NATURAL ANALOG STUDIES AT PENA BLANCA AND SANTORINI

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Abstract. Natural analog studies have been conducted at the Nopal I deposit in the Peña Blanca uranium district, Chihuahua, Mexico, and at the Akrotiri archaeological site on the island of Santorini, Greece. The objectives of the studies were to identify and evaluate processes likely to occur at the proposed geologic repository for high-level nuclear waste (HLW) at Yucca Mountain, Nevada, USA, to support performance assessments of the Yucca Mountain repository, and to evaluate the utility of natural analog studies in support of geologic disposal of nuclear waste.

The Nopal I uranium deposit is hosted in a cuesta of silicic volcanic rocks above the water table in a semiarid environment. In the deposit, primary uraninite, an analog of spent nuclear fuel, has been mostly oxidized to a suite of uranyl minerals dominated by schoepite, soddyite, and uranophane. Also, uranium has been mobilized from the deposit and deposited with secondary opal, calcite, and ferric oxides and oxyhydroxides. Studies of uraninite alteration provide source term constraints for performance assessment. Uranium transport studies provide constraints for performance assessment on mechanisms and rates of radionuclide transport and deposition.

The Akrotiri archaeological site is a Minoan city which was buried in volcanic ash approximately 3600 years ago. The primary objective of natural analog studies at Akrotiri was to test predictive modeling methodologies for contaminant transport over long time periods by comparing model results based on site characterization data to the distribution of contaminants derived from buried artifacts. Positive contributions were provided, and problems were encountered in each study.

General conclusions from both studies are that contaminant transport processes are slow, and that these systems are relatively stable over thousands to millions of years with respect to release and transport of chemical species. Processes of release are slow and transport fluxes of chemical species are low even though the Nopal I and Akrotiri sites are near the ground surface, lack engineered barriers, and occur in wetter climates than the proposed repository at Yucca Mountain. Transport processes at both analog sites are transient and heterogeneous. Simple models for contaminant release and transport may fail to capture important effects of heterogeneities and variations identified by natural analog studies.

Introduction

Natural analog studies have been conducted at the Nopal I deposit in the Peña Blanca uranium district, Chihuahua, Mexico, and at the Akrotiri archaeological site on the island of Santorini, Greece. The studies were supported by the US Nuclear Regulatory Commission and were conducted by the Center for Nuclear Waste Regulatory Analyses at Southwest Research Institute. The objectives of the studies were to identify and evaluate processes likely to occur at the proposed geologic repository for high-level nuclear waste (HLW) at Yucca Mountain, Nevada, USA, to support performance assessments of the Yucca Mountain repository, and to evaluate the utility of natural analog studies in support of geologic disposal of nuclear waste.

Selection of the Yucca Mountain site by the US government for HLW repository investigations permitted identification of natural analog sites that have physical characteristics pertinent to that site. Yucca Mountain is a cuesta composed of silicic volcanic rocks in a semiarid environment. The proposed repository horizon is above the water table in rocks that are partially saturated with water. The system is oxidizing: the gas phase is rich in oxygen, and the rocks have minimal reducing components.

Like Yucca Mountain, the Nopal I uranium deposit is hosted in a cuesta of silicic volcanic rocks above the water table in a semiarid environment. In the deposit, primary uraninite, a chemical and mineralogic analog of spent nuclear fuel, has been mostly oxidized to a suite of uranyl minerals dominated by schoepite, soddyite, and uranophane. Also, uranium

has been mobilized from the deposit and deposited with secondary opal, calcite, and ferric oxides and oxyhydroxides. Extraordinary geological, geochemical, hydrologic, and climatic similarities between the Nopal I site and the proposed repository at Yucca Mountain assure that data from Nopal I will play a role in evaluating the proposed repository. Primary objectives of studies at Peña Blanca were to determine mechanisms and rates of oxidative alteration of uraninite and to examine the timing and mechanisms of uranium transport from the primary deposit and deposition in surrounding rock. Studies of uraninite alteration provide source term constraints for performance assessment. Uranium transport studies provide constraints on mechanisms and rates of radionuclide transport and deposition for performance assessment.

The Akrotiri archaeological site is a Minoan city which was buried in volcanic ash approximately 3600 years ago. The rocks are silicic tuffs, the climate is Mediterranean and relatively dry, and artifacts in the ancient city were buried above the water table. In addition to sharing analogous physical characteristics to the proposed repository at Yucca Mountain, the Akrotiri site offers the opportunity to examine contaminant transport from spatially well constrained, exotic materials over a well constrained time scale of several millennia. The primary objective of natural analog studies at Akrotiri was to test predictive modeling methodologies for contaminant transport over long time periods, such as those used in repository performance assessment. This objective was pursued by comparing model results based on site characterization data to the distribution of contaminants derived from buried artifacts.

Detailed results of the Peña Blanca and Santorini analog projects are published in the following literature: Murphy and Pearcy (1992), Leslie et al. (1993), Pearcy et al. (1994), Pearcy et al. (1995), Prikryl et al. (1996), Pickett et al. (1996), and Murphy et al. (1996). Also, see the companion paper by Pickett and Murphy in these proceedings. The object of this presentation is to provide an overview, update, and evaluation of major findings of these studies.

Peña Blanca

Although primary genesis of the uranium deposit at Nopal I is not the focus of the analog study, establishing the conditions of original uranium mineralization provides the framework to evaluate subsequent alteration and transport processes that are relevant to repository performance. Detailed mineralogical investigations reveal that primary uraninite was deposited in association with quartz, kaolinite, and pyrite. The primary mineralization process is interpreted to be due to mixing of reducing groundwater derived from underlying organic rich carbonate rocks with oxidizing water circulating in the silicic tuff. Mixing could occur in a fractured, high permeability zone that drains water from both sources. The reducing water contained Fe^{2+} and HS^- derived from the carbonates, and the oxidizing water contained UO_2^{2+} derived from the tuff. Reaction of these fluids with feldspar of the tuff can be generalized by

$$UO_2^{2+} + Fe^{2+} + 2\ HS^- + H_2O + 2\ KAlSi_3O_8$$

$$\rightarrow UO_2 + FeS_2 + Al_2Si_2O_5(OH)_4 + 4\ SiO_2 + 2\ K^+$$

which is driven by reduction of uranium, partial oxidation of sulfur, and incongruent acid dissolution of feldspar. This general reaction produces the observed primary mineral assemblage, uraninite + pyrite + kaolinite + quartz. Bulk mineralized rock analyses indicating U/Fe molar ratios greater than one suggest that reductants other than sulfur (e.g., CH_4) were important but are only indirectly reflected in the primary mineral assemblage.

Uraninite (UO_2 to $UO_{2.33}$) at Nopal I contains chemical impurities comparable in quantity (but not chemistry) to impurities in spent fuel. Based on the lead contents of late crystallizing uraninite with relatively low impurity cation contents, its age is estimated at 8±5 Ma. Consequently, alteration products of uraninite at Peña Blanca are not dominated by lead chemistry as in many older uranium deposits.

Oxidation of uraninite as an analog of spent nuclear fuel is a primary focus of the study because the Yucca Mountain site is oxidizing. Oxidation of uraninite at Nopal I resulted in formation of schoepite (including dehydrated schoepite and probably metaschoepite), soddyite, and uranophane, generally in that sequence, and lesser quantities of other uranyl minerals including ianthinite, becquerelite, weeksite, and boltwoodite. Formation of the predominant secondary minerals can be represented by

$$2\,UO_2 + 4\,H_2O + O_2 \rightarrow 2\,UO_3 \cdot 2H_2O \text{ (metaschoepite)}$$

$$2\,UO_2 + 2\,H_2O + O_2 + SiO_2 \rightarrow (UO_2)_2SiO_4 \cdot 2H_2O \text{ (soddyite)}$$

$$2\,UO_2 + 7\,H_2O + O_2 + 2\,SiO_2 + Ca^{2+}$$

$$\rightarrow Ca(UO_2)_2\,[SiO_3(OH)]_2 \cdot 5H_2O \text{ (uranophane)} + 2\,H^+$$

These reactions illustrate oxidation and hydration of uraninite with progressive incorporation of environmental components, SiO_2 and Ca^{2+}.

Generally the same paragenetic sequence of uranyl mineral formation was observed in experiments designed to approximate conditions in the proposed repository at Yucca Mountain (Wronkiewicz et al., 1993). Similarity of results in long (geologic) and short (experimental) time scales bracketing the repository time scale provides confidence in geochemical expectations for the behavior of spent fuel in a repository at Yucca Mountain.

Observations of uraninite alteration at Peña Blanca indicate that the source term for release of uranium and much of the radionuclide inventory in a repository at Yucca Mountain is likely to be controlled by the properties of secondary uranyl minerals. Petrographic and field relations indicate that uraninite oxidation and transformation to secondary uranyl silicate minerals have been rapid relative to mass transport of uranium out of the Peña Blanca system. A maximum limit on the average rate of uraninite oxidation at Peña Blanca can be calculated using geologic constraints on the amount of oxidation and the available time. Maximum values for the average rate of uranium oxidation and release from the Nopal I deposit were estimated to be 3.2×10^4 g of UO_2 per year using estimates of mass of oxidized uranium, water flux, aqueous uranium solubility controlled by equilibrium with uranophane, and duration of oxidizing conditions. In this derivation the period of time of oxidizing conditions was conservatively estimated from geologic constraints to be greater than 10,000 years. Seven U/Pb dates on 3 uranophane samples are in the range of 3 to 3.5 Ma. A revised value of the maximum average oxidation rate is 1.4×10^2 g/year using a time period of 3 million years. For the longer period of time, the maximum amount of uranium estimated to have been removed from the deposit by dissolution is 30 percent of the amount remaining in the deposit based on conservatively large solubility and water flux. However, consistent uranophane ages near 3 Ma indicate that oxidation occurred much more rapidly over a shorter period of time than the estimated maximum average oxidation rate. Much or most oxidative alteration at Nopal I is likely to be a consequence of an episode of low or moderate temperature hydrothermal activity in the late Pliocene.

Prior to mining, the uranium deposit at Nopal I was exposed at the ground surface along the eastern face of a basin and range structural block. In this position, the deposit has been subject to weathering and consequent transport of uranium. Uranium rich opal that covers uranophane in the deposit has been dated using uranium series techniques at about 54 Ka. In addition, uranium-rich caliche located about 3 m beyond the limit of present uranophane occurrence has been dated by uranium series isochron techniques at 54 Ka. These dates indicating near surface weathering of the deposit and remobilization of uranium for at least that period of time.

Gamma radiation intensity patterns, mineral distributions, fracture patterns, and uranium series isotopic activities at the Nopal I site indicate that some uranium has been remobilized from the area of original mineralization and transported away from the deposit mainly along fracture paths. Geologic mapping of the deposit shows that uranium

mineralization exposed at the surface is limited to a small well-defined area (about 18 m by 30 m in horizontal dimensions). This area has a roughly annular pattern of gamma intensities that corresponds to variations in mineralogy. The high gamma intensity annulus contains abundant uranophane, whereas the interior of the deposit has lower gamma intensity and no observable uranium minerals. X-ray diffraction analyses of samples from the interior of the deposit show abundant alunite [$KAl_3(SO_4)_2(OH)_6$], which is not present in the outer high-gamma intensity ring. Alteration of primary pyrite by interaction with oxidizing fluids is a likely mechanism leading to formation of alunite and low pH solutions. Conditions under which alunite forms (oxidizing, low pH) are also conditions that tend to mobilize uranium. Mobilization of uranium associated with alunite formation is a probable explanation for the spatial correspondence among low gamma intensities, the absence of uranium minerals, and the presence of alunite in the central portion of the deposit.

If the zone of visible uranium mineralization at Nopal I represents a source area, then areas of anomalous uranium beyond the mineralized zone may be interpreted to reflect flow paths. Transport vectors are likely to have resulted from anastomosing flow with a strong vertical component.

Contact gamma measurements of the surface exposures indicate relatively little transport away from the area of visible uranium mineralization to the W (2 to 3 m maximum) and only limited transport to the N (about 20 m). Anomalous gamma intensities extend well beyond the area of visible uranium mineralization along a SE-trend that corresponds to the downslope direction of the pre-mining surface. Tuff in the area of this extension is heavily weathered to clay (mainly smectite with minor kaolinite). These patterns suggest uranium remobilization by meteoric fluids infiltrating the fractured rock within and surrounding the deposit resulting in the greatest transport in the downslope direction.

There is a strong correspondence between the distribution of anomalous uranium concentrations and the locations of major fractures indicates that individual major fractures. This relation indicates that fracture pathways were more important to the long distance transport of uranium from the deposit than was the general fracture network comprising thousands of less continuous fractures. Studies of fracture densities and interconnectedness indicate that uranium transport has been largely independent of the general fracture network pattern. Matrix transport of uranium has been quite limited at the Nopal I deposit. Transport of uranium away from individual fractures into homogeneous, unfractured (at optical microscopy scales) matrix appears limited to distances less than 1 mm.

Constraints on uranium transport derived from uranium-series disequilibrium systematics at Nopal I are discussed elsewhere in this volume and summarized here. The key observation is that isotopic disequilibrium (i.e., in terms of activity, $^{230}Th \neq {}^{234}U \neq {}^{238}U$) is observed outside the deposit in both generally fractured tuff and in secondary phases filling fractures. This observation demonstrates that uranium has been subjected to open system processes over the past few hundred thousand years. $^{234}U/^{238}U$ activity ratios greater than one are consistent with uranium transport from the ore body in percolating, oxidizing meteoric waters and deposition in fracture minerals. This conclusion is supported by $^{234}U/^{238}U$ activity ratios ranging from 2.2 to 5.1 in present-day unsaturated zone waters. The observation of $^{230}Th/^{234}U$ activity ratios greater than one further implies that these rocks have been subjected to more recent partial depletion in uranium. Spatial patterns and sequential leaching results indicate that shifts in uranium deposition and removal regimes were episodic and complex. Combined with the U/Pb data mentioned above, the isotopic data suggest the following history: (i) uraninite deposition several million years ago, (ii) uraninite oxidation and deposition of uranyl phases (due to a shift to oxidizing conditions) during a period around 3 Ma, and (iii) complex episodic uranium mobilization and remobilization in the past few hundred thousand years. The episodicity of recent uranium transport is likely related to environmental factors affecting groundwater flux. Because of the strong analogy between conditions at Peña Blanca and Yucca Mountain, the demonstrated timing, episodicity, and complexity of transport at Peña Blanca suggest that such conditions can be expected at Yucca Mountain and are likely to be important to radionuclide transport at the proposed repository.

The following general conclusions relevant to a Yucca Mountain repository have been derived from the Peña Blanca natural analog study.

- Oxidation of UO_2 was rapid relative to migration of uranium out of the site, which occurs at a geologic pace.

- Secondary uranyl minerals control the distribution of uranium and releases of material from the deposit .

- Uranium that is transported away from the primary site is strongly associated with ferric iron minerals.

- Relatively long distance transport of uranium is controlled by discrete fractures; matrix transport is minimal.

- Uranium transport processes into the geologic environment are characterized by shifting episodes of deposition and remobilization.

The following important problems are associated with use of data from Peña Blanca in support of geologic disposal of HLW at Yucca Mountain.

- Much uraninite alteration and transport at Nopal I occurred under unknown water flow and saturation conditions. The system was likely to have been saturated during a portion of the alteration history, which is unlikely for the Yucca Mountain repository.

- Spatial definition of the source volume for transported uranium around the Nopal I deposit is only approximate.

- Pyrite oxidation and sulfuric acid production may have contributed to mineral alteration and redistribution at Nopal I, whereas no pyrite is expected to be present in the proposed repository.

Santorini

A challenging goal of natural analog studies is to contribute to validation of quantitative transport models for repository performance assessments. Although many natural systems, such as the Nopal I uranium deposit, provide valuable information on mechanisms and timing of transport processes, often the data are ambiguous or subject to alternative interpretations. In particular, initial and boundary conditions, forcing functions, and parameter values for geologic systems are often unknown, or known to be complicated or transient. To some extent these problems can be mitigated by natural analog studies at archaeological sites. The Akrotiri archaeological site has been studied as a natural analog system where metal transport has occurred from buried artifacts. The system is relatively well constrained. The Akrotiri site is in a relatively dry Mediterranean climate, artifacts are buried in a hydrologically unsaturated and chemically oxidizing environment, and the rocks enclosing and underlying the ancient city are silicic tuffs. These characteristics are similar to the Yucca Mountain repository site. The timing of the volcanic eruption that buried the city is well established at around 1628 BC, providing a 3600 year period for alteration of artifacts and transport of contaminants. Through information provided by site archaeologists and archival photographs belonging to the National Geographic Society, a particular room (Δ3) was identified where several large bronze artifacts were excavated in 1970 only centimeters above the tuffaceous bedrock floor of the building. These artifacts provide a spatially and temporally well defined and exotic source of contaminant material.

A limited characterization program was conducted using common techniques for repository site characterization. The site was mapped for geologic and hydrologic

characteristics, and hydrologic properties of the rocks were measured in the field. Rock samples were collected and examined in the laboratory for chemical and mineralogical composition and hydraulic properties. Samples of corrosion products developed on bronze artifacts were collected and analyzed. The extensive archaeological literature was reviewed to help fill gaps in understanding of transport processes at the site.

The site characterization data were used to support development of a numerical flow and transport model of the type used to support performance assessments. Many of the major assumptions and approximations employed in model development are of the same type as those used in performance assessments. Essential features of the model were one dimensional, isothermal, vertical flow, infiltration with an annual cycle, steady-state unsaturated flow below a few centimeters from the ground surface, source term controlled by equilibrium with secondary copper minerals, copper solubility and sorption coefficients estimated using a thermodynamic model for groundwater chemistry and literature data, and a one bar pressure head boundary condition at the groundwater table.

Primary model results are that steady-state flow and transport conditions are achieved within a short time period relative to the 3600 years since volcanism buried the artifacts. Sensitivity studies using reasonable ranges of hydraulic parameters confirm this model result. Given solubility limited concentrations at the source and constant distribution coefficients for each medium, concentrations of aqueous and sorbed species are predicted to achieve constant values for each medium within a few hundred years. Contaminant flux is determined in the model by water flux (infiltration) times solubility.

Independently of the model development, data were collected on contaminant transport and deposition at the site. Holes were dug and samples collected from a three dimensional array of packed earth (the Minoan floor) and underlying bedrock from an area approximately 1 m^2 and 0.5 m deep immediately below the area where the artifacts were found. Samples were crushed and subjected to selective leaching to extract trace elements, including copper, lead, and zinc, that may provide evidence of a contaminant plume. Leaching data indicate that excess copper, lead, and zinc are associated with the packed earth material, and copper and zinc show a pattern of decreasing concentration with depth. Also, elevated concentrations of contaminants are associated with a fracture adjacent to samples taken from one hole.

Details of the site characterization and copper chemistry at Akrotiri are not specifically relevant to the Yucca Mountain repository. The question addressed is the extent to which the tools of the practice of performance assessment modeling provide reliable information on the long term behavior of chemical transport in geologic systems. This question can be judged by comparison of model results to evidence for a contaminant plume at Akrotiri.

Results of the comparison are mixed. Many qualitative model results correspond well to the field data. The extent of copper migration is small. The artifacts are visibly corroded, which supports the model approximation that oxidation to secondary phases is rapid relative to transport of copper out of the system, and that solubility of secondary oxides controls the source term. Although altered, bronze artifacts remain largely intact indicating minimal removal of material. Detailed decorations and structural features can be easily recognized. Low estimated copper solubility and low water fluxes in the model are consistent with a small quantity of copper mobilization. Many other features of the buried city are in an excellent state of preservation after 3600 years, including paintings, pottery, and metallic tools, which generally attests to limited mobility of metals in a relatively arid, hydrologically unsaturated environment. In contrast, organic materials such as wooden structural units and furniture have been consumed leaving voids in the enclosing tuff.

Although absolute quantities of leached copper from the underlying tuff exceed predicted amounts by factors of about 2 to 4, uncertainties in estimated solubilities and sorption coefficients in the model, and uncertainties in the leaching process are at least of this magnitude.

The constant quantity of sorbed copper predicted by the model is in conflict with data for leached copper and other species from field samples. Field data for the contaminant plume show heterogeneous distributions for which reasonable explanations can be postulated. copper and zinc show patterns of generally decreasing concentrations with depth which is consistent with a diffusive, rather than advective, primary mode of transport. This is in sharp contrast

with model results in which diffusive transport is predicted to be negligible relative to advective transport. Diffusion control of contaminant transport suggests that moisture redistribution occurs by gas phase transport downward when the ground surface is wet and upward at other times. Gas phase moisture redistribution was neglected in the model.

Elevated concentrations of contaminants in one particular hole adjacent to a major vertical fracture in the tuff suggest focused recharge and contaminant transport along this fracture. Although the fracture was easily recognized in the field and samples were deliberately taken next to it, the fracture was neglected in the one dimensional model because of practical computational and resource limitations. Comparable simplifications characterize performance assessment models. Nevertheless, the field data indicate that transport is strongly affected by heterogeneities that were not accommodated in the model.

The following general conclusions have been derived from Santorini natural analog study relevant to the Yucca Mountain repository.

- Processes of alteration and transport in an unsaturated, oxidizing environment are slow on a time scale of thousands of years.

- Simplified models can miss primary characteristics controlling transport in a system analogous to the Yucca Mountain system.

The following important problems are associated with use of data from Santorini in support of geologic disposal of HLW at Yucca Mountain.

- The Akrotiri system differs from the Yucca Mountain in many regards. The archaeologic site is close to the ground surface, and to an intermittent stream; infiltrating water is affected by sea spray; the most promising site available was excavated a decade before the analog study, and some site information was unavailable.

- Definitive characterization of a contaminant plume is challenging in an unsaturated environment even under relatively well constrained chemical, physical, and temporal conditions. Ambiguities in the chemical leaching process contribute to uncertainties in characterization of the contaminant plume.

Generalizations

Alteration processes are slow, fluxes of material are small, and systems are fairly stable on time scales of thousands to millions of years in environments analogous to the proposed repository at Yucca Mountain. Processes of release of chemical species are slow even though the Nopal I and Akrotiri sites are close to the ground surface, lack engineered barriers, and occur in wetter climates than the proposed repository at Yucca Mountain.

Transport processes are transient and heterogeneous at both analog sites. Simple models for contaminant release and transport may fail to capture important heterogeneities and variations. Proper identification of the heterogeneities and complexities of processes and system characteristics that control long term release and transport will be required to accurately represent repository performance.

Analog studies are useful primarily to identify system characteristics of significance to long term geologic isolation of nuclear waste.

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