PEÑA BLANCA NATURAL ANALOGUE PROJECT: SUMMARY OF ACTIVITIES

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The inactive Nopal I uranium mine in silicic tuff north of Chihuahua City, Chihuahua, México, was studied as a natural analogue for an underground nuclear-waste repository in the unsaturated zone. Site stratigraphy was confirmed from new drill cores. Data from site studies include chemical and isotopic compositions of saturated- and unsaturated-zone waters. A partial geochronology of uranium enrichment and mineralization was established. Evidence pertinent to uranium-series transport in the soil zone and changing redox conditions was collected. The investigations contributed to preliminary, scoping-level performance assessment modeling.

I. INTRODUCTION

The Peña Blanca region, 50 km north of Chihuahua City, Chihuahua, México, was a target of uranium exploration and mining by the Mexican government. After mining ceased in 1981, researchers became interested in this region as a study area for subsurface uranium migration with relevance to geologic disposal of nuclear waste. Many studies related to this concept were conducted at the Nopal I mine site located on a cuesta (hill) of the Sierra Peña Blanca. This site has geologic, tectonic, hydrologic, and geochemical similarities to Yucca Mountain, Nevada, a formerly proposed site for a high-level nuclear-waste repository.

The U.S. Department of Energy (U.S. DOE), Office of Civilian Radioactive Waste Management (OCRWM), sponsored studies at Nopal I in the 1990s and supported the drilling of three research wells – PB1, PB2, and PB3 – at the site in 2003 (Fig. 1). Beginning in 2004, the Peña Blanca Natural Analogue Project was undertaken by U.S. DOE, OCRWM to develop a three-dimensional conceptual model of the transport of uranium and its radiogenic daughter products at the Nopal I site.

II. SITE STRATIGRAPHY AND DEPOSITIONAL HISTORY

Four main stratigraphic units were penetrated by the continuously cored PB1 well (Fig. 2). These units comprise all of the unsaturated zone and about 25 m of the saturated zone. Depths and elevations of contacts and the water table for the Nopal wells are shown in Table I. Cretaceous limestone represents carbonate deposits east (basinward) of the Cretaceous reef complex bordering the Chihuahua Trough.[1] The Pozos Formation is a continental molasse deposited on the margins of terrane uplifted during early to mid-Tertiary time. The age of the unit is constrained between 54 and 44.8 million years (Ma).[2] The Coloradas Formation unconformably overlies the Pozos Formation.[3] Constraints on the age of this unit are the same as for the Pozos Formation. The 44.8-Ma Nopal Formation is the uppermost preserved stratigraphic unit of the cuesta.

III. URANIUM DISTRIBUTION IN THE NOPAL WELLS

The most complete information about uranium distribution in the subsurface comes from borehole natural gamma logs and hand-held scintillometer survey of the continuous core from PB1 (Fig. 2). Both surveys measured gross gamma counts. High gamma counts predominantly reflect the presence of uranium.

High gamma counts associated with the main ore deposit are strongest in PB1, where values between 500 and 100,000 counts per second (cps) were recorded from the surface to about 113-m depth. The corresponding interval in PB2 is from the surface to 86-m depth (≤ 3,000 cps) and in PB3 from 17- to 119-m depth (≤ 4,500 cps). Natural gamma values in the 125-m-deep well PB4, within limestone 1.3 km SE of Nopal I, do not exceed 175 cps.

Deeper gamma anomalies (beneath the Nopal I ore body) are mostly within the Pozos conglomerate (Table II). The strongest anomalies are in the lower Pozos and, only in PB3, in Cretaceous limestone. This anomaly is strongest in PB3. The higher gamma values associated with the deep anomaly in PB3 seem to correlate with the higher uranium content of the groundwater sampled in that well.[4] The uranium content of groundwater in the shallow saturated zone may be influenced by rock-water interaction in the vicinity of the water table.

IV. URANIUM ENRICHMENT AND MINERALIZATION HISTORY

Rock units at the Nopal cuesta have a complex and incompletely understood alteration history. This section highlights selected alteration processes that contributed to uranium transport or sequestration.

IV.A. Early Silicification and Uranium Enrichment in the Pozos Formation

Various types of silicification are present in sediments throughout the Pozos Formation in PB1 core. The green ultraviolet-stimulated fluorescence of the secondary silica attests to a significant uranium content, and variations in the intensity of fluorescence help identify distinct clasts and multiple generations of silica cement.

Clasts of silicified limestone in the conglomerates preserve evidence of the earliest silicification, inherited from the parent limestone because many calcareous clasts are not altered. Silica-altered volcanic clasts also are common. Microquartz cement is present in clasts and as *in situ* cement. The recycled quartz cement is locally distinguishable from *in situ* cement by differences in intensity of ultraviolet fluorescence.

Uraniferous silica enrichment occurred through the processes of erosion and redeposition of silicified rock and *in-situ* silica cementation, possibly resulting in the formation of a silcrete. One sample of silica-cemented sediment contains 475 parts per million uranium.[5]

IV.B. Uranium Mineralization in the Nopal and Coloradas Formations

Initial Stage 1 uraninite precipitated in the Nopal and Coloradas Formations from 45-55ºC fluids that interacted with the welded tuffs. Mineralization occurred at 32±8 Ma under reducing conditions within a vertical breccia zone at the intersection of two faults.[5] Within the uncertainty of the uraninite age, mineralization was approximately contemporary with the earliest phase of Basin and Range extension at no more than about 29 Ma.[6] Primary uraninite was preserved only within a strongly silicified breccia.

The alteration of Stage 1 minerals resulted in the precipitation of Stage 2 uranium minerals. Uranophane, schoepite/dehydrated schoepite, and weeksite are the dominant Stage 2 uranium minerals, with minor amounts of colloform uraninite. Ages of these minerals include 3.1±0.5 Ma for uranophane (oxidizing event), 1.6±0.5 Ma for colloform uraninite (reducing event), and 85±8 ka for schoepite/dehydrated schoepite. Weeksite/boltwoodite from near the margins of the deposit gives an age of 41±5 ka. The two most recent mineral-precipitation events occurred under oxidizing conditions.[5]

IV.C. Uranium Mineralization in the Pozos Formation

The alteration that produced the ore body in the Nopal and Coloradas Formations probably affected the Pozos Formation as well. The ore body and surrounding rocks experienced subsequent hydrothermal modification and low-temperature alteration accompanying redistribution of uranium. The Pozos Formation also has experienced recent uranium transport and deposition.

Uraninite is present in a uranium-rich portion of tuffaceous Pozos conglomerate located nearly 100 m below the main ore body and 30 m above the water table.[5] The silicified rock described above contains disseminated grains of TiO_2 and TiO_2 replacements of highly altered sphene, as well as pyrite. Some of the titaniferous material is the TiO_2 polymorph anatase with rims of uraninite.

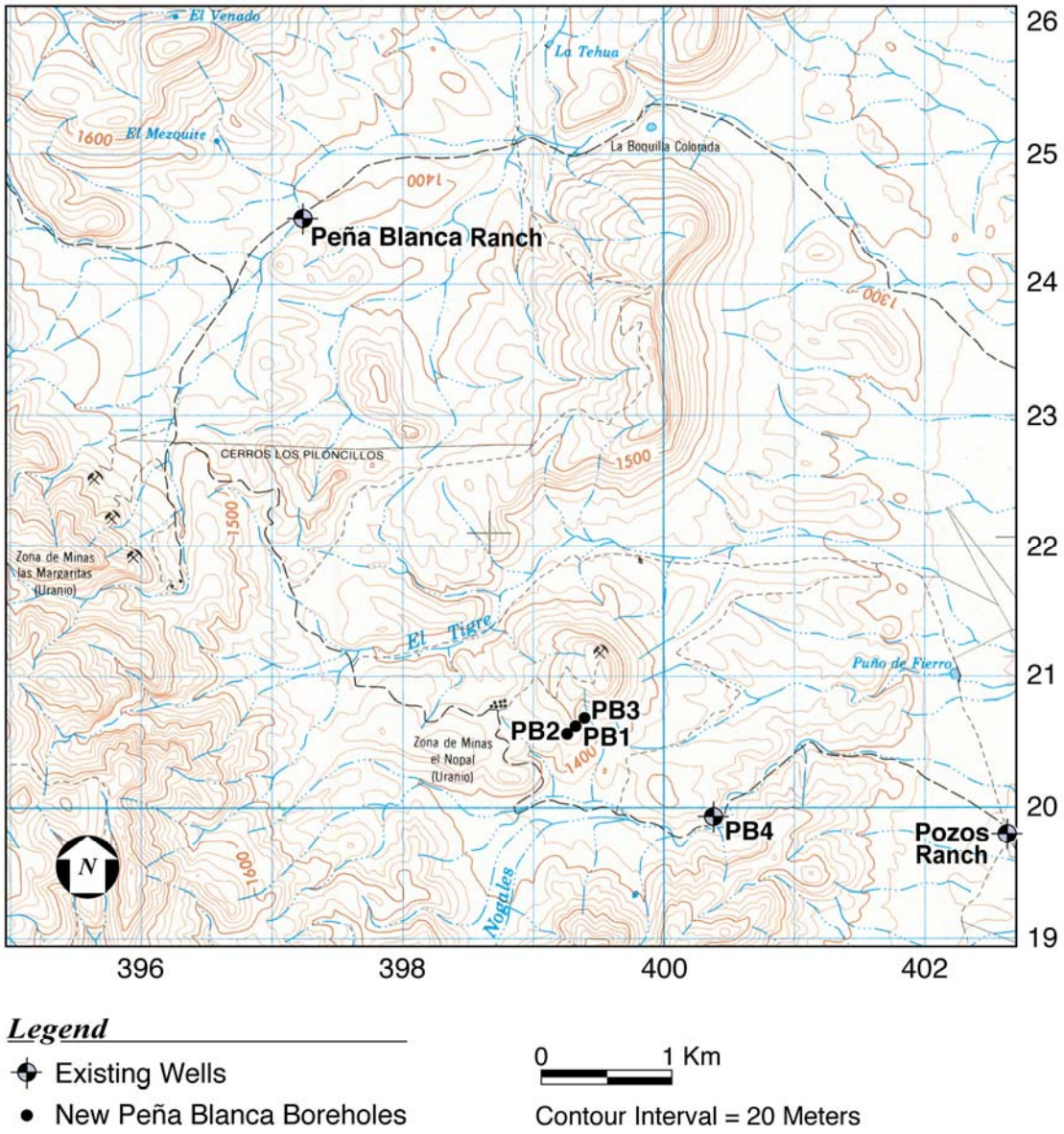

Fig. 1. Location of wells in this study. Base map adapted from Instituto de Estadística, Geografía e Informática 1:50,000 El Sauz topographic map (H13C46; 2004). Numbers along the border of the figure are Universal Transverse Mercator coordinates using NAD27 datum (the coordinates for PB1 are 3220596 N, 399322 E).

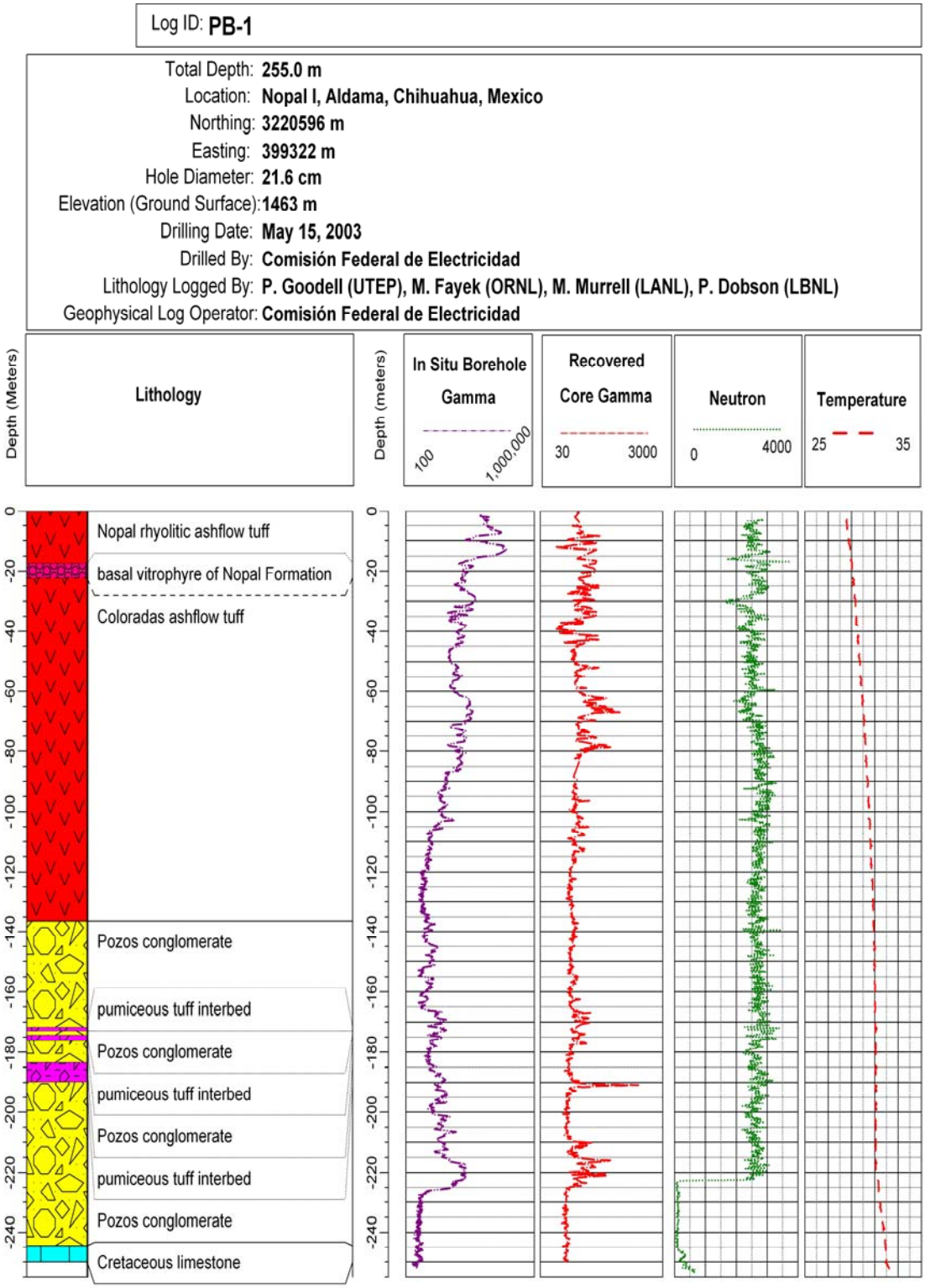

Fig. 2. Stratigraphic column and geophysical logs for well PB1. Borehole and core gamma values are in counts per second (plotted on a log scale), neutron measurements are in API units, and temperatures are in degrees Celsius.[2]

TABLE I. Depths and elevations of contacts and water table in Nopal I boreholes

	PB1 (core)	PB2 (cuttings)	PB3 (cuttings)
Ground elevation (masl)	1463	1463	1452
Depth/elevation of Nopal-Coloradas contact (m)	22.3/1440.7	22/1441	17/1435
Depth/elevation of Coloradas-Pozos contact (m)	136.38/1326.3	142/1321	130/1322
Depth/elevation of Pozos-limestone contact (m)	244.4/1218.6	242/1221	227/1225
Depth/elevation of water table (m)	222.8/1240.2	228.4/1234.6	213.8/1238.2
Total depth of well (m)	255.0	253.7	243.0

Sources: Refs. 2 and 7; Dobson, unpublished data.

TABLE II. Borehole gamma anomalies below the Nopal I main ore deposit

Well PB1			Well PB2			Well PB3		
Depth interval (m)	Gamma counts/second	Stratigraphic interval	Depth interval (m)	Gamma counts/second	Stratigraphic interval	Depth interval (m)	Gamma counts/second	Stratigraphic interval
144.1-151.7	500-1,000	All Pozos	163.1-165.4	500-2,000	All Pozos	130.0-204	500-3,000	Pozos
166.1-179.1	500-2,000		173.6-177.6	500-1,000				
			180.1-184.3	500-2,000				
189.1-226.7[a]	500-5,000		194.5-199.3	500-2,000		204-242.8[c]	500-32,300	Pozos and Cretaceous limestone
			211.5-235.5[b]	500-11,000				

[a]Highest values are in the 216- to 225-m interval. [b]Well was not surveyed below 235.5 m. [c]Highest values are in the 204- to 219-m interval. Source: Ref. 8.

The U-Pb age of the uraninite is < 1 Ma. The depositional temperature is estimated to have been 10º to 20ºC. Anatase is actively sequestering uranium from fluids that have interacted with the conglomerate.

General mineralogic trends over time toward increasing silica crystallinity through dissolution and reprecipitation would suggest that at least some of the uranium originally present in the silcrete has been released and redistributed.

IV.D. Sulfide Deposition and Alteration

Granular uraninite commonly is intergrown with syngenetic pyrite in the main ore body (not intersected by PB1).[9] This paragenesis, representing the initial uranium mineralization, was preserved by silicification. The underlying limestone may have provided sedimentary sulfides and organic matter favoring reducing conditions and uraninite/pyrite precipitation.[10] Rare sulfide occurrences in the Coloradas Formation mostly are restricted to xenoliths. Sulfides are common but not abundant in the Pozos Formation below a depth of about 180 m and in the underlying limestone.

There is a textural association of uraninite with titanium-rich minerals and pyrite in a PB1 core sample from the 191-m depth in the Pozos conglomerate.[5] The uraninite is less than one million years old, but this provides no constraint on the age of the pyrite and other sulfides. Some of the sulfide in the Pozos Formation has been replaced by iron oxy-hydroxide.

IV.E. Uranium Transport in the Shallow Unsaturated Zone

A water-collection system, consisting of 240 separate 30-cm^2 compartments that are each connected to a 125-mL bottle, was installed in April 2005 within the +00 m (arbitrary vertical reference datum) adit of the mine to collect water that had infiltrated from the +10 level (surface) and seeped into the adit.[4,11] The seepage

pathway from the ground surface to the collection system, assuming vertical fluid flow through fractures, is about eight meters. Analyses of seepage-water samples provide information about uranium-dissolution rates and uranium-isotopic compositions of the seepage water.

Seepage analyses confirm that uranium is being dissolved from the vicinity of the ore body under present conditions. Evidence of Quaternary-age uranium and uranium-daughter mobilization, transport, and sequestration has been observed in fractures exposed on the +10 surface of the mine.[12,13,14] The uranium is associated with iron oxides-oxyhydroxides probably derived from sulfide alteration. There are examples of both mobilization (primarily for U and Ra) and long-term sequestration (mostly for Th and Pa).

Quaternary-age uranium transport documented by the fracture fillings would have occurred in the shallow vadose zone. The hand-held gamma survey of PB1 drill core detected "hot spots" associated with iron oxide-oxyhydroxide mineralization. The iron-mineralized fractures with uranium have approximately the same vertical distribution as the ore body.

V. SITE GEOHYDROLOGY

Regional groundwater flow is from west to east, with the highest present-day water-table levels (>1500 masl) to the west and lower levels (~1200 masl) to the east.[15]

Previous studies of the Nopal I uranium deposit postulated that the ore body formed in the saturated zone and that oxidative mineralogic alteration and mobilization of uranium began when the deposit was uplifted into the unsaturated zone.[16] Paleohydrology and present-day geohydrology of the site have not been systematically studied, but some inferences can be made from existing information.

V.A. Late Cenozoic-Quaternary Topography and Drainage

The existing topography around the cuesta hosting the Nopal I deposit places bounds on possible high stands of the regional static water level at this location. As a result of uplift, mass wasting, and stream incision, the cuesta is bounded on the north, east, and south by erosional features at elevations of no more than about 1380 to 1400 masl (Fig. 1). A channel in the alluvium on the west side of the west-dipping cuesta occupies elevations of about 1420 to 1440 masl. The regional static water level below the cuesta, measured in borehole PB1, is at an elevation of about 1240 masl (Table I).

At the PB1 drill site, the uneroded remnant of the uranium deposit extends from the ground surface at 1463 masl to about 1350 masl. Thus, the present-day static water level is about 110 m below the base of the deposit. The existing state of erosion around most of the cuesta would preclude the static water level from reaching a position higher than about 1380 to 1400 masl, which just above the level of the base of the ore deposit.

A general inference from the Cenozoic-Quaternary history of more thoroughly studied sections of the Basin and Range Province (e.g., Ref. 17) is that the present topography and drainages of the Sierra Peña Blanca, including the Nopal I cuesta, began to develop 9-13 Ma and are likely to be at least several million years old. The maturity of the dissected landscape around the Nopal cuesta implies that all or most of the Nopal I ore body has been in the unsaturated zone for at least the last several million years.

VI. EVIDENCE OF CHANGING REDOX CONDITIONS

The groundwater beneath the Nopal cuesta contains dissolved oxygen, an indication of present oxidizing conditions in the shallow saturated zone and, presumably, the unsaturated zone. The evolution of redox conditions beginning at the time of initial uranium mineralization is inferred from the record of uranium-mineral deposition and alteration and from the distribution of sulfide minerals.

VI.A. Uranium Mineralogy

The uranium minerals of Nopal I record a long history of varying redox conditions.[5] Initial ore-body uraninite precipitation occurred at 32±8 Ma under reducing conditions in the Nopal and Coloradas Formations. A major oxidation event involving uranophane deposition occurred at about 3.1 Ma. This event was followed by a reducing event at about 1.6 Ma in which a second generation of uraninite, with a colloform texture, was formed. The ages of schoepite (about 85 ka) and weeksite/boltwoodite (about 41 ka) document more recent oxidizing conditions in the ore deposit.

Uraninite within Pozos conglomerate, about 100 m below the main ore body and 30 m above the water table, is less than one million years old. Anatase (TiO_2) is actively sequestering uranium as uraninite from fluids that have interacted with the conglomerate under reducing conditions.[5]

VI.B. Sulfide-Mineral Distribution

Sulfides formerly existed throughout the stratigraphic section penetrated by borehole PB1. The shallowest surviving sulfides include pyrite protected by silicification in the Nopal Formation[9] and sulfides primarily within lithic inclusions in the Coloradas Formation. Sulfides are common only below a depth of about 180 m in the Pozos Formation and Cretaceous limestone. Pseudomorphous iron oxide-oxyhydroxide replacements of sulfides are present in the Nopal and Coloradas Formations.

A tuffaceous layer within the Pozos Formation includes an interval between 173.20- and 175.22-m depths in which Liesegang bands are distributed around mostly low-angle fractures. Liesegang bands are distinct bands of mineral that form when coprecipitating ions interdiffuse in a porous rock, leading to discontinuous precipitation bands, usually of iron oxide or oxyhydroxide. The precipitation results from the reaction of oxygenated groundwater with soluble ferrous iron. An oxidized/reduced boundary with sulfide pseudomorphs at 210-m depth documents the derivation of iron oxide-oxyhydroxide from iron sulfide. These features, though not dated, probably record conditions in effect during the last few million years.

The downward transition from rare to common sulfide preservation at the 180-m depth in the Pozos Formation may mark the stable position of a former water table. This depth also corresponds to a downward transition to lower-permeability rocks.[2] Both factors could have restricted the interaction of rock sulfides with oxygenated water in what is now the deep unsaturated zone.

VII. URANIUM-SERIES GEOCHEMISTRY OF NOPAL I GROUNDWATERS

Groundwaters at Nopal I and nearby regional wells were analyzed for uranium-series disequilibria.[4] Unsaturated-zone seepage waters were collected between 2000 and late 2006. The waters were analyzed for chemical and isotopic suites including total uranium, $^{234}U/^{238}U$, total thorium, ^{230}Th, ^{226}Ra, and ^{239}Pu.

Seepage waters from the front of the adit, where the ore body is located, generally have $^{234}U/^{238}U$ activity ratios near unity (0.9 to 1.5) and uranium concentrations between 0.7 and 422 parts per billion (ppb). These attributes result from dissolution of rock with a preponderance of uranium in secular equilibrium, as well as longer water-rock interaction times and interaction of water with uranium-rich rock. Analyses of high-grade uranium ore from Nopal I confirm that uranium-series radionuclides high in the decay chain are close to secular equilibrium.[18] Waters from the rear of the adit, away from the ore body, have higher $^{234}U/^{238}U$ activity ratios ranging from 2.6 to 5.2 and mostly lower uranium concentrations (0.1 to 14.9 ppb). These differences reflect preferential dissolution of recoil-related ^{234}U and uranium concentrations affected by varying fluid flux and/or water-rock interaction times. Seepage-water samples from the middle of the adit have $^{234}U/^{238}U$ activity ratios and uranium concentrations overlapping the values for the front and back of the adit. Stable-isotope data for seepage samples show evidence of evaporation during the dry season that would have increased the concentrations of solutions.[19] The effects on uranium-isotopic systematics are described below.

Uranium concentrations and isotopic ratios were measured for shallow saturated-zone waters from Nopal I and regional wells.[4] For most wells, samples were collected from 2003 to 2006. Uranium concentrations in the Nopal wells have decreased over time as the local effects of contamination from drilling have diminished. During the last year of sampling (2005-2006), concentrations were between 23 and 61 ppb in PB1 and between 370 and 1077 ppb in PB3. Concentrations in the smaller sample set for PB2 are between 11.9 and 137.2 ppb. From 2003 to 2006, uranium varied between 0.1 and 15 ppb in PB4 and between 5.0 and 5.8 ppb at the Pozos Ranch well. Uranium in the Peña Blanca Ranch well varied from 9.5 to 10.0 ppb between 2003 and 2005.

Activity ratios of $^{234}U/^{238}U$ for all analyzed PB1 and PB2 water samples are between 1.005 and 1.102. Ratios for PB3 are between 1.363 and 1.974. The activity-ratio data, uranium-concentration data, and hydrologic-testing results for the three wells all indicate good connectivity between PB1 and PB2 and poor connectivity between these wells and PB3. Hydraulic testing also revealed that PB3 has higher permeability than the other two wells, as it experienced almost no drawdown during pumping.[20] Activity ratios for the regional wells are mostly >2.

The near-unity $^{234}U/^{238}U$ activity ratios and uranium concentrations less than 100 ppb in the PB1 and PB2 groundwaters presumably were acquired by percolating water, like the front-adit seepage, that interacted with bedrock containing abundant uranium in secular equilibrium. Waters of this composition could have traveled unmodified from the uraniferous source rock to the shallow saturated zone. However, considerable mixing could occur with minimal effect on the near-unity $^{234}U/^{238}U$ activity ratios if high-uranium waters (e.g., > 100 ppb) mix with low-uranium waters (e.g., < 10 ppb) of any observed activity ratio.

Percolation in the shallow unsaturated zone is highly variable with regard to uranium content and $^{234}U/^{238}U$

activity ratios. For the cuesta as a whole, shallow percolation is likely to have relatively low uranium concentrations and activity ratios in the range of about two to five, like seepage from the back of the adit, because the ore body occupies only a small part of the cuesta.

Groundwaters in the Nopal wells have higher colloid contents (~0.4 to 2 mg/L) than regional well waters (~0.001 to 0.09 mg/L). Differences may reflect both the recent drilling and more intense host-rock alteration of the Nopal wells. Uranium-concentration measurements of unfiltered and ultrafiltered water from PB1 and PB4 indicate that ~93 to 97 % of uranium present is truly dissolved.[4]

VII.A Uranium-Series Constraints on Radionuclide Mobility

The retardation factor, $R_f(^{238}U)$, was estimated to vary between ~30 and 7200 in the saturated zone, with a mean value of 1300, by combining long-lived and short-lived nuclide data (Ref. 21). ^{239}Pu values below a well defined detection limit, combined with measured uranium systematics for the Nopal and PB4 waters, were used to estimate that plutonium mobility is at least three orders of magnitude lower than uranium mobility in the saturated zone.[4] Average values of saturated-zone retardation factors range from 10^3 to 10^7, and decrease in the order $^{239}Pu \approx {}^{210}Po \approx {}^{230}Th > {}^{210}Pb > {}^{238}U \approx {}^{226}Ra$. Using the mean value for $R_f(^{238}U)$ in the saturated zone, estimated retardation factors for U-series daughter nuclides in the unsaturated zone range from ~10 to 230 for ^{226}Ra and from ~1300 to 260,000 for ^{230}Th.

VII.B. Modeling Uranium-Isotopic Systematics of Seepage Water

An analytical model was constructed based on the concept that intermittent seasonal infiltration and percolation in the shallow unsaturated zone leads to a linear relationship between reciprocal uranium concentration and $^{234}U/^{238}U$ ratio in percolating waters.[22] Seepage waters from various locations in the adit define distinct linear trends based on wet-versus-dry-season collection time. The underlying cause of this difference is the accumulation of recoil-produced ^{234}U on rock-pore and fracture surfaces during the dry season, followed by preferential uptake of the surficial ^{234}U by percolating water during the wet season. Other factors contributing to the trends include rock-water interaction (or water-transit) time, uranium-dissolution rate, and the rate of recoil that supplies ^{234}U to the rock surfaces. Longer periods of low infiltration and lower uranium-dissolution rates lead to high $^{234}U/^{238}U$ ratios in the percolating water.

Seepage waters with $^{234}U/^{238}U$ ratios close to unity and relatively high dissolved uranium content typify the front of the adit where the water interacted with uranium-ore rock.[4] Uranium-dissolution inputs for fractures with water-rock interaction times of ~0.5 days are ~1 ppb dissolved uranium/day. The dissolution of abundant uranium close to secular equilibrium masks the effects of processes that otherwise would produce waters with higher $^{234}U/^{238}U$ ratios.

VIII. RADIONUCLIDE TRANSPORT IN SOIL AT THE NOPAL CUESTA

Surficial weathering of the uranium ore body exposed at the surface of the Nopal cuesta has potential importance as a source of uranium transported into the unsaturated zone. Aspects of this process could be relevant analogues to extreme nuclear-waste release scenarios involving exposure or deposition of contaminated material on the ground surface.

At Nopal I, the natural soil was removed during the mining operation. A site with natural soil was studied close to where blocks of uranium ore were stockpiled in the 1980s. One residual ore block was selected to study the migration of uranium and its daughters into the underlying soil over a period of about twenty-five years.[18] Samples from the ore block and underlying soil profile were analyzed by gamma spectroscopy to identify radionuclide peaks from the ^{238}U-decay series.

Gross-gamma counts on two incomplete soil-sample suites show the radioactivity generally decreasing with depth to low levels below six to seven centimeters.[23] These findings are consistent with similar studies elsewhere.[24] The ore-block sample was relatively close to secular equilibrium for daughter/parent pairs $^{230}Th/^{234}U$ and $^{226}Ra/^{230}Th$, with activity ratios of 1.222 and 1.206, respectively.[23] A majority of the soil samples have non-equilibrium $^{230}Th/^{234}U$ activity ratios in the range of 1.80 to 2.18. The two shallowest samples from two soil suites have activity ratios between 1.18 and 1.50, probably due to incorporation of ore-block fragments into the soil. All soil samples had non-equilibrium $^{226}Ra/^{230}Th$ activity ratios in the range of 1.70 to 2.71.

Data for a complete eight-interval suite from below the ore block show that background radiation levels are reached at depths of about 10 to 15 cm.[25] Curves were fitted to the depth-activity data using a standard advection-dispersion equation with radioactive decay and ingrowth.[26] The best fits were achieved by setting the advection term (velocity of water flow) to zero. Although the equation assumes steady-state flow, the arid climate and wet/dry precipitation cycles cause soil conditions to

vary from no flow to downward flow following major precipitation to upward flow as the soil dries.

IX. PERFORMANCE ASSESSMENT MODELING

The simulation of radionuclide transport at Nopal I was conducted by the Yucca Mountain Project.[27] A numerical model was used to analyze the mobilization and groundwater transport of radionuclides potentially released from the ore deposit. The goal of the investigation was to estimate whether further investigations at the Nopal I site would provide a basis for a natural-analogue comparison with the expected performance of the Yucca Mountain site.

The numerical analysis used as input a combination of Yucca Mountain process models and Nopal I site-specific data such as the vertical section of rock units in the unsaturated zone, porosity and permeability data for rock units, the inferred eastward direction of regional groundwater flow, and the dimensions of the ore body. Estimates of the uranium content of the original ore deposit and the inventory of uranium species and daughter products were based on published studies. The model assumed that uranium oxide (uraninite) is analogous to spent nuclear fuel.

The model simulated the processes of meteoric-water infiltration into the unsaturated zone, downward percolation and dissolution of the uraninite, fluid mixing in the unsaturated and saturated zones, and eastward transport in the saturated zone. Radionuclide concentrations were captured for the saturated zone 150, 600, and 1,300 m downgradient from the ore body. The only corroborating data available were water analyses for well PB4 1,300 m SE of Nopal I.

A base-case analysis was run, along with sensitivity analyses to investigate the effects of reduced infiltration, variations in the solubility of the ore body, and variations in sorption coefficients. The study concluded that, even with strong sorption, uranium could be transported from the vicinity of Nopal I in amounts that would be detectable in PB4 water. The simulations projected concentrations of ^{99}Tc in the groundwater derived predominantly from natural fission of ^{238}U. The pertechnetate anion behaves as a conservative non-sorbing species in water, unlike uranium. Model results suggested that ^{99}Tc concentrations would be very low, but detectable (~10^{-8} mg/L).

The model results indicated that uranium concentration at an observation point varies directly with the quantity of infiltration and the solubility of the ore body. In this context, solubility was taken to mean the combined effects of mineral solubility and changes in surface area available for dissolution.

The numerical model was updated as the Peña Blanca Natural Analogue Model.[28] The PBNAM simulated the release and transport of radionuclides from the Nopal I ore deposit. The results were calibrated to uranium concentrations reported for 2003 water samples from the shallow saturated zone in boreholes PB1, PB2, and PB3.

Examples of model results include a base-case simulation for ^{238}U transport for 100 realizations of the uncertain dissolution parameters, but not including sorption. The observed uranium concentrations in Nopal well waters are bracketed by the range of results obtained in the simulations and within the uncertainty of the source-dissolution parameters. This result remains generally valid even with the trend of gradually diminishing uranium content shown by more recent water samples. The updated PBNAM predicted a ^{99}Tc concentration of 2.8×10^{-2} ppb in groundwater directly beneath the ore body. However, analytical results of Nopal well waters from two laboratories determined that no ^{99}Tc was observed above a detection limit of about 6×10^{-5} ppb.

After the PB wells were drilled at Nopal I, elevated uranium concentrations in the wells provided an opportunity for an informal saturated-zone tracer test in which higher uranium concentrations might be detected in well PB4. The three-year record of water chemistry for PB4 contains no clear evidence of water with elevated uranium content. The record of decreasing uranium concentrations and the concentrations of short-lived uranium-series daughters in the Nopal wells were used to estimate upper-limit groundwater velocities of 5 to 15 m/y, so that a tracer plume could take hundreds of years to reach PB4.

X. DISCUSSION

At the beginning of this study, the prevailing conceptual model for the onset of uranium migration at Peña Blanca was that the Nopal I uranium deposit remained largely intact under reducing conditions in the saturated zone until about three million years ago. The site then was uplifted into the unsaturated zone, and uranium began to migrate away from the primary deposit under oxidizing conditions. Our results generally are compatible with this concept. However, our studies indicate that, after three million years, the rock units have not fully equilibrated with the mostly oxidizing environment.

The conceptual model of uranium migration begins with the dissolution of uranium in the Nopal I ore body by

downward-percolating water. The uranium experiences some sorption in the unsaturated zone, but a sufficient amount arrives at the water table to form a detectable plume in the shallow saturated zone. This model is based on an assumption that no other significant uranium sources exist along the flow paths. Our observations of high uranium content and the presence of uraninite less than one million years old in parts of the Pozos Formation suggest that this unit may play a more complex role in water-rock interaction in the deep unsaturated zone. Additional lines of evidence are discussed below.

A basic unknown in unsaturated-zone transport at the Nopal cuesta is whether percolation that acquires its uranium-geochemical characteristics from the ore body controls the distinctive isotopic signature of the shallow saturated-zone water beneath the cuesta. The Pozos Formation is a potential additional locus of uranium exchange with percolating water. Variations in uranium-isotopic systematics in this unit are not known. The distance from uranium-rich Pozos rock to the water table is about 30 m versus a minimum of about 110 m between the ore body and the water table. Local precipitation, dissolution, and exchange of uranium could affect the isotopic composition of water in ways that are not easily distinguishable from the effects of processes in the overlying ore deposit.

The consistently higher uranium content of PB3 saturated-zone water relative to PB1 and PB2 demonstrates restricted mixing due to the existence of a flow barrier between the wells. Higher uranium concentrations exist in PB3 water even though this well does not intersect the ore deposit, whereas lower concentrations exist in water from PB1, which penetrated the edge of the deposit. The highest gamma counts for the uranium-rich portions of the Pozos Formation (below the Nopal I ore body) were encountered in the PB3 well, thus indicating that uranium sources in addition to the ore deposit may strongly contribute to the local water chemistry. The existence of a component of preferential and isolated lateral transport from the ore deposit toward the water table at PB3 would be an alternative or additional factor to account for the compositional differences.

XI. CONCLUSIONS

The Peña Blanca Natural Analogue Project studies reported here will support the development of hypotheses regarding the roles of the Nopal I uranium ore body and the uraniferous Pozos Formation in unsaturated-zone uranium transport.

The first direct dating of uranium minerals established an age of 32±8 Ma for the primary uranium mineralization. Alteration and uranium-mineral deposition at 3.1±0.5 Ma may mark the beginning of a transition from a reducing environment under saturated conditions to an oxidizing environment under unsaturated conditions. Local reducing environments have survived.

Initial uranium enrichment in the Pozos may date from a period of early diagenesis. Authigenic uraninite less than one million years old in the Pozos documents recent water-rock interaction involving the redistribution of uranium.

In the unsaturated zone, seasonal and lateral variations in the uranium content and $^{234}U/^{238}U$ activity ratios of seepage water result from differences in uranium content of the rock, fluid flux, and water-rock interaction times. Activity ratios near unity result from dissolution of rock with uranium mostly in secular equilibrium, as well as longer water-rock interaction times and interaction of water with uranium-rich rock. Seepage waters from less uranium-rich rock have activity ratios from 2.6 to 5.2, and may be more representative of shallow unsaturated-zone waters.

Shallow saturated-zone waters at PB3 are somewhat isolated from the waters at PB1 and PB2. Derivation of saturated-zone uranium predominantly from the Nopal ore body could occur if highly uraniferous waters from the ore body mix only with percolating waters of much lower uranium content. An alternate hypothesis, that uranium in the shallow saturated zone is derived in part from the Pozos Formation below the ore body, could be evaluated only with additional study.

Studies were conducted of uranium-series radionuclide migration from a block of uranium ore into underlying undisturbed soil. Radionuclides were found to have penetrated to depths of about 10 to 15 cm, with the highest abundances encountered in the upper 7 cm.

Simulations of aqueous uranium transport from the Nopal I ore body downward through the unsaturated zone and eastward within the shallow saturated zone predicted that elevated uranium concentrations would be detectable 1,300 m downgradient. Calculated water-transport rates suggest this transit would require hundreds of years. Overall, both water and nuclide transport rates are very low, thus providing positive implications for siting a nuclear-waste repository in this type of environment.

The rock units at Nopal I had a long alteration history in the saturated zone before beginning the transition to unsaturated conditions about three million years ago. This site would be a relevant analogue for nuclear-waste disposal in the unsaturated zone or in the saturated zone

with multi-million-year changes culminating in unsaturated, oxidizing conditions.

ACKNOWLEDGMENTS

Our co-investigators who contributed to this project include Shangde Luo, Alfredo Rodríguez-Pineda, Katrina Pekar, Diana French, Michael Murrell, Teamrat Ghezzehei, Paul Cook, Ron Oliver, Michael Rearick, Elizabeth Anthony, Minghua Ren, Steve Harder, and John Walton. We would like to thank our colleagues at the Autonomous University of Chihuahua, particularly Professors Ignacio Reyes-Cortés, Victor Reyes, and Lourdes Villalba. Our thanks also to Mabel Amador for Spanish translation and facilitating communications in México. John Dinsmoor led the field effort for our August 2006 site visit, and Alan Mitchell participated in earlier site visits. Ardyth Simmons guided the project through its important early stages, including the drilling at Nopal I. Peer reviews by Scott James and Ellen Leitschuh are greatly appreciated.

Funding for this work was provided by the Office of Civilian Radioactive Waste Management, U.S. Department of Energy, under Contract Number DE-AC02-05CH11231. The views expressed in this paper are those of the authors and cannot necessarily be taken to represent the views of the United States Government or any agency thereof, or The Regents of the University of California.

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