced form as pyrite and U occurs mostly as U(IV) complexed by organic matter or as U(IV) phases of carbonate complexes. Moreover, U–O distances of ~2.05 Å were noted, indicating the potential formation of other poorly defined U(IV/VI) species. We also noted a small contribution from U=O at 1.79 Å, which indicates that U is partially oxidized. There is no apparent U–S or U–Fe interaction in any of the U spectra analyzed. However, SEM analysis of thin sections prepared from the same core material reveals surficial U associated with pyrite which is probably a minor fraction of the total U present as thin coatings on the surface of pyrite.

Our data show the presence of different structurally variable uranium forms associated with the mined cores. U associated with carbonaceous materials is probably from the original U mobilization that accumulated in the organic matter-rich areas under reducing conditions during shallow burial diagenesis. U associated with pyrite represents a small fraction of the total U and was likely deposited as a result of chemical reduction by pyrite. Our data suggest that areas rich in carbonaceous materials had limited exposure to the lixiviant solution, continue to be reducing, and still hold significant U resources. Because of their limited access to fluid flow, these areas might not contribute significantly to post-mining U release or attenuation. Areas with pyrite that are accessible to fluids seem to be more reactive and could act as reductants and facilitate U reduction and accumulation, limiting its migration.

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1. Introduction

* Corresponding author. Tel.: +1 505 667 8749. *E-mail address:* wgiday@lanl.gov (G. WoldeGabriel). The sandstone-type roll-front deposits located in the southern part of the Powder River Basin in Wyoming host the largest *in-situ* recovery

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(ISR) operation in the United States, the Smith Ranch–Highland facility, presently operated by Cameco Resources (Fig. 1) (Flores et al., 1999; Freeman and Stover, 1999). The principal U minerals associated with arkosic sandstone beds were identified as pitchblende and coffinite, and in most cases occur with pyrite, marcasite, hematite, ferroselite, native selenium, and calcite (Dahlkamp, 1993; Stewart et al., 2000; Heinrich, 2012). U was derived from multiple sources, including granitic rocks located to the south of the Powder River Basin and from the alteration and leaching of granitic sediments and late Tertiary volcanic ashes deposited in the area. The U subsequently migrated in oxygen- and carbonate-rich waters until it encountered an oxidation-reduction interface within organic-rich and organic-poor arkosic sandstone units, where it formed roll-front ore zones (Seeland, 1976; Santos, 1981; Freeman and Stover, 1999; Norris and Drummond, 2000). ISR is a solution mining technique that involves the injection of oxygen- and CO₂-fortified water (Miller et al., 1990; Norris and Drummond, 2000) to economically extract U from low-grade ore deposits while avoiding the production of tailings and U-bearing dust associated with conventional open-pit and/or underground mining operations. The main challenges with ISR mining are the inability to access all of the U in the ore zone, and, more importantly, the difficulties in restoring groundwater to baseline conditions and the mined host rocks to chemically reducing conditions capable of immobilizing any residual U or other contaminants.

During ISR mining, U(IV) minerals are oxidized to form highly soluble uranyl hydroxycarbonate species $(UO_2(CO_3)_n(OH)_m^{2-2n-m})$ or calcium–uranyl–carbonate species (CaUO₂(CO₃) n^{4-2n}), which is facilitated by the presence of excess carbonate. The U(VI)-rich solutions are pumped to the surface for U recovery using ion-exchange methods. Following ISR mining, restoration is typically conducted by sweeping the mined ore body with aquifer water drawn in from outside the ore zone (groundwater sweep) and then treating the ore zone water by reverse osmosis (RO). Treatment with reducing agents (H₂, Na₂S and H₂S) has also been used to restore the reducing capacity of the aquifer in an attempt to reverse the oxidizing conditions created during ISR mining (Mays, 1994; Hall, 2009). Bioremediation, through the stimulation of native metal reducing bacteria within the aquifer has also been considered for the restoration of mined sites (Mays, 1994; Reimann and Huffman, 2005; Luo et al., 2007; Hall, 2009; Yi et al., 2009; Hu et al., 2011). An understanding of the chemical speciation of U in postmined ore material and of the nature of associated minerals and geochemical conditions would help in evaluating and applying these and other remediation strategies.

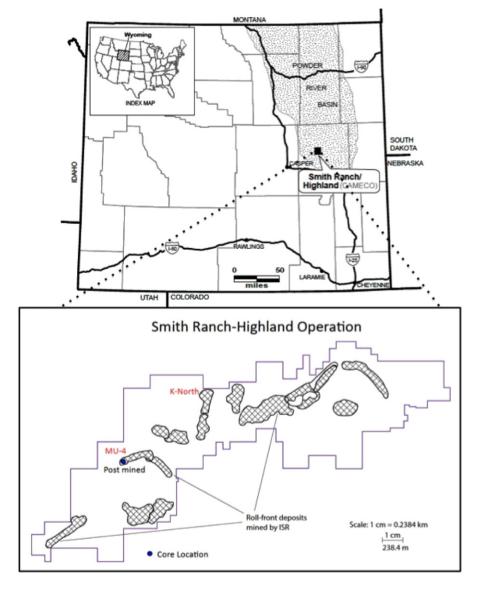


Fig. 1. Map showing the location of the Smith Ranch–Highland mining site and the core hole (Easting 348,744.59 ft, Northing 867,055.62 ft, elevation 5456.81 ft, and water level at 259.3 ft) at mine unit (MU)-4 in the southern part of the Powder River Basin in east-central Wyoming.

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