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August 28, 2002

Robert Latta/Jack Parrot  
US Nuclear Regulatory Commission  
1551 Hillshire Drive  
Suite A  
Las Vegas NV 89134

Dear On-Site Representatives:

Please find enclosed the requested copy of the TRAC report. The report consists of a CD-ROM and an executive summary in hard copy form. If you have any questions, please contact me at (775) 687-3744

Sincerely,

A handwritten signature in cursive script, reading "Susan W. Lynch".

Susan W. Lynch  
Administrator of Technical Programs

Encl.

NM5507  
wm11

**From:** "MARTA ADAMS" <MAADAMS@ag.state.nv.us>  
**To:** <JDP1@nrc.gov>  
**Date:** Friday, September 06, 2002 2:49:21 PM  
**Subject:** TRAC Report

Please be advised that the report prepared by TRAC for the State of Nevada was done pursuant to a contract with the Nevada Attorney General's office.

Marta Adams  
Sr. Deputy Attorney General  
(775)684-1237

**CC:** <bloux@nuc.state.nv.us>

## YUCCA MOUNTAIN MONOGRAPH

### EXECUTIVE SUMMARY

A group of international scientists working for the past two years under the direction of Dr. Charles Archambeau, President of TRAC Corp., presented a 688- page monograph to Attorney General Frankie Sue Del Papa on July 3, 2002. The monograph fulfills a contract between TRAC and the State of Nevada for documentary evidence that may be used in a licensing proceeding before the Nuclear Regulatory Commission as proof that Yucca Mountain is an unsuitable, if not a dangerous, site for development as a repository for spent nuclear fuel rods and high-level radioactive waste. The monograph addresses in three Parts the nature, origin and ages of widespread water borne mineralization at the ground surface and deep into the vadose zone at Yucca Mountain. The topic has been the subject of controversy since the site was selected as the only site for study as a potential repository site because of its show-stopping potential.

The heart of the controversy is whether the mineral deposits were formed by upwelling hydrothermal fluids or by rainwater. The prevailing view of the Department of Energy and United States Geologic Survey researchers that advise the DOE assumes that the minerals were deposited by percolating rainfall. A suitability decision was recently made by the Secretary of Energy and approved by the President based, entirely, on this assumption. The authors of the monograph present findings that prove beyond a reasonable doubt the deposits were deposited by episodes of upwelling water which began millions of years ago and have continued essentially to the present.

The underlying concern which prompted the research project is that releases of radioactivity from the proposed facility into the biosphere may result from contact at the surface of hot, disposal canisters with significant quantities of water. If a gas- or vapor-assisted hydrothermal plume of water originating deep within Yucca Mountain were to erupt through permeable conduits that feed into the facility, the result would be the complete breakdown of the natural barriers and the rapid deterioration of the integrity of all engineered barriers. The releases of radioactivity, directly into the atmosphere, could be very large, potentially attaining catastrophic proportions.

The monograph presents evidence that has forced the DOE's researchers to concede that the mineralization was deposited by hot water. Originally they denied this fact. After the unavoidable concession, the USGS scientists fabricated a hypothesis that the interior of Yucca Mountain remained hot for at least five million years and the growth of deposits from the percolation of rainwater through a hot rock mass in the vadose zone was exceedingly slow (on the order of mm per Ma). The monograph debunks their hypothesis. As a fall back position, the DOE researchers maintain that the mineral deposits are too old to be of concern. The monograph discusses in mathematical detail why their age-dating techniques are flawed.

#### *Part I*

The authors used the extensive database compiled by the DOE in support of the anticipated licensing case before the NRC in addition to their own research. Part I, authored by Jerry Szymanski, from Las Vegas, and Dr. Tim Harper, from England, discusses late Cenozoic volcanism, thermodynamics and the active tectonic environment

of Yucca Mountain, as expressed through the present-day structural and geologic framework, and the geophysics of the area. Data from a wide range of field observations were assembled and synthesized and provide the background for the formulation of a conceptual model of the long-term behavior of the geohydrologic system underlying Yucca Mountain. Part I lays the basis for understanding how an earthquake can initiate seismic pumping of water from deep beneath Yucca Mountain to the ground surface in response to complexly coupled heat dissipative structures and dissipative fault structures in a dynamically evolving, non-equilibrium, geologic system.

The conceptual model introduced in Part I regards the horst-bounding, Paintbrush and Solitario Canyon faults, as time-dependent, planar sources of heated, gaseous fluids. The conceptual model implies that the vadose (unsaturated) zone is episodically subjected to an upward flux of hydrothermal plumes of hot, gas-charged water. These plumes originate near the transition between the brittle and ductile crust, where major earthquakes nucleate and detachment faults are present. The plumes transport accumulated heat to the atmosphere. Thus, they intermittently cool the crust by the episodic eruptions of upwelling hydrothermal water.

## **Part II**

Part II, was written by Jerry Szymanski, Dr. Harper and Dr. Yuri V. Dublyansky from the United Institute of Geology, Geophysics and Mineralogy Studies, Siberian Branch of the Russian Academy of Science, in Novosibirsk, Russia. Part II introduces a set of expectations regarding the long-term behavior of the geohydrologic system, which was derived from the upwelling model introduced in Part I. Part II introduces multiple lines of evidence, which support the authors' conceptual model and question DOE's rainwater hypothesis. The question whether the ages of the mineral assemblage can be dated with sufficient accuracy to identify the most recent episode of deposition of the minerals is addressed. Lines of evidence are presented, which interpret recently acquired mineralogic, geochemical, geochronological and fluid inclusion data from the Exploratory Studies Facility.

The conceptual model enlarged upon in Part II recognizes both ascending and descending water in the vadose zone with a minor rainwater component. It predicts a space and time evolution of the chemistry of the fluids and of the depositional environment. The model requires that the mineral-forming solutions be composed of two end-members giving rise to a mixture, which varies and dilutes in space and time. One end-member consists of reducing solutions that accumulate inside the deep-seated dilating hypocentral regions before their introduction into the vadose zone. These fluids are transported by *seismic pumping*. The other end-member consists of more oxidizing and fracture-based solutions which reside in the regional aquifer. The different environments to which the two end-members were initially exposed result in contrasting chemical, stable isotope and radiogenic isotope properties. The resulting properties of the fluids at a particular time and location are inherited by the precipitated minerals.

Calcite, quartz, chalcedony, fluorite, apatite, heulandite, strontianite, and gypsum, were deposited in the ESF during one or more hydro-tectonic eruptions. The author's conceptual model explains the observed characteristics of these minerals. The DOE's rainwater hypothesis does not provide an alternative, coherent and contradiction-free explanation for the ESF minerals.

The model implies that the fluid residing in pores in the vadose zone rock would have been introduced during one or more episodes of hydraulic mounding induced by past seismic and hydrothermal processes. These fluids have likely been introduced into the interstitial space, in the past, by dissipation and diffusion of warm or hot fluids from a hydraulic mound into the vadose zone. An eight- to ten-fold enrichment of the pore water deposits in calcium and magnesium and the enrichment in trace elements, including rare earth elements and base and noble metals, indicates a hydrothermal source consistent with the model. The pore fluids show a relative enrichment in thorium (Th) and lead (Pb) but are depleted in uranium (U). The significantly higher Pb/U and Th/U ratios imply that the pore fluid did not evolve in a near-surface environment.

The proposed conceptual model has important implications concerning the interpretation of the properties of the stable isotopes of oxygen and carbon and the radiogenic isotopes of strontium, uranium, thorium and lead in authigenic minerals at Yucca Mountain, including the ESF mineral aggregates. The conceptual model predicts the observed trends of changing stable isotope characteristics with evolving surface lithofacies, consisting of increasingly more distal (both in space and time) deposits. They include the most proximal authigenic breccia cements, then bedrock veins, followed by calcretes, and, finally, the most spatially and temporally distal "pedogenic" carbonates. The temporal variability of the  $^{234}\text{U}/^{238}\text{U}$  ratio is an essential expression of the underlying hydro-tectonic processes. The observed variability reflects the evolution of the radiochemistry of the fluids toward the present-day radiochemistry in response to changing sources of the fluid and the changing physico-chemical environment. The  $^{234}\text{U}/^{238}\text{U}$  ratios increase systematically, as the calculated apparent  $^{230}\text{Th}/\text{U}$  ages diminish. Thus, the calculated ages reported by the DOE researchers do not reflect the corresponding actual ages. The opaline deposits in particular have acquired time varying amounts of U, Th and Pb isotopes and, therefore, age dating of the opals is in violation of qualifying assumptions of the  $^{230}\text{Th}/\text{U}$  and  $^{207}\text{Pb}/^{235}\text{U}$  dating methods. The erroneous  $^{207}\text{Pb}/^{235}\text{U}$  ages have lead the DOE researchers to a variety of questionable conclusions, not the least of which are the rainwater hypothesis and its progeny.

The authors of Part II identified a present-day analog of the hydraulic mounding of water predicted by the conceptual model. It is located at the southern end of Yucca Mountain in wells drilled under the auspices of the Nye County Early Warning Drilling Program (EWDP). The EWDP drill holes revealed that the water table is at a depth of 17m from the ground surface in drill holes EWDP-1s and -1d and at a depth of 30m in drill hole EWDP-9s. These levels are 50m to 100m higher than those which were expected by hydrologists based on water tables in the area. The temperature of the water in well EWDP-1s ranges between 27°C and 30°C. This temperature is 9-12 °C higher than the mean annual surface temperature of 18°C. The chemical and isotopic properties of the water match water, which deposited minerals in the ESF and Trench 14. Thus, the mounding provides a present-day analog in support of the conceptual model. The data indicate that the anomaly is not only an expression of a local upward flux of groundwater, but suggests that the phenomenon of tectonically-controlled upwelling of groundwater is widespread in the Yucca Mountain area.

### ***Part III***

Part III was written by Dr. Dublyansky, Jerry Szymanski, Dr. Sergey Smirnov, Dr. Sergey Pashenko and Dr. G. P. Palyanova; the last three authors are also from the United Institute of Geology, Geophysics and Mineralogy in Novosibirsk. Contributions were also provided by other members of the Institute. Part III presents detailed multifaceted tests of the veracity of the authors' conceptual model and subjects the DOE's rainwater hypothesis to a critical evaluation, which renders it moribund.

Part III contains eight chapters subdivided into two general topics. The first deals with chemical and mineralogical alteration of the tuffs based on a variety of laboratory analyses of rock cores extracted from the interior of Yucca Mountain down to a depth of approximately 2.0 km. The origin of secondary minerals in the vadose zone of Yucca Mountain and at its surface is analyzed based upon the two competing conceptual models. The extensive mineralogical database established from characterization studies of Yucca Mountain permits an examination of the temporal and spatial distribution of epigenetic minerals, both at the surface and in the interior of the mountain. This four-dimensional distribution provides for an accurate analysis of the geologic record, which has been imprinted on the tuffs in response to the long-term behavior of the hydrologic sub-system.

The second topic of Part III concerns the calcite-silica deposits, which occur at the ground surface, in the vadose zone, and in the phreatic zone. The origin of these deposits is considered based on their stable and radiogenic isotope signatures, the data on fluid inclusions, the trace element signatures, and the results of thermodynamic modeling. The hydrothermal origin of the calcite-silica deposits is firmly established and the proposed conceptual model of hydrothermal upwelling water is validated.

#### Chapter 1

Chapter 1 of Part III deals with secondary mineralization of the vadose zone. Zeolite species within Yucca Mountain are distributed into two chemically distinct alteration aureoles. The alkali (Na and K) alteration aureole was produced by the Timber Mountain hydrothermal metamorphism, about 10–11 Ma ago. The Timber Mountain hydrothermal episode produced a temperature of more than 100 °C, which persisted at a depth in excess of 800-m, for a period spanning between 1.0 and 1.5 Ma and fueled the hydrothermal flow. The prolonged thermal exposure of the tuffs caused a pervasive alteration until the glass in the tuffs was depleted.

The alkaline earth (Ca and Mg) alteration aureole is younger, and overprints the pervasive Timber Mountain hydrothermal metamorphism. It has the shape of double inter-grown mushroom stems, which are rooted in the Paintbrush fault zone and in the NW-SE trending shear zone in conformity with the proposed conceptual model. The juxtaposed west and southwest pileuses of this double mushroom are located in the area of the proposed facility. The corresponding tuffs host the calcic zeolite species (stellerite, heulandite, and laumontite) in the form of both the fracture-lining minerals and the matrix alteration minerals. In addition, these tuffs also host products of the (earlier) Timber Mountain hydrothermal metamorphism such as some of the siliceous species (clinoptilolite and mordenite), orderly interstratified clays, analcime, and albite. Crosscutting relationships among the fracture-lining calcic species provide evidence for multiple episodes of the deposition of stellerite and heulandite. In addition, these

relationships indicate that these zeolite species are contemporaneous with the fracture-lining calcite-opal-quartz-fluorite assemblages.

The Paintbrush fault zone and the NW-SE trending shear zone served as pathways for the ascent of hydrothermal fluids and for the later ascent of alkaline earth solutions. Upon reaching the overlying tuff units, the water diffused into the tuff units and intermixed with the local alkali metal aquifer water. The lateral diffusion of the ascended water is expressed by a systematically changing chemistry of the altered tuffs. The highly asymmetric spatial distribution of the alkaline earth alteration aureole is thick in the north and east sectors of Yucca Mountain, where it extends to the phreatic zone. In the west and south sectors of Yucca Mountain, it is both thinner and shallower. The asymmetry creates the illusion that in the south and west sectors of Yucca Mountain the alteration and the corresponding calcite-silica vein mineralization are restricted to the vadose zone but, in fact, it extends well into the phreatic zone in the northern and eastern sectors.

The clinoptilolite K/Ar ages validate the inference that the alkaline earth alteration aureole overprints (post-dates) the alkali alteration aureole. These ages, therefore, can be regarded as recording the episodic ascension of alkaline earth solutions, from the basement into the overlying tuffs over the past 8-9 Ma.

### Chapter 2

Chapter 2 deals with the thermodynamic modeling of a hypothetical interaction of percolating, meteoric water with the rhyolitic tuffs and calcretes of Yucca Mountain suggested by DOE's rainwater hypothesis. Thermodynamic calculations by Palyanova et al. (2001) show that solution resulting from such an interaction would not precipitate smectite or any substantial amounts of Ca-Na zeolites. Ca and K, in alteration assemblages, would be concentrated in carbonates, accessory zeolite, authigenic muscovite, and microcline. The formation of such mineral phases as calcite, quartz, amorphous silica, fluorite, and other mineral assemblages, which are observed in the Yucca Mountain vadose zone, would be thermodynamically impossible. The DOE's conceptual alternative to the proposed hydrothermal upwelling model is physically impossible and must necessarily be discarded as an invalid hypothesis.

### Chapter 3

Chapter 3 evaluates the alluvial, colluvial and soil deposits on the surface of the environs on and around Yucca Mountain, which are extensively coated and cemented with calcium-bearing deposits. In some places, horizons of eolian sand and coarse colluvium, which may be two to three meters thick, are cemented with micritic calcite and to a lesser degree by opaline silica. Transport of the minerals to the topographical surface are variously explained by DOE researchers as the result, solely, of pedogenic processes associated with rainfall while the authors discuss evidence that validate their view that the carbonate-silica minerals were formed by the discharge of water that ascended along fault zones from great depth before being influenced by pedogenic processes.

The calcite-silica deposits at the topographic surface of Yucca Mountain are expressed in the form of four morphologically distinct members, or lithofacies. These are the so-called AMC (authigenic-mineral-cemented) breccias, veins in the bedrock, slope-parallel calcretes within and on top of the overburden, and carbonate coatings of alluvial and colluvial clasts. In the sequence, each lithofacie is a more distal (in terms of time and

space) derivative of the previous one. In addition, since emplacement of all lithofacies occurred at or near the topographic surface, all of the lithofacies are likely to be affected, in some degree, by supergene (pedogenic) processes, which increase from proximal to distal facies.

The calcite-silica deposits occur along youthful faults, which are readily recognizable in the field by displaced tuff units, slickensided surfaces of scarps, and brecciated and mineralized zones. In places, where faults are exposed by trenching, the deposits occur in the form of sub-vertical seams or veins along or near the fault planes, which grade into sub-horizontal aprons, or layers of calcrete at the ground surface. Several of the deposits are located along major faults: the Trench 14 and Busted Butte exposures along the Paintbrush fault zone (including the Bow Ridge fault); the WT-7 exposure along the Solitario Canyon fault zone; and the so-called Wailing Wall exposure along a splay from the Stagecoach Road fault system.

It is known with certainty that the deposits are of a late Quaternary age based on their directly observable stratigraphic relationships, therefore, the origin of the deposits will be a critically important issue in a licensing proceeding.

All lines of evidence in support of the contention that the calcite-silica deposits are products of infiltrating rainwater were examined. None was shown to have scientific merit. In this regard the progressive and concurrent changes in slope carbonates as a function of distance away from fault zones hosting the Paintbrush and Solitario Canyon geothermal anomalies were persuasive. These changes involve the macroscopic textures and the SiO<sub>2</sub>-abundance. These lines of evidence alone validate the authors' conceptual model.

Several independent lines of evidence suggest that some of the calcite-silica deposits were formed under reducing conditions transported by hydrothermal fluids. These are the presence of iron-sulfides (FeS<sub>2</sub> and CuFeS<sub>2</sub>), the absence of the negative Ce anomaly, and the predominantly CH<sub>4</sub> composition of gases trapped in water-rich fluid inclusions in calcite.

#### Chapter 4

Chapter 4 discusses the epigenetic mineralization expressed as mineral crusts lining the walls of open fractures and lithophysal cavities. These deposits consist of two distinct generations of minerals. The earliest comprises minerals deposited during the vapor-phase diagenetic alteration of tuffs. The later generation, deposited by mineralized fluids, overgrew the earlier vapor-phase alteration minerals.

The authors' observations of secondary minerals in fracture and lithophysal cavity linings from the Exploratory Studies Facility (ESF) and the east-west cross-drift (ECRB), is supplemented, where appropriate by the analysis of data published in the scientific literature. Observations were performed on thin and thick sections of hand specimens by means of optical microscopy, as well as on crystals and fragments of mineral aggregates by means of scanning electron microscopy (SEM) and with an energy-dispersive X-ray spectrometer (EDS). The chemistry of minerals was studied by means of an electron microprobe (EMP) with special focus on the compositional variations in different growth zones of calcite and fluorite. Trace- and rare-earth elements in a number of fluorite



samples from several ESF locations were studied by instrumental neutron activation analysis (INAA).

The method of study was ontogenic mineralogy, which addresses the origin of mineral individuals and aggregates with the objective of reconstructing the complete history of their mineral development starting from the act of nucleation to the latest stages of development. The method is widely used for reconstructing the history of mineral deposition based on principles of crystal growth.

The data demonstrated that the deposition of secondary minerals in open cavities in the rhyolite tuffs of the vadose zone occurred when these cavities were filled, partly or entirely, with water. This environment persisted for periods of time sufficient for the crystallization of relatively large (centimeter-sized) crystals.

The features observed in the secondary minerals at Yucca Mountain are explained by deposition in a short-lived, ~30 to 90°C temperature, hydrothermal system. Upwelling water in such a system would have acquired a mineral load within the Paleozoic carbonate sedimentary rocks and at deeper levels. The mineralogic features are thus compatible with the authors' conceptual model.

A critical evaluation of DOE's hypothetical deposition of secondary minerals from films of meteoric water percolating along interconnecting fractures in the vadose zone, in view of the ontogenetic analysis of different features of the mineralization led the authors to conclude that the hypothesis does not withstand scrutiny. Many processes that are postulated by the rainwater hypothesis are physically impossible (e.g., the crystallization of large euhedral crystals and crystals with scepter morphology from water films). The mineral compositions of crusts, particularly the presence of quartz, fluorite, heulandite and strontianite, are not compatible with the deposition of minerals from meteoric waters. The geochemistry of minor elements (notably, the elevated abundances of Mg, Sr, and As) are also incompatible.

An important aspect of the mineralogy of the deposits involved the colloidal nature of opals. Data reported in the published literature in connection with detailed work, relating to U-Th and U-Pb age dating, indicated that opals from the ESF typically contain between 50 and 280 ppm uranium, whereas the contents of uranium in co-genetic calcite range between <0.01 and 0.5 ppm. The conspicuous uranium enrichment of opals, and in some cases chalcedonies, requires at least two conditions:

1. Opals and chalcedonies should have formed from colloidal solutions; and
2. The mineral-forming fluids must have carried 5 to 100,000 times the content of U in modern waters from the Yucca Mountain region (consistent with the conceptual model).

More evidence regarding the origin of these deposits were obtained during radon, stable isotope and fluid inclusion studies described, *infra*.

### Chapter 5

Chapter 5 deals with the stable isotopes of strontium, oxygen and carbon.

Early studies of the drill core demonstrated that the calcite-silica minerals in the Yucca Mountain subsurface occur in the form of two distinct isotopic facies, and that the

vadose-zone deposits differ from those in the phreatic zone in terms of their textures, trace element compositions, and isotope properties.

The analyses of stable isotopes became a very powerful tool in the Yucca Mountain studies when micro sampling techniques were employed instead of less discriminating techniques, and a more detailed record was obtained from different parts of the mineral crusts. The authors employed a method of Gas Chromatography – Isotope Ratio Mass Spectrometry (GC-IRMS). This *in situ* technique permitted a spatial resolution of about 90-200  $\mu\text{m}$ . Employed in combination with the appropriate software, this fine-resolution method permitted the authors to perform an "isotopic mapping" of a number of samples.

Samples taken from different parts of the repository block and from dissimilar fracture and lithophysal cavity deposits exhibited remarkably similar behavior. The  $\delta^{13}\text{C}$  ratio gradually decreased while the  $\delta^{18}\text{O}$  ratio increased in response to crystal growth. The variability of the isotopic ratios within individual samples was, generally, quite large. The variability of  $\delta^{18}\text{O}$  ranged between 2 to 6 ‰, whereas for  $\delta^{13}\text{C}$  the range of variations was as large as 12-16 ‰. Each sample exhibited an evolution of crystal morphology. Early calcite was represented by granular, platelet, platelet+scepter, or blocky morphologies. The latest in the sequence was blocky calcite associated with opal—universally recognized as the latest member of the mineral paragenesis at Yucca Mountain.

The heavy  $\delta^{13}\text{C}$ –light  $\delta^{18}\text{O}$  values ( $\delta^{13}\text{C}$  of 8 to 10 ‰ PDB and  $\delta^{18}\text{O}$  of 9 to 10 ‰ SMOW) are typical of the earliest parts of the calcite from the central part of the repository block. These values, however, are not uniquely related to a specific morphologic type of calcite but are found in granular, platelet-bladed, and blocky (without opal) calcite. The heavy  $\delta^{13}\text{C}$ –light  $\delta^{18}\text{O}$  calcite does not represent a specific mineral-deposition event; rather, it is one end-member of a continuum, evolving as crystals grew, toward the light  $\delta^{13}\text{C}$ –heavy  $\delta^{18}\text{O}$  ( $\delta^{13}\text{C}$  of -5 to -8 ‰ PDB and  $\delta^{18}\text{O}$  of 13 to 17 ‰ SMOW) end-member. This resulted in a prominent negative correlation between  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$ .

The spatial scale of the isotopic evolution described above varied within at least one order of magnitude. Complete transitions from heavy  $\delta^{13}\text{C}$ –light  $\delta^{18}\text{O}$  to light  $\delta^{13}\text{C}$ –heavy  $\delta^{18}\text{O}$  calcite were documented in a 1.2 mm-thick mineral crust as well as across a 1.4 cm-thick crust. The latest, in the paragenetic sense, blocky calcite with opal had characteristically light  $\delta^{13}\text{C}$ –heavy  $\delta^{18}\text{O}$  properties. Inside this group, however, a negative correlation between  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  was absent.

The DOE researchers interpret the evolving  $\delta^{18}\text{O}$  properties of secondary calcite from the vadose zone as reflecting an increasing temperature with depth from the surface and, at a given depth, a decreasing temperature as a function of time. They interpret the evolving  $\delta^{13}\text{C}$  properties as reflecting changes in the vegetation at the mountain's surface.

The authors believe the  $\delta^{18}\text{O}$  ratio reflects the changing temperatures of the mineral-forming fluids, at least for some of the morphologic types of calcite. The  $\delta^{18}\text{O}$  ratio, however, is not likely a function of the temperature in the latest morphologic type – blocky calcite with opal.

Changing vegetation does not provide an explanation of the  $\delta^{13}\text{C}$  pattern. The authors interpret the decreasing  $\delta^{13}\text{C}$  ratios of calcite with time as reflecting the overall

change of the redox properties of the depositing fluids. Early fluids injected into the vadose zone came from the deep anoxic parts of the Earth's crust where they resided for substantial periods of time. In such an environment, the isotopic fractionation and enrichment of the dissolved oxidized carbon with "heavy" carbon isotopes may have reached advanced stages. These fluids, upon reaching super-saturation with respect to calcite, deposited  $\text{CaCO}_3$  with heavy positive  $\delta^{13}\text{C}$  ratios. The conceptual model developed in Parts I and II, requires that a more-prolonged circulation of fluids, which inevitably involves oxidized waters of the upper part of the hydrosphere, follow the initial "injection" of the deep-seated fluids. The increasing role of such oxidized waters, which would carry "lighter"  $\delta^{13}\text{C}$  ratios, they believe, are responsible for the gradual decrease in the  $\delta^{13}\text{C}$  ratio of the calcite deposited by such waters.

#### Chapter 6

Chapter 6 describes the fluid inclusion studies. It presents, perhaps, the most definitive proof of the conceptual model. The effort by the DOE and USGS researchers to force fit a rainwater hypothesis to describe the hot saline water in the inclusions is reduced to nothing more than nonsense.

The fluid inclusion record includes thousands of measurements obtained from several tens of samples by independent research teams. Two-phase fluid inclusions suitable for thermometry were found in secondary calcite, fluorite and (rarely) quartz from the proposed repository horizon in the ESF and ECRB. Inclusions in all three types of minerals yielded consistent homogenization temperatures ranging from  $85^\circ\text{C}$  to as low as  $35^\circ\text{C}$ . Mono-phase all-liquid inclusions suggesting temperatures less than approximately  $35\text{-}50^\circ\text{C}$  were also found.

The highest fluid inclusion temperatures were found in the northeastern part of the ESF near the north portal and along the north ramp, as well as in the southeastern part (south ramp). The coolest temperatures were measured in samples from the area near the north bend (north central part of the ESF). The highest fluid temperatures were found in zones where more concentrated fluid flow should have occurred (i.e., close to the horst-bounding Paintbrush fault located east of the Bow Ridge fault) and the lowest temperatures were found in the zone where a diffuse lateral flow is expected (close to the central part of the repository block).

The paleo temperatures measured in the fluid inclusions indicate the presence of very high paleo heat flows in the proposed repository area, which is only possible in the setting of a hydrothermal system. Like the homogenization temperature spatial zonation, the calculated paleo heat flow field was seen to exhibit a strong east-west gradient with values increasing in the direction of the horst bounding Paintbrush fault.

The measured salinities of the mineral forming fluids ranged from  $<1,800$  ppm to 21,000 ppm, which encompasses the three categories – fresh, brackish and saline waters. The measured salinities are inconsistent with a meteoric source for the depositing fluids.

Gases trapped in inclusions were analyzed by means of bulk methods (mass spectrometry, gas chromatography) and as individual inclusions (crushing, selective adsorption, Raman spectrometry, and gas chromatography). Inclusions were found to contain significant quantities of methane and, possibly, some quantities of heavy

aromatic hydrocarbons. The logical source of these gaseous hydrocarbons is the Paleozoic sedimentary rocks underlying the Yucca Mountain rhyolitic tuffs.

Overall, the gas chemistry data are readily explained by a model whereby thermal fluids acquire dissolved hydrocarbons during their upwelling through the Paleozoic carbonate rocks underlying the Yucca Mountain rhyolitic tuffs. If the gas chemistry data is considered in conjunction with the elevated fluid inclusion temperatures, the inference regarding the phreatic hydrothermal origin of the minerals is compelling and incompatible with a postulated rainwater origin.

### Chapter 7

Chapter 7 departs from considerations of the origin of the calcite-silica deposits and returns to the question of their ages. The authors present a model, which describes the role of radon and colloids in the accumulation of additional radiogenic lead in opals from Yucca Mountain. This additional lead is in excess of the *in situ* decay of U and Th in the opals and as a consequence may cause significant distortion of the age dates computed by DOE and UNLV researchers using the "conventional" U-Pb radiometric dating equations. The authors point out that the U-Pb method cannot be used for dating minerals that: (a) crystallize in large (~1 cm) open cavities; (b) crystallize from, or in the presence of colloidal solutions, and (c) are young (Miocene or younger). Their model is described in detail in the chapter. Calculations derived from the model show that if the "conventional" equations of the U-Pb dating are applied to the minerals that are constrained as noted, the resulting ages will appear excessively older. For example, minerals formed 1000 years ago may produce "conventional" U-Pb radiometric ages of several million years.

The authors submit that minerals growing in an open cavity are exposed to a flux of additional radiogenic Pb isotopes, which are not accounted for by the "common lead" correction used in conventional calculations. The parents for these isotopes are U and Pb, which reside in the surrounding rock. The decay chains of U and Pb contain radon. Being a gas, the radon readily diffuses into the cavity ("radon emanation"). After several decays, radon produces stable isotopes of Pb, which take up residence in the fluid filling the cavity. The authors' modeling shows that concentrations of this Rn-derived radiogenic Pb in relatively large cavities will be substantially higher than in thin (< .1 mm) fractures.

The Rn and Pb resulting from its decay in the fluid is transported by diffusion and becomes adsorbed on the surface of the growing mineral. The authors calculated the characteristic diffusion fluxes of Rn-derived Pb on the surface of opals, assuming the rock contains 5ppm of U and 14 ppm of Th (typical values for Yucca Mountain rhyolitic tuffs), and found that this mechanism alone could produce over a period of time on the order of 1Ma, in equivalent amounts of radiogenic Pb comparable to the amounts of Pb present in opal specimens from Yucca Mountain.

Opals are commonly formed through the coagulation of colloids. This process is considered responsible for the formation of the uraniferous opals (Yucca Mountain opals with 200+ ppm of U fall into this category). If colloidal solutions are present in the cavity, the accumulation of Rn-derived Pb isotopes occurs on the micelles, which possess a very large surface area. Upon coagulation and sedimentation, the micelles become

incorporated in the opal. The mechanism responsible for the concentration of the "excess" radiogenic Pb on the micelles is orders of magnitude more efficient than the net diffusion mechanism referred to above. The authors' calculations show that concentrations of Pb on the order of hundreds of ppb, typical of the Yucca Mountain opals, may be acquired by micelles of silica over a period of several days. Controlled by the velocity of water exchange in the cavities, the absolute quantities of Pb contained in the Yucca Mountain opals from individual cavities could accumulate within 100 to 1000 years.

The authors concluded, after a detailed analysis, that the numeric isotope data reported by the USGS researchers are of a high quality, but in both the USGS and UNLV age dating efforts the use of "conventional" equations for the U-Pb dating without first having been determined to be applicable in the specific setting (i.e., the deposition of opals in cavities from colloidal solutions) was inappropriate. The tuffs at Yucca Mountain do contain U and Th and the emanation of Rn does occur, and therefore, additional Rn-derived Pb must be present in the system. Therefore, the ages calculated without accounting for the additional Rn-derived lead cannot be accurate.

It is important to note that the Rn derived Pb is in addition to the contribution of Pb and other daughters of U carried by the mineral depositing fluids described in Part II. The two effects, taken together, cause the age dating results of the DOE researchers to overstate the actual ages by up to millions of years.

### Chapter 8

Chapter 8 considers the source of energy that heated the waters that deposited minerals at hydrothermal temperatures in the vadose zone. The application of thermal modeling techniques to establish the extent and timing of the heating of the vadose zone in the vicinity of the proposed repository by a cooling magma chamber that resided under the Timber Mountain Caldera was considered. Problems associated with the quantitative thermal modeling of the magma chamber and the thermal regimes of the Yucca Mountain vadose zone were addressed.

The high-level magma chamber just north of Yucca Mountain continually replenished heat and matter by discharging eruptions for almost 5 million years (between at least 14.9 and 10 Ma). The preheating of the Earth's crust before the formation of Yucca Mountain (12.7-12.8 Ma) lasted for about 2 million years, and certain magma gains occurred for almost 3 million years, thereafter. Unfortunately little is known with any degree of certainty regarding the volumes of magma involved, the compositions, depth, shapes of magma chambers, rates of magma convection, or differentiation. This lack of information is the major source of uncertainty in any attempt to employ quantitative thermal modeling for evaluating the thermal history of Yucca Mountain.

The authors were forced to conclude that gaps in relevant information regarding important input parameters and boundary conditions, such as gains and losses of igneous heat in the vicinity of Yucca Mountain, and potential uncertainties associated with these gaps were too large to produce reliable numeric results. In addition, the geometry of the model was not favorable for obtaining reliable solutions. Under such circumstances, any results of numeric thermal modeling must be interpreted with extreme caution. They

rejected thermal models proposed by the DOE researchers because they made unreasonable assumptions in their selection of input parameters.

The ultimate verification of the results of thermal modeling must come from establishing consistent trends with the known thermal history of the area, which have been independently determined on the basis of the geologic record. Two thermal events are considered well established with respect to the Yucca Mountain geological history. The first event was the deposition of the ash-flow tuffs, which built up layers of the mountain. Following this deposition approximately 12.7 Ma ago, the tuff "pile" was hot, and cooling to ambient temperatures took time. The second was the so-called Timber Mountain Caldera hydrothermal event that occurred ~10-11 Ma ago (e.g., Bish and Aronson, 1993). It is thought that a hydrothermal convection system was set off by a granitic magma body, which resided under the Timber Mountain caldera, some 8-10 km to the north of Yucca Mountain. The Timber Mountain event is held responsible for pervasive zeolitic (alkali), montmorillonitic and carbonate alteration of the rhyolitic tuffs and the deposition of abundant calcite and silica below a depth of  $>1.2$  km under the surface of Yucca Mountain.

The authors concluded that the secondary minerals sampled in the ESF cannot be related to either of the two thermal events that are known to have occurred at Yucca Mountain because the appearance of fluids with elevated temperatures in the vadose zone of Yucca Mountain occurred after both had long been exhausted. The authors' conceptual model provides a coherent explanation of all of the observed features of secondary mineralization at Yucca Mountain. According to the model the source of heat is the deep-seated part of the Earth's crust. Heat accumulated there was transported advectively to the vadose zone of Yucca Mountain and to the topographic surface by thermal waters welling up along fault-associated permeable zones.

#### **CONCLUDING OBSERVATION**

Based upon the findings and conclusions reached in the monograph, the authors are of the opinion that the DOE's application for a license to receive and dispose of spent fuel and high-level radioactive waste at Yucca Mountain will be denied by the Nuclear Regulatory Commission. Their opinion assumes that the issues will be vigorously litigated before a fair and impartial licensing board empaneled with administrative law judges that permit a full explication of the scientific facts addressed in this monograph. In such a forum, politics is expected to defer to science rather than overshadowing it, as has been the case to date.