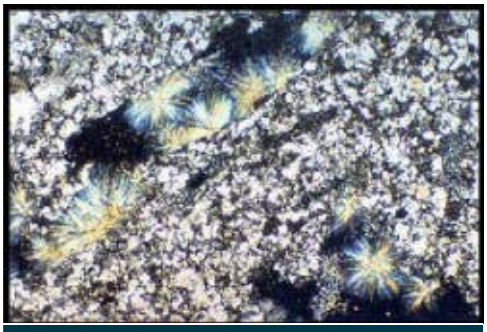
The Pena Blanca Natural Analog Project: Natural Analog Studies for Geologic Disposal of Nuclear Waste

Introduction*



Transmitted light photomicrograph of clusters of acicular weeksite crystals precipitated in open spaces in brecciated, silicified tuff. Scale is 1.3 mm across.

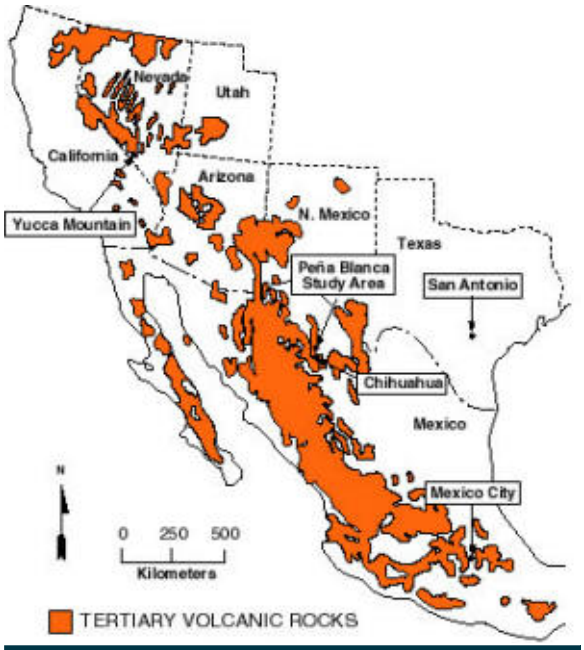
Nuclear power and nuclear weapons industries generate waste products with formidable toxicity, which will persist for periods of time that exceed the known duration of human civilization. The requirement for safe, permanent isolation of these wastes invokes contemplation of geologic time scales and geologic systems. A world-wide technical and social consensus has grown in support of permanent disposal of high-level nuclear wastes (HLW) in geologic repositories located hundreds of meters below the ground surface. Assessment of the ultimate safety of geologic repositories for HLW will require unprecedented and challenging predictions or estimations of the behavior of these complex systems for thousands or ten of thousands of years into the future. One approach to identify and characterize processes that operate on this time scale and to judge the validity of predictive assessments is through the study of analogous natural systems. In this context, natural analogs are geologic or archaeologic systems in which processes of significance to nuclear waste isolation have occurred on time scales (and space scales) relevant to geologic systems for permanent waste isolation.

Reasoning by Analogy

Reasoning by analogy is an exercise of induction in the sciences, notably in the earth sciences where enormous time and space scales are prevalent themes. General principles governing the evolution of geologic systems are based on available data, e.g., from the present, and routinely applied to the understanding of analogous geologic phenomena that are remote in time. For example, the history of the early Earth, for which no accessible record exists, has been largely ascertained through investigations of the present state of meteorites and the moon. The elementary concept of permanent geologic repositories for HLW is implicitly based on analogy to natural systems that have been demonstrably stable for millions of years. Analogous geologic or archaeologic systems offer the only opportunity for direct study of relevant chemical isolation and transport phenomena over the long time scales appropriate to nuclear waste disposal.

National High-Level Nuclear Waste Disposal Program

In the Nuclear Waste Policy Act of 1982, as amended in 1987, the Congress and President of the United States mandated investigation of Yucca Mountain, Nevada, as the unique national candidate site for a HLW repository. The statutes provide for the federal government to take title to spent nuclear fuel generated by utility nuclear reactors, and to dispose of it, along with certain high-level defense wastes, in geologic repositories to be designed, constructed, and operated by the U.S. Department of Energy. Environmental standards for HLW disposal are to be promulgated by the U.S. Environmental Protection Agency. These standards are to be implemented by the U.S. Nuclear Regulatory Commission (NRC), which has responsibility for public radiological health and safety, and authority to license the repository.



Map of tertiary volcanic rocks from Mexico City to Nevada

View of the Yucca Mountain region looking southwest

Yucca Mountain is located 150 kilometers northwest of Las Vegas on federal land that includes part of the Nevada Test Site, the national site for nuclear weapons testing. The repository is to be designed to isolate safely up to 70,000 metric tons of HLW for 10,000 years or longer after its closure some time in the 21st century. The Center for Nuclear Waste Regulatory Analyses (CNWRA®) at Southwest Research Institute (SwRI) conducts research and provides technical assistance to support the HLW program of the NRC. Scientists at the CNWRA have identified and are studying two natural analogs to the Yucca Mountain repository system. One of these is an archaeologic site on the island of Santorini, Greece, where a volcanic

eruption 3,600 years ago buried artifacts of the Minoan civilization in tuffaceous rocks. Migration of trace elements from these artifacts during the past millennia is considered to be analogous to migration of trace radioactive contaminants in the tuffaceous rocks of Yucca Mountain [editor's note: see Murphy et al., 1998]. The other site under investigation is the Nopal I uranium deposit in the Sierra Peña Blanca near Chihuahua, Mexico. The Peña Blanca natural analog is described in detail in this article.

Relevance of Natural Analog Studies to Nuclear Waste Disposal

Studies of natural analogs can be used to support predictive models of repository performance in two general ways: identification of processes and events, and calibration and validation of models. Observations and interpretations associated with repository site characterization will identify many important aspects of the geology (e.g., hydrology, geochemistry, petrology, and tectonics) that could affect waste isolation in the future. However, other significant phenomena are not expected to be manifested at the natural site, including chemical and hydrologic processes associated with introduction of foreign materials and with radiation and thermal effects. Studies of analogous systems should permit identification and appraisal of processes and events likely to influence the evolution of the perturbed geologic repository system and its capacity to isolate nuclear wastes. The development of complete and realistic conceptual models and scenarios for performance assessments will depend on data from analog systems.

Strict validation or proof of predictive models for repository performance is impossible because of the large time and space scales and the geologic and engineering complexity of the repository system. Nevertheless, a judgment must be made of the accuracy and applicability of the models. Both qualities can be evaluated using the degree of correspondence between model results and observable features of natural analog systems. Correct predictions of the characteristics of natural analog systems will help to demonstrate model accuracy over the range of characteristics, and over time and space scales that are inaccessible in the laboratory. Iterative modeling of systems that can be directly observed with progressive refinement is a routine method of model calibration. Predictions of processes and events in analog systems that are representative of specific repository conditions will show applicability of the models. Elements of model validation derived from analog studies may be largely qualitative because the representation of the repository system provided by analogs is approximate and because some quantitative features of natural systems are difficult to obtain (e.g., initial conditions). Derivation of quantitative data is enhanced by examination of analog systems and processes within those systems that can be well defined, well studied, and reasonably well understood, and that have characteristics closely resembling those of interest in the repository system.

Despite their unique potential, the limitations of natural analogs are indubitable. Chief among these is the incompleteness of the geologic record at any site with respect to details of relevant processes and events. Even the limited geologic record available is generally a manifestation of many complicated and overlapping processes that may be difficult or impossible to unravel. Partial resemblances between analogous systems and geologic repositories lead to uncertainties in the transfer of information and in the relevance of the natural analog data. For example, the existence of uranium ore deposits that are millions of years old does not necessarily prove that uranium is immobile. Genesis of ore deposits requires massive transport and concentration of chemical species by natural processes, and we can only infer or imagine the existence of former uranium or other ore deposits that are now dispersed in the environment. An additional problem is that some engineered components of repository systems such as special metal alloys have no reasonable analogs in nature. At best, processes in a natural analog system are a partial representation of those likely to occur in a repository system.

Numerous natural analog studies conducted in recent years have yielded information regarding the stability of materials and the migration of elements. The behavior of uranium in natural environments has been a primary focus because uranium constitutes a major component of nuclear waste. For example, two billion years ago, several remarkable uranium deposits in Oklo, Gabon, were so concentrated and rich in uranium-235 that they achieved criticality and sustained nuclear chain reactions as natural fission reactors for a few hundred thousand years. Researchers have been able to deduce the behavior of a rare mixture of uranium, plutonium, and nuclear fission products in the geologic environment at Oklo, in part through analysis of their daughter products. Other natural analogs include igneous contact zones and hydrothermal systems, which have been studied to evaluate thermal effects on repository environments due to the emplacement of heat-generating radioactive wastes. Another class of analogs has been studied to assess the durability of nuclear waste forms and container materials in geologic environments. These include, for example, volcanic glasses from mid-ocean ridge basalts, uraninite at Cigar Lake, Canada, native copper deposits on the Keweenaw Peninsula of Michigan, and metallic artifacts such as Cronan's bronze cannon extracted from the Baltic Sea.

The Peña Blanca Natural Analog and the Yucca Mountain Repository

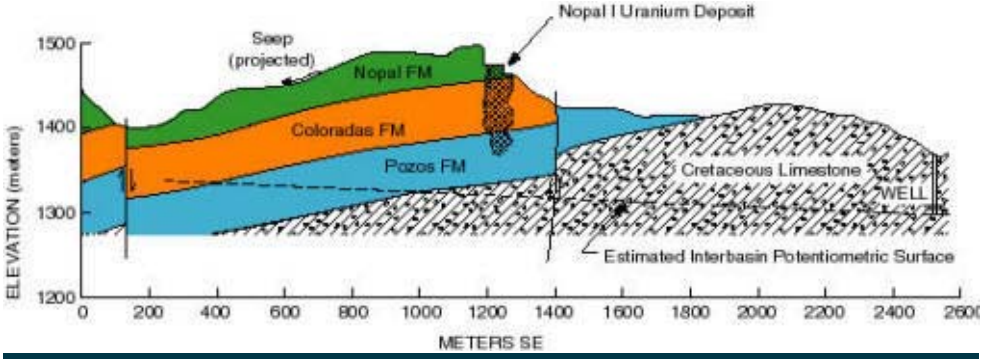
Remarkable physical similarities make the Sierra Peña Blanca an attractive natural analog to the proposed Yucca Mountain repository system. Both geologic terrains are large rotational fault blocks in the Basin and Range province of Western North America. Both are composed of volcanic rocks of Tertiary age: 10 to 14 million years old at Yucca Mountain, and 35 to 44 million years old at Peña Blanca. Although there are important variations at both sites, the pyroclastic textures and chemical compositions of the rocks are similar. The volcanic deposits were initially created by ash falls and ash flows that emanated in caldera-forming eruptions. The magma was rich in silica (~75%), alumina (~12%),

and potassium oxide (~5%), and when solidified, it formed rocks called rhyolite. At both sites, rapid cooling of the deposits quenched some magma to glass, especially at the upper and lower margins of individual flows. Where the rocks cooled more slowly, the magma crystallized, forming the dominant minerals feldspar, quartz, and cristobalite. Subsequently, through reactions with groundwater, some of the primary phases, and mainly the glass, were altered to clay and zeolite minerals, including smectite and clinoptilolite. Thermal contraction and tectonic deformation since deposition of the rocks have resulted in rock fracturing at both Yucca Mountain and Peña Blanca, although in both sites the rock layers are only slightly tilted relative to their original horizontal orientation.

View of the Sierra Peña Blanca

Groundwater characteristics are a key factor affecting geologic isolation of nuclear waste. The groundwater tables at Yucca Mountain and at Peña Blanca are both hundreds of meters below the elevated ground surface. Both the proposed repository horizon at Yucca Mountain and the uranium ore deposit at the Nopal I uranium deposit at Peña Blanca are in the unsaturated zone well above the water table. However, rocks in the unsaturated zone are not dry because capillary and sorptive forces retain water in small pores and on surfaces. The porosity may be about 15% of rock volume and 60% saturated with water, so although they look dry, the rocks are likely to approach or exceed 10% water by volume. Arid to semi-arid climates at Yucca Mountain and Peña Blanca lead to annual rain-fall averaging 15 and 24 centimeters per year, respectively. Most of the precipitation evaporates or is transpired by plants, so groundwater recharge and flow through the unsaturated zone are probably very low. Nevertheless, ephemeral, discontinuous zones of perched water, and episodic, local fracture flow are likely to occur at both sites.

View of the Nopal Formation in the Sierra Pe?lanca



Geologic cross section through the Nopal I uranium deposit (modified from Pearcy et al., 1994)

Groundwater flow and chemical transport in unsaturated rocks are complicated functions of recharge, suction pressures, pore size and configuration, and the interactions of chemical species among the solid, aqueous, and gas phases, and mineral surfaces. Generally, these phenomena are not fully understood. For example, the chemistry of water from the unsaturated zone is difficult to evaluate because it is almost impossible to sample without modification. Nevertheless, the similarities in climate, vegetation, and rock type between Yucca Mountain and Peña Blanca have probably led to generation of similar water compositions in the unsaturated zones of the two environments. The waters are most likely dilute, oxidizing, silica-rich, sodium-calcium-potassium bicarbonate solutions, with a near-neutral or slightly alkaline pH. The gas phase in the unsaturated zone at Yucca Mountain has been analyzed to be air (mostly nitrogen and oxygen), saturated with respect to water, and enriched in carbon dioxide relative to the atmosphere. By analogy one would expect similar ground gas at Peña Blanca.

One major, significant difference between Peña Blanca and Yucca Mountain is that several stages of hydrothermal activity and near-surface weathering at Peña Blanca have produced and subsequently altered numerous rich deposits of uranium. The Peña Blanca district is one of the most important uranium ore reserves in Mexico. Uranium was extracted from several sites in the district, and the ore deposit at Nopal I was exposed at the surface before mining ceased in the early 1980s because of economic and other social factors. Petrographic observations indicate that the uranium oxide mineral uraninite was originally deposited at the Nopal I site from hydrothermal fluids flowing in a permeable, intensely fractured (breccia) zone. The exact conditions of the ore formation are unknown, but clues can be derived in the study of inclusions of liquids and/or gases trapped in microscopic pores in stable minerals that formed in association with the ore. Observations of fluid inclusions with a petrographic microscope equipped with a heating and cooling stage suggest that the temperature at the

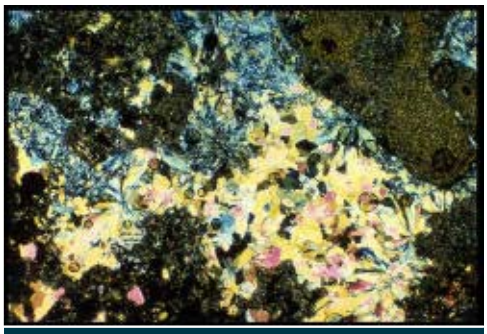
View of the Nopal I uranium deposit. This view shows the +10 m level of the deposit before clearing of loose rock and debris.

time of uraninite deposition was approximately 200°C, at pressures that were sufficiently high to maintain a liquid aqueous phase, but that also possibly permitted subterranean boiling to produce a carbon dioxide rich vapor.

Photograph of a thin section of uranium ore from Nopal I showing transition from reduced, black-colored U minerals to oxidized, yellow-colored U minerals in brecciated, tan-colored tuff. Scale is 40 mm across.

Natural uraninite such as that at Nopal I is compositionally and structurally similar to spent nuclear fuel, the waste form generated by commercial nuclear energy production in the United States, and one of the dominant waste forms proposed for disposal in the Yucca Mountain repository. Uraninite, which contains uranium in the reduced four-plus oxidation state, is thermodynamically unstable in the oxidizing geologic environment of Peña Blanca, just as spent nuclear fuel would be unstable in Yucca Mountain. Primary uraninite of the ore deposits at Peña Blanca has been almost entirely transformed to a suite of more stable, oxidized mineral phases through several stages of groundwater alteration, which may have occurred over a period of millions of years. These minerals are composed of uranium in the six-plus valence state combined with other major components of the geochemical environment. Some of the important uranium alteration minerals, and the main ore minerals at Peña Blanca, are soddyite (hydrated uranyl silicate), uranophane (hydrated calcium uranyl silicate), and weeksite (hydrated potassium uranyl silicate).

By analogy, the processes that could affect spent fuel alteration and contaminant transport in a Yucca Mountain repository should resemble processes affecting the uraninite ore body at Nopal I. The great similarity in the present physical environments of the two sites prompts particular interest in processes that have occurred recently in the Peña Blanca system, i.e., in the past thousands or tens of thousands of years. Textural relations among the different minerals, which can be observed at the field scale, in hand specimens, or by microscopic examination, are being used to identify processes governing mineral alterations, and to provide an indication of the relative timing of mineralogical changes. In addition, radioisotope geochemical analyses are being used to study the absolute timing of these processes. In a closed system, the radioactive isotopes in the uranium and thorium series decay chains evolve toward a state of secular equilibrium in which the activities of all isotopes in a chain are equal. The time required to reach (or closely approach) secular equilibrium depends on the initial conditions at the time of closure of the system and the half lives of the radioisotopes. Systems that have not achieved secular equilibrium must have been open with regard to one or more of the decay-series radioisotopes within a time period of several half lives or less of the disequilibrium isotope(s). These uranium and thorium series disequilibrium relations should permit identification of chemical and mineralogical changes that have occurred at Peña Blanca in geologically very recent times. These processes can reasonably be inferred to have occurred under conditions that resemble the present environment, and by analogy, under conditions similar to those in a geologic repository at Yucca Mountain.



Transmitted light photomicrograph of oxidized uranium minerals filling open space in brecciated tuff from Nopal I. The coarser yellow-colored crystals are beta-uranophane and the finer, blue-green cyrstals are uranophane. Scale is 2.6 mm across.

Two technical issues of particular concern in performance assessment modeling of a geologic repository for nuclear waste are the source term and the processes of contaminant transport. The source term is the rate of supply of the contaminant at its source in the transport system. Source-term limits are provided in general by the dissolution rate of the waste form, the solubilities of solids that control the aqueous concentrations of contaminant species at the source, and/or the migration rate of waste species out of engineered containment structures. Petrographic and field relations at Nopal I indicate that the processes of uraninite oxidation, dissolution, and transformation to secondary oxidized uranium minerals have been rapid relative to mass transport of uranium out of the deposit. The rate-limiting process for uranium removal is likely to be advective transport in groundwaters with uranium contents controlled by interactions with uranyl silicate minerals such as uranophane or weeksite. Examination of the secondary minerals at Peña Blanca can provide strong indications of the controls and compositions of unsaturated zone groundwaters under uranium-contaminated and uncontaminated conditions. Groundwater compositions affect both source and transport processes. Furthermore, a maximum limit on the rate of uraninite oxidation at Peña Blanca can be evaluated using geologic constraints on the maximum amount of oxidation that has occurred and the minimum available time.

Processes influencing the transport of uranium can be addressed at the Peña Blanca site by identification of fluid flow, diffusion, and chemical retardation mechanisms. The distribution of uranium among fracture coatings and rock matrix materials can contribute to an understanding of the relative significance of fracture flow and matrix flow in the unsaturated system. The time scales of transport mechanisms may be revealed by investigations of the openness of fracture and matrix systems with regard to species in the uranium decay series. The significance of diffusion as a transport mechanism can be judged by observing concentration gradients on small (e.g., mineral grain) scales. Geochemical retardation is the diminution of the transport rate of a contaminant species relative to the fluid flow rate and the aqueous diffusion rate due to its transient incorporation in or on immobile solids. Minerals that retain uranium can be identified (e.g., by autoradiographic analysis) to elucidate the retardation processes in the Nopal I system. If water samples can be obtained and analyzed, a knowledge of the aqueous speciation and uranium concentrations can contribute to an understanding of the solid-liquid distribution coefficients, which lead to retardation effects, and the solubilities of solid phases, which may control the source term.

Conclusion

The geologic record shows that particular geologic systems have remained effectively closed with respect to elemental migration for millions of years. By analogy, carefully selected geologic repositories could be capable of safely isolating high-level radioactive wastes, which will remain toxic over geologic time scales. Assuring the safety of geologic repositories requires assessments of processes that will occur over periods of time that far exceed those accessible to laboratory studies. The study of analogous natural systems can reveal the processes that are likely to occur in a nuclear waste repository and permit their characterization. In addition, natural analog studies can aid the development of models used in repository performance assessments, and can serve as testing grounds for the calibration and validation of models that represent processes related to those anticipated in repository systems over the long time scales required for nuclear waste isolation.

The Nopal I uranium ore deposit in the Sierra Peña Blanca, Mexico, provides an exceptional natural analog to the proposed high-level nuclear waste repository at Yucca Mountain, Nevada. Uraninite, analogous to spent nuclear fuel, has been oxidized in this deposit, where present geological, geochemical, and hydrological conditions closely resemble those at the Yucca Mountain site. Issues of contaminant source and transport over periods of time and under conditions relevant to nuclear waste isolation at Yucca Mountain can be addressed at Peña Blanca. Research at the Center for Nuclear Waste Regulatory Analyses is continuing to evaluate mechanisms of uraninite alteration, controls on groundwater and mineral compositions, and processes affecting elemental migration in this environment, and to apply the results in support of performance assessment modeling of the proposed Yucca Mountain repository.

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*Text is taken from Murphy, 1992, *Technology Today®,* Southwest Research Institute, San Antonio, Texas, June, p. 16-21.

For more information about the Peña Blanca Natural Analog Project, or how you can contract with Southwest Research Institute (SwRI), please contact James D. Prikryl at jprikryl@swri.org, or call (210) 522-5667.

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