THE ORIGIN AND HISTORY OF ALTERATION AND CARBONATIZATION OF THE YUCCA MOUNTAIN IGNIMBRITES

Volume I

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**Las Vegas, Nevada
April 1992**

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This report was prepared by Jerry S. Szymanski while still employed by the U.S. Department of Energy, Yucca Mountain Project. The report presents information not published, presented or discussed by the Department. The State of Nevada believes that this report raises further questions about the viability of Yucca Mountain, and thus should be read by all parties interested in and concerned about high-level nuclear waste.

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COMMENTS ON "THE ORIGIN AND HISTORY OF ALTERATION AND CARBONATIZATION OF THE YUCCA MOUNTAIN IGNIMBRITES"

This letter is to document my current views regarding the subject report. Having read each section as it has been developed, and a more recent version in its entirety, I wish to compliment you on the results of your efforts. This report is worthy of the most thorough and detailed reading by all parties interested in and concerned about high-level nuclear waste. I find that the technical and logical analyses are sound and cannot be proven wrong to any significant degree on the basis of the data presented. From my familiarity with the presently available geochemical data, I find that there are no significant omissions. The field, petrological, and geochemical evidence all demonstrate the fact that the tuffs of Yucca Mountain have been epigenetically and metasomatically altered by hypogene fluids. Indeed, some of the scientists of the Yucca Mountain Site Characterization Project (YMP) have recognized this for a number of years. This fact cannot be contested. The geochronological data indicate that this alteration has occurred repeatedly since the time that the tuffs were deposited in the Miocene and has continued through the Pleistocene and, most likely, into the Holocene. This latter point is, however, contested in various ways by a number of the project scientists.

That metasomatic alteration is not trivial at Yucca Mountain can be made even more compelling by examining the full suite of major elements reported in Tables A and E of Broxton et al., 1986 (Los Alamos National Laboratory Report LA-10802-MS). I have taken the liberty of doing so and include with this letter histograms and cross-plots of those reported major element analyses common to both existing glass and altered tuff at Yucca Mountain. Upon examination, the histograms indicate the following:

1. A substantial portion of the altered rocks are 4 to 10 percent lower in silica than is glass.
2. Most altered rocks are 0.1 to 0.5 percent richer in titania than is glass.
3. Most altered rocks are richer in alumina by at least 0.5 percent than is glass, and a substantial portion is richer by 2 to 5 percent.

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4. Most altered rocks are richer by 0.5 to 4.5 percent of iron oxide than is glass.
5. Most altered rocks are richer by 0.25 to 1.75 percent of magnesia than is glass.
6. Almost all altered rocks are 0.5 to 5 percent richer in lime than is glass.
7. Most altered rocks are 0.5 to 2.5 percent lower in soda than is glass, though a few are enriched by 2.5 percent.
8. Most altered rocks are lower in potash by 0.5 to 3 percent than is glass, although a few are richer by 2.5 percent.

The same relations can be seen on the cross-plots of silica versus alumina, iron oxide versus titania, lime versus magnesia, soda versus potash, and lime versus iron oxide. It may be possible that the strong correlation of iron oxide and titania for altered tuff is the result of an unrecognized analytical interference. Even so, because the glass represents the original composition of the tuffs, it is clear that silica, soda, and potash have been depleted by the altering solutions while iron oxide, alumina, magnesia, and lime have been enriched. As has been noted by others, this is a case of open system chemical alteration or, in the jargon of petrologists and economic geologists, metasomatism. It is clear that fluids have carried these enriched components from reservoirs other than the tuffs of Yucca Mountain or other rocks of similar composition. An important point is that both the altered tuffs and the veins (even some breccias), which occur epigenetically in the tuffs, are calcic-magnesian in character. This establishes that both have a similar or common genesis.

Though several project scientists state that the alteration is due to early deuteric processes during the cooling stages of the ash flow eruptions or to later diagenesis by locally infiltrating rainfall, I have neither heard of nor seen any compelling evidence that these more benign processes are responsible for the totality of alteration at Yucca Mountain. Therefore, from a public safety point of view, the more hazardous scenario of an episodic hypogene genesis ought to be seriously addressed. This is especially true for those alteration phases which yield Pliocene or Quaternary ages by isotopic and/or stratigraphic analyses.

Other data presented by Broxton et al. indicate that the alteration products of the tuffs, namely zeolites, vary substantially in their chemical composition. As you have so clearly noted in your report, this indicates that the altering solutions have come from more than a single source. That is, most certainly, evidence for polygenesis and is to be expected in a complex, nonlinear geodynamic system such as southern Nevada. It occurs to me that the widely acknowledged fact of metasomatic alteration at Yucca

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Jerry S. Szymanski

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Mountain must be the keystone of any geologically-based suitability analysis. It is my understanding that none of the currently employed hydrological models used by Yucca Mountain scientists include the effects of geothermal and tectonically-induced upwelling.

As I have severely criticized your efforts in the past, I am pleased to note that this report does not ". . . seriously, if not grossly, underestimate . . ." the conditions at Yucca Mountain. Your report is a clear and compelling logical analysis of contrasting opinions regarding the nature and degree of epigenetic alteration of the site. Those with unvested and unbiased interests in and concerns about high-level nuclear waste should concede that your arguments are sound and compelling. These concerns will persist!

The analyses and conclusions contained in this memorandum are my own. They have not received technical or policy review, and should not be interpreted as representing the policy of YMP or the Office of Civilian Radioactive Waste Management:

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○ Altered Tuff
□ Glass

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○ Altered Tuff
□ Glass

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ABSTRACT

In the Great Basin, the flow of terrestrial heat through the crust is effected in part by the flow of fluids (magma, aqueous solutions and gases). At Yucca Mountain, the role of fluids in crustal heat transport is manifested at the surface by youthful calcretes, sinters, bedrock veins, hydrothermal eruption breccias and hydrothermal alteration. The main focus of the present work is the subsurface, whence specimens of vadose-zone interstitial fluid, well water in ignimbrite and carbonate horizons, glasses and altered ignimbrites, alteration minerals, calcite veins and fluid inclusions have been analyzed. The results are integrated to assemble a four-dimensional image of the post-Miocene bathing experienced by the Yucca Mountain ignimbrites. The earliest hydrothermal episode produced alkali earth metasomatism (Ca + Mg ~10-25%, cf. ~ 3% for glass) which is pervasive in the lower part of the stratigraphic section. Associated alteration minerals include clinoptilolites with K/Ar ages ranging from 9.5 to 10.5 million years, contemporaneous with the Timber Mountain caldera. Both the strontium isotopic ratio (~ .709) of whole-rock samples and spatially correlative calcites, and the carbon isotopic ratio ($\delta^{13}\text{C}$ from -2 to 4.5 per mil wrt PDB) of the calcites are indicative of parent fluids having resided in the underlying Paleozoic carbonates. Hydrothermal activity subsequent to the Timber Mountain episode differs spatially, chemically, isotopically, and in duration. The more recent metasomatism observed higher in the stratigraphic section is less pervasive, and appears to be confined to aureoles typically associated with faults and fractures. The whole-rock Ca + Mg substitution is greater, ranging above

50%. Clinoptilolites have (mixed) K/Ar ages ranging from 2 to 8.5 million years. The strontium isotopic ratio ($\sim .712$) of whole-rock samples and spatially correlative calcites, and the carbon isotopic ratio ($\delta^{13}\text{C}$ from -10 to -3) of the calcites are both indicative of parent fluids from deep-seated sources, the Precambrian basement and igneous gas respectively. In contrast to the prolonged (1 million year) Timber Mountain metasomatism, hydrothermal flow over the past 8.5 million years has been intermittent, has spanned a much greater depth range, and has been associated with faults and fractures. Remnants of late hydrothermal fluid residing in vadose-zone pore space have been separated from cores from two shallow dry-drilled boreholes. Relative to well water in the ignimbrites, this interstitial water is strongly enriched not only in alkali earth elements, but also in transition, base and noble metals, and rare earth elements (REE), indicative of hydrothermal origin. In addition to the overall enrichment of REE, there is an unusual enrichment of heavy REE relative to light REE that is not shared by the host ignimbrites. Relative enrichment of heavy REE is observed elsewhere for hydrothermal solutions that are concentrated and rich in CO_2 ; carbonate anion complexing is the mechanism believed responsible. The chemistry of the interstitial fluids is comparable with that of the mosaic breccia cements. Similar solutions have evidently caused explosive fragmentation of bedrock associated with the late carbonatization, in addition to late alkali earth metasomatism and calcic zeolitization. Over the last several hundred thousand years, episodes of calcite emplacement, contemporaneous with local mafic volcanism, have occurred at intervals that are not long in comparison with the isolation time required for a HLRW repository. This site

condition poses a hazard that cannot be mitigated and reduced to prudent and acceptable levels.

1.0 Introduction

The main purpose of this report is to test an independently formulated expectation that, at Yucca Mountain, there is intermittent diffusion of deep-seated solutions into the local vadose zone. This expectation has been formulated as a result of interpretation of a spate of geodynamic data and observations concerning contemporary in-situ stress and hydrothermal conditions, Szymanski (1989). This interpretation has been subjected to a two-year review by the Special DOE Review Panel. The overall results of this review may be accurately summarized by the following quotation from Appendix I of Archambeau and Price (1991). *"The JS model, as interpreted by the panel in its position statements on key processes (which is based on tectonically- driven upwelling fluids) is considered a valid hypothesis by all panel participants. There was divided opinion on whether the field evidence confirms that the water table has reached the ground surface in the geologically-recent past. The key evidence to consider is the vein and fracture-filling material throughout the unsaturated zone at Yucca Mountain."* Similarly, Price (1991) has made the following statement. *"The members of this committee are in complete agreement regarding the Szymanski Model: they consider that, beyond reasonable doubt, the model is generally valid. Where we did not reach agreement is with regard to the magnitude of the effects that this model will engender."*

The evident absence of consensus regarding the magnitude of the expected long-term hydrologic effects is the main factor that motivated the research undertaken herein. The following quotation from Archambeau and Price (1991) describes the adopted investigatory approach. *"The second approach is fundamental, and that is to evaluate geologic data from the area to look for evidence of past upward flow and mineral deposition. Broadly speaking, one would expect to see evidence of relatively rapid depositions of minerals in veins and, in some cases, along the surface, with such deposition being episodic and strongly correlated with near vertically oriented fracture zones and faults since the flows are considered to be triggered by relatively infrequent major tectonic events and to be focused in, if not largely confined to, highly fractured or faulted zones. In addition, the mineralization itself should reflect the chemical composition of the water and strata at depth; in the case of Yucca Mountain a major constituent should be calcium from the paleozoic limestones beneath the mountain. Likewise, the isotopic character of the mineral deposits should be compatible with water at considerable depth; or at least compatible with a mixture of the water from greater and shallower depths since the isotopic properties of the water vary with depth. We should also expect mineral deposition in veins to be observed throughout the depth section above the present water table, as well as on the surface, if major up-welling of water has occurred frequently in the past. Further, the ages of such minerals should span a considerable period, from at least several millions of years ago up to essentially the present. In addition, in any restricted area, the ages*

should be distributed in a reasonably uniform manner if we are to have confidence in the 'frequent' occurrence of this up-welling depositional process and its extrapolation to future occurrences. Finally, one might expect to see evidence of hydrothermal alteration and brecciation of the country rock along faults and in fracture zones, particularly if volcanism has in the recent past been responsible for upward intrusions of water. Again, the ages of any such occurrences would be critical in assessing whether the processes responsible for such high temperature phenomena are still active."

The subsequent considerations of the long-term behavior of the Yucca Mountain hydrosphere, as expressed in the local geologic record, are presented in four distinct parts. The first part, Section 2.0, is concerned with the contemporary geodynamic configuration of the Yucca Mountain region. In this part, radiometric ages of local mafic and rhyolitic magmatic suites are considered together with a variety of seismic velocity data. The main focus is on developing an understanding of the observed evolutionary trends pertaining to local geodynamic developments during the last 10^7 years. This understanding, in turn, is used to evaluate (1) geothermal conditions of the region during the last 10^7 years, and (2) the potential for the local occurrence of hydrothermal processes at the present time. The second part, Sections 3.0 and 4.0, deals with the results of direct field examinations of surficial outcrops and rock cores extracted from the Yucca Mountain boreholes. In this part, an attempt is made to construct a picture that

would have emerged in the mind of a reasonably competent and sincere geologist, say, after a few weeks-long reconnaissance of the Yucca Mountain area. The third part, Sections 5.0 through 8.0, is concerned with a gamut of geochemical, mineralogical, and isotopic data obtained from a variety of samples of local fluids and various authigenic minerals. In this part, four different and broad issues are considered. These are: (1) the origin of the vadose zone interstitial fluids, Section 5.0; (2) the origin and age of the Yucca Mountain "mosaic" breccias, Section 6.0; (3) the origin and age of the Yucca Mountain zeolitic and montmorillonitic alteration, Section 7.0; and (4) the origin and age of the Yucca Mountain carbonatization, Section 8.0. The overall conclusions are presented in the fourth part, which is Section 9.0.

2.0 Volcanic Activity in the Yucca Mountain Region, and Geothermal Implications

2.1 General

In many tectonically active regions of the world, volcanism is closely associated with and, often for prolonged periods of time, followed by hydrothermal circulation of fluids. A common causative factor for both of these processes is excessive terrestrial heat flow. In tectonic settings similar to that of Yucca Mountain, the onset of orogenic activity is typically preceded by an increased conductive and advective heat input from the upper mantle into the crust. This heat input augments the intracrustal radiogenic heat production, and ultimately leads to excessive heating of the crust. After a certain threshold is attained (sufficiently high geothermal gradient), thermal disequilibrium is locally introduced into the crust, and heat transport mechanisms other than conductive heat flow become operational. One such mechanism is volcanism (advective heat transport involving molten rock), and hydrothermal activity (convective heat transport involving crustal fluids) is another. Because of commonly occurring linkage between these two processes, the review of local volcanism is a logical first step in addressing the issue of hydrothermal activity and its history. This review may provide a useful background and perspective for the subsequent hydrothermal considerations and interpretations.

Review of volcanic activity around Yucca Mountain is presented in four parts. Section 2.2 deals with current views regarding the origin of rhyolitic and mafic magmas. Locally, both of these magmatic suites, as well as their various derivatives, are intrinsic elements of the latest geodynamic developments. The distribution and geologic characteristics of the local basaltic and rhyolitic magmatism, during the last 10^7 years, are briefly discussed in Section 2.3. Considerations regarding radiometric ages of the corresponding igneous rocks are also included. In this section, the main focus is on formulating a conceptual understanding of the contemporary geodynamic configuration of the Yucca Mountain region. Section 2.4 is concerned with the local P-wave velocity structure. The geographic distribution of P-wave velocities are considered for both the lower crust and the upper mantle. The main purpose of these considerations is to test the conceptual understanding of the contemporary geodynamic configuration, as inferred from observations concerning the character of local magmatism. Finally, in Section 2.5, an attempt is made to identify geothermal implications that stem from this conceptual picture, formulated and tested independently. In this section, the main focus is on evaluating the possibility of fairly young hydrothermal activity at Yucca Mountain.

2.2 The Origin of Basaltic and Rhyolitic Magmas

During the past several decades, significant advances have been made in enhancing our knowledge of the melting and fractionation behavior of materials comprising the crust and the upper mantle. From these advances, a general consensus has emerged as to both the processes involved in the generation of mafic and rhyolitic magmas and the nature of parent materials for these magmas. The following three quotations are believed to provide an accurate representation of the current understanding.

While commenting upon the origin of basaltic magmas, Green and Ringwood (1969) made the following statement. *"Although a number of processes have been advocated as the prime cause of melting in the upper mantle (e.g., relief of pressure or localization of energy during earthquakes, accumulation of radioactive heat, etc.), the authors favor a mechanism related to 'convective' or 'advective' movement in the gravitationally unstable zone of the upper mantle (i.e., <200-300 km depth). Under suitable rheological conditions, a source mass of solid pyrolite may begin to rise diapirically (in the manner of a salt dome) within the upper mantle, probably beginning from the Low Velocity Zone. The rising diapir is large enough in relation to its velocity that it cools adiabatically and does not interact appreciably by thermal conduction with the surrounding mantle. The adiabatic gradient, approximately 0.3° Celsius/km, is much smaller than the gradient of the pyrolite*

solidus, approximately 3° Celsius / km, so that partial melting may occur in the diapir as its P-T curve intersects the pyrolite solidus. The degree of partial melting increases as the crystal and liquid diapir rises further, although the P-T curve is deflected from the adiabatic gradient by the effects of latent heat of melting. Up to this stage, although the amounts and compositions of crystalline and liquid phases change with changing pressure, it is considered that chemical equilibrium is maintained between liquid and residual crystals of the parent pyrolite; i.e., the basaltic liquid is continually buffered by residual olivine and pyroxene. At some depth, called the depth of magma segregation, the degree of partial melting is extensive enough (20-40 percent) and the tectonic environment is such that the liquid segregates from residual crystals. From this stage, the magma is no longer in equilibrium with the residual crystals with which it was originally saturated. Instead, it may fractionate independently by cooling and crystal settling as it rises toward the earth's surface. It has been shown that the degree of partial melting and the depth of magma segregation are the two prime factors determining the composition and magma type of basalts derived directly by partial melting of the pyrolite of the upper mantle."

While commenting upon the origin of rhyolitic volcanism, McBirney (1969) made the following statement. *"The extreme composition and great volume of rhyolitic ignimbrites places a severe constraint on the possible mechanisms by which they can be explained. An origin by crystal fractionation is ruled out by the huge*

volume of basic magma required to produce such a differentiate. Most recent workers have concluded that large-scale melting of the crust offers the only plausible source, but there is also evidence that at least some rhyolites, such as those of ancient shield areas, may have been derived directly from a primitive mantle. There is considerable experimental evidence to demonstrate the possibility of producing large volumes of siliceous magma by partial melting of a wide range of common crustal rocks. Such an origin is supported by inclusions in ignimbrites of partly fused basement rocks or xenocrysts of strained quartz and other minerals demonstrably derived from older rocks. The high potassium, alumina, and silica content of most rhyolites can also be explained as the result of fusion of clay or mica-rich quartzose sediments or metamorphic rocks, although other explanations can also account for the major element composition, and a unique interpretation may not be possible on the basis of present experimental data. Another clue to the origin of rhyolites is found in their rubidium and strontium isotopic composition. The Rb/Sr and $^{87}Sr/^{86}Sr$ ratios of ignimbrites are notably higher than those of intermediate and basic volcanic rocks and correspond more closely to those of sialic crustal material. Studies of the rhyolites of Central America have shown a correlation between the proportion of radiogenic strontium in rhyolitic ignimbrites and the character of the basement rocks; high $^{87}Sr/^{86}Sr$ ratios are found only in regions underlain by Paleozoic metamorphic and plutonic rocks having high rubidium contents and similar high $^{87}Sr/^{86}Sr$ ratios. The oxygen isotopic composition of the same rhyolitic ignimbrites resembles that of metamorphic and

plutonic rocks and is notably richer in ^{18}O than basic igneous rocks or rhyolites believed to be the products of differentiation of basic magma."

Lastly, the following quotation from Hughes (1982) expresses the general consensus regarding the genetic linkage between the two considered magmatic suites. *"Anatexis of continental crustal material might occur where large volumes of mafic magma had been emplaced in the crust within a short period of time, or in other areas of exceptionally high thermal gradients in the crust. Resultant melts should have minimum compositions, approaching ideal granite compositions the more felsic the crustal material being remelted. Rocks crystallized from such melts would also occur in a volcanic association but would not have trace-element contents indicative of prolonged fractionation from a mafic progenitor. Initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratios should be variable and high, equalling those of the rocks being remelted."*

2.3 The Distribution and Radiometric Ages of the Latest Igneous Rocks

The outcrop distribution along with radiometric ages and some important characteristics of the latest igneous rocks, as identified in the Yucca Mountain region, are summarized in Figures 2-1 through 2-5b. The outcrop distribution of mafic rocks, intruded and extruded during the last $\sim 10^7$ years, is presented in

Figures 2-1a and 2-1b. The corresponding K/Ar and $^{40}\text{Ar}/^{39}\text{Ar}$ laser fusion ages are tabulated and plotted in Figures 2-2a and 2-2b, respectively. Figure 2-3 presents some evolutionary trends associated with the local basalts. Specifically, for these basalts, this figure shows temporal changes in both eruptive volume and trace element concentrations (Rb, Sr, and relative rare earth element abundances). The outcrop distribution of silicic volcanics, extruded during the last $\sim 10^7$ years, is presented in Figure 2-4. The corresponding K/Ar, $^{40}\text{Ar}/^{39}\text{Ar}$ laser fusion, and fission track ages are tabulated and plotted in Figures 2-5a and 2-5b, respectively.

Examination of Figures 2-1 through 2-5b leads to the formulation of the following four observations/interpretations. First, in the Yucca Mountain region (within a radius of, say, 100 miles), mafic volcanism has persisted over the last 10^7 years. As shown in Figures 2-1a and 2-1b, outcrops of fairly young basalts are scattered all around Yucca Mountain. A variety of emplacement forms has been identified, including lava flows, cinder cones, dikes, sills, etc. Samples of the corresponding igneous materials yield K/Ar and $^{40}\text{Ar}/^{39}\text{Ar}$ laser fusion ages ranging from about 10^7 to as little as 0.1×10^6 years B.P., Figures 2-2a and 2-2b. In terms of mineralogical and chemical composition, the local late basalts are predominantly of alkalic composition with a groundmass of olivine and calcic clinopyroxene. Alkali basalts, hawaiites, hypersthene hawaiites, and basaltic andesites are the major lithologic types represented. Vaniman et al. (1982) and Crowe (1990) have shown that, during the last 10^7 years, the local mafic magmatism has undergone

significant transformation. One aspect of this transformation is a decrease in the rate of magma generation. As shown in Figure 2-3a, during the last 4×10^6 years the rates of mafic magmatism have decreased by a factor of about 10. Although the eruption frequency has remained fairly constant, volumes erupted during individual episodes have noticeably decreased. For the older basalts, the representative single-episode volume exceeds 3 km^3 . For the younger basalts, however, the corresponding volume is typically less than 1 km^3 . Another aspect of the temporal transformation is a tendency toward more mafic composition with decreasing age. The older basalts exhibit a fairly wide range of chemical compositions, from basaltic to andesitic. The younger basalts, however, are compositionally more uniform, ranging from alkali basalts to hawaiites. Among the local Plio-Quaternary mafic centers, only the Buckboard Mesa basalt contains a sufficiently high SiO_2 content to warrant the basaltic-andesite classification. Both of the above evolutionary trends appear to have been accompanied by progressive changes in trace element concentrations. As shown in Figure 2-3, such changes involve: (1) relative rare earth element abundances (the young basalts are enriched in light rare earth elements, Figure 2-3c); (2) concentration of strontium (relative to the older basalts, the younger basalts are enriched in strontium, Figure 2-3b); and (3) concentration of rubidium (relative to the older basalts, the younger basalts are depleted in rubidium, Figure 2-3b). Vaniman et al. (1982) have proposed that the observed evolutionary trends may be explained by postulating that these trends reflect a time-progressive decrease in the degree

of partial melting. The absence of a noticeable trend toward more alkaline and Rb-rich basalts, along with the inferred reduction in the degree of partial melting, was accounted for by assuming that phlogopite is present in the refractory residuum. It was suggested that the high potassium and rubidium content of phlogopite moderates both the alkali content and the rubidium content of mafic magma liberated from the phlogopite-bearing residuum.

Second, in the Yucca Mountain region, significant silicic volcanism has persisted, at least, to the end of Pliocene time. Locations of some of the late silicic volcanic centers (younger than the Ammonia Tanks Member of the Timber Mountain Tuff) are shown in Figure 2-4. Noble et al. (1991) considered that the latest silicic magmatism is represented by two spatio-temporally distinct suites. The older suite consists of high-silica tuffs and associated silicic and intermediate lava flows. The corresponding rock units have been informally referred to as: (1) the tuffs and lavas of Bullfrog Hills; (2) the rhyolitic tuffs and lavas of Rainbow Mountain; (3) the tuff of Cutoff Road; and (4) the rhyolites of Shoshone Mountain. As shown in Figures 2-5a and 2-5b, samples of these rocks yield K/Ar ages ranging from 10.4×10^6 to about 9.3×10^6 years B.P. Although these ages are up to 1.5×10^6 years younger than those of the Ammonia Tanks Member of the Timber Mountain Tuff, Noble et al. (1991) regarded the corresponding rocks as a part of the Timber Mountain Caldera magmatism. Two lines of evidence were put forth in support of this interpretation. These are: (1) close lithologic and petrographic affinity

between the corresponding rocks and the Rainier Mesa Member of the Timber Mountain Tuff, and (2) occurrence of these rocks around and within the Timber Mountain Caldera proper. In contrast to the Timber Mountain Tuff (erupted in association with the caldera-forming magmatism), however, the subsequent tuffs and lavas seem to have been erupted from fissure vents, contemporaneously with movements along nearby faults. The younger suite of post-Timber Mountain Tuff silicic rocks is mostly associated with the Black Mountain and Stonewall Mountain volcanic centers. As shown in Figure 2-4, both of these centers are situated some 30-70 miles northwest of Yucca Mountain. The corresponding rocks include subalkaline and peralkaline ash-flow tuffs and highly potassic, silicic to intermediate lava flows, alkaline to peralkaline in composition. They comprise two stratigraphic units, namely: (1) the Thirsty Canyon Tuff, and (2) the Stonewall Flat Tuff. As shown in Figures 2-5a and 2-5b, samples of the corresponding rocks yield K/Ar and $^{40}\text{Ar}/^{39}\text{Ar}$ laser fusion ages ranging from 8.8×10^6 to about 7.4×10^6 years B.P. Importantly, in the Yucca Mountain region, silicic volcanism did not cease with the emplacement of the Stonewall Flat Tuff. Two examples of still younger rhyolites are locally known. On the one hand, an interesting example of fairly young rhyolites has been reported by Carr (1982), from the southern edge of Crater Flat. At this location, a few km southwest of the Yucca Mountain exploratory block, a bed of silicic pumice was found in a sandy, poorly indurated alluvium underlying the 3.7×10^6 years B.P. old basaltic flow. The pumice occurs as numerous light-colored and somewhat rounded clasts,

locally up to 10 inches in diameter. As shown in Figures 2-5a and 2-5b, zircon from the pumice clasts yielded a fission-track age of $6.3 \pm 0.8 \times 10^6$ years B.P. Because the pumice correlates neither petrographically nor temporally with any of the local ignimbrites, Carr (1982) suggested that this pumice may express the presence of some unknown rhyolites, buried somewhere within Crater Flat proper. On the other hand, still younger silicic volcanism has been identified in the Mount Jackson area, southwest of Goldfield, Nevada. As shown in Figure 2-4, this volcanic field is situated some 65 miles northwest of Yucca Mountain. Within this field, volcanic domes composed mainly of phenocryst-rich rhyolite lavas occupy an area of about 75 square miles, McKee et al. (1989). Several rhyolitic domes cut across young basalts, which were erupted from common vents. Importantly, some of the Mount Jackson volcanics range in composition from high-silica rhyolites to dacites. Some of these rocks exhibit petrographic evidence for mixing of silicic and intermediate magmas. In the Mount Jackson volcanic field, individual domes are aligned along a NE-NNE direction, and some domes are elongated accordingly. Both of these observations indicate that emplacement of the Mount Jackson domes was controlled by NE-NNE striking faults and fissures. A glassy specimen, representing the high-silica Mount Jackson rhyolite, yielded an $^{40}\text{Ar}/^{39}\text{Ar}$ laser fusion age of $2.93 \pm 0.16 \times 10^6$ years B.P., Figures 2-5a and 2-5b. Radiometric ages of less than about 4.5×10^6 years B.P. have also been obtained from samples of silicic tuffs occurring some 10-20 miles west of Mount Jackson. McKee et al. (1989) suggested that aphyric rhyolitic lavas of the Montezuma Range may be of

similar age. An important feature associated with the local silicic volcanism is that both the eruptive volume and the eruptive frequency appear to have been diminishing with time. The former trend parallels that associated with the local mafic volcanism, and has important geodynamic implications.

Third, in the Yucca Mountain region, both the silicic magmatism and the mafic magmatism are clearly related and, thus, may reasonably be regarded as expressing a common, yet fundamental-in-character, geodynamic process. Such a state of affairs is indicated by two lines of evidence. These are: (1) spatio-temporal coexistence of both of the local magmatic suites, as expressed through the respective radiometric ages and the observed stratigraphic relationships, and (2) intermixing of mafic and silicic magmas, as expressed by the alkaline-peralkaline affinity of the corresponding silicic-intermediate lavas and ash-flow tuffs. The apparent affinity, together with the current viewpoints regarding the origin of mafic and silicic magmas, lead to the development of a reasonable interpretation of the contemporary geodynamic configuration of the Yucca Mountain region. In accordance with this interpretation, the locally observed bi-modal magmatism expresses a convective or advective ascent of matter from the Low Velocity Zone, or Asthenosphere. During ascent, the diapirically rising asthenospheric matter undergoes a reduction in temperature. The cooling, however, mimics the adiabatic temperature gradient ($dT/dz \sim 0.3^\circ$ Celsius per 1 km of depth) which is much smaller than the gradient of the solidus

for pyrolite ($dT/dz \sim 3^\circ$ Celsius per 1 km of depth), Green and Ringwood (1969).

The difference in the respective gradients causes the rising asthenospheric matter to gradually drift toward solidus conditions. Upon attaining these conditions, the rising material begins to melt. The degree of partial melting is influenced by a number of factors, such as pressure, temperature, availability of free H_2O , etc.

The attainment of liquidus conditions, however, is prevented by the withdrawal of latent heat of fusion. At some depth, the degree of partial melting is extensive enough and the melting environment is such that liquid segregates from a refractory residuum and forms discrete bodies of mafic magma. Because of their relative buoyancy, such bodies are gravitationally unstable and have a tendency to rise toward the Earth's surface. If both the magmatic volume and the residence time are fairly small, bodies of liberated mafic magma pass through the crust without experiencing extensive fractionation and without significant assimilation of silic matter. The result of such behavior is an extrusion of fairly pure mafic magma, such as that solidifying to form alkali basalts and hawaiites. Either larger magmatic volumes or slower ascents may be accompanied by some assimilation of silic matter from the crust. The resulting composite melt exhibits higher SiO_2 content, and its composition is shifted toward that of parent magmas for basaltic andesites or latites. If large volumes of mafic magma are introduced into the crust within a relatively short period of time, however, the resulting magmatic heating causes locally extensive melting (anatexis) of the affected crust. Upon eruption, the resulting volatile-charged silicic magma forms a variety of

rhyolitic-rhyodacitic lavas and tuffs. Because mafic magmas erupt as well, these lavas and tuffs are spatio-temporally correlative with the mantle-derived mafic volcanics. A spatially concentrated melting of the crust leads to the accumulation of silicic magma with sufficient volume and volatilities to support caldera-forming pyroclastic magmatism. This type of magmatism leaves behind a multitude of spectacular forms of expression and, therefore, is readily visible in the local geologic record. More spatially diffuse melting of the severely extended crust, however, may be accommodated less dramatically, through fault- and fissure-controlled vents and associated feeder dikes. In the geologic record, products of such magmatism are much more subtly expressed. Identification of the late fault- and fissure-controlled rhyolites is often difficult, particularly in regions with prior caldera-forming magmatism, and requires extensive radiometric age dating.

Fourth, for the Yucca Mountain region, the observed decline of silicic volcanism may reasonably be taken as reflecting the decreasing influx of mantle-derived mafic magma into the crust. The latter decrease, in turn, may be attributed to the diminishing degree of partial melting of mantle matter contained in the rising asthenospheric diapir. It is unlikely, however, that these interrelated temporal trends indicate either a relaxing thermal state of the local upper mantle or a change in the overall geodynamic configuration. While searching for a possible cause that may be responsible for the inferred reduction in the degree of partial

melting, it may be useful to recognize two points. On the one hand, it is highly unlikely that the primitive mantle matter is completely devoid of water, or anhydrous. Ringwood (1966), for example, has inferred that the mantle has probably retained at least three times as much water as has been degassed to produce the terrestrial oceans; this would suggest an average content of H_2O in the mantle of about 0.1 percent. On the other hand, for the primitive pyrolite, the solidus curve is strongly influenced by the presence of H_2O . The influence of H_2O on both the solidus temperature and the degree of partial melting is illustrated in Figure 2-6. It may be observed that the absence of H_2O elevates the solidus temperature and, at the same time, lowers the degree of partial melting. The relationship indicates that, for a constant temperature slightly above the solidus, lowering the availability of H_2O has the effect of reducing the degree of partial melting. It is possible that, for the Yucca Mountain region, the diminishing degree of partial melting reflects the diminishing availability of H_2O . A factor that may ultimately be responsible for this is a time-progressive ascent of the upper thermal boundary layer of a sub-lithospheric convective system, or of an advective asthenospheric diapir, Figure 2-7. Apart from causing progressive corrosion and thinning of the lithosphere (see for example Fleitout and Yuen, 1984, and Olson et al., 1988), the ascent of the upper thermal boundary layer also causes the segregation of rising asthenospheric matter (mafic melt and refractory residuum) to occur at progressively shallower depth and lower pressure. This depth (pressure)-varying segregation, or melting, has important implications

regarding the availability of H_2O . A critical factor in this process is the stability of hydrous mineral phases, primarily amphibole and to a lesser degree phlogopite. For the Yucca Mountain region, the inferred overall effect of the depth-varying melting of the primitive pyrolite is summarized in Figure 2-8. During early stages of asthenospheric ascent, fractional melting occurred relatively deep, say below a depth of about 60 km. At this depth (pressure of about 20 kb), and for a temperature near the corresponding solidus temperature, hydrous amphibole is unstable and transforms into eclogite plus H_2O . Consequently, during melting below a depth of 60 km, water occurred as a free-phase, "*mostly along mineral grain boundaries and in small bubbles*," Ringwood (1969). The resulting solidus temperature was relatively low and the corresponding degree of partial melting was relatively high. As the upper thermal boundary layer migrated upward, however, fractional melting occurred at progressively lesser depth and pressure. Green and Ringwood (1967) have shown that, in an olivine-rich environment, amphibole is a stable mineral phase for temperatures not far below the dry solidus temperature and for pressures ranging from 15 to about 20 kb. During melting at a depth of about 35-60 km, this fairly narrow stability range causes small amounts of available H_2O to be bound mostly in stable amphibole crystals. Relative to the prior and deeper magma source region, the availability of H_2O is much smaller. The result is that, for the contemporary and shallower source region, the solidus temperature is relatively high and the degree of partial melting is relatively low.

2.4 The P-Wave Velocity Structure

The foregoing conceptual understanding of the contemporary geodynamic configuration of the Yucca Mountain region may be tested by using observations regarding propagation velocities of seismic waves. For the purposes of this report, two topics are of particular relevance. On the one hand, the local presence of small-scale Quaternary mafic magmatism implies that, in the Yucca Mountain region, the upper mantle is in a quasi-stable state of incipient-partial melting. Such conditions must be associated with abnormally low velocities of seismic waves. For the upper mantle of stable cratonic regions, for example, the P-wave velocity is usually within the 8.2 ± 0.2 km/sec range. Significantly lower P-wave velocities, say 7.6-7.8 km/sec, are observed only in magmatically active regions. Commonly, such velocities are regarded as evidence for incipient-partial melting of the upper mantle, see, for example, Archambeau et al. (1969) and Anderson and Sammis (1970). On the other hand, the incipiently or partially melted upper mantle is also (1) gravitationally unstable, relative to its surroundings, and (2) has low viscosity, say 10^{19} - 10^{20} poises. Both of these inferences imply that, for the Yucca Mountain region, either convective or advective motions of upper mantle material must be intrinsic to the contemporary geodynamic configuration. Such motions may assume the form of either plumes with cylindrical cross-sections or elongated rolls with sheet-like ascending and descending limbs.

The available P-wave velocity data from the Yucca Mountain region are presented in Figures 2-9 through 2-11b. Figure 2-9 is a seismic tomography map for the western United States. This map shows the distribution of P-wave velocities in the uppermost mantle. It is based on travel-times of earthquake-generated waves, as compiled by the International Seismological Centre. The velocity mapping was performed, employing the tomographic back-projection method, by T. Hearn (of New Mexico State University), N. Beghoul, and M. Barazangi (both at the Institute of the Study of the Continents at Cornell University).

For the Yucca Mountain region, the crust and upper mantle P-wave velocity structure down to a depth of 230 km is presented in Figures 2-10b through 2-10d. This structure has been defined by Monfort and Evans (1982), and is based on a study of teleseismic P-wave travel-time residuals. The seismic data employed for this study were obtained by recording 98 teleseismic events at as many as 53 monitoring stations. The data were analyzed by means of linear inversion, employing the Aki-Christofferson-Husebye damped-least-squares technique. The portion of the Earth covered by the monitoring array was subdivided into five layers. These layers are at the following depth intervals: 0 to 15 km, 15 to 31 km, 31 to 81 km, 81 to 131 km, and 131 to 231 km. Each of the layers was then subdivided into rectangular blocks, 35 km long. For each of these blocks, the relative P-wave velocity was computed using damped-least-squares inversion, and assuming that the average velocity for each layer increases with depth. The

relative velocities were computed as ratios of the observed velocity in each block to the assumed average velocity in a layer hosting that block. Because relative P-wave travel-time residuals were used, the assumed average velocity may be different from the actual velocity in the corresponding layer. In other words, the technique allows for detection of velocity contrasts but does not produce absolute velocity information.

Figure 2-11a presents the average amplitude vs. travel-time decay curve for a single seismic profile of the local crust. This curve has been constructed from the results of a seismic reflection survey along line AV-1. As shown in Figure 2-11b, this seismic survey line is located in the Amargosa Desert, about 20 km south of Yucca Mountain. Of particular relevance is the presence of an unusually high-amplitude reflection from the lower crust. The corresponding "bright spot" is situated at a seismic depth of about 8.4 sec.

Examination of Figures 2-9 through 2-11 leads to the formulation of the following three observations/interpretations. First, for the Yucca Mountain region, the P-wave velocity in the uppermost mantle is abnormally low. As shown in Figure 2-9, the velocity is as low as 7.6 km/sec. For the western United States, similarly low seismic velocities were obtained only at a few locations. Examples of such locations are the Snake River Plain, the Yellowstone "hot spot," the Rio Grande rift, and the northwestern margin of the Great Basin. Without exception, the low

P-wave velocity regions are characterized by fairly young magmatism and fairly high heat flow. Both of these observations indicate that abnormally high temperature, resulting in incipient-partial melting of the upper mantle, is the principal factor expressed through the imaged low P-wave velocities. For the Yucca Mountain region, the P-wave velocity image confirms that, indeed, the material comprising the uppermost mantle remains in a quasi-stable state of incipient-partial melting.

Second, in some portions of the Yucca Mountain region, temperatures in the lower crust are sufficiently high to produce near-solidus thermal conditions. Such a state of affairs is suggested by the results of a seismic reflection survey reported by Brocker et al. (1989). Specifically, this survey revealed the presence of a "bright spot" in the local lower crust. On the explosive source profile for line AV-1, this "bright spot" occurs at a seismic depth of about 8.4 sec, and manifests itself through unusually high-amplitude reflections from the lower crust. On the common-shot gather, the amplitude of these reflections is ~9 dB above the average amplitude vs. travel-time decay curve, Figure 2-11a. According to Brocker et al. (1989), *"the Amargosa Desert 'bright spot' is similar to the lower crust spots imaged by COCORP in Death Valley, California, and near Socorro, New Mexico, in the Rio Grande rift. Both of these bright spots have been attributed to reflections from thin and discontinuous magma chambers."*

Third, around Yucca Mountain, the lower crust exhibits a pronounced seismic heterogeneity. As is the case with the local upper mantle, the lateral P-wave velocity gradients in the crust are surprisingly large. As shown in Figure 2-10c, these gradients may be as large as $\Delta V_p/V_p \sim 6$ percent per 50 km. The overall pattern is one of low velocity areas alternating with relatively high velocity areas. Some of the former areas are fairly large, and their areal extent is measured in 10^3 km^2 . Both the alternating pattern and the size of the low velocity areas suggest that, in the local lower crust, lateral compositional differences are not the sole factor expressed through the P-wave velocity image. Rather, at least in part, this image seems to be expressing the effects of laterally varying temperature on elastic properties of the lower crust. This, of course, implies that, at a given depth, the distribution of lower crustal temperature lacks lateral uniformity. Relative to the high velocity areas, the low velocity areas are hotter. It is also possible that, within the hotter low velocity areas, near-solidus thermal conditions prevail. As a matter of fact, such a possibility is suggested by two spatial associations. On the one hand, the Amargosa Basin "bright spot" occurs within the extensive low velocity area, Figures 2-10c and 2-11b. On the other hand, all of the locally known examples of post-Timber Mountain Tuff silicic magmatism, including the Mount Jackson volcanic field, are spatially associated with presumably hotter low velocity areas, Figures 2-4 and 2-10c. The interpretation that the local P-wave velocity image, at least in part, expresses the lateral distribution of crustal temperatures is supported by conclusions drawn by Evans

and Smith (1992). Based on the results of teleseismic tomography studies of the Yucca Mountain region, these investigators made the following statement.

"Mid-crustal silicic magma chambers 4 km across and larger probably can be ruled out within 10 km of the repository block. However, the weak middle and lower crustal low-velocity anomaly beneath Crater Flat is significant. It may be caused by many phenomena, including piston-like caldera collapse juxtaposing middle-and lower-crustal rocks, alteration by Miocene volcanism, or crustal heating, possibly related to current basaltic activity. It is quite similar to the Mineral Mountains anomaly in Utah, which lies beneath vents of Quaternary rhyolite flows and near the Roosevelt Hot Springs geothermal area. This similarity supports the crustal heating hypothesis at Crater Flat."

2.5 Expected Geothermal Conditions in the Yucca Mountain Region

Implications resulting from the conceptual understanding of the geodynamic configuration of the Yucca Mountain region, as formulated and successfully tested in the preceding two sections, are numerous and profound. For the purposes of this report, some of the implications may be summarized by the following five points. First, at the base of the Yucca Mountain hydrologic system, situated say at a depth of the Conrad discontinuity (~15 km), the average flux of terrestrial heat is fairly high, Figure 2-13. By analogy with regions of comparable

lithospheric thickness (30-50 km), this flux may reasonably be judged to be 100 percent higher ($\Delta HF \sim 40\text{-}50 \text{ mW/m}^2$) than that in cratonic regions ($HF \sim 42 \text{ mW/m}^2$, Figure 2-14a). The total inferred flux of terrestrial heat is as high as 80-100 mW/m^2 (2.0-2.5 HFU), Figure 2-14a. For the southern Great Basin, the 2.0-2.5 HFU heat flow estimate is in fairly satisfactory agreement with the results of more direct heat flow estimates. Swanberg and Morgan (1978), for example, have estimated that, in the Great Basin, the regional heat flow is as high as about 100 mW/m^2 , or about 2.5 HFU. A similar viewpoint was expressed by Sass and Lachenbruch (1982). With specific reference to the Yucca Mountain region, these authors have stated that *"the regional heat flow from beneath the zone of hydrologic disturbance may be same as that characteristic of the Great Basin in general (80 mW/m^2 or 2 HFU), or it could be as high as 100 mW/m^2 , or 2.5 HFU."* A conductive heat flow ranging from 80 to 100 mW^2/m is not trivial by terrestrial standards. As a matter of fact, the southern Great Basin heat flow rivals that of the most active continental regions of the world. Examples of such regions are: (1) the Andean orogenic arc, HF no more than 120 mW/m^2 ; (2) the East African Rift, HF no more than 100 mW^2/m ; and (3) the East Asiatic back-arc basins, HF no more than 130 mW^2/m .

Second, it may reasonably be inferred that, along the base of the Yucca Mountain hydrologic system, the terrestrial heat input lacks spatial uniformity. In other words, deep within the crust, where convective heat flow is effectively inoperative

because of scarcity of free fluids, there are lateral gradients of heat flow, Figure 2-13. In part, these gradients reflect flow patterns in the upwelling upper mantle. As shown in Figure 2-14b, over the ascending limbs of mantle plumes, the intensity of heat flow may be up to about $25 \text{ mW}^2/\text{m}$ higher than the corresponding intensity over the descending limbs. Further amplification of the heat input heterogeneity may be caused by episodic advective heat transfers from the mantle, and the resulting local magmatic heating of the lower crust. As illustrated in Figure 2-14c, the introduction of a body of mafic magma into the crust produces a transient heat pulse. The intensity and duration of such a pulse depends on a number of factors, including the thermal state of the crust, availability of fluids, size of the magmatic intrusion, etc. Thin and discontinuous magma chambers, such as those expected to be present around Yucca Mountain, may be presumed to be associated with fairly subtle increases of the intensity of heat flow, say less than $20\text{-}30 \text{ mW}/\text{m}^2$. Locally, the total lateral differences in the input of terrestrial heat may be expected to be occasionally as high as $\Delta \text{HF}_{(x)} \sim 40 \text{ mW}/\text{m}^2$. Because of the fairly small size of the local mantle plumes, as implied by the sharp P-wave velocity gradients, lateral heat input differences, with the stated magnitudes, may reasonably be presumed to be causing the emergence of very substantial lateral temperature gradients in the lower crust. These temperature gradients, in turn, will be reflected through (1) spatially varying rheological properties of the lower crust, and (2) likewise varying propagation velocities of seismic waves. Importantly, in the middle and lower crust immediately beneath

Yucca Mountain, there are significant lateral differences in P-wave velocity and, presumably, in the corresponding crustal temperature. Such a state of affairs is clearly indicated by the three-dimensional P-wave velocity image, as reported by Evans and Smith (1992). This image shows that, at Yucca Mountain, two sharply contrasting seismic regions adjoin one another. To the west (Crater Flat area), a low-velocity seismic region is present; the corresponding P-wave velocities are two to three percent lower than the assumed reference velocity. To the east (eastern Yucca Mountain and western Jackass Flat area), however, a high-velocity region is apparent; the corresponding P-wave velocities are a few percent higher than the assumed reference velocity. If the observed east-west P-wave velocity gradient does indeed reflect a lateral temperature gradient in the lower crust, then it follows that as far as local convective circulations of intracrustal fluids are concerned, there are profound implications. In that regard, Yucca Mountain seems to be situated within the most unfortunate thermodynamic circumstances. Locally, not only the actual Rayleigh number is fairly high, but also the critical Rayleigh number is vanishingly small. The increased flux of terrestrial heat, and the resulting elevated geothermal gradient, is responsible for the former; and the lateral temperature gradient is responsible for the latter. Under such circumstances, therefore, convective circulation of the local intracrustal fluids, either continuous or intermittent, is a thermodynamic necessity.

Third, the history of magmatic activity, as formulated in Section 2.3, may be

interpreted to indicate that, in the Yucca Mountain region, the thermodynamic state of the crust is gradually evolving. As shown in Figure 2-15, such evolution is of two-fold character. On the one hand, the inferred decrease in the degree of partial melting of the rising asthenospheric matter must have been accompanied by a similarly decreasing influx of mantle-derived mafic magma. Consequently, the advective heat transport from the mantle, and the resulting localized magmatic heating of the crust, may be presumed to have been diminishing with time. It follows then that, with passage of time, large-scale and localized melting of the crust becomes less important and the thermal state of the crust becomes more spatially uniform (i.e., the lateral temperature gradients become smaller). On the other hand, however, the decrease in the magma segregation depth, and the concurrent increase in melting temperature, must have been accompanied by steadily increasing conductive heat flow across the Moho discontinuity. It follows then that, although both the advective mantle --> crust heat transport and the localized magmatic heating of the crust diminish with time, there is a steady increase in the average crustal temperature, and likewise steady increase in the overall geothermal gradient. Excessive rise of the geothermal gradient, however, will be prevented by augmenting conductive heat flow with convective heat flow. To become effective, the latter requires the participation of crustal fluids. It is important to recognize that, for the Yucca Mountain region, the contemporary presence of convective heat flow manifests itself through a locally known geothermal paradox. This paradox becomes apparent if one recognizes that, in

part, the Yucca Mountain region is situated within the so-called Eureka Low, Sass et al. (1976). Within this low, the intensity of terrestrial heat flow, computed from downhole measurements of in-situ temperature assuming exclusively conductive heat transfer, is $\sim 65 \text{ mW/m}^2$, or 1.5 HFU. This computed value is substantially lower than the regional heat flow estimate derived through (1) consideration of both the volcanic setting of the region and the measured P-wave velocities, and confirmed by (2) consideration of silica content of local groundwaters, Swanberg and Morgan (1978). One way to explain the discrepancy is to presume that, within the Eureka Low, the assumption of exclusively conductive heat flow is not valid. It is possible that, for the Eureka Low and including Yucca Mountain, convective heat flow is of widespread importance, and augments conductive heat flow. As a matter of fact, this resolution of the paradox is supported by the results of detailed geothermal investigations performed in the immediate vicinity of Yucca Mountain. These results revealed the presence of at least two independent hydrothermal circulation centers. One of these centers is located around well UE-25a#3, some 15 km east of Yucca Mountain, Sass et al. (1980). The other center occurs below Yucca Mountain itself. It has been identified in well UE-25p#1, at a depth of about 1.2 km, Sass et al. (1983) and Szymanski (1989).

Fourth, for the Yucca Mountain region, both fairly high intensity of the mean heat flow and lateral heat flow heterogeneity virtually assure that Bernard-Rayleigh

instabilities are intrinsic elements of the long-term behavior of the local hydrosphere. The former gives rise to elevated geothermal gradients which, in turn, introduce non-equilibrium fluid density configurations. After a certain thermal stability limit (related to hydraulic conductivity) is attained, however, these configurations tend to dissipate through convective circulation of intracrustal fluids. The latter gives rise to lateral temperature gradients and, thus, assures that the thermal stability limits are very small indeed. For the local hydrosphere, two varieties of thermal instability of fluids may be expected. On the one hand, recognizing the potential for local presence of intra-crustal magma bodies, one may expect that strongly disequilibrated and sustainable Bernard-Rayleigh instabilities are present, particularly in enlarged spatio-temporal domains. Such instabilities are spatially stationary and temporally prolonged. This is so because they are anchored to concentrated and readily replenishable heat sources. For these instabilities, convectively enhanced heat losses may be compensated readily, via convective circulation within the magma body itself. Magmatic center-based convective systems, therefore, may remain active for a fairly long period of time. Typically, such systems leave behind strong and readily visible imprints in the geologic record. They are of particular interest to mineral exploration geologists. This is so because prolonged heating and circulation of mineral-rich solutions cause extensive alterations of host rocks and, typically, lead to the development of various hydrothermal ore deposits. On the other hand, recognizing both the substantial conductive heat input from the

upwelling mantle and the episodic enhancement of extrinsic hydraulic conductivity by ongoing tectonic straining (and the resulting decrease in the critical Rayleigh number), one may expect that more intermittent, or transient Bernard-Rayleigh instabilities are also present. Being developed away from concentrated and readily replenishable heat sources, such instabilities assume the form of intermittent, or ephemeral, plumes. For these plumes, intermittently enhanced heat losses exhaust the thermal source that fuels the instability. In other words, the loss of thermal energy is not being adequately compensated by slow conductive heat flow from the far-field. Being deprived of thermal energy, the Bernard-Rayleigh instability loses its far-from-equilibrium status and ceases to exist. This cessation, however, is only temporary. It is followed by a period of thermal recovery, during which heat conduction from the far-field slowly restores the original thermally unstable state. Owing to their intrinsically ephemeral character, hydrothermal systems driven by conductively replenished heat sources are expected to be more subtly expressed in the geologic record. For such systems, multi-pass and almost incessant hydrothermal circulations are absent. Instead, the initial eruptive burst is relatively rapidly followed by a period of decaying activity and the eventual cessation of such activity. The associated heating of host rocks is spatially restricted and, consequently, the corresponding alteration and mineralization aureoles are fairly narrow and meager. Similarly, the absence of multi-pass hydrothermal circulation results in the accumulation of trace elements being both low and spatially restricted. It is entirely reasonable to

expect that, in the geologic record, intermittent hydrothermal systems are principally expressed through accumulations of gangue minerals, such as opaline silica and calcite. This is so because, for rapidly ascending solutions (1) initial concentrations of these gangue minerals may be fairly high, and (2) during ascent, the solubility of both of these minerals is drastically reduced. The rapidly dropping fluid temperature causes reduction in the solubility of silica, and assures precipitation of various textural and mineralogical forms of silica. The rapid CO₂-degassing of ascending fluids, in turn, causes reduction in the calcite solubility, and assures precipitation of a variety of textural forms of calcite. It is small wonder that, for some geologists accustomed to dealing with magmatic center-based hydrothermal systems, the subtle expressions of hydrothermal systems driven by conductively replenished heat sources are particularly confusing and unfamiliar. Often, experience or detailed field and laboratory investigations are required to tell them apart from, for example, pedogenic expressions of supergene processes.

Fifth, for the Yucca Mountain region, the previously inferred gradually evolving thermodynamic state of the crust may be expected to have been accompanied by a likewise evolving character of local convective heat transport. Specifically, it may be presumed that, during the earlier period of high influx of mantle-derived mafic magma, convective heat transport assumed predominantly the stationary-prolonged form (magmatic center-based hydrothermal system). This is the reason

why the majority of locally known hydrothermal ore deposits are temporally and spatially correlative with local caldera-forming magmatic centers. As the advective mantle --> crust heat input became less intense, however, magmatic center-based hydrothermal systems also became less important and intense. The inferred steadily increasing average temperature of the crust, however, may be presumed to have been preventing a complete cessation of local hydrothermal activity. Rather than being completely eliminated, the convective heat transport may be presumed to have assumed the ephemeral form (hydrothermal system driven by conductively replenished heat sources). If this presumption is correct, then it follows that in addition to bearing imprints of the earlier magmatic center-based hydrothermal activity, the local geologic record ought to contain expressions, or imprints, of fairly young and distinctly more subtle hydrothermal activity. The remainder of this report is devoted to establishing whether or not such is the case indeed. The area considered is fairly small in size, less than, say, 100 square miles, and corresponds to Yucca Mountain itself. This small area, however, has been subjected to over ten years of very intense geologic exploration. A multifaceted and very extensive data base has been established and will be used for the stated purposes.

3.0 Mineralization and Alteration of the Yucca Mountain Area, Based on the Results of Direct Examination of Surficial Exposures

Even a brief geologic reconnaissance of the Yucca Mountain area is sufficient for one to recognize that, sometime in the past, the local ignimbrites were affected by hydrothermal solutions, see, for example, Figures 3-1 through 3-5. For a geologist experienced with siting and licensing of nuclear facilities, such reconnaissance should also be sufficient for recognition that, potentially, hydrothermal and related processes constitute a hazard which needs to be accounted for in considering the suitability of Yucca Mountain to accommodate a high-level nuclear waste repository. This is so because the observed hydrothermal mineralization and alteration features exhibit four troublesome characteristics. These are: (1) some of these features appear to be epigenetic; (2) they are present in the latest locally known ignimbrites, which are the Ammonia Tanks and Rainier Mesa Members of the Timber Mountain Tuff; (3) sometime in the past, hydrothermal solutions were discharging at the contemporary topographic surface; and (4) in a few instances, the observed hydrothermal features appear to be fairly young, say, Plio-Quaternary in age.

After, say, a two-week long reconnaissance of Yucca Mountain, an experienced field geologist will have the following four general impressions firmly imprinted in his mind.

Calcretes and Sinters

First, the local occurrence of calcretes, silcretes, and/or sinters is widespread. These topographic surface-conformable depositional features consist of varying amounts of aeolian, alluvial, and colluvial detritus firmly cemented by calcium carbonate and opaline silica. In places, they are up to a few meters thick and sometimes contain plant root casts, or rhizoliths. Their development is clearly an intermittent process. In the Busted Butte area, for example, at least three distinct generations of calcretes may be observed in the local sand ramps. Stratigraphically, the Busted Butte sand ramps occur above the locally present Bishop Ash. It is clear, therefore, that the development of all three generations of calcretes took place after emplacement of the Bishop Ash, about 7×10^5 years B.P.

Importantly, the Yucca Mountain calcretes occur in the form of five texturally distinct varieties. These are: (1) GS-textured calcretes--allogenic clasts are in direct contact and authigenic cement occurs in undilated pore space, Figure 3-6; (2) F-textured calcretes-- allogenic clasts "float" in authigenic cement, Figures 3-7 and 3-8; (3) M-textured calcretes--authigenic cement (either calcite or opal) constitutes more than 90 percent of a specimen's total volume, Figure 3-9; (4) intercalated M-textured calcretes--bands or laminae of calcite alternate with bands or laminae of opal, Figure 3-10; and (5) laminated M- and GS-textured

calcretes--laminae of fine cemented and GS-textured sand separate laminae or bands of detritus-free authigenic cement, Figure 3-11. Both the presence of rhizoliths and conformity with the topographic surface indicate that the local calcretes and sinters are of pedogenic origin, i.e., formed at or near the topographic surface, locally within the soil horizon.

Among the Yucca Mountain Site Characterization Project investigators, it is commonly understood, or implied, that fluids responsible for pedogenic accumulations of calcretes and/or silcretes are necessarily supergene fluids, or infiltrating rainwater. In other words, for these fluids, the intraformation residence time in either soil or surficial bedrock is very short, and the depth of their infiltration is very small, no more than a few meters. Such understanding, however, is erroneous. Secondary carbonate and opal may accumulate through varied processes to form either calcic-silicic soils, or other deposits that resemble such soils, see, for example, Goudie (1973) and Machette (1985).

Properly understood, pedogenic accumulations of carbonate and/or opaline silica are the result of topographic surface desposition (through evaporation, evapo-transpiration, run-off, and infiltration) of meteoric fluids regardless of their intraformation residence times and histories. For example, in many arid and semi-arid regions, calcretes and silcretes are commonly regarded as formed by upward movement of calcium- or silicon-rich fluids from the water table, via

capillary or other mechanisms, and then by ongoing chemical, biochemical, or evaporative mineral precipitation. Examples of such pedogenic mineral accumulations are common throughout the world, see, for example, Summerfield (1983) and Carlisle (1983). Clearly, in the case of the Yucca Mountain calcretes, the issue is not whether these deposits are pedogenic. Rather, the issue is: what does their presence indicate about the long-term stability of the Yucca Mountain hydrosphere? There are two aspects to this issue. First and foremost, there is the question as to whether the Yucca Mountain calcretes represent supergene accumulations or, conversely, whether these deposits were formed in response to fundamental hypogene processes.

In the supergene model, calcium and/or silicon, either introduced at the topographic surface by rainfall and airborne dust or released as the result of various weathering processes, is dissolved in upper soil horizons, translocated through these horizons, and then precipitated at depth. In contrast, the hypogene model relies on subsurface fluids, typically deep-seated. For such fluids, dissolved calcium and/or silicon is derived through leaching of the respective reservoir rocks. Precipitation of calcium carbonate and opaline silica is the result of some combination of several factors. Among such factors are: (1) evaporation and/or evapo-transpiration; (2) reduction in fluid temperature; (3) reduction in CO₂ partial pressure; (4) change in fluid pH, etc.

While being engaged in rational evaluations of both of these alternatives, an inquiring geologist is soon confronted with two important issues. On the one hand, the geologist must find a rational explanation for the observed **textural diversity** of the Yucca Mountain calcretes. To account for it, a supergene advocate must rely on a dubious concept, that enlargement of pore space may be caused by authigenic cement crystallization pressures. If such an advocate is reasonably inquisitive, however, soon he will become confronted with two seemingly unanswerable questions, namely: (1) why do the calcretes with clearly comparable ages exhibit the observed textural diversity?--presumably, the authigenic cement crystallization pressures exert their dilatory influence indiscriminately; and (2) why are the M-textured calcretes laminated with the GS-calcretes?--presumably, the authigenic cement crystallization pressures act in all directions, but not selectively normal to the topographic surface. When dealing with both of these questions, however, a hypogene advocate is blessed with a decisive advantage. Such an advocate will immediately point out that: (1) for a number of reasons, the rates of formation of authigenic cement vary in time and space; (2) authigenic cement precipitates both directly at the topographic surface and below the topographic surface (ejected hypogene fluids may flow either on the topographic surface or through pre-existing alluvial, colluvial, or aeolian deposits); and (3) the formation of authigenic cement occurs in association with ubiquitous introduction of allogenic clasts, be it by wind action, run-off, or flash floods. Clearly, the spatio-temporally varying rates of formation of authigenic cement,

relative to the rates of introduction of allogenic clasts, may be held responsible for the observed textural diversity. Rapid precipitation on the topographic surface may be presumed to yield both M-textured calcretes and laminated M- and GS-textured calcretes. A slower, but still direct, surface precipitation may be presumed to lead to the formation of F-textured calcretes. Here, the rates of formation of authigenic cement are comparable to the rates of introduction of allogenic clast. Finally, GS-textured calcretes may be presumed to be produced when (1) either the ejected hypogene fluids flow through pre-existing accumulations of various detritus, or (2) the rates of formation of authigenic cement are smaller than the rates of introduction of allogenic clasts.

On the other hand, the inquiring geologist must also find a rational explanation for the observed **compositional diversity** of some of the Yucca Mountain M-textured calcretes. The fact that these calcretes are composed of bands of calcium carbonate alternating with bands (up to four inches thick) of opal may reasonably be taken as indicating that either the chemistry of parent fluids or the conditions of formation of authigenic cement were alternating between those favoring precipitation of opal and those favoring precipitation of micritic calcites. Alternatively, it may also be assumed that the opal bands represent an epigenetic replacement of earlier micritic calcites. In any event, one may reasonably conclude that either the parent fluids for the Yucca Mountain calcretes or the conditions of formation of these calcretes were fluctuating, either episodically or

continuously. Whether episodic or continuous, fluctuations involving both of these factors are not expected characteristics of supergene pedogenic processes. Here again, the supergene advocate is confronted with another seemingly unanswerable question. For a hypogene advocate, however, finding an answer to this question presents little challenge. Immediately, such an advocate will point out that both the fluctuating chemical composition of fluids and the fluctuating conditions of mineral deposition are typical features of most geothermal systems discharging at the topographic surface, or in the shallow subsurface. During certain periods of activity of such systems, for example, the associated fluids may carry high values of CO_2 partial pressure. Precipitation of calcite is favored because of the rapid escape of CO_2 from ascending solutions. During other periods of activity, however, CO_2 partial pressure is lower and, in this case, progressive cooling of these solutions dominates the mineral formation process. Precipitation of opal is then favored, because of the prograde solubility of SiO_2 with temperature.

Bedrock Veins

Second, at Yucca Mountain, the occurrence of various bedrock veins is ubiquitous. These veins range in thickness from a few millimeters to about 1 meter. They occur in all of the locally known stratigraphic units (including the Ammonia Tanks Member of the Timber Mountain Tuff and the overlying unconsolidated

Quaternary deposits), and in all of the locally known lithologic rock types (including welded and unwelded ignimbrites, and poorly consolidated or loose detritus of various origin). Based on visual examination of hand-specimens, two broadly distinct varieties of veins may readily be distinguished.

The older variety consists of clearly hydrothermal veins and impregnations, Figures 3-12 and 3-13. Such veins are composed of calcites and multi-colored opals and, typically, are accompanied by narrow alteration aureoles. Although, for the corresponding veins, a hydrothermal origin is indisputable, the appropriate question is (1) whether these veins represent deuteric mineralization (i.e., formed in association with the cooling, syn-depositional devitrification and solidification of the host ignimbrites) or, conversely, (2) whether they represent subsequent epigenetic mineralization.

Based on visual observations alone, it is difficult to resolve this dilemma satisfactorily. There are, however, a few field observations which indicate that at least some of the hydrothermal veins are epigenetic. In support of this statement, two particularly convincing examples may be put forth. On the one hand, along the northeastern flank of Harper Valley, a jasper-opal vein a few inches thick was observed to have been emplaced along a fault contact between stratigraphically distinct ignimbrites, namely the Tiva Canyon and the Topopah Spring Members of the Paintbrush Tuff, Figures 3-19a and 3-19b. When traced southwardly, into the

upper vitrophyre of the Topopah Spring Member, the silicified fault contact was observed to be accompanied by a narrow, yet very intense, devitrification of the formerly vitric footwall. In this case, the presence of jasper vein in the Tiva Canyon Member indicates that both the vein mineralization and the fault-based alteration of the Topopah vitrophyre occurred after the cooling and solidification of the Paintbrush Tuff and, therefore, the mineralization and alteration are the result of epigenetic hydrothermal processes.

On the other hand, across the Lathrop Wells mafic center, a few fairly thick siliceous veins were observed to have been emplaced along local fractures and small faults. At this location, the host rock for these veins is the latest locally known pyroclastic unit, namely the Ammonia Tanks Member of the Timber Mountain Tuff. The corresponding unwelded ignimbrites do not exhibit any indication of having experienced syn-depositional (deuteric) devitrification, as numerous vitric pumice fragments may still be observed. Epigenetic character of the observed veins is indicated by the fact that, in one instance, the fault-based vein is accompanied by a small apron, or siliceous sinter. This sinter consists of vein-equivalent siliceous matter cementing rounded cobbles. It occurs directly at the topographic surface, in-filling a local fault scarp. Both the formation of sinter and the emplacement of vein must have occurred after the development of the local topographic surface, including the development of a local valley, clearly a long time after the eruptive emplacement of the Ammonia Tanks Member. Here

again, the epigenetic character of the observed silification is an indisputable fact.

The younger variety of Yucca Mountain veins consists of micritic calcite intercalated with milky opaline silica. The corresponding veins range in thickness from a few centimeters to about 1 meter, Figures 3-14 through 3-16. They occur in all of the locally known stratigraphic units, including the aeolian deposits of Busted Butte and Harper Valley, Figures 3-5, 3-17, and 3-18. Importantly, the Busted Butte aeolian deposits occur above the locally present Bishop Ash, so there is little doubt that at least some of the calcite-opaline silica veins are younger than the $\sim 7 \times 10^5$ years B.P. age of the Bishop Ash. Compositionally, texturally, and spatially, the younger veins display indisputable affinity with the local calcretes. Clearly, both the calcretes and the equivalent veins must have been precipitated from common parent solutions. The results of extensive uranium, strontium, oxygen, and carbon isotopic investigations, as performed by the USGS investigators during the last seven years, confirm that such is the case indeed.

While dealing with the question of the origin of the younger veins, the inquiring geologist is again confronted with his old dilemma; either both the calcretes and the equivalent veins represent supergene mineralization brought about by infiltrating rainwater or, alternatively, these deposits represent pedogenic expression of fundamental hypogene processes. Early in his attempts to find a rational resolution to this dilemma, this geologist will realize that, in selecting the

rainwater possibility, he is confronted with a further difficulty, in addition to the previously identified unanswerable questions. Specifically, why do the observed veins with fairly large apertures exhibit M-textures (in the sense that they consist of pure mineral phases, conspicuously lacking internal impurities such as clays, silts, sands, pebbles, and small boulders)? After all, one is hard pressed to envisage either an open or progressively opening bedrock (or sand) fissure that, while being in-filled at vanishingly small rates by rainwater-borne precipitants, remains isolated from the overlying soil cover or detritus being transported at the topographic surface by wind action and run-off, including occasional flash floods. To get away from the apparent paradox, it is necessary to assume the presence of a non-delinquent "cleaning agent" that removes the perpetual supply of wind- and water-borne detritus. In sharp contrast to occasionally descending rainwater, ascending hypogene fluids would satisfactorily fulfill this necessary role. Having recognized that by selecting the rainwater possibility he is facing as many as three unanswerable questions, the inquiring geologist should not be satisfied with this possibility. It may be recalled that these unanswerable questions are: (1) why do the calcretes exhibit the observed textural diversity? (2) why do both the calcretes and the equivalent veins exhibit the observed compositional diversity? and (3) why do the presumably slowly forming veins exhibit the observed purity?

Hydrothermal Alteration

Third, at Yucca Mountain, examples of hydrothermally altered rocks are not particularly hard to find. Most commonly, the observed bedrock alterations occur in the form of fracture- and fault-based alteration aureoles, with thickness ranging from fractions of a centimeter to a few meters. Examples of such aureoles may be observed at several locations, including: (1) in the vicinity of Stage Coach Road, Figure 3-1; (2) along the eastern flank of Harper Valley, Figures 3-19 and 3-20; (3) along the trace of the Solitario Canyon fault, Figure 3-12; and (4) in the Trench #14b exposure. Both welded and unwelded ignimbrites are affected. Importantly, all of the locally known pyroclastic units, including both members of the Timber Mountain Tuff, exhibit more or less pronounced indications of having been subjected to hydrothermal alteration processes.

Based on visual examinations of hand specimens, four types of alteration may be observed. These are: (1) silicification; (2) "weak" argillic alteration; (3) locally "strong" oxidation; and (4) fracture- and fault-based devitrification of the Topopah Spring Member vitrophyre. Although for some of the Yucca Mountain surficial ignimbrites, a hydrothermally altered state is an indisputable fact, one may appropriately ask: (1) am I observing deuteric alteration? (i.e., alteration developed during the cooling and solidification of the host ignimbrites) or, alternatively, (2) am I observing subsequent epigenetic alteration? Based on

visual examinations alone, it is difficult to answer these questions with total certainty. There are, however, a few observations which, if taken collectively, indicate that at least some of the alteration features are epigenetic. Among these are: (1) epigenetic hydrothermal mineralization is locally known; (2) the youngest locally known unwelded ignimbrites (i.e., the Ammonia Tanks Member of the Timber Mountain Tuff) are affected; (3) devitrification of the Topopah Spring Member vitrophyre is restricted to pockets and bands centered on brittle deformational features, such as brecciated and slickensided fault zones; and (4) across fault contacts, stratigraphically distinct ignimbrites are equally affected.

Even assuming most conservatively that the deuteric possibility is on an equal footing with the epigenetic possibility, one is hard pressed to cite a single observation that would require to regard the former as superior to the latter. Of course, by conveniently selecting the former possibility, one may allay troublesome questions regarding the age of apparent hydrothermal activity. But to what end? In contrast, however, by either prudently reserving judgment or conservatively selecting the latter possibility, one brings these questions into sharp focus. In view of hydrothermal expectations as set forth in Section 2.5, particularly troublesome is the possibility that, at Yucca Mountain, the observed alteration represents overprinting of several hydrothermal episodes. Some of the suspected epigenetic alteration may represent the hydrothermal stage of activity of the Timber Mountain Caldera. Considering the local presence of both apparently

young calcretes and equivalent bedrock veins, however, the inquiring geologist may prudently suspect that not only some of the observed alteration is epigenetic but also that some of this alteration is significantly younger than the Timber Mountain hydrothermal episode. To set aside these suspicions, however, it is essential to perform microscopic and radiometric studies of the local alteration minerals.

Mosaic Breccias

Fourth, a truly striking feature, associated with the Yucca Mountain ignimbrites, is the widespread occurrence of very peculiar looking breccias. Such breccias are present in almost every large exposure. Examples of these breccias may be observed at numerous locations, including: (1) area around Stage Coach Road, Figures 3-20b and 3-21; (2) head of the Solitario Canyon, Figure 3-22; (3) the Trench #14 exposure, Figure 3-23b; (4) east and west flanks of Busted Butte, etc.

In a hand-specimen, breccias are composed of angular to sub-rounded clasts of various sizes firmly cemented by opaline silica with lesser involvement of micritic calcite. Both GS-textured breccias (Figure 3-21) and F-textured breccias (Figures 3-22 and 3-23) are present. The most areally extensive breccias occur in welded and stiff ignimbrites, such as those comprising the Tiva Canyon and the Topopah Spring Members of the Paintbrush Tuff. Typically, they are tens or hundreds of

meters wide, forming irregular but interconnected "pods" centered on several of the local faults. Breccia clasts appear to be authigenic in the sense that, compositionally, they resemble the unfragmented adjacent wallrock. Compositionally and texturally, the authigenic breccia cement is identical to the material contained in both the bedrock veins and the calcretes and sinters.

Much less commonly observed are occurrences of breccias in soft, unwelded ignimbrites. Typically, such breccias are developed as discontinuous dikes, up to 0.3m thick, Figures 3-24 and 3-25. They are particularly numerous and well developed along the eastern flank of Harper Valley. In all of the observed cases, the breccia dikes are GS-textured and contain allogenic and polymictic clasts. For some breccia dikes, compositional clasts are well rounded; for other dikes, however, these clasts are angular. The authigenic breccia cement consists mainly of opaline silica with lesser presence of micritic calcite. A remarkable feature associated with the breccia dikes is the width/length ratio. In one instance, the observed width/length ratio is as large as 0.2, see, for example, Figure 3-2a.

While considering the origin of the Yucca Mountain breccias, the inquiring geologist will first note that they occur in both the local pyroclastic rocks and the local sedimentary rocks, specifically the Paleozoic carbonates. Evidently, the eruptive emplacement of the local pyroclastic rocks, including their cooling and solidification, is not a factor involved in the formation of these breccias. Rather

than being auto-breccias (i.e., formed syn-depositionally with the host ignimbrites), these breccias appear to be epigenetic. As a next logical step, the inquiring geologist will note that breccias do not appear to have been formed solely via shear fragmentation associated with ordinary fault slip. This is so because, in contrast to typical fault breccias, the Yucca Mountain breccias lack any shear fabric. Furthermore, these breccias: (1) occur as irregular pods and discontinuous dikes; (2) in places, exhibit very large volumetric strain, ranging from several percent to tens of percent; and (3) as a rule, exhibit a volumetric strain that is remarkably isotropic; i.e., the observed dilation is similar in all three perpendicular directions. Having eliminated both the syn-depositional origin and the shear-fragmentation origin, the inquiring geologist is left with little choice but to reckon that the Yucca Mountain breccias are either "denudation" breccias (i.e., formed through in-filling of independently formed bedrock openings by erosionally derived clasts) or "explosive" breccias (i.e., produced as the result of large and rapid build-up of fluid/gas pressure), or both.

As far as the authigenic breccia cement is concerned, the inquiring geologist will note that, compositionally and texturally, this cement is very much like the material that occurs in both the bedrock veins and the calcretes and sinters. With reference to the origin of this cement, therefore, the geologist is confronted with his two old choices, namely: (1) either the authigenic cement was precipitated from infiltrating rainwater, or (2) the cement has been precipitated from

ascending hypogene fluids. Having selected the rainwater possibility, he is confronted with the difficulties identified previously. With specific reference to the local breccias, these difficulties are: (1) how to account for the observed F-textures? (2) how to account for the observed compositional alternations? and (3) how to account for the observed purity of the authigenic cement?

In contrast, having selected the upwelling possibility, the inquiring geologist is confronted with a clean and difficulty-free solution. In one broad sweep, this possibility accounts for all of the field observations made, including: (1) the observed textural and compositional diversity of the local calcretes and sinters; (2) the observed compositional diversity and purity of the bedrock veins; and (3) the observed compositional diversity and purity of the authigenic breccia cements. Within the context of upwelling possibility, there is no single field observation that either (1) is difficult to explain, or (2) has been left unaccounted for. There is no rational reason, therefore, to regard the upwelling possibility as either inappropriate or inferior to the rainwater possibility. Even if, most conservatively, both of these possibilities are regarded as being on an equal footing, and considering the potentially catastrophic consequences that may result from a malfunction of the proposed nuclear facility, it is clear that the upwelling possibility must be seriously considered. As a first step in such considerations, it is appropriate to examine, visually and microscopically, rock cores extracted from the Yucca Mountain vadose zone. The results of such examinations are briefly

summarized in the following section.

4.0 Mineralization and Alteration of the Yucca Mountain Ignimbrites, Based on the Results of Visual and Microscopic Examination of Rock Cores Extracted from the Vadose Zone

After becoming somewhat acquainted with conditions at the topographic surface, a logical next step for the inquiring geologist is to get familiar with the local subsurface conditions. At Yucca Mountain, an extensive drilling program has already been implemented, and several thousand meters of rock cores are currently available. These cores may be used to establish whether or not the vadose zone ignimbrites display any sign of having been influenced, sometime in the past, by epigenetic hydrothermal processes.

Figures 4-1 through 4-9 are photographs of some of the Yucca Mountain rock cores. Without exception, these cores represent ignimbrites occupying the local vadose zone. Stratigraphically, these ignimbrites belong to two units of the Paintbrush Tuff, specifically the Topopah Spring Member and the overlying Yucca Mountain-Pah Canyon Members. Examination of the enclosed photographs reveals that, in the Yucca Mountain vadose zone, expressions of hydrothermal processes are clearly apparent. Hydrothermal mineralization is represented by a variety of gangue veins emplaced in both unwelded and welded ignimbrites. Some of the observed veins do not appear to be associated with any discernible staining and alteration of the wallrock, Figures 4-1 and 4-2. Some of these veins enclose

breccia dikes composed of variably devitrified clasts, Figure 4-3. For some other veins, however, alteration of the wallrock is visible. In some cases, this alteration involves fracture-based devitrification of the vitric country rock, Figures 4-4 and 4-5. For other cases, however, the alteration consists of narrow bleaching of welded and devitrified ignimbrites, Figure 4-6. In addition to the emplacement of veins, past localized circulations of hydrothermal solutions manifest themselves through larger scale alteration aureoles. Examples of such aureoles are depicted in Figures 4-7 and 4-8 (oxidation of unwelded ignimbrites) and in Figure 4-9 (bleaching of welded and devitrified ignimbrites).

For the lower parts of the Yucca Mountain vadose zone, fracture-lining minerals were investigated by Carlos et al. (1990). These investigations were based on 15-20 samples selected from each of five of the Yucca Mountain boreholes, specifically USW G-1, G-2, GU-3, G-4, and UE-25a#1. Locations of these boreholes are shown in Figure 4-10. For all of the samples examined, the host rock is the Topopah Spring Member of the Paintbrush Tuff. The selected samples were first examined using a binocular microscope, and then sub-samples were taken for both thin-section analysis and scanning electron microscope (SEM) analysis. The fracture-coating minerals were also investigated employing the X-ray powder diffraction (XRD) method. Mineral identifications were accomplished by comparing the obtained XRD patterns with the JCPDS and the Los Alamos standard patterns. The results of the XRD analyses are summarized

in Figures 4-11 and 4-12.

The results of microscopic examinations of fracture-lining minerals, reported by Carlos et al. (1991), may be taken as indicating that, in the Yucca Mountain vadose zone, both a deuteric mineralization episode and a subsequent epigenetic mineralization episode, or episodes, are expressed. According to Carlos et al. (1990), the deuteric episode is represented by mineral species precipitated from both vapor and liquid phases. Typically, these species occur in lithophysal cavities, as well as along cooling and lithophysal fractures. They include tridymite (which may or may not be transformed into cristobalite or quartz), some hematite, fine-grained manganese oxides (rancieite, lithiophorite, and todorokite), and fine-grained zeolites (mainly mordenite). The subsequent epigenetic mineral species may be identified on the basis of the paragenetic relationships reported by Carlos et al. (1990). Among such mineral species are drusy quartz, fluorite, smectite, coarse-grained zeolites (heulandite, clinoptilolite, stellerite, and mordenite), and calcite. Epigenetic character of some drusy quartz is indicated by the fact that, in borehole USW GU-3, this mineral was observed to occur over the presumed deuteric tridymite and over pseudomorphs after such tridymite. Similarly, the reported paragenetic relationships indicate that, in borehole USW GU-3, fluorite was formed after tridymite and quartz. The occurrence of smectite appears to be ubiquitous. Typically, it occurs as a late alteration mineral, forming white powder-coatings over slickensides, fresh fractures, and zeolites. The

reported paragenetic relationships also indicate that, in a few boreholes, coarse-grained zeolites were formed after the presumed deuteritic manganese oxides and after the presumed deuteritic fine-grained mordenites. Furthermore, many of epigenetic zeolites are euhedral, neither crushed nor slickensided. In a few instances, such zeolites were observed to be developed over slickensided fractures. Evidently, some of the euhedral zeolites were formed late in Yucca Mountain deformational history. Calcite appears to be the latest mineral formed. In borehole USW G-2, it was observed to occur as two distinct generations, separated by the deposition of heulandite.

The epigenetic character of some of the vadose zone mineral species, as interpreted based on the reported paragenetic relationships, is in accord with viewpoints expressed by Weiss (1990). This investigator reported his impressions after non-destructive examination of Yucca Mountain rock cores. Intervals totaling about 1850 m were inspected. The following quotation from Weiss (1990) summarizes the most relevant observations and comments. *"1) In general, most of the visibly altered rocks contain alteration mineral assemblages (calcite, zeolites, montmorillonitic clay, chlorite(?), magnetite, pyrite) characteristic of propylitic alteration. 2) Quartz and quartz-calcite veins, and fracture-fillings are relatively uncommon in the intervals examined, particularly at shallower levels, although numerous thin silica- (opal, chalcedony), calcite/silica- and calcite-filled fractures and veinlets are present. In general, very little pervasive silicification or quartz*

veining was observed. 3) Pyrite is commonly present as disseminated grains within the groundmass and lithic fragments of ash-flow units in the deeper parts of several holes and is also present as irregular veins and replacements in some lithic fragments. A significant amount of disseminated and vein pyrite, and less common fluorite were found in somewhat silicified Paleozoic carbonate rocks recovered from drill hole UE-25p#1. We estimate the abundance of disseminated groundmass pyrite at a trace to <1 percent; pyrite is more commonly present and is more abundant in lithic fragments (generally as much as about 5 percent, but some are nearly completely replaced). Much of the pyritic, lithic-rich ash-flow tuff is probably part of the Lithic Ridge Tuff. If this is the case, average original FeO was about 1.20 wt. percent (Broxton et al., 1989) and the more pyritic intervals reflect a substantial addition of sulfur and an addition and (or) redistribution of iron. 4) Fluorite is present as veins and fracture-fillings and as irregular, drusy coatings lining lithophysal and (or) relict pumice cavities and is not confined to great depths, or the northernmost part of Yucca Mountain, but was observed in fractures at depth as shallow as 318 m and 362 m in drill hole USW GU-3. 5) A metallic mineral with a color similar to chalcopyrite, tarnished pyrite and gold, and with a form and habit unlike typical pyrite, was observed in core from a depth of about 1035 m in USW-G1. Positive identification of this material could be accomplished by scanning electron microscopy (non-destructive testing). 6) The presence of extensive and pervasive propylitic alteration, ±fluorite, in otherwise fresh tuffs of Miocene age clearly implies the existence of a large fossil

hydrothermal system in Yucca Mountain, and supports our earlier contention that zeolitic alteration may not be entirely of a diagenetic or deuteritic origin as is commonly believed."

In accounting for the rock core-based observations, two conflicting viewpoints may be put forth. On the one hand, considering both the manifestly magmatically active character of the region and the intended utilization of Yucca Mountain, one may elect to start with a more prudent and cautious viewpoint. In accordance with this viewpoint, and short of having unequivocal facts to the contrary, one may reasonably suspect that, in the Yucca Mountain vadose zone, most of the observed mineralization and alteration phases are epigenetic. Based on the inferred thermal evolution of the local crust, as set forth in Section 2.5, one may presume that these phases record, in part, the hydrothermal stage of activity of the Timber Mountain Caldera and, in part, subsequent intermittent activity of a hydrothermal system driven by conductively replenished heat sources. Based on more detailed studies of the local geologic record, it should be feasible to either confirm or reject this suspicion. In that regard, particularly helpful are the results of radiometric analyses of the local alteration and mineralization phases. However, the casual dismissal of the field observations giving rise to this suspicion, as provisionally explicable in a more inconsequential manner, can not possibly be regarded as a responsible and wise course of action.

On the other hand, it has also been proposed that, at Yucca Mountain, the observed alteration and mineralization phases represent a combination of processes ranging in origin from deuteric, through epigenetic-hydrothermal, to supergene, or diagenetic (i.e., brought about by infiltrating rainwater). This viewpoint is being aggressively advocated by various investigators associated with the Yucca Mountain Site Characterization Project. The indisputable hydrothermal mineral species (montmorillonitic clays, jasperoidal opal, drusy quartz, chalcedony, sparry calcite, fluorite, albite, analcime) are regarded as formed either during the cooling and solidification of the local ignimbrites or, shortly thereafter, during the hydrothermal stage of activity of the Timber Mountain Caldera, see, for example, Bish (1989) and Bolivar et al. (1990). The younger mineral species (micritic calcite, uraniferous opal, sepiolite, smectite, calcic zeolites), however, are regarded as having been produced with exclusive involvement of infiltrating rainwater. The alteration minerals (smectite and calcic zeolites) are regarded as "diagenetic," i.e., produced via glass $\leftrightarrow$ rainwater interactions, see, for example, Broxton et al. (1986) and WoldeGabriel (1990). The vein minerals (micritic calcite, uraniferous opal, sepiolite) are considered as having been precipitated from infiltrating rainwater, and as representing an introduction of rainwater-dissolved windblown dust into the interior of Yucca Mountain, see, for example, Szabo and Kyser (1990), Stuckless et al. (1991), and Peterman et al. (1991). This somewhat novel understanding of the subsurface vein-forming processes is rooted solely in the a priori belief that, at Yucca

Mountain, the contemporary water table is a permanent feature. However, if the objective is to rationally evaluate stability of the local water table then it is absolutely essential to demonstrate unequivocally, and independently of the tested premise, that the local vadose veins and associated wallrock alterations are either very old or their formation is the result of infiltrating rainwater, i.e., a newly discovered phenomenon. Short of that, the syllogism "the water table is a permanent feature and, consequently, all young veins from the vadose zone must have been precipitated from rainwater" is based on circular logic (premise becomes conclusion), and can not be considered to be serious and valid scientific reasoning.

The remainder of this report is devoted to evaluating of the respective merits of the above-stated viewpoints. A gamut of mineralogical, chemical, isotopic, and radiometric data is already available, and will be used for this purpose. The subsequent discussion is presented in five parts. Section 5.0 is concerned with interstitial fluids from the Yucca Mountain vadose zone. Recently, the results of both major and trace element analyses and stable and radiogenic isotope analyses were made available for samples of interstitial fluids. These results will be used to interpret the origin of the vadose zone interstitial fluids. Section 6.0 deals with the so-called "mosaic" breccias. Recently, samples of these breccias were analyzed for their base and noble metal concentrations, fission-track ages, stable isotope contents, and mineral composition. The results of these analyses will be used to interpret the age and origin of the Yucca Mountain "mosaic" breccias. Section 7.0

is concerned with the pro-grade zeolitic and montmorillonitic alteration of the Yucca Mountain ignimbrites. Spatial distributions of both the alteration grade and the chemical composition of alteration minerals are considered, together with the corresponding K/Ar ages. These data will be used to interpret the age and origin of the Yucca Mountain metamorphism. Section 8.0 deals with the Yucca Mountain carbonatization. The corresponding fluid inclusion, isotopic, and radiometric data are presented and considered. These data will be used to interpret the age and origin of the Yucca Mountain carbonatization. Finally, overall conclusions are presented in Section 9.0.

5.0 The Origin of the Vadose Zone Interstitial Fluids

5.1 General

For the Nevada Test Site area, differences in chemical compositions between interstitial fluids and fracture-based fluids have been observed by several investigators. Initially, such differences were noted by Keller (1960 and 1962). Using the interpretations of downhole electric logs, and the results of laboratory measurements of the electric conductivity of cores saturated with water of differing salinity, Keller concluded that the specific conductance of interstitial fluids may be 25-30 times as great as that of fracture-based fluids. For interstitial fluids, the estimated concentrations of total dissolved solids ranged from 3700 to 5100 ppm. In contrast, for fracture-based fluids the corresponding concentrations are commonly less than 500 ppm.

Differences in chemical compositions between fracture-based and interstitial fluids were also observed by White et al. (1980), and later by Russell (1987). These authors have shown that, relative to fracture-based fluids, interstitial fluids carry significantly higher concentrations of dissolved chloride and sulfate anions. White et al. (1980) stated that "*the mechanism responsible for differences in anion composition between interstitial and fracture waters is not obvious.*" These authors examined a few possibilities that could account for elevated sulfate and chloride

concentrations in the interstitial fluids. The first possibility was that drilling fluids may have contaminated these fluids. After considering both the results of dye tests and the results of laboratory measurements of permeability, however, this possibility was discounted. The second possibility was that the chemical composition of interstitial fluids reflects the composition of relict fluids, high in chloride and sulfate, that were present at the time of deposition of the local ignimbrites. The preservation of such fluids, over the time span of about 10^7 years, would require that a large percentage of pore space be insulated from the overall hydrologic system. Considering the relatively high interstitial permeability of the ignimbrites involved, however, this proposition was not regarded as a plausible one. The third possibility was that, for interstitial fluids, the type and extent of ionic exchange reactions with the host ignimbrites were different from those for fracture-based fluids. No prior reason was apparent, however, why chloride and sulfate anions would be concentrated preferentially in the interstitial fluids. Also, the fact that the major cation ratios, for both types of fluid, were similar was regarded as not supportive of the hypothesis of varient dissolution mechanisms.

Based on the results of analysis of a variety of data and observations from the Yucca Mountain area, Szymanski (1989) put forth a testable hypothesis regarding the origin of interstitial fluids from the local vadose zone. In his 1989 report, Szymanski made the following statement. "*The assumption that the Yucca*

Mountain groundwater system is developed in a deforming fractured medium, and that it involves a heat-fluid coupled flow, leads to another important issue: the origin and chemistry of interstitial waters in the Yucca Mountain vadose zone. If interpretations performed in this report are correct then it follows that interstitial waters represent, at least in part, relict hydraulic mounds developed in association with high groundwater stands at the end of tectonic cycles. Chemistry of such waters should be different from that of 'ordinary' meteoric waters infiltrating through the vadose zone. . . . As far as it could be determined, there are no published data concerning chemistry of interstitial water from the Yucca Mountain vadose zone. Consequently, there is no basis to either confirm or reject the hypothesis that interstitial waters are relict waters that originated from below the water table."

Recently, the results of chemical and isotopic analyses of samples of vadose zone interstitial fluids were made available. The analyzed fluid samples were extracted from rock cores obtained in the Yucca Mountain boreholes UE-25 UZ-4, and UZ-5. The main purpose of the following discussion is to present and evaluate the newly acquired isotopic and geochemical data. The discussion is presented in three parts. Section 5.2 describes the methodology employed in extracting fluid samples from rock cores. The newly acquired isotopic and chemical data are presented and analyzed in Section 5.3. In this section, an evaluation is made as to whether or not these data provide any factual support for the viewpoint that, at Yucca

Mountain, the vadose zone interstitial fluids are modified deep-seated fluid phases which were injected into the vadose zone. The overall summary and conclusions are presented in Section 5.4.

5.2 Interstitial Fluid Extraction Methods

All of the samples used in studies of the vadose zone interstitial fluids were extracted from rock cores obtained in the Yucca Mountain boreholes UE-25 UZ#4 and UE-25 UZ#5. Both of these boreholes were drilled to a depth slightly more than 100m, using air as a cooling agent. Use of the dry-coring technique prevents contamination of the interstitial fluids by drilling fluids. To obtain fluid samples for analyses, three different extraction techniques were employed. These are: (1) direct and vacuum distillations; (2) squeezing in a multiple-cycle triaxial compression apparatus; and (3) high-speed centrifuge extraction. The distillation technique involves heating of rock-core samples, under vacuum or otherwise, and capturing the vaporized interstitial fluids in a cold trap. The technique yields distilled fluids and consequently its application is limited to extracting samples for deuterium, tritium, and oxygen-18 isotopic analyses. The triaxial compression extraction technique involves squeezing core samples, in a compression chamber, by applying axial and confining pressures incrementally to force fluids and gases out of the pore space. Yang et al. (1988) have demonstrated that concentrations of

the extracted dissolved solids are fairly independent of the applied axial pressure. This observation suggests that the triaxial squeezing does not promote dissolution of the rock matrix and does not change the original composition of the extracted fluids. The extracted fluid samples are suitable for both ion concentration analyses and isotopic analyses. The high-speed centrifuge extraction technique employs a centrifuge to drive the interstitial fluids out of the pore space. The centrifuge extraction technique yields the largest quantities of interstitial fluids. The resulting fluid samples are also suitable for both ion concentration and isotopic analyses.

A potential shortcoming of the Yucca Mountain data base is that, thus far, no samples of the vadose zone fracture-based fluids have been obtained and analyzed. In a strict sense, therefore, both the chemical composition and the isotopic composition of these fluids remain unknown. However, it is highly unlikely that, in terms of chemical and isotopic compositions, the vadose zone fracture-based fluids are significantly different than the corresponding fluids from immediately below the contemporary water table. This is true, in particular, for shallow fluids containing significant relative concentrations of dissolved carbon-12 and carbon-14. For the purpose of interpreting the origin of the vadose zone interstitial fluids, these fluids may be used as an adequate reference.

5.3 Origin of the Vadose Zone Interstitial Fluids

The newly acquired isotopic and geochemical data, from samples of the vadose zone interstitial fluids, are presented in Figures 5-1 and 5-2. The relative concentrations of deuterium and oxygen-18, and the absolute concentrations of tritium, are summarized in Figures 5-1a through 5-1c. The analyzed samples were extracted employing either the distillation method or the triaxial compression method. For the stable isotope ratios, measurements were made employing the USGS Research Laboratory mass spectrometer. For these measurements, the analytical error is ± 0.2 per mil_{smow} for values of the $\delta^{18}\text{O}$ ratio, and ± 1.0 per mil_{smow} for values of the δD ratio. The tritium activity values were determined at the Tritium Laboratory of the University of Miami, Florida. The reported values are measured values adjusted back to the date of sample acquisition. For the tritium activity determinations, the analytical error is ± 5.0 T.U. Figure 5-2 presents the results of chemical analyses of six samples of the vadose zone interstitial fluids. Two different data sets are depicted. Figure 5-2a presents the concentrations of cations (alkali earth elements, alkali metals, transition metals, noble metals, and rare earth elements), as reported by Smith (1991). These concentrations were measured employing the inductively coupled plasma atomic emission spectrometry method. The analyzed samples were extracted using the high-speed centrifuge method. Figure 5-2b presents the concentrations of cations and anions, as reported by Yang (1989). These concentrations were determined

using the inductively-coupled plasma atomic emission spectrometry method for cations and the ion chromatography method for anions. Only limited quantities of the interstitial fluid were extracted. These quantities were sufficient neither for complete chemical analyses nor for duplex chemical and isotopic analyses. For all of the measured ion concentrations, the analytical error is less than ± 10 percent. In addition to the dissolved ion concentrations, Figure 5-2b also presents apparent carbon-14 ages and values of the $\delta^{13}\text{C}$ ratio, for two samples of the vadose zone interstitial fluids. Both the carbon-14 concentrations and the relative carbon-13 concentrations were determined employing the Tandem Accelerator Mass Spectrometer at the Department of Physics of the University of Arizona. For the $\delta^{13}\text{C}$ ratio, the analytical error is ± 0.2 per mil_{PDB}; for the carbon-14 concentration, the analytical error is ± 0.7 percent.

For fracture-based fluids from below the water table, the matching data set is presented in Figure 5-3. The represented fluids carry apparent carbon-14 ages ranging from 3.8×10^3 to about 30×10^3 years B.P. Figure 5-3a presents concentrations of the dissolved solids (major constituents, alkali earth elements, alkali metals, transition metals, noble metals, and rare earth elements) reported by Smith (1991) for samples of fluids from the local wells J-12 and J-13. These concentrations were determined using the ion chromatography method for anions and the inductively coupled plasma atomic emission spectrometry method for cations. Figures 5-3b and 5-3c present the results of standard chemical and

isotopic analyses of fluid samples obtained from sixteen wells drilled in the Yucca Mountain area. These analyses were made and reported by Benson and McKinley (1985).

Isotopic and chemical fluid composition data, that are judged to be pertinent to interpreting the origin of the vadose zone interstitial fluids, are also summarized in Figures 5-4 through 5-8. Figure 5-4 presents a relationship between the standard "meteoric water line" and the isotopic composition of the vadose zone interstitial fluids. This figure also specifies the weighted average values of the $\delta^{18}\text{O}$ ratio for local atmospheric precipitation. Comparisons of the chemical compositions, for both vadose zone interstitial fluids and fracture-based fluids, are presented in Figure 5-5. Two different comparisons are shown. For Figure 5-5a, the comparison reference is the chemical composition of fracture-based fluids from the "saturated" zone, and includes fluids residing in both the Tertiary ignimbrites and the Paleozoic carbonates. For Figure 5-5b, however, the comparison reference is constructed based on the chemical compositions of three distinct fluid phases from the Paleozoic carbonates alone. The distinct chemical characters of these fluid phases were attributed to the corresponding differences in the fluid intraformation residence time. The chondrite-normalized rare-earth-element (REE) abundance patterns are shown in Figure 5-6. These patterns were constructed based on the REE abundances, as determined for samples of both the J-12 and J-13 fracture-based fluids and the vadose zone interstitial fluids. Figure

5-7 presents the chondrite-normalized REE abundance patterns for three stratigraphic members of the Crater Flat Tuff (reservoir rocks for the Yucca Mountain fracture-based fluids). These patterns were constructed based on the average results from four samples of each member.

The REE abundance patterns for hydrothermal solutions from France and Bulgaria are shown in Figures 5-8a and 5-8b. Figure 5-8a represents the CO₂-rich fluids from Vals-Les-Bains, France. These fluids emerge from closely spaced springs, located along traces of the Sandron Valley faults in the Massif Central. They are produced through intermixing of superficial poorly-evolved meteoric fluids with upwelling, highly mineralized hydrothermal solutions. The latter solutions well up from a granitic body and are regarded as having been equilibrated at a temperature of about 170° Celsius. Importantly, the Vals-Les-Bains fluids occur as more diluted and more concentrated varieties, yielding contrasting REE abundance patterns. This offers an opportunity for evaluating factors that could have been involved in producing the REE abundance pattern as observed, at Yucca Mountain, for the vadose zone interstitial fluids. Figure 5-8b presents results for hydrothermal solutions from Bulgaria. These solutions exhibit: (1) head-temperatures ranging from 47° to as much as 97° Celsius; (2) alkalinity ranging from 1.68 to as much as 21 meq per kg; and (3) equilibration temperatures ranging from 45° to 130° Celsius. They occur as high-alkalinity and low-alkalinity varieties, yielding contrasting REE abundance patterns. Again, the

Bulgarian hydrothermal solutions may be used to illustrate the potential significance of the REE abundance pattern as observed, at Yucca Mountain, for the vadose zone interstitial solutions.

Examination of Figures 5-1 through 5-8 leads to the formulation of the following five observations/interpretations. First, relative to the standard "meteoric water line," the vadose zone interstitial fluids exhibit a very pronounced "oxygen isotopic shift." As shown in Figure 5-4, the magnitude of this shift ranges from 0.5 to about 2.5 per mil_{SMOW}. If the regression line through the observed δD and $\delta^{18}O$ values is extended back to the "meteoric water line", the resulting intercept occurs at a $\delta^{18}O$ value of about -14.5 per mil_{SMOW}. Ingraham et al. (1990) have shown that, for land surface altitudes comparable to those of the Yucca Mountain area, contemporary atmospheric precipitation yields weighted average values of the $\delta^{18}O$ ratio ranging from -11.3 to -11.7 per mil_{SMOW}. The apparent discrepancy between the observed input values (-11.3 to -11.7 per mil_{SMOW}) and the -14.5 per mil_{SMOW} intercept value indicates that, for the vadose zone interstitial fluids, the observed "oxygen isotopic shift" may not be attributed solely to the evaporative oxygen-18 enrichment of the local surficial and year-round recharge. While dealing with this discrepancy, Yang (1989) explained the observed isotopic shift by assuming that, at Yucca Mountain, only winter precipitation (rain or snowmelt) infiltrates into the vadose zone. The surficial oxygen-18 evaporative enrichment of such recharge satisfactorily accounts for the observed isotopic

compositions of the vadose zone interstitial fluids. Based on the stable isotope data alone, however, this interpretation may or may not be correct. An alternate explanation, one that equally well explains the observed stable isotope data, is that the observed "oxygen isotopic shift" has been produced through in-situ evaporative oxygen-18 enrichment of the interstitial fluids (after introduction of these fluids into the pore space). As a matter of fact, this interpretation is suggested by two observations. On the one hand, fracture-based fluids, from both the vadose zone and from below the water table, plot at or very near the "meteoric water line", Russell (1987) and Kerrisk (1987). Evidently, for the Nevada Test Site recharge, surficial oxygen-18 evaporative enrichment is not a significant factor. On the other hand, for the Yucca Mountain area, evaporative removal of water from the vadose zone has been actually observed. Thorstenson et al. (1989), for example, reported that, during a 30-month long monitoring period of the Yucca Mountain borehole UZ-6S, *"there has been a net flux to the atmosphere of approximately 40m³ of liquid water and 1150 kg of carbon."*

Second, the tritium activity data, if considered together with the carbon-14 data, indicate that at Yucca Mountain the vadose zone interstitial fluids were produced through intermixing of fluid phases carrying differing radiometric ages. As shown in Figure 5-2b, two of the analyzed samples of interstitial fluids yielded relative concentrations of carbon-14 of 55.1 and 88.6 percent of the modern carbon standard. The corresponding carbon-14 ages are 4.9×10^3 and 1×10^3 years B.P.,

respectively. Importantly, the vadose zone interstitial fluids do contain measurable concentrations of tritium. As shown in Figures 5-1a and 5-1b, the majority of analyzed samples yielded tritium activity values that significantly exceed the combined activity values attributable to both analytical error and expected background levels. If considered in isolation, the tritium activity data would lead to the conclusion that the interstitial fluids were introduced into the vadose zone pore space during the last 30-40 years. Based on the carbon-14 data, however, we do know that this conclusion is erroneous. Short of questioning the integrity of either the tritium activity data or the carbon-14 data, there is no alternative but to conclude that the vadose zone interstitial fluids are multi-component solutions. Evidently, the intermixing of young tritium-carrying fluids with some older fluids is responsible for the observed dichotomous radiometric ages.

Third, relative to fluid phases resulting from "ordinary" ionic exchange reactions between rainwater and the local ignimbrites, the vadose zone interstitial fluids are strongly discordant. Such discordance is evident through a comparison of the respective concentrations of dissolved solids. As shown in Figure 5-5a, the average concentrations of ionic pairs $\text{Ca}^{++} + \text{Mg}^{++}$ and $\text{Cl}^- + \text{SO}_4^-$, dissolved in the interstitial fluids, are a factor of about 10 higher than the corresponding concentrations for fracture-based fluids from the local ignimbrites. Importantly, the latter fluid phases carry apparent carbon-14 ages ranging from 3.8×10^3 to

about 27×10^3 years B.P., Figure 5-3b. Evidently, these fluid phases were undergoing ionic exchange reactions with the local ignimbrites for a fairly long time. Even greater compositional discords are evident in a comparison of the trace elements dissolved in the respective fluid phases. As shown in Figures 5-2a and 5-3a, relative to the J-12 and J-13 fracture-based fluids (with apparent carbon-14 ages of 9.1×10^3 and 9.9×10^3 years B.P., respectively), the vadose zone interstitial fluids contain significantly higher concentrations of dissolved alkali earth elements (Ca, Mg, Sr, and Ba). The concentrations of dissolved alkali metals (Na, K, Rb, and Cs), however, are comparable. The other most obvious compositional discords are: (1) a factor of 20-30 difference in concentrations of iodine and chloride; (2) a factor of 300-500 difference in concentrations of molybdenum and tungsten; (3) a factor of 10-100 difference in concentrations of lanthanum and yttrium; (4) significantly different concentrations of dissolved transition metals (Mn, Ni, Cu, and Zn); and (5) significantly different concentrations of dissolved noble metals (Au and Pt). Recognizing the above compositional discords, Smith (1991) concluded that *"the compositional differences . . . indicate that the pore waters have evolved by significantly different processes."* Specifically, the vadose zone interstitial fluids appear to be composed of two genetically very distinct fluid phases. One of these fluid phases, of course, is infiltrating rainwater. Its presence is clearly expressed through both the above-background tritium activity values and the strongly negative values of the $\delta^{13}\text{C}$ ratio. As shown in Figure 5-2b, the latter values range from -20.0 to as low as -26.7 per mil_{PDB}. Such

low values indicate that the main source of carbon, incorporated in the vadose zone interstitial fluids, is the local biosphere. The origin of the remaining fluid phase, however, is truly enigmatic. The observed elevated concentrations of both the dissolved major constituents and the dissolved trace elements suggest that one is dealing with remnant, highly-evolved hydrothermal solutions, either deuteric (relict) or epigenetic (remnant).

Fourth, in terms of the dissolved major constituents (expressed as concentrations of $\text{Na}^+ + \text{K}^+$, $\text{Ca}^{++} + \text{Mg}^{++}$, and $\text{Cl}^- + \text{SO}_4^-$ ionic pairs), the interstitial fluids from the Yucca Mountain vadose zone exhibit strong geochemical affinity with the typical calcium-magnesium fluid phases residing in the local Paleozoic carbonates. Two lines of evidence may be put forth in support of this statement. On the one hand, the interstitial fluids carry high concentrations of dissolved alkali earth elements (Ca, Mg, Sr, and Ba), relative to dissolved alkali metals (Na, K, Rb, and Cs). This relative abundance suggests that the interstitial fluids, rather than having acquired their dissolved solids through ionic exchange reactions with the alkali ignimbrites, may have acquired them through ionic exchange reactions with the underlying carbonate rocks instead, possibly in a CO_2 -rich environment. On the other hand, for the vadose zone interstitial fluids, observed concentrations of $\text{Ca}^{++} + \text{Mg}^{++}$ and $\text{Cl}^- + \text{SO}_4^-$ ionic pairs are identical to those for the Paleozoic carbonate-based fluids, Figure 5-5a. In the Yucca Mountain area, the latter fluids well up along the Tertiary-Paleozoic fault contact, and were encountered in well

UE-25p#1 at a depth of about 1200m, Szymanski (1989). Furthermore, in terms of the Chebotarev groundwater evolution sequence, the vadose zone interstitial fluids appear to represent the intermediate evolutionary stage. Such evolutionary affinity may be illustrated using, as a reference, the results of aqueous geochemistry studies performed by Maxey and Mifflin (1966). As shown in Figure 5-5b, for the vadose zone interstitial fluids the relative concentrations of the index ionic pairs ($\text{Na}^+ + \text{K}^+$, $\text{Ca}^{++} + \text{Mg}^{++}$, and $\text{Cl}^- + \text{SO}_4^{--}$) are identical to the corresponding concentrations for fluid phases from the so-called regional flow system. This is in sharp contrast to fluid phases from the so-called local flow system. For this system, the intraformation residence time is fairly short, and the corresponding fluid phases carry much lower concentrations of the dissolved $\text{Na}^+ + \text{K}^+$ and $\text{Cl}^- + \text{SO}_4^{--}$ ionic pairs. The geochemical affinity between the vadose zone interstitial fluids and the evolved Paleozoic carbonate-based fluids is important. It is telling us that, as far as the Yucca Mountain vadose zone interstitial fluids are concerned, the preserved hydrothermal component does not represent relict deuteric fluids. Instead, this component appears to be epigenetic, or remnant.

Fifth, for the vadose zone interstitial fluids, the observed chondrite-normalized REE abundance pattern is unexpected and highly unusual. Two observations may be put forth in support of this statement. On the one hand, relative to the host ignimbrites, the interstitial fluids carry the "reverse" REE abundance pattern. As shown in Figure 5-6, for the vadose zone interstitial fluids the chondrite-

normalized REE abundance pattern shows a very pronounced enrichment in heavy rare earth elements (HREE), and a depletion in light rare earth elements (LREE). The value of the Ce/Yb ratio is uncommonly low, less than 10^{-1} . In contrast, samples of the local ignimbrites yield a chondrite-normalized REE abundance pattern similar to that typical of most terrestrial rocks. As shown in Figure 5-7, such an abundance pattern is characterized by: (1) LREE relative enrichment; (2) a negative Eu anomaly; and (3) slight HREE relative depletion. The value of the Ce/Yb ratio is substantially higher, about 10^1 . The observed discords between these REE abundance patterns strengthen earlier suspicions that (1) the interstitial fluids are not relict deuteric fluids, and (2) the interstitial fluids did not acquire their chemical composition through "ordinary" rock $\leftrightarrow$ meteoric fluid ionic exchange reactions. On the other hand, the REE abundance pattern observed for the vadose zone interstitial fluids is similar to that associated with certain hydrothermal solutions. As shown in Figures 5-8a and 5-8b, the HREE relative abundance is a feature observed in association with CO₂-rich, highly-evolved, and concentrated hydrothermal solutions. In particular, in the pristine state and prior to CO₂ degassing near the topographic surface, such solutions exhibit high values of both CO₂ partial pressure and alkalinity. These features may be regarded as providing an indication of high relative abundances of REE complexing anions, such as mono- and bi-carbonates. An inclination of the smoothed REE curve, whether positive or negative, indicates that fractionation of the dissolved REE has occurred during fluid $\leftrightarrow$ rock ionic exchange reactions.

This fractionation, in turn, reflects the solubility of the LREE complexes relative to the HREE complexes. Empirical observations have led Michard and Albarede (1986) and Michard et al. (1987) to conclude that the relative abundances of the complexing carbonate anions promote the entrainment of HREE by hydrothermal solutions. Such solutions, therefore, are characteristically enriched in HREE and carry the lowest values of the Ce/Yb ratio. The opposite is true for solutions that carry very low concentrations of complexing carbonate anions. For the Yucca Mountain interstitial fluids, the observed chondrite-normalized REE abundance pattern further strengthens the earlier suspicion that, partially, these fluids are remnant (epigenetic) hydrothermal solutions. In the pristine state, these solutions could have been carrying high values of CO₂-partial pressure and high concentrations of complexing carbonate anions.

Putting together all five of the above observations/ interpretations leads to the construction of a realistic and complete interpretation of the chemical and isotopic compositions of interstitial fluids, as observed in the Yucca Mountain vadose zone. In accordance with this interpretation, the interstitial fluids contain two genetically distinct fluid phases. One of these phases is rainwater that, during episodic recharge events, is being introduced into the vadose zone pore space. Its presence is clearly manifested through both above-background tritium activity values and strongly negative values of the $\delta^{13}\text{C}$ ratio. The other fluid phase seems to represent an oxygen-18 enriched and chemically-evolved hydrothermal

solution. Its presence is manifested through: (1) elevated concentrations of the dissolved alkali earth elements; (2) abnormally high concentrations of the dissolved transition and noble metals; and (3) relative HREE enrichment and overall high abundance of REE. It is possible that, in part, the hydrothermal fluid component has acquired its chemical character through dissolution of the Paleozoic carbonates in a CO₂-rich environment. In contrast to the rainwater component, however, the latter component must have been introduced into the vadose zone pore space sometime in the past. The intermixing with rainwater, and the resulting isotopic contamination (both the acquisition of tritium and biogenically-derived carbon and the evaporative oxygen-18 and deuterium enrichment) appears to be occurring in-situ, and involves both the vapor phase and the liquid phase. In part, the intermixing and the resulting fluid exchange may be facilitated by the observed, convective or otherwise, circulations of vadose zone gases. During no-recharge periods, these circulations may be presumed to cause the local and differential "drying" of the vadose zone rocks. A net flux to the atmosphere of the vadose zone water, as reported by Thorstenson et al. (1989), may be the result of such "drying." The process involves vapor phase and, consequently, only pure (distilled) water is removed. The dissolved solids, however, remain behind. During episodic recharge events, the evaporative losses are being compensated for by in-flux of chemically very immature rainwater. The result of this back and forth fluid exchange seems to be of a two-fold character. On the one hand, the vadose zone interstitial fluids evolve isotopically with time

becoming very much like stagnant and shallow rainwater. Accordingly, these fluids exhibit: (1) evaporative oxygen-18 and deuterium enrichments; (2) above background tritium activity values; and (3) strongly negative values of the $\delta^{13}\text{C}$ ratio. On the other hand, being strongly held by capillary forces, the vadose zone interstitial fluids remain immobile and while exchanging vapor phase components, maintain their original concentrations of dissolved solids. In spite of their isotopically youthful-meteoric appearances, these fluids also exhibit sharply contrasting geochemical characteristics. These are: (1) elevated concentrations of dissolved alkali earth elements, relative to dissolved alkali metal concentrations; (2) abnormally high concentrations of dissolved transition and noble metals; and (3) an unusual REE abundance pattern.

The foregoing conceptual understanding of the origin of the vadose zone interstitial fluids may be tested using the stable isotope data. The results of this test are presented in Figure 5-9. It may be observed that, for the vadose zone interstitial fluids, the postulated fluid exchanges do satisfactorily explain the observed relative concentrations of deuterium and oxygen-18. In constructing the stable isotope test, two reasonable assumptions were made. It has been assumed that the rainwater component plots at the "meteoric water line," and its isotopic composition may be represented by the weighted average δD and $\delta^{18}\text{O}$ values for the local contemporary atmospheric precipitation. For the hydrothermal component, it has been assumed that there was a slight "oxygen isotopic shift,"

caused by geothermal rock <--> fluid isotopic exchange reactions, and its isotopic composition was similar to that of the Paleozoic carbonate-based fluids from the local well UE-25p#1.

5.4 Summary and Conclusions

The analyses of isotopic and geochemical data, as performed in the preceding section, lead to the formation of provisional views regarding the origin of interstitial fluids encountered in the Yucca Mountain vadose zone. For the purposes of this report, these views may be summarized by the following five points. First, as expected, the chemical character of the vadose zone interstitial fluids is distinctly different from that observed for the corresponding fracture-based fluids below the water table. Relative to the latter fluids, the vadose zone interstitial fluids carry significantly higher concentrations of: (1) dissolved alkali earth elements; (2) dissolved $\text{Cl}^- + \text{SO}_4^{2-}$ ionic pair; (3) dissolved transition and noble metals; and (4) dissolved lanthanum and yttrium. The compositional discords are sufficiently strong to justify the conclusion drawn by Smith (1991). This conclusion is that, relative to the corresponding fracture-based fluids, the vadose zone interstitial fluids "*must have evolved by significantly different processes.*"

Second, in terms of isotopic composition, the vadose zone interstitial fluids are largely controlled by surficial processes, including biological activity (carbon) and human activity (tritium). The observed relative deuterium and oxygen-18 enrichments are readily attributable to evaporation of stagnant and shallow waters. The isotopic character of carbon incorporated in the vadose zone interstitial fluids is uniquely characteristic of biogenically recycled carbon. The above background tritium activity values indicate that the vadose zone interstitial fluids are being contaminated with less than 30- to 40-year old rainwater.

Third, in terms of the major dissolved constituents, the vadose zone interstitial fluids exhibit strong geochemical affinity with calcium-magnesium fluid phases. At the Nevada Test Site, including the Yucca Mountain area, the latter fluid phases typically reside in the Paleozoic carbonates. The noted geochemical affinity is expressed through similarities of the relative and absolute concentrations of dissolved $\text{Na}^+ + \text{K}^+$, $\text{Ca}^{++} + \text{Mg}^{++}$, and $\text{Cl}^- + \text{SO}_4^-$ ionic pairs. This affinity is also expressed through the abundance of dissolved alkali earth elements, relative to dissolved alkali metals. In terms of the Chebotarev groundwater evolution sequence, the vadose zone interstitial fluids seem to be associated with the intermediate evolutionary stage. Relative to the least evolved local fluids, the interstitial fluids carry order-of-magnitude higher concentrations of the dissolved $\text{Ca}^+ + \text{Mg}^+$ and $\text{Cl}^- + \text{SO}_4^-$ ionic pairs. For the interstitial fluids, the geochemical affinity with the Paleozoic carbonate-based fluids is important. It clearly indicates

that the vadose zone interstitial fluids were not produced through intermixing of rainwater with relict deuteric fluids.

Fourth, the observed isotopic and chemical characteristics of the vadose zone interstitial fluids may be adequately explained by assuming that these fluids result from intermixing of rainwater with remnant hydrothermal solutions.

Presence of the latter solutions is suggested by several lines of evidence. Among these are: (1) elevated concentrations of transition metals; (2) elevated concentrations of some noble metals; (3) elevated concentrations of REE; and (4) the relative abundance of HREE. The observed REE abundance pattern may be explained by assuming that the hydrothermal component has evolved under conditions promoting fractionation of REE. This fractionation in turn may be explained by assuming that, at least in part, the hydrothermal component has acquired its chemical character through dissolution of the Paleozoic carbonates in a CO₂-rich environment. The resulting high abundance of the complexing carbonate anions caused substantial enhancement of HREE solubility relative to LREE solubility.

Fifth, if the foregoing interpretations are either partially or completely correct, then it follows that at Yucca Mountain the vadose zone ignimbrites were influenced by epigenetic hydrothermal processes. Fluid phases participating in such processes seem to have been calcium-magnesium in bulk composition, and

could have contained large quantities of dissolved CO₂. During their ascent through the alkali and still vitric ignimbrites, such fluid phases must have left behind a multitude of other expressions. One may reasonably speculate that, among such expressions are: (1) "explosive" fragmentation of the vadose zone bedrock, brought about by rapid CO₂ degassing; (2) subsequent cementation of the fragmented bedrock; (3) localized enrichment of base and noble metals and path-finder elements; (4) metasomatic carbonatization and zeolitization of the vitric ignimbrites; (5) stratigraphy-independent pro-grade hydrothermal metamorphism; (6) transgressive emplacement of various hydrothermal veins, relative to stratigraphic boundaries, etc. All of the above forms of expression of hydrothermal activity are rather conspicuous; commonly experienced geologists have little trouble in recognizing them. Considering the intended utilization of the Yucca Mountain area, and taking advantage of the very extensive local data base, it is imperative to examine, objectively and thoroughly, the local geologic record. The sole purpose of such an examination is to establish whether or not this record contains any indications that the expected expressions of epigenetic hydrothermal processes are indeed present. The remaining sections of this report are devoted to this end. The discussion is focused on the calcium-magnesium fluid phases, and, in particular, on the expected alteration-mineralization products of these fluid phases.

6.0 The Origin and Age of the Yucca Mountain "Mosaic" Breccias

6.1 General

While traversing Yucca Mountain, an observer is immediately confronted with several types of thought-provoking lithic materials. One such type occurs along the traces of locally-mapped faults, and consists of so-called "mosaic" breccias. The combined thickness of these "mosaic" breccias, relative to the total length of the traverse, is surprisingly large. In a hand specimen, "mosaic" breccias consist of either angular or subrounded fragments of the local ignimbrites embedded in a matrix composed of a mineral assemblage of calcite, opaline silica, and sepiolite. Remarkable characteristics associated with these breccias are: (1) their "matrix" support textures, i.e., individual breccia clasts are not in direct contact with each other; (2) very large volumetric strain (dilation), ranging from several percent to tens of percent; and (3) very isotropic character of the volumetric strain, i.e., the amount of dilation is comparable in all three mutually perpendicular directions.

To account for the observed widespread occurrence of "mosaic" breccias, two sharply contrasting viewpoints have been put forth. On the one hand, based on field inspections of the Trench #14 exposure, Hanson et al. (1986) have diagnosed "mosaic" breccia as "*hydrothermal eruption breccias*." Similarly, hypogene origin has been proposed by Szymanski (1989) who, after extensive field examinations,

concluded that "mosaic" breccias are polygenetic mixtures of so-called "explosive" breccias and so-called "fragmentation" breccias. The phrase "explosive breccia" was employed to denote a breccia for which formations of clasts and matrix are linked to a common hypogene process and, in the case of clasts, require large and very fast build-up of fluid/gas pressure (e.g., hydrothermal eruptions, gas-assisted seismic pumping, ordinary seismic pumping, wallrock fragmentation by vibratory ground motion, etc.). In contrast, the phrase "fragmentation breccia" was employed to denote a breccia for which formations of clasts and matrix are caused by two independent processes (one supergene and another hypogene) operating at the same time and in the same space (e.g., [1] mass wasting and filling up of a bedrock opening within which minerals are precipitating from supersaturated solutions; [2] rapid cementation of a mass wasting infill of a topographic depression, whether structural or erosional, by mineral-rich solutions, etc.). The proposed hypogene interpretation was largely based on the observed matrix-support textures. Such textures were taken as indicating that (1) authigenic cement was formed concurrently with the introduction of breccia clasts, and (2) rates of both the clast introduction and the cement precipitation were comparable. For a sizable fault-scarp, such as that observed in the Trench #14 exposure, the rates of clast introduction, resulting from both land surface transport and bedrock-scarp degradation, were judged to be fairly significant. Under such conditions, the comparably high rates of precipitation of authigenic cement could be accounted for only by assuming the involvement of mineral-rich

solutions, discharging from adjacent faults and fissures. The hypogene affinity of such hypothetical solutions was inferred based on two additional observations. First, some breccia clasts were found to exhibit alteration fringes, or rims. Second, both of the brecciated stratigraphic units juxtaposed by faulting, specifically the Tiva Canyon Member of the Paintbrush Tuff and the Rainier Mesa Member of the Timber Mountain Tuff, were found to exhibit clear indications of having been subjected to epigenetic hydrothermal alteration.

On the other hand, it has also been proposed that the Yucca Mountain "mosaic" breccias owe their existence solely to supergene processes. This viewpoint is being advocated by some investigators associated with the Yucca Mountain Site Characterization Project. The following quotations, from Levy and Naeser (1991), are believed to provide an accurate representation of this viewpoint. *"CTM (crushed-tuff-matrix) breccias at Busted Butte, interpreted as having formed by settling of the host pyroclastic unit shortly after deposition, are not directly related to recent fault movement. AMC (authigenic- mineral-cemented) breccias probably formed by varying combinations of surface erosion and deposition (including possible aeolian input) and recent near-surface fault movement. Most of the authigenic mineral cementation of AMC breccias is related to surficial pedogenic processes, as shown by the abundance of plant remains, but the origin of some fracture and pore fillings is ambiguous. . . . Interpretation of the hydrothermally*

altered CTM breccia in the upper Topopah Spring tuff at Busted Butte is based on texture, mineralogy, and the field-scale distribution of alteration. The altered breccia is located in the transition zone from vitric to devitrified tuff. Another transition zone occurs in the lower part of the unit marking the downward change from devitrified tuff to vitrophyre. This zone is characterized by fracture-controlled devitrification and moderate- temperature hydrothermal alteration. Secondary minerals in this lower zone include quartz, chalcedony, opal, smectite, zeolites, and iron oxyhydroxides. Lower Topopah Spring transition-zone alteration is nearly ubiquitous but the volume of rock affected and the abundance of secondary minerals show extreme lateral variations. The alteration is attributed to downward percolation of meteoric water along fractures in a still-warm pyroclastic deposit; alteration was localized in the transition zone because the glass had cooled too quickly for complete devitrification but retained enough heat that downward-moving water could induce partial devitrification, glass dissolution, and secondary mineral deposition."

The main purpose of the discussion contained in this section is to evaluate the respective merits of the above-stated viewpoints. A variety of geochemical and isotopic data is currently available and will be used for this purpose. The discussion is presented in three parts. Section 6.2 summarizes the results of thin-section examinations of samples of the "mosaic" breccia, from the Trench #14 exposure and from the Busted Butte area. Section 6.3 is concerned with the origin

of "mosaic" breccias. The presented interpretation is based on the results of various analyses, including field and thin-section observations, base and noble metals and path-finder element assays, oxygen and carbon isotopic analyses, and fission-track analyses of zircon crystals incorporated in authigenic cements of "mosaic" breccias. The overall summary and conclusions are presented in Section 6.4.

6.2 Summary of the Results of Thin-Section Examinations

Textural and compositional characteristics of the Yucca Mountain "mosaic" breccias have been investigated using electron microscopy. The samples examined represent "mosaic" breccias from the Busted Butte area and from the Trench #14 exposure. The results of these investigations were reported by Levy and Naeser (1991); all of the thin-section observations presented and discussed in this section are based on their work. An ISID-500 scanning electron microscope was used to produce back-scattered and secondary-electron images of carbon-coated polished sections. Mineral identification in SEM images was aided by qualitative examination of energy-dispersive X-ray spectra.

Based on textural and compositional characteristics, "mosaic" breccias were found to consist of two distinct varieties. The so-called crushed-tuff-matrix (CTM) breccias consist of bedrock clasts in a matrix of finely-crushed bedrock, and

contain little authigenic cement. In contrast, the so-called authigenic-mineral-cemented (AMC) breccias contain very little of the finely-crushed bedrock, and the matrix is composed almost entirely of authigenic cement. The CTM breccias contain no plant remains, whereas the AMC breccias contain abundant root casts, or rhizoliths.

For the Busted Butte CTM breccias, clasts are composed of fragments of the densely-welded Topopah Spring Member (devitrified) and aggregates of vapor-phase minerals, mostly tridymite. Morphologically, breccia clasts range from sub-angular to sub-rounded. Within the finely-crushed bedrock matrix, there is a considerable grain size diversity. For small domains, some 500 micrometers across, the grains may be as small as 2.0 to about 20 micrometers. More commonly, however, the grains range in size from 5.0 to about 500 micrometers. Some breccia clasts have textures in which crystals are coarser near the edges of clasts and appear to have grown beyond the original boundaries of the clasts. Other breccia clasts, however, have no coarse alteration fringes or possess incomplete fringes, broken off in places. The CTM breccia phase contains small amounts of authigenic cement. The silica, calcite, and sepiolite mineral assemblage is the main constituent of this cement. Silica phases are the most abundant of authigenic minerals. In smaller fractures and pores, silica occurs in the form of a simple isopach cement. In larger fractures, the texture of silica in-fillings is more complex and includes botryoidal forms. In addition to silica,

fractures and pores are in-filled with calcite. In one specimen of the CTM breccia, calcite was observed to occupy central voids of the silica-lined pores. Sepiolite was found to be present mainly in pore space; it has not been observed as a fracture-filling mineral. The pore-filling sepiolite occurs as plumose aggregates, which are composed of elongated ribbons, about one micrometer thick.

For the AMC breccia phase, clasts are composed of a variety of lithic materials, including both the Topopah Spring Member (Busted Butte) and the Tiva Canyon Member (Trench #14), CTM breccia fragments, and chunks of lithophysal cavity-lining minerals (tridymite and drusy quartz). Accessory clasts include both allogenic and authigenic varieties. The allogenic variety includes fragments of muscovite and zircon crystals. The authigenic variety includes: (1) calcitic ooids; (2) fragments of laminated opal-calcite aggregates; and (3) sepiolite pods. Both GS-textured breccia specimens and F-textured breccia specimens have been observed. Authigenic cement was found to occur either as in-filling of the breccia pore space or in-filling of dilated fractures. Similar to the CTM breccia phase, the AMC breccia cement was found to be composed of the mineral assemblage of silica, calcite, and sepiolite. The silica component was found to exhibit three textural varieties. Some vesicles and root casts were observed to be lined with spherulitic silica; individual spherulites are about 0.01 mm in diameter. Within the breccia pore space, silica was commonly observed to display fibrous texture. Individual fibres, about 0.05 mm in length were observed to exhibit a tendency to be aligned

Numerous mineralogical analyses of rock cores have revealed that, at Yucca Mountain, several zeolitic species are present. Among these species are mordenite, heulandite (= clinoptilolite), chabazite, erionite, phillipsite, stilbite (= stellerite), laumontite, and analcime (= wairakite), Broxton et al. (1986), Bish and Chipera (1989), and Carlos et al. (1990). Pressure and temperature conditions under which these species crystallize have been measured in active hydrothermal regions of the world. Figures 7-4a through 7-4h present the results of these measurements. The attached quotations are from a recent book by R. W. Tschernich entitled "Zeolites of the World." A review of these quotations is a useful first step in interpreting the origin of the Yucca Mountain zeolites.

7.3 The Spatial Distribution of Alteration Minerals

For the Yucca Mountain area, the currently available mineralogical data, including chemical composition data, are summarized in Figures 7-5 through 7-10. These data represent the results of various analyses of core samples extracted from a number of local boreholes. Locations of these boreholes are shown in Figure 7-5. For twelve of the local boreholes, the relative abundances of preserved glasses, and of authigenic and alteration minerals, are presented in Figure 7-6. These abundances were determined using quantitative X-ray powder diffraction analyses. The results of these analyses were summarized by Bish and Chipera (1989). Figure 7-7 presents the results of X-ray powder diffraction analyses of

perpendicular to the walls of pores. The innermost parts of some pores were observed to contain aggregates of botryoidal silica. Generally, calcite was found to be less abundant than silica. Three textural varieties of calcite have been observed. Micritic calcite was found as a common coating of both breccia clasts and root casts. Coalescence of the individual root cast coatings was found to have been involved in forming aggregates of authigenic cement with an overall vesicular texture. Subsequently, the vesicles have been filled with silica, causing the authigenic cement to have nil porosity. In local areas, sparry calcite has been observed instead of, or in addition to, micritic calcite. Similar to the former textural variety, sparry calcite was found to occur in both the intra-clast pore space and the fractured and dilated breccia clasts. Most of the sparry calcite crystals are fairly small, less than 0.1 mm in diameter. In a few instances, however, poikilitic calcite crystals, up to 5 mm in diameter, were observed. The remaining fracture- and vesicle-filling calcite is acicular in texture. Sepiolite was found to occur as discrete pods, scattered along fracture traces. Individual sepiolite pods were commonly observed to be elongated and aligned along planes of host fractures. The pod-forming material was found to exhibit platy-fibrous texture. Individual fibres were observed to be aligned sub-parallel to the elongation of sepiolite pods themselves. In all of the examined specimens of the AMC breccia phase, solidified plant remains have been observed. Typically, these remains occur in the form of root casts, coated and in-filled with silica, calcite, and sepiolite. The largest root casts, as observed in the examined thin-section slides,

were about 0.2 mm in diameter. The most abundant root casts, however, were found to be less than 0.01 mm in diameter. Typically, root casts were found to be concentrated along breccia clast surfaces, forming coalescing masses completely in-filling the intra-clast space. Carbonate root casts, together with the silica coatings and in-fillings, were found to be the major constituent of authigenic breccia cement. In a few instances, root casts were observed to have radiaxial coatings of silica crystals, indicating that either they or their predecessors have acted as the sub-strata for silica precipitation.

6.3 Origin of the "Mosaic" Breccias

Available geologic and geochemical data, judged to be pertinent to interpreting the age and origin of the Yucca Mountain "mosaic" breccias, are summarized in Figures 6-1 through 6-7. Figures 6-1 and 6-2 are a geologic map and cross-section, respectively. Both of these figures show the locations of local faults, traces of which are known to be associated with the occurrence of "mosaic" breccia. Figure 6-2 may be used to estimate the combined volume of "mosaic" breccias, relative to the total volume of exposed bedrock. The results of assays of base and noble metals and path-finder elements are presented in Figures 6-3a and 6-3b. This figure presents trace element concentration data obtained by two different

research teams. For the Trench #14 "mosaic" breccias, these data may be used to estimate trace element concentrations, relative to the stratigraphically equivalent background.

Figure 6-4 presents the relative concentrations (values of the $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ ratios) of carbon-13 and oxygen-18 incorporated in three samples of authigenic breccia cement. This figure also shows the corresponding concentrations for local calcretes and equivalent bedrock veins. The observed isotopic composition vs. litho-facies gradient may be used to deduce relative conditions of precipitation (temperature and relative degree of evaporative oxygen-18 and diffusional carbon-13 enrichment) for these compositionally equivalent facies. These relative conditions, in turn, may be used to infer the origin of the parent solutions (i.e., direction of fluid flow). Figure 6-5 presents the results of a comparative analysis using carbon isotopes to evaluate the origin of "mosaic" breccia cement. This figure may be used to evaluate whether or not the isotopic character of carbon incorporated in breccia cement is equivalent to that expected to be dissolved in local supergene fluids. Figure 6-6 shows the results of chemical analyses of magnesian clays (sepiolite) incorporated in samples of "mosaic" breccias. Chemical compositions were determined employing the electron microprobe method. They may be used to estimate the chemical character of fluid phases involved in the formation of authigenic breccia cements.

Finally, Figure 6-7 presents the results of fission-track radiometric analyses of fragments of zircon crystals separated from the HD-41-4 and HD-74-2 samples of "mosaic" breccias. These samples represent AMC breccias from the Trench #14 exposure and the Busted Butte area, respectively. The external detector method was used to date the individual zircon grains, Naeser (1976 and 1979). These grains were mounted in teflon and etched in a eutectic melt of KOH-NaOH. The teflon mounts were then covered with a muscovite detector and irradiated along with neutron dose monitors. The fission-track ages were calculated employing the Zeta method, Hurford and Green (1983). Neutron dosimetry was determined by measuring track densities in the muscovite detectors used to cover the dose monitors during neutron irradiation. The fission-track radiometric method is used to date various mineral phases and to study their thermal histories. This method is based on the phenomenon of so-called "damage track" formation. This phenomenon results from spontaneous fission of ^{238}U atoms contained in a solid compound. When charged particles, resulting from fission, travel through a solid medium they leave a trail of damage caused by transfer of energy from these particles to atoms of this medium. The "damage tracks" may be enlarged through etching and then counted by microscopic examination. The density of damage tracks is a function of the age of specimen and its uranium content. Because the fission tracks fade by annealing at elevated temperatures, the fission-track method yields a cooling age, corresponding to the time elapsed after specimen temperature dropped below the annealing temperature. For various mineral

phases, the annealing temperature varies within a wide range. Zircon is fairly refractory and anneals at relatively high temperatures. Harrison et al. (1979), for example, have estimated these temperatures to be about 175° Celsius; Hurford (1986), however, gives the zircon annealing temperature as $240 \pm 50^\circ$ Celsius.

Examination of Figure 6-1 through 6-7, and consideration of the discussion in the preceding section, leads to the formulation of the following eight observations/interpretations. First, relative to the total volume of exposed bedrock, the Yucca Mountain "mosaic" breccias constitute a fairly significant component. As shown in Figure 6-1, these breccias were mapped along the traces of five local faults. For the 14 km wide explored segment, the combined mapped thickness of "mosaic" breccias is as large as about 2.0 km, Figure 6-2. The mean volumetric strain represented by "mosaic" breccias (equal to the volumetric proportion of authigenic cement) is conservatively estimated to be $\Delta V/V \sim 10$ percent. Combining this estimate with the total thickness of the mapped breccias leads to a conclusion that "mosaic" breccias record a significant "widening" of the Yucca Mountain area. For the 14 km wide segment, the total "widening" appears to be as large as $\Delta L \sim 200$ m, or about 1.5 percent. By any standard, this is not a trivial amount of accretionary strain. Refinement of the estimate of the amount of the accretionary strain will not alter this conclusion.

Second, the Yucca Mountain "mosaic" breccias represent epigenetic deformational

phases. Such a state of affairs is indicated by two lines of evidence. On the one hand, both types of "mosaic" breccia contain brecciated fragments of the mineral species produced during deuteric vapor phase mineralization. This indicates, of course, that the formation of both types of breccia occurred subsequent to the cooling, syn-depositional devitrification and solidification of the host ignimbrites. This conclusion is further reinforced by two additional observations, namely: (1) the "mosaic" breccias are not bound to a single ash-flow unit (instead, they transgress stratigraphic boundaries), and (2) their development is not restricted to the pyroclastic complex ("mosaic" breccias were also found to occur in the Paleozoic carbonates). On the other hand, samples of the AMC breccia variety were found to contain allogenic detritus, specifically fragments of muscovite and zircon crystals. As shown in Figure 6-7, the breccia incorporated zircons yield a wide spectrum of fission-track ages, from 59×10^6 to as little as 4.8×10^6 years B.P. The strong age discords between the host ignimbrites and the breccia incorporated zircons confirm that at least the AMC breccia variety is epigenetic. Levy and Naeser (1991) considered, however, that the CTM breccia phases are auto-breccias, i.e., syn-depositional with their host rock. Specifically, these phases were regarded as *"formed by settling of the host pyroclastic unit shortly after deposition."* The observed fact that *"many breccia fragments (clasts) have devitrification textures in which the crystals are coarser around the edges of the fragments and have grown beyond the original boundaries of the fragments"* was taken as supportive evidence for this interpretation. The devitrification textures were

correctly taken as indicating that *"some brecciation occurred while the tuff was still at least partly glassy or while devitrification was in progress."* However, it was erroneously implied that the CTM breccia phases must have been formed exclusively during the syn-depositional devitrification of the Topopah Spring Member. Clearly, such is not necessarily the case. Observing that (1) parts of the Topopah Spring Member still retain their vitric state, and (2) the CTM breccia phase contains clasts with and without devitrification textures, one may reasonably propose an alternate interpretation. The CTM breccia phase could have been formed after the cooling and syn-depositional devitrification of the Topopah Spring Member. This breccia phase, however, contains polymictic clasts; those with devitrified fringes represent initially vitric clasts derived from the Topopah Spring Member vitrophyre; those without devitrified fringes represent clasts derived from the syn-depositionally devitrified interior of the Topopah Spring Member.

Third, the Yucca Mountain "mosaic" breccias are polygenetic deformational phases, formed during multiple episodes of bedrock fragmentation and subsequent cementation. Such a state of affairs is indicated by two lines of evidence. On the one hand, the AMC breccia specimens were found to contain solidified plant remains. In contrast, however, the CTM breccia specimens were found to be free of such remains. Evidently, the CTM breccias were formed below the biological root zone, when the contemporary topographic surface was covered by younger

rock units. Subsequent erosion has produced the contemporary topographic surface, exposing the CTM breccias. In the case of the AMC breccias, however, abundant plant remains indicate that these breccias are younger, and produced in a near-surface environment. On the other hand, some specimens of the CTM breccia display evidence of having been formed with the involvement of moderate-temperature fluid phases. Such fluid phases were also involved in (1) fracture-based devitrification of the Topopah Spring Member vitrophyres, and (2) precipitation of epigenetic mineral species, such as drusy quartz, chalcedony, opal, zeolites, and oxyhydroxides. In contrast, the AMC breccias do not exhibit any indications of having been formed under elevated geothermal conditions. The difference in formation temperature confirms that, indeed, the Yucca Mountain "mosaic" breccias are composed of polygenetic deformational phases.

Fourth, in thin-section examinations, samples of the AMC breccias were found to be cemented by a compositionally and texturally diverse mixture of mineral species. Compositionally, the authigenic breccia cements were found to be composed of the calcite, silica, and sepiolite mineral assemblage. The individual mineral species, however, are clearly demarked from neighboring mineralogically distinct species. This mineral phase demarcation has the effect of producing the overall laminated texture of the authigenic breccia cement. The compositional diversity is accompanied by textural diversity. Authigenic calcite, for example, was found to occur as four texturally distinct varieties, including micritic, sparry,

poikilitic, and acicular. Similarly divergent are the authigenic silica species. For these species, three texturally distinct varieties have been identified, including spherulitic, fibrous, and botryoidal. To account for the observed textural and compositional diversity, one is left with little choice but to assume that, during the formation of authigenic cement, the mineral precipitation environment was undergoing substantial fluctuations. Several facets of this environment could have been involved, including CO_2 partial pressure, fluid chemistry, and precipitation temperature. For mineral-rich hypogene solutions, emerging from faults and fissures, substantial fluctuations involving various combinations of the above facets are not surprising, and are to be expected. In contrast, for supergene pedogenic processes, stimulated solely by infiltrating rainwater, such fluctuations are not expected characteristics. Based on these contrasting expectations, it seems reasonable to suspect that the observed compositional and textural diversity of authigenic cements is telling us that a supergene pedogenic origin of these cements is doubtful at best.

Fifth, samples of AMC breccias from the Trench #14 exposure contain elevated and anomalous concentrations of trace and path-finder elements. As shown in Figure 6-3, relative to the stratigraphically equivalent ignimbrites, the epigenetically brecciated and cemented ignimbrites contain decisively higher concentrations of both base and noble metals and the path-finder elements. To account for the observed concentrations, Castor and Tingley (1990) concluded that

"the anomalous samples could represent a very distant halo around a concentration of base metals in the deep subsurface." Similar conclusions were also drawn by Weiss et al. (1991). Commenting upon the observed concentrations of base and noble metals, these authors stated, *"It would seem to us that the three most plausible sources of elevated As, Au, Bi, Zn, and Pb, and the correlation of Au with As, in rocks exposed at the surface of Yucca Mountain are: (1) distal, meteoric water-dominated hydrothermal fluids, (2) meteoric water moving downward and (or) laterally through the Paintbrush Tuff, and (3) pedogenic processes of leaching and accumulation. The low concentrations of these metals in fresh tuff, and the lack of adjacent or overlying metal-rich rocks nearby leads us to favor a hydrothermal source of metal enrichment."* Both of these seemingly reasonable and straightforward interpretations, however, are not being accepted by investigators associated with the Yucca Mountain Site Characterization Project. While commenting upon the observed base and noble metal concentrations in Trench #14, for example, Stuckless et al. (1991) made the following statement. *"The anomalously high concentrations of some metals in the silicified breccias are not surprising, and simply serve to further emphasize the differences between the early formed breccias and the later formed, laminated calcite/silica deposits. Element mobility during late-stage cooling of pyroclastic units is commonly focused by fractures, faults, or other openings such as lithophysal cavities and is the norm rather than the exception in felsic volcanic piles. This fact was demonstrated long ago in a study of fumarolic mounds and ridges in the Bishop Tuff."* However, it is

imperative to recognize that, as far as can be determined, there is no factual basis whatsoever to support the convenient belief that the Trench #14 concentrations of some metals are related to *"late-stage cooling of pyroclastic units."* Quite the contrary, several lines of evidence may be put forth as being in direct conflict with it. Among these are: (1) the Trench #14 breccias are clearly epigenetic; (2) the Trench #14 breccias contain abundant plant remains; (3) isotopic characters of carbon and oxygen, incorporated in the Trench #14 breccia authigenic cement, are comparable to those contained in the late Quaternary calcretes (Figure 6-4); and (4) the Trench #14 breccias contain zircon crystals yielding fission-track ages substantially younger than the age of *"late-stage cooling of pyroclastic units,"* Figure 6-7.

Sixth, the isotopic characters of carbon and oxygen incorporated in the authigenic breccia cements, from both the Trench #14 exposure and the Busted Butte area, may reasonably be interpreted as indicating a hypogene origin of these cements. The following reasoning constitutes the foundation for this statement. As shown in Figure 6-4, the relative concentrations of carbon-13 and oxygen-18 incorporated in the authigenic breccia cements are comparable to those contained in samples of local calcretes and associated bedrock veins. For all three litho-facies, the observed $\delta^{13}\text{C}$ vs. $\delta^{18}\text{O}$ fields are sufficiently similar to assume that these compositionally and texturally equivalent litho-facies were precipitated from common solutions. It seems reasonable to further assume that, in the isotopically

pristine state, these solutions carried similar values of the $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ ratios. If the litho-facies are of hypogene origin, then one may expect that: (1) the degree of departure from the pristine state, resulting from both oxygen-18 evaporative enrichment and carbon-13 diffusional enrichment, was lowest for the parent solutions of the authigenic breccia cements, somewhat higher for the parent solutions of the bedrock veins, and still higher for the parent solutions of the local calcretes, and (2) the mineral precipitation temperature was highest for the authigenic breccia cements, somewhat lower for the bedrock veins, and still lower for the calcretes. From Figure 6-4, it may be observed that values of both the $\delta^{13}\text{C}$ ratio and the $\delta^{18}\text{O}$ ratio become greater with increasing "topographic exposure" of the respective litho-facies. These values are lowest for the authigenic breccia cements, somewhat higher for the bedrock veins, and still higher for the calcretes. The observed isotopic compositions vs. litho-facies gradient may reasonably be attributed to: (1) for the oxygen-18 gradient, the combined effect of oxygen-18 evaporative enrichment and temperature-controlled isotopic fractionation, and (2) for the carbon-13 gradient, carbon-13 diffusional enrichment. This, in turn, leads to the following two-fold inference. On the one hand, the observed $\delta^{18}\text{O}$ vs. litho-facies gradient, from 19.5 to 21.0 per mil_{smow} (Figure 6-4b), may be taken as indicating that, for the distal calcretes, both the isotopic fractionation effects (brought about by the lower precipitation temperature) and the degree of oxygen-18 evaporative enrichment were higher than those for the proximal authigenic breccia cements. For the hypogene mode of deposition, both

of these conditions are not surprising, and are to be expected. The opposite is true, however, for the supergene mode of deposition. On the other hand, the observed $\delta^{13}\text{C}$ vs. litho-facies gradient, from -7.0 to -5.0 per mil_{PDB} (Figure 6-4b), may be taken as indicating that, for the distal calcretes, diffusional carbon-13 enrichment (resulting from CO_2 degassing of hypogene solutions) was higher than that for the proximal authigenic breccia cements. Again, in contrast to the per descensum mode, for the per ascensum mode of deposition such is the rationally-expected effect.

Seventh, as far as can be determined, there is no factual basis to support the viewpoint that *"most of the authigenic mineral cementation of AMC breccias is related to surficial pedogenic processes (ones stimulated solely by rainwater), as shown by the abundance of plant remains,"* Levy and Naeser (1991). Quite the contrary, two lines of evidence may be put forth as being in direct conflict with it. On the one hand, the isotopic character of carbon incorporated in the authigenic breccia cements, from both the Trench #14 exposure and the Busted Butte area, is different from that expected to be dissolved in local supergene pedogenic fluids. Specifically, parent solutions for the authigenic breccia cements carried values of the $\delta^{13}\text{C}$ ratio that are much too high to permit direct genetic association with local supergene pedogenic fluids alone. A complete justification for this statement is provided in Figure 6-5. Although significantly different from those expected for local supergene deposits, the authigenic breccia cements do carry values of the

del¹³C ratio that are identical to those incorporated in carbonate gangue associated with many known and unquestionable hydrothermal deposits, see, for example, Figures 8-8 and 8-9. On the other hand, the presence of both carbonate matter and magnesian clays indicates that calcium-magnesium fluid phases were heavily involved in the formation of authigenic cements. Relative to fluid phases indigenous to the alkali ignimbrites, however, these calcium-magnesium fluid phases are allogenic, or metasomatic. Contemporary fluid phases residing in the local ignimbrites, for example, carry very low concentrations of Ca + Mg cations, from 0.82 to no more than 20 ppm, Figure 5-3, and Benson and McKinley (1985). Early in their intraformational residence history, these fluid phases must have been analogues to those held responsible for the supergene "pedogenic" formation of the authigenic breccia cements. Assuming a highly evaporatively enriched state, the hypothetical supergene "pedogenic" fluids could have produced calcium-magnesium solid phases, but only with vanishingly low rates of deposition, say fractions of a gram per cm² per 10³ years, Machette (1985). For the authigenic breccia cements, however, the inferred precipitation rates are fairly large and continuous. Such a state of affairs is indicated by both the F-textures of breccia specimens and the poikilitic and sparry textures of some of authigenic calcites. Levy and Naeser (1991) considered that the abundance of plant remains "shows" that the authigenic cements owe their existence to "*surficial pedogenic processes*," engendered solely by infiltrating rainwater. By no stretch of the imagination, however, is such the case. The observed abundance of plant remains

indicates, of course, that authigenic cements were precipitated in an environment supportive of plant life. Such an environment occurs near the topographic surface, possesses sufficient moisture and, in terms of fluid chemistry and temperature, is not prohibitively hostile to biological activity. To restrict such an environment solely to rainwater-activated pedogenic processes, however, is sheer nonsense.

Eighth, samples of "mosaic" breccias, both from the Trench #14 exposure and from the Busted Butte area, were found to contain fragments of zircon crystals incorporated in authigenic cements. As shown in Figure 6-7, the separated zircons yielded a wide spectrum of fission-track ages, from 59.7×10^6 to as little as 4.8×10^6 years B.P. Such a wide spectrum indicates that the enclosed zircons originated either from distinct igneous sources or from a differentially-heated source, or sources. Importantly, for both of the breccia specimens examined, there are fission-track ages significantly older and significantly younger than the K/Ar ages of the host ignimbrites. For the Tiva Canyon Member and the Topopah Spring Member of the Paintbrush Tuff, the representative K/Ar ages are 13.0×10^6 and 13.5×10^6 years B.P., respectively. To account for both the presence of zircons and the anomalous fission-track ages of these zircons, two contrasting viewpoints may be put forth. On the one hand, one may conclude that, during their ascent, allogenic calcium-magnesium fluids (parent solutions for the zircon-enclosing cements) have brought zircons from a deep sub-stratum. If such is the case indeed then both the presence of zircons and their fission-track ages may be taken as

indicating three important points. These are: (1) the age of formation of some of the AMC "explosive" breccias, or in terminology employed by Hanson et al. (1987) "*hydrothermal eruption breccias*," must be equal to or younger than the 4.8×10^6 years B.P. fission-track age of the youngest zircons; (2) prior to or during emplacement, some of the enclosed zircon crystals were heated sufficiently to cause fission-track annealing; and (3) the calcium-magnesium fluid phases must have ascended with sufficient velocity to carry the high-density zircons. On the other hand, one may also stubbornly keep insisting that the breccia-incorporated zircons were introduced from the topographic surface, by either wind or run-off action. It is possible, one may presume, that zircons from different igneous and/or hydrothermally-altered bodies are being redistributed on the land surface by either aeolian or fluvial processes. If such is the case indeed, then both the presence of zircons and the fission-track ages of these zircons may be taken as indicating two points. These are: (1) the age of formation of some of the AMC "fragmentation" breccias must be equal to or younger than the 4.8×10^6 years B.P. fission-track age of the youngest zircons, and (2) locally, there is a fairly young source of either primary (juvenile) zircons or thermally-reset zircons. The latter point requires the local presence of exposures containing either fairly young rhyolites or fairly recently hydrothermally-altered ignimbrites. It is imperative to recognize that, as far as can be determined, there is no factual basis to support the belief that the allogenic zircons were introduced from the topographic surface. To the contrary, several facets of the zircons' setting may be regarded as

effectively arguing against this belief. Among these are: (1) the spatial association of zircons with deformational phases, which independent experts regard as "*hydrothermal eruption breccias*;" (2) the spatial association of zircons with dilated and hydrothermally-altered fault zones; and (3) the genetic association of zircons with authigenic mineral phases containing elevated concentrations of both base and noble metals and path-finder elements.

6.4 Summary and Conclusions

The preceding analyses of both the thin-section observations and the assay, isotopic, and fission-track data, lead to the formation of provisional views regarding the age and origin of the Yucca Mountain "mosaic" breccias. For the purposes of this report, these views may be summarized by the following four points. First, the currently available data do not provide any factual basis to support the viewpoint that the CTM phase of the Yucca Mountain "mosaic" breccias was formed shortly after the eruptive emplacement of the local ignimbrites, during the late-stage cooling of these ignimbrites. To the contrary, the presence of both incorporated fragments of vapor-phase mineral species and epigenetic hydrothermal mineral species is in direct conflict with this viewpoint.

Second, the currently available data do not provide any factual basis to support the viewpoint that the AMC breccia phase was formed and cemented by

infiltrating rainwater as the sole agent. Quite the contrary, several indirect lines of evidence may be put forth as arguing against this viewpoint. Among these are: (1) microscopically-observed textural and compositional diversity of the authigenic breccia cements; (2) elevated concentrations of both base and noble metals and the path-finder elements; (3) the isotopic character of carbon incorporated in the authigenic breccia cement; and (4) the spatial association of the AMC breccias with dilated and altered fault zones. The fact that the AMC breccia phase contains abundant plant remains can not be regarded as uniquely supportive of the supergene pedogenic viewpoint.

Third, the currently available data do not provide any factual basis to support the viewpoint that the AMC breccia phase is genetically distinct from the CTM breccia phase. To the contrary, the fact that both of these deformational phases share the same calcite, opaline silica, and sepiolite mineral assemblage, as a cementing agent, may reasonably be regarded as effectively arguing against this viewpoint. The observed compositional differences (which are the relative abundances of plant remains and authigenic cement), however, may satisfactorily be explained by recognizing the polygenetic character of formation of the Yucca Mountain "mosaic" breccias. From a genetic point of view, both the CTM phases and the AMC phases may reasonably be regarded as representing a common deformational and depositional process. Specifically, it may be assumed that both of these breccia phases were formed through in-filling of so-called "large-scale wallrock

separations," Szymanski (1989). In association with this process, breccia clasts are derived through a variety of bedrock fragmentation modes, including: (1) explosive or eruptive fragmentation, (2) subsequent secondary explosive fragmentation, and (3) surficial denudation and mass wasting. Concurrently with the introduction of clasts, the authigenic cements are formed via mineral precipitation from hypogene solutions that occupy the wallrock separation and are discharging from it. Along the depth-dimension of a wallrock separation, rates of clast introduction, relative to rates of authigenic cement formation, vary from relatively small at the top to much larger toward the bottom. Differences in clast supply rates cause the deeper breccias to develop the observed GS-textures, and the shallower breccias to develop the observed F-textures. In light of the preceding remarks, the respective differences between the CTM and AMC breccia phases have a logical explanation. In accordance with this explanation, the CTM breccia phases are older. They were formed deeper, below the biosphere (below the plant root zone), when the contemporary topographic surface was covered by younger rock units. Subsequent erosion has formed the contemporary topographic surface and, concurrently, has exposed the CTM breccia phases. The relative sparsity of authigenic cement indicates that (1) relative to the rates of formation of authigenic cement, the rates of introduction of allogenic clasts were fairly large, and (2) the allogenic clasts varied in size, and included a fairly large proportion of fines. In contrast, the AMC breccia phases are younger. They were formed in a near surface environment, when the older CTM phases were erosionally exposed.

Both the abundance of plant remains and the relative abundance of authigenic cement are readily attributable to such a depositional environment.

Fourth, the currently available isotopic and geochemical data, however, do provide fairly firm factual support for the hypogene viewpoint expressed, as early as 1987, by Hanson et al. In accordance with this viewpoint, the Trench #14 "mosaic" breccias, and by association the texturally and compositionally equivalent breccias from the Busted Butte area, are "hydrothermal eruption breccias." Several lines of evidence may be put forth as being supportive of this viewpoint. Among these are: (1) the observed compositional and textural diversity of the authigenic breccia cements; (2) the spatial association of the "mosaic" breccias with dilated and hydrothermally-altered fault zones; (3) the isotopic characters of carbon and oxygen incorporated in the authigenic breccia cements; (4) elevated concentrations of both base and noble metals and the path-finder elements; and (5) the incorporation of zircon crystals yielding a wide spectrum of fission-track ages. The formation of the Yucca Mountain "mosaic" breccias appears to be an intermittent process. Fission-track ages, from the breccia-incorporated zircon fragments, indicate that some of the breccia specimens were cemented after the 4.8×10^6 years B.P. age of the youngest zircons. The apparent compositional, textural, and isotopic affinity between the authigenic breccia cement and the clearly late Quaternary calcretes and sinters further suggests that hypogene processes, that evidently were involved in the formation of "mosaic" breccias, have

remained operational during the Plio-Quaternary time span. It is eminently reasonable to conclude that, at Yucca Mountain, both the hydrothermal component of the vadose zone interstitial fluids and the parent fluids for the authigenic breccia cements share the same hypogene origin. This is so because both of these fluid phases exhibit common characteristics, either observed or inferred. Among such characteristics are: (1) the calcium-magnesium bulk composition; (2) elevated concentrations of the base and noble metals; and (3) high values of CO₂ partial pressure.

7.0 The Origin and Age of the Yucca Mountain Zeolitic and Montmorillonitic

Alteration Minerals

7.1 General

An important compositional feature of the Yucca Mountain ignimbrites is the occurrence of mineral species from two pro-grade alteration facies, specifically (1) the zeolitic alteration facies, and (2) the montmorillonitic alteration facies. The occurrence of the corresponding low-grade alteration minerals (clinoptilolite, mordenite, and smectite) is ubiquitous. These mineral species occur throughout the entire ~2 km depth of the locally-explored stratigraphic section. The higher-grade alteration minerals (analclime, laumontite, albite, alleverdite clays, kalkberg clays, discrete illite), however, are known to be present mostly in deeper parts of the section, typically below a depth of about 0.5 km.

To account for the local presence of both of the observed pro-grade alteration facies, two contrasting viewpoints may be put forth. On the one hand, recognizing the manifestly magmatically active character of the region, one may reasonably propose that these facies represent so-called propylitic alteration, Figure 7-1 and Weiss (1990). Such hypogene alteration results from a variety of wallrock <--> fluid (hypogene) exchange reactions which, if operating for prolonged periods of time, lead to the formation of a variety of epigenetic minerals, including

carbonates, opals, montmorillonitic clays, hydromicas, zeolites, iron oxides, sulfides, feldspars, etc. In ore deposits where hydrothermal alteration is zoned, propylitic alteration is usually a fringe effect. The propylitic mineral assemblage is commonly developed in igneous rocks, particularly those containing solid aluminosilicates, such as volcanic glasses and feldspars. In such rocks this alteration assemblage includes, as a significant component, pro-grade zeolitic and montmorillonitic facies. A contemporary case of such alteration has been investigated at Wairakei, New Zealand. Results of these investigations were published by Steiner (1955) and Coombs et al. (1959), and are summarized in Figure 7-2. The observed alteration sequence starts with a surficial acid-leached zone of kaolinitic clays followed by montmorillonitic clays. The latter clays occur in regions where major steam loss and cooling of fluids takes place. Such regions are associated with temperatures ranging from 100° to 150° Celsius. Next in the alteration sequence is the low-grade zeolitic zone, containing mainly mordenite. Temperatures associated with the highest levels of occurrence of mordenite range from 130° to about 160° Celsius. The higher-grade zeolites, such as wairakite, along with albite, occur deeper. They are associated with temperatures ranging from 160° to about 224° Celsius. Still deeper, there is an adularia and hydromica alteration zone. The major feature of this zone is the replacement of plagioclase by adularia and hydromica, and the presence of minor albite at higher levels.

On the other hand, it has also been proposed that, at Yucca Mountain, the

alteration facies represent a variety of temporally distinct alteration minerals, ranging in origin from deuteric, through epigenetic (hypogene), to diagenetic (supergene). This viewpoint is being advocated by various investigators associated with the Yucca Mountain Site Characterization Project. The higher-grade alteration minerals are regarded as formed either during the cooling, syn-depositional devitrification and solidification of the local ignimbrites (deuteric alteration) or, shortly thereafter, during the hydrothermal stage of activity of the Timber Mountain Caldera (epigenetic alteration). However, the younger and lower-grade alteration minerals, particularly those from the vadose zone, are regarded as having been produced exclusively in a supergene environment, through leaching and dissolution of the vitric ignimbrites by infiltrating rainwater (diagenetic alteration). The following quotation from Levy (1991) is believed to provide an accurate representation of the currently held viewpoints. *"Most zeolites and zeolitized tuffs appear to be products of diagenetic alteration in which the original glass dissolved and the zeolites precipitated at ambient temperatures in a water-rich environment. The distribution of diagenetically altered zeolitic rocks is important evidence for the paleohydrologic interpretations in this paper. Yucca Mountain also contains zeolites of moderate-temperature hydrothermal origin linked to post-emplacement cooling of a pyroclastic unit Researchers studying zeolitization in the Yucca Mountain region have made a variety of inferences about the hydrologic regimes in which zeolitization occurred. Most studies predating the Yucca Mountain Project attribute zeolitization to unsaturated-zone hydrologic*

processes: either from localized active recharge creating a zone of near-saturation and rock alteration above the SWL, or from permeability barriers responsible for the formation of local or regional perched water tables and associated alteration. Yucca Mountain researchers recognize a few probable examples of perched-water alteration but tend to favor alteration at or below a SWL. The Yucca Mountain glass- and zeolite-distribution data provide no support for the existence or former existence of a regional perched water table. Known examples of probable perched-water zeolitization are of much smaller vertical and lateral extent than the main mass of zeolitized tuffs. The working hypothesis is that most zeolitization occurred around or below the SWL in place at the time of alteration Even without a consensus on the relationship between the transition zone and the SWL position at the time of alteration, researchers generally agree that zeolitization required the presence of abundant water over a long period of time. The underlying assumption is that glass in nonwelded tuffs is preserved only where the rocks have not been subjected to prolonged saturation. This could be a valuable interpretive tool for the Yucca Mountain Project because it may be possible to estimate the highest elevation ever occupied by the SWL."

The purpose of the discussion presented in the subsequent sections is to evaluate the respective merits of the above-stated viewpoints. A variety of mineralogical, geochemical, and radiometric data is currently available, and will be used for these purposes. The evaluation consists of four parts. Section 7.2 presents

background information concerning circumstances of formation of pro-grade zeolitic and montmorillonitic facies. For the Yucca Mountain area, this information will be used to estimate the local alteration temperatures, based on the observed distribution of mineral species representing a particular grade of alteration. Geochemical and mineralogical data regarding spatial distribution of the alteration facies, including chemical composition of both the original or unaltered ignimbrites (represented by the locally preserved glasses) and the alteration products (whole-rock ignimbrites and separated alteration minerals), are presented and analyzed in Section 7.3. The K/Ar ages of the Yucca Mountain alteration minerals are presented and analyzed in Section 7.4. The overall conclusions and summary are presented in Section 7.5.

7.2 Background

Pro-grade metamorphic facies are particularly useful for the purposes of performing various paleo-geothermal reconstructions. The performance of such reconstructions requires that (1) spatial distribution of mineral assemblages, representing various metamorphic grades, is known; and (2) thermal stability limits for these assemblages are also known. In the case of Yucca Mountain, the spatial distribution of alteration minerals is known based on fairly detailed mineralogical analyses of core samples, extracted from a number of local boreholes. For both of the locally present pro-grade alteration facies, the

respective thermal stability limits may be estimated based on the results of studies of appropriate contemporary analogs elsewhere. Alteration temperatures associated with various grades of the montmorillonitic alteration are presented in Figure 7-3. This figure is a summary diagram of a relationship between alteration temperature and the corresponding extent of the smectite --> illite transformation. It has been constructed based on field data obtained in regions undergoing active burial or hydrothermal metamorphism. The diagram shows that if an adequate supply of potassium exists, the expandable smectite transforms, through a series of steps, to the non-expandable illite. The transformation is influenced by several factors, including reaction time, temperature, water chemistry, etc. Below the 90-100° Celsius temperature range, the illite/smectite fraction is randomly interstratified (the so-called RO type of interstratification) and possesses a fairly low percentage of collapsed layers. Above this temperature range, however, the percentage of collapsed layers increases and a higher ordering of interstratification appears, the so-called RI or alleverdite type of interstratification. Hower and Altaner (1983), and later Thompson and Jennings (1985), have concluded that below a temperature of about 150° Celsius, the RO --> RI transformation is kinetically controlled. For time spans in excess of 10^7 years, the alleverdite interstratification is formed at a temperature ranging from 90 to 100° Celsius. Over reaction times shorter than $\sim 10^6$ years, however, the RO --> RI transformation temperature ranges from 130 to 150° Celsius. At temperatures in excess of 150° Celsius, the smectite --> illite transformation ceases to be kinetically

controlled. With increasing temperature, both the percentage of collapsed layers and the interstratification ordering increase fairly independently of the duration of thermal exposure. At a temperature of about 175° Celsius, the so-called kalkberg type of interstratification appears. The corresponding percentage of collapsed layers ranges from 85 to about 95 percent. At a temperature of about 260° Celsius, the smectite --> illite transformation is complete. In addition to alteration temperatures, fluid chemistry also plays an important role in controlling the extent of the smectite --> illite transformation. Increasing concentration of potassium tends to promote the smectite --> illite transformation, Whitney and Northrop (1987). The predominance of other cations (Ca, Mg, and Na), however, tends to inhibit this transformation, Howard (1981).

For the zeolitic alteration facies, the corresponding alteration temperatures are presented in Figure 7-4. This figure is a summary diagram showing a relationship between alteration temperature and the resulting zeolitic alteration minerals. It is based on the results of investigations of a number of zeolitized silicic tuff sequences by Iijima (1975, 1978, and 1980). It has been shown that a typical pro-grade zeolitic sequence consists of four distinct mineralogical, or alteration, zones. The lowest-grade, Zone I, includes a zeolite-free rock and contains volcanic glass shards and their low temperature derivatives, such as opal and smectite. Below this zone, the zeolitic Zone II occurs. This volcanic glass-free zone contains the lowest-grade zeolitic alteration minerals, such as clinoptilolite,

mordenite, and heulandite. The underlying zeolitic Zone III is characterized by the presence of analcime. Typically, this alteration mineral replaces the precursor zeolites, which often coexist as relicts. The highest-grade zeolitic alteration is represented by Zone IV. This zone is characterized by the presence of albite, replacing analcime. Several lines of evidence have been employed to establish linkage between interfaces separating the recognized zeolitic zones and the corresponding alteration temperatures. The results of such considerations are presented in Figure 7-4. The top of zeolitic Zone II has been associated with alteration temperatures ranging from 25° to about 60° Celsius. This fairly wide temperature range is attributed to sensitivity of the glass → zeolite reaction to fluid chemistry. Hay (1978), for example, has shown that fluid pH is an important factor controlling the rate of this reaction. At pH ~9.5, fairly advanced zeolitization may take place in just a few thousand years. Alteration temperatures associated with the tops of zeolitic Zones III and IV vary with chemical composition of fluid phases that promote the glass → zeolite reactions. A particularly important factor appears to be the concentration of sodium. In the case of weak alkaline geothermal fluids (Na concentrations of ~ 10³ ppm), the alkali clinoptilolite → analcime conversion occurs at a temperature ranging from 90° to about 100° Celsius. Under similar circumstances, the transformation of the analcime + quartz + H₂O predecessor assemblage into the resulting albite + quartz + H₂O assemblage occurs at a temperature ranging from 175 to 200° Celsius.

samples of the illite/smectite fractions. For four of the local boreholes, this figure gives the depth distribution of: (1) percentage of illite layers; (2) type of the illite/smectite interstratification; (3) ethylene glycol-smectite complex thickness; and (4) basal spacing at 20 ± 10 percent relative humidity. The corresponding X-ray powder diffractions patterns, and the interpretation of these patterns, were obtained and reported by Bish and Chipera (1988) and Bish (1989). Figure 7-8 presents the results of chemical analyses of samples of the Yucca Mountain clinoptilolites. These samples were obtained from eleven of the local boreholes. They were analyzed by Broxton et al. (1986), employing the electron microprobe method. Figure 7-9 presents the results of chemical analyses of whole-rock samples of altered ignimbrites. These samples were obtained from six of the Yucca Mountain boreholes. They were analyzed by Broxton et al. (1986), employing a combination of analytical methods, including X-ray fluorescence, atomic absorption, and plasma-source emission spectrophotometry. Finally, Figure 7-10 presents the results of chemical analyses of samples of the locally preserved volcanic glasses. These samples were obtained from seven of the local boreholes. They were analyzed by Broxton et al. (1986), employing the electron microprobe method.

For samples of the Yucca Mountain alteration minerals, mineralogical and chemical data are also summarized and interpreted in Figures 7-11 through 7-20. In these figures, an attempt is being made to define the spatial distribution of the

zeolitic and montmorillonitic alteration grades and, thereupon, the spatial distribution of the corresponding alteration temperatures. Figure 7-11 presents two cross-sections, each showing the spatial distribution of the zeolitic alteration grades. Figure 7-12 is a similar diagram. It depicts the spatial distribution of the montmorillonitic alteration grades. Figure 7-13 presents the configuration of the zeolitic Zone I/Zone II interface, relative to both the contemporary water table and the local stratigraphic boundaries. Figure 7-14 presents two cross-sections, each showing the spatial distribution of chemical compositions of the local clinoptilolites. Figure 7-15 is a triangular diagram. For samples of the vadose zone clinoptilolites, this diagram shows the relative concentrations of major cations. Figures 7-16 and 7-17 present a comparison of the chemical compositions of the vitric phases and the corresponding whole-rock altered ignimbrites. Figures 7-18 and 7-19 present the results of paleo-geothermal reconstructions, based on the spatial distributions of both of the alteration facies considered. Finally, Figure 7-20 presents a planar distribution of the inferred alteration temperatures, at a constant depth of 3000 feet.

Examination of Figures 7-11 through 7-20 leads to the formulation of the following five observations/interpretations. First, at Yucca Mountain, the observed depth distribution of alteration minerals lacks a discrete zoning, Figures 7-11 through 7-13. This is expressed by the fact that, commonly, the observed depth distribution of low-grade alteration minerals overlaps the distribution of

equivalent higher-grade alteration minerals. Mineral species representing the low-grade zeolitization and montmorillonitization (clinoptilolite, mordenite, heulandite, and smectite) appear to be ubiquitous. The occurrence of these species extends from the highest explored levels of the vadose zone (depth of ~350 feet in borehole USW G-4, Figure 4-12) to the deepest explored levels of the "saturated" zone (depth of ~6000 feet in borehole USW G-1, Figure 7-6e). Intermixed with the ubiquitous low-grade alteration background, there are local occurrences of higher-grade alteration minerals, such as analcime, laumontite, albite, alleverdite clay, kalkberg clay, and discrete illite. The observed overprinting of higher-grade mineralization by lower grade mineralization indicates that alteration of the Yucca Mountain ignimbrites is a polygenetic, or multi-stage, process. Specifically, lower-grade mineral species appear to have been produced both prior to and after the formation of higher-grade alteration minerals. This interpretation is consistent with observations made by Bish (1989). This investigator has reported that sample USW G2-5369, for example, contained both the high-temperature discrete illite and the low temperature RO clays. The polygenetic character of the low-grade alteration is important. It indicates that, at Yucca Mountain, the formation of the corresponding minerals is not related solely to deuteric alteration, during the cooling, syn-depositional devitrification and solidification of the local ignimbrites. Instead, at least in part, these minerals appear to be either epigenetic (hypogene) or diagenetic (supergene).

Second, at Yucca Mountain, the isogradal surfaces from both of the local alteration facies lack a consistent relationship to the local stratigraphic boundaries. This means that, in different boreholes, the top of a particular alteration grade occurs at different stratigraphic levels, Figures 7-11 and 7-13. From these figures it may be observed that in boreholes J-13, USW G-1, and G-2, the top of the analcime zone is situated within the Prow Pass Member of the Crater Flat Tuff. In boreholes UE-25p#1 and 25a#1, however, the equivalent isogradal surface is situated within the underlying Tram Member. Importantly, in borehole J-13, analcime occurs above the water table, at a depth of 608 feet in the Tiva Canyon Member of the Paintbrush Tuff, Broxton et al., 1986 (Appendix G, pg. 147). The lowest stratigraphic level of occurrence of the top of the analcime zone is known from borehole USW G-3, where it is situated below the Crater Flat Tuff within the Lithic Ridge Tuff. In boreholes UE-25a#1 and USW G-1, the top of the albite zone is situated within the Tram Member of the Crater Flat Tuff. In borehole USW G-1, however, the equivalent isogradal surface occurs deeper, within the Lithic Ridge Tuff. Similarly varying are stratigraphic positions of the isogradal surfaces from the montmorillonitic facies, Figures 7-12 and 7-13. The highest stratigraphic position of the alleverdite clays is known from borehole USW G-2. In this borehole, the RO/RI interface occurs within the Bullfrog Member of the Crater Flat Tuff. In boreholes USW G-1 and UE-25p#1, however, the equivalent isogradal surface occurs in the underlying ignimbrites. Specifically, this surface was identified within the Lithic Ridge Tuff, in borehole USW G-1, and within the

pre-Lithic Ridge Tuff units, in borehole UE-25p#1. The highest stratigraphic position of the kalkerberg clays is known from borehole USW G-2. In this borehole, the kalkerberg clays were first encountered within the Tram Member of the Crater Flat Tuff. In borehole USW G-1, however, the equivalent isogradal surface is situated much deeper, within the pre-Lithic Ridge Tuff units. The observed significant discordances between the stratigraphic boundaries and the isogradal surfaces are important. They provide clear and unequivocal testimony that, at Yucca Mountain, the higher-grade alteration minerals are not the result of deuteritic alteration. Considering that the corresponding alteration temperatures are equal to and exceed 100° Celsius, it is totally unreasonable to regard the higher-grade alteration minerals as the products of supergene-diagenetic alteration. Consequently, there is little choice but to admit that, at Yucca Mountain, the higher-grade alteration minerals must have been formed in response to hypogene processes, specifically in response to epigenetic hydrothermal metamorphism. The fact that in borehole J-13 analcime occurs a few hundred feet above the contemporary water table is important. It clearly indicates that, at Yucca Mountain, higher-grade hydrothermal metamorphism is not confined to deeper parts of the stratigraphic section. Instead, such metamorphism is also expressed in the local vadose zone.

Third, at Yucca Mountain, the top of the volcanic glass-free zone lacks a consistent relationship to the contemporary water table. As shown in Figures 7-13 and 7-17,

glass-free ignimbrites occur both above and below the contemporary water table. In six of the local boreholes (USW G-1, G-2, G-3, G-4, USW H-4, and H-6), such ignimbrites occur within basal parts of the vadose zone. In three other boreholes (J-13, UE-25a#1, and USW VH-2), however, vitric ignimbrites occur well below the contemporary water table. In two cases (boreholes J-13 and USW VH-2), preserved glasses represent the dense vitrophyre of the Topopah Spring Member. For the remaining two cases (boreholes J-13 and UE-25a#1), however, the preserved glasses occur within the unwelded tuffs of Calico Hills and the likewise unwelded Prow Pass Member of the Crater Flat Tuff. The observed cases of preservation of glass below the contemporary water table are important. Specifically, for the Yucca Mountain alteration environment, such cases seem to suggest that under ordinary circumstances (i.e., (1) low ambient temperature; (2) fluids with near neutral pH; and (3) low concentrations of dissolved cations), the glass --> low-grade zeolite conversion proceeds with exceedingly low rates. To speed up the conversion process, special circumstances seem to be required. Of course, such circumstances would be associated with hypogene processes, specifically ascent of warmer and chemically more complex hypogene solutions. Is it possible that at Yucca Mountain, the low-grade alteration minerals (clinoptilolites, mordenites, heulandites), particularly those that were encountered in the vadose zone, are not the sole result of supergene-diagenetic alteration (i.e., brought about by infiltrating rainwater)? Is it possible that, similar to the higher-grade alteration minerals, the lower-grade alteration minerals are also

related to epigenetic hydrothermal metamorphism?

Fourth, at Yucca Mountain, the local clinoptilolites display a surprisingly high degree of chemical diversity, Figures 7-14 and 7-15. In the upper parts of the stratigraphic section, these alteration minerals contain fairly large concentrations of alkali earth elements (Ca + Mg). This is true for the entire area shown in Figure 7-5. In deeper parts of the section, however, there are important discords. For the northwestern segment (boreholes USW G-1, G-2, G-3, USW H-4 and H-5), the corresponding clinoptilolites contain fairly high concentrations of alkali metals (Na + K), Figure 7-14b. This is in sharp contrast to the southeastern sector (boreholes J-13, UE-25p#1, 25a#1, 25b#1, and USW G-4), Figure 7-14a. For these clinoptilolites, the concentrations of alkali earth elements are fairly high, particularly deeper in the section. Importantly, the chemical diversity of the Yucca Mountain clinoptilolites is also clearly expressed in the vadose zone. As shown in Figure 7-15, in terms of major cation composition, the vadose zone clinoptilolites range from sodic-potassic to calcic-magnesian. Underneath the western segment of the repository block, the vadose zone clinoptilolites are predominantly sodic-potassic in composition. Underneath the eastern segment, however, the circumstances are markedly different. Here, the vadose zone clinoptilolites are predominantly calcic-magnesian in composition. Importantly, at Yucca Mountain, the calcic clinoptilolite-bearing intervals also contain other zeolites from the calcic series. Such zeolites have been identified in the following

boreholes: J-13 (heulandite and chabazite), UE-25p#1 (heulandite, laumontite, and erionite), UE-25a#1 (heulandite and erionite), and USW G-4 (heulandite), Figure 7-14. The observed chemical diversity of the low-grade alteration minerals, developed in the same stratigraphically ignimbrites (which presumably, in the pristine state, were chemically homogenous), is important. To account for it, one is left with no choice but to reckon that, at Yucca Mountain, the low-grade alteration minerals were produced with a spatially selective involvement of two chemically distinct fluid phases. Importantly, one of these fluid phases must have been carrying relatively high concentrations of dissolved alkali earth elements (Ca + Mg). Relative to the alkali ignimbrites, however, such fluids are allogenic in the sense that they must have acquired their chemical composition from outside of the ignimbritic complex. It seems reasonable to presume that, as is the case with both the hydrothermal component of the vadose interstitial fluids (Section 5.0) and the parent fluids for the "mosaic" breccia cements (Section 6.0), the calcium-magnesium fluid phases (those responsible for the (Na + K) glass --> (Ca + Mg) clinoptilolite conversion) have acquired their chemical composition, in part, through dissolution of the underlying Paleozoic carbonates. The implications resulting from this presumption are two-fold. On the one hand, the calcic composition of some of the vadose zone clinoptilolites seems to confirm the conclusion drawn previously that, some time in the past, the contemporary vadose zone was inundated by hypogene fluids. Prior to ascent, these fluids seem to have resided either in the Paleozoic carbonates or in the underlying Pre-cambrian

basement. On the other hand, at least in part, the glass --> clinoptilolite conversion is clearly metasomatic. Importantly, the metasomatic introduction of large quantities of alkali earth elements is also indicated by chemical compositions of the whole-rock ignimbrites. As shown in Figures 7-16a through 7-16c, in the present altered state, the Yucca Mountain ignimbrites contain fairly significant concentrations of alkali earth elements. However, for the locally preserved glasses, the corresponding concentrations are typically very low, Figure 7-17a through 7-17c. Because the present-day chemical composition of these glasses reflects the chemical composition of the original igneous matter (that now comprises the whole-rock altered ignimbrites), there is no choice but to attribute the noted compositional discords to epigenetic alkali earth metasomatism. Large volumes of the local ignimbrites have been affected and, consequently, it is totally unreasonable to insist that these processes are of the supergene-diagenetic origin. For the Yucca Mountain calcic zeolites, the observed geochemical affinity with the metasomatically altered whole-rock ignimbrites is important. It provides clear and convincing testimony that some of the low-grade zeolitization is neither deuteric nor supergene-diagenetic. As is the case with the higher-grade alteration minerals, some of the lower-grade zeolites are directly attributable to epigenetic hydrothermal metamorphism.

Fifth, at Yucca Mountain, the planar distribution of alteration temperatures, as interpreted based on the observed spatial distribution of the higher-grade

alteration minerals, appears to be somewhat systematic, Figures 7-18 through 7-20. This distribution, if considered together with the spatial distribution of the calcic zeolites (Figures 7-14a and 7-14b), allows for a recognition of at least two spatio-chemically distinct alteration aureoles. One such aureole is situated in the northwestern sector of Yucca Mountain, Figure 7-20. It occurs near the southern margin of the Timber Mountain Caldera and, in deeper parts, is associated with the predominantly sodic-potassic clinoptilolites (boreholes USW G-1, G-2, G-3, USW H-4, and H-5), Figure 7-14b. The other alteration aureole is situated in the southeastern sector of Yucca Mountain, Figure 7-20. It seems to be centered near the NE striking trace of the Stage Coach Road fault. The corresponding zeolites are predominantly calcic in composition (boreholes J-13, UE-25p#1, UE-25a#1, and 25b#1, and USW G-4), Figure 7-14a. Is it possible that, at Yucca Mountain, the chemically distinct alteration aureoles were formed in association with spatially distinct hypothermal systems, active at different times? To explore this possibility further, it is necessary to consider K/Ar ages from the respective alteration minerals.

7.4 Radiometric Ages of the Yucca Mountain Alteration Minerals

K/Ar radiometric analyses have been reported for a total of 24 samples. These samples were obtained from six of the Yucca Mountain boreholes, specifically, USW G-1, G-2, GU-3, G-3, G-4, and UE-25p#1, Figure 7-5. They represent

metasomatically altered ignimbrites both from the vadose zone and from the "saturated" zone. The analyzed samples represent the clinoptilolite fraction (1-3 micrometers) and the smectite/illite fraction (0.1-0.35 micrometers). Exclusively, these samples were obtained from zones of fairly advanced alteration. For example, the samples analyzed for clinoptilolite contained more than 30 percent of this fraction, in bulk analysis.

The results of the K/Ar radiometric analyses were reported by Aronson and Bish (1987) and WoldeGabriel (1991). The K_2O weight concentrations were determined using acid solutions of beads fused in lithium metaborate and employing a flame photometer. The concentrations of radiogenic argon were measured using an MS-10 mass spectrometer on line with the argon extraction system, and a bulb-pipetted tracer. The argon tracer had been calibrated using the LP-6 interlaboratory standard, with the concentration of radiogenic argon known to be 19.3×10^{-10} mol per gram.

All of the currently available K/Ar ages for samples of the Yucca Mountain alteration minerals are presented in Figures 7-21 through 7-23. Figure 7-21 presents the K/Ar ages from samples of the smectite/illite fraction. This figure also identifies stratigraphic positions of the dated samples, along with the grade of the smectite --> illite transformation. Figure 7-22 presents the K/Ar ages from samples of the clinoptilolite fraction. Stratigraphic positions of the dated samples,

along with the associated authigenic minerals, are presented in Figure 7-23.

The available K/Ar ages are also plotted and interpreted in Figures 7-24 through 7-31. Figure 7-24 shows a relationship between the K/Ar ages of clinoptilolites and the corresponding K/Ar ages of the host ignimbrites. Figure 7-25 presents a relationship between the K/Ar ages of the alteration minerals (both clinoptilolite and smectite/illite fractions) and the corresponding depths of sample acquisition. Figure 7-26 is also a depth diagram. It shows the K/Ar ages of clinoptilolites as a function of depth of the corresponding samples, relative to the contemporary base of the glass-bearing zone. A relationship between the K/Ar ages of clinoptilolites, the corresponding depths of sample acquisition, and the "representative" percentage of major cations is presented in Figure 7-27. For the K/Ar dated-clinoptilolites, this figure may be used to estimate the major cation compositions (i.e., percentage of either Ca + Mg cations or K + Na cations). A relationship between the K/Ar ages of clinoptilolites and the corresponding "representative" percentage of major cations is shown in Figure 7-28. Figures 7-29 and 7-30 present an interpretation of the observed spatial distribution of the Yucca Mountain alteration minerals. This interpretation has been constructed based on the observed chemical (major cation composition) and temporal (K/Ar age) characteristics of these minerals. Finally, Figure 7-31 presents a history of the zeolitic and montmorillonitic alteration, as interpreted for the Yucca Mountain ignimbrites. This history has been constructed based on the K/Ar ages of both the

local alteration minerals and the local igneous rocks.

Examination of Figures 7-24 through 7-30 leads to the formulation of the following nine observations/interpretations. First, relative to the K/Ar ages of the host ignimbrites, the K/Ar ages of the Yucca Mountain clinoptilolites are strongly discordant. As shown in Figure 7-24, the radiometric age discords are expressed in a two-fold manner. On the one hand, even the oldest ages of the local clinoptilolites are significantly younger than the K/Ar ages of the corresponding ignimbrites. This observation confirms the conclusion drawn previously that, at Yucca Mountain, the local clinoptilolites are not the result of deuteritic alteration. Instead, these clinoptilolites appear to be exclusively either epigenetic (hypogene) or diagenetic (supergene), or both. On the other hand, for a given stratigraphic unit, samples of clinoptilolites yield a wide range of radiometric ages. For the Tuffs of Calico Hills, for example, the K/Ar ages of clinoptilolites range from 4.7×10^6 to as little as 2.0×10^6 years B.P. For the Crater Flat Tuff, this variability is even greater, from 10.6×10^6 to about 2.4×10^6 years B.P. The observed range of radiometric ages confirms the conclusion drawn previously that, at Yucca Mountain, the low-grade zeolites are polygenetic, i.e., formed during repeated, or intermittent, episodes of alteration.

Second, in the deeper parts of the Yucca Mountain ignimbritic complex (boreholes USW G-1, G-2, and G-3, Figure 7-20), the K/Ar ages of clinoptilolites are

equivalent to the K/Ar ages of montmorillonitic clays. As shown in Figure 7-25, four samples of the deeper clinoptilolites yielded K/Ar ages ranging from 9.5×10^6 to about 10.6×10^6 years B.P. For alleverdite-kalkberg clays and discrete illites, the corresponding K/Ar ages are about 11×10^6 years B.P. The close correspondence of the K/Ar ages, from samples with different alteration minerals, is of two-fold importance. On the one hand, it is evident that at least some of the Yucca Mountain clinoptilolites are suitable for the purpose of K/Ar geochronometric analysis. In the local alteration environment, these clinoptilolites, although they are excellent molecular sieves that are prone to various ionic exchange reactions, seem to be retaining their potassium and argon contents adequately. Clinoptilolite $\leftrightarrow$ fluid ionic exchanges do not appear to be causing loss of radiogenic argon, and the resulting resetting of the K/Ar radiometric clock. On the other hand, the K/Ar ages indicate that, at Yucca Mountain, some of the sodic-potassic clinoptilolites are neither deuteric nor supergene-diagenetic. Instead, as is the case with the higher-grade and calcic zeolites, some of these clinoptilolites appear to be of epigenetic hydrothermal origin. Such a state of affairs is clearly indicated by the fact that both montmorillonitic clays (indisputable products of hydrothermal metamorphism) and the spatially corresponding clinoptilolites yield equivalent K/Ar ages. The observed range of K/Ar ages, from 9.5×10^6 to 11.0×10^6 years B.P., further indicates that, for the deeper parts of the northwestern segment of Yucca Mountain, both montmorillonitic clays and sodic-potassic clinoptilolites record the prolonged

activity of a hydrothermal system driven by the Timber Mountain heat source.

Third, at Yucca Mountain, the K/Ar ages of clinoptilolites increase with increasing depth below the topographic surface. As shown in Figure 7-25, the youngest clinoptilolites occur higher in the stratigraphic section, above and immediately below the contemporary water table. For such clinoptilolites, the K/Ar ages range from about 8.5×10^6 to as little as 2.0×10^6 years B.P. Deeper clinoptilolites, however, are clearly older. Samples of such clinoptilolites yield K/Ar ages ranging from 9.5×10^6 to about 10.6×10^6 years B.P., Figure 7-25. The clearly expressed K/Ar age vs. depth gradient may be presumed to indicate two important points. On the one hand, the availability of ignimbritic matter in the vitric state seems to have been a factor that controlled timing and duration of the low-grade zeolitization. Such a state of affairs is suggested by the relationship depicted in Figure 7-26. It may be observed that, immediately below and near the contemporary base of the vitric zone, the indigenous clinoptilolites yield the youngest K/Ar ages. These ages range from 2×10^6 to about 4.6×10^6 years B.P. At greater depths below the contemporary base of the vitric zone, the K/Ar ages of clinoptilolites become progressively older. Clinoptilolites, sampled 400-800 feet below the base of the vitric zone yield K/Ar ages ranging from 4.5×10^6 to about 8.5×10^6 years B.P. Still further below the vitric zone, the oldest clinoptilolites are present. On the other hand, the availability of ignimbritic matter in the vitric state seems to have been diminishing differentially in time and space. The oldest

K/Ar ages of the deep and clearly epigenetic clinoptilolites suggest that, at a depth in excess of ~1.0 km, fairly high alteration temperatures associated with the Timber Mountain hydrothermal episode have persisted for a fairly long period of time. The result of this prolonged thermal exposure was fairly intense and pervasive alteration of the deeper ignimbrites. At the corresponding depth, the vitric reservoir has been completely depleted, and further low-grade zeolitization has been effectively arrested. Higher in the stratigraphic section, however, the Timber Mountain hydrothermal episode was much more subtly expressed. Evidently, the rate of vitric reservoir depletion was substantially lower and much of the vitric matter has survived. Subsequent alteration events could have consumed some of the remaining glass to produce the younger clinoptilolites. The newly produced clinoptilolites seem to have grown over the older clinoptilolites. The resulting radiometric age overprinting, being a depth-variant effect, is expressed in the observed K/Ar age vs. depth gradient.

Fourth, at Yucca Mountain, the K/Ar ages of the younger clinoptilolites must be regarded as "mixed" ages, reflecting the polygenetic character of the local zeolitic alteration. Deeper in the stratigraphic section, the relative proportions of the younger clinoptilolites are fairly small. Here, the indigenous clinoptilolites were largely formed during the Timber Mountain Caldera hydrothermal episode. For these clinoptilolites, the K/Ar ages may be regarded as accurately recording the

waning stages of this episode. Higher in the stratigraphic section, however, the low-grade zeolitic alteration appears to be an intermittently active process. Here, the relative proportions of the younger clinoptilolites vary as a function of distance below the contemporary base of the vitric zone. The observed mean K/Ar age vs. depth gradient seems to be a composite expression of two factors, namely: (1) depthward diminishing overprinting of the young clinoptilolites, and (2) depthward increasing rates of paleo-depletion of the vitric reservoir. As far as the latest episode of the low-grade zeolitization is concerned, however, it is not possible to reliably specify an upper age limit because of the inferred radiometric age overprinting and the relatively small number of dated samples. It is fairly safe to conclude, however, that this episode must be younger than about 2×10^6 years B.P.

Fifth, the Yucca Mountain clinoptilolites consist of at least four chemico-temporally distinct sets. As shown in Figure 7-28, the oldest set includes the epigenetic clinoptilolites formed during the Timber Mountain hydrothermal episode. Such clinoptilolites are predominantly sodic-potassic in composition and yield K/Ar ages ranging from 9.5×10^6 to about 10.6×10^6 years B.P. They were encountered in the northwestern sector of Yucca Mountain, in deeper segments of boreholes USW G-1, G-2, and G-3. The second oldest set was identified in the southeastern sector of Yucca Mountain. It was encountered in deeper segments of boreholes UE-25p#1 and USW G-4. The corresponding clinoptilolites are predominantly calcic-magnesian in composition and yield K/Ar ages ranging from 7.3×10^6 to about

8.4×10^6 years B.P. The remaining two younger sets occur higher in the stratigraphic section. The predominantly sodic-potassic set is slightly older. Samples of the corresponding clinoptilolites yield K/Ar ages ranging from 3.8×10^6 to about 7.0×10^6 years B.P. The youngest set contains a fairly high proportion of the calcic-magnesian clinoptilolites. Such clinoptilolites yield K/Ar ages ranging from 4.7×10^6 to as little as 2.0×10^6 years B.P.

Sixth, the Yucca Mountain clinoptilolites display a well expressed tendency toward becoming younger with increasing concentration of alkali earth elements. As shown in Figure 7-28, the most alkalic clinoptilolites, those containing only 10-15 percent of Ca + Mg cations, yield an average K/Ar age of about 7.8×10^6 years B.P.; the chemically intermediate clinoptilolites, those containing 20-25 percent of Ca + Mg cations, yield an average K/Ar age of about 5.8×10^6 years B.P.; and the most calcic clinoptilolites, those containing 30-45 percent of Ca + Mg cations, yield an average K/Ar age of about 4.7×10^6 years B.P. Clearly, the observed average K/Ar age vs. alkali earth content relationship is of two-fold importance. On the one hand, this relationship indicates that, at Yucca Mountain, clinoptilolites are polygenetic mixtures of two compositionally distinct end-members. One end-member consists of predominantly alkalic (Na + K) clinoptilolites. The development of this end-member, however, was a multi-stage process. Deeper in the stratigraphic section, this process was initiated and terminated during the Timber Mountain Caldera hydrothermal episode. Higher in the section, however,

the production of the alkalic clinoptilolites continued to at least about 4×10^6 years B.P. The observed relatively low concentrations of alkali earth elements indicate that, at Yucca Mountain, formation of the predominantly alkalic zeolites involved fluid phases that were carrying significant concentrations of dissolved alkali metals. To some perhaps large degree, such fluids could have acquired dissolved solids through ionic exchange reactions with the alkali ignimbrites. However, the observed elevated alkali earth concentrations for both whole-rock altered ignimbrites and separated clinoptilolites, relative to the corresponding glasses (Figures 7-14, 7-16, and 7-17), strongly suggest that these fluids were produced by intermixing of sodium-potassium fluid phases (from the alkali ignimbrites) with calcium-magnesium fluid phases (from the underlying Paleozoic carbonates). The other end-member of the clinoptilolitic mixture consists of the predominantly calcic clinoptilolites, Figure 7-14. Spatially, the occurrence of these clinoptilolites is accompanied by the occurrence of other species from the calcic zeolitic series. Specifically, such accessory species include heulandite, erionite, chabazite, stellerite, and laumontite. As with the alkalic zeolites, development of the calcic zeolites was a multi-stage process. It was initiated at or after 8.5×10^6 years B.P., and has continued throughout Pliocene time and, most probably, into Quaternary time. In contrast to the former zeolites, however, the production of the calcic zeolites did require almost exclusive involvement of calcium-magnesium fluid phases. Relative to the alkali ignimbrites (Figure 7-17), however, such fluid phases are allogenic, and must have acquired their chemical character from

outside of the ignimbrites. On the other hand, the observed average K/Ar age vs. alkali earth content relationship further confirms the earlier conclusion that, at Yucca Mountain, most of low-grade zeolitization is the result of epigenetic (hypogene) hydrothermal metamorphism. The older predominantly alkalic clinoptilolites display clear spatio-temporal association with the Timber Mountain hydrothermal system. To account for the presence of these clinoptilolites, therefore, there is no rational reason to call for supergene-diagenetic alteration. The observed overprinting of the earlier alteration by the younger calcic zeolites, however, was produced by allogenic calcium-magnesium fluid phases. To associate such fluid phases with infiltrating rainwater, however, is not a rational proposition. In sharp contrast, the observed metasomatic overprinting, including its timing, is readily attributable to intermittent ascent of deep-seated and calcium-magnesium fluid phases, either from the Paleozoic carbonates or from the underlying Pre-cambrian basement. In terms of the major dissolved cations, these fluid phases appear to have been analogous to those that (1) were observed in the pore space of the contemporary vadose zone, Section 5.0, and (2) were inferred to have been involved in the precipitation of the authigenic cements for the local "mosaic" breccias, Section 6.0. Importantly, the post- 4.8×10^6 years B.P. age of ascent of calcium-magnesium fluids, as interpreted from fission-track ages of allogenic zircons, is independently confirmed by the K/Ar ages of calcic clinoptilolites.

Seventh, at Yucca Mountain, the spatio-chemically distinct alteration aureoles identified previously (Figure 7-20) appear to have been formed at different times, most probably in response to broadly different hydrothermal processes. Such a state of affairs is indicated by both the K/Ar ages of alteration minerals and the observed spatial distribution of these minerals. For the northwestern segment of Yucca Mountain (boreholes USW G-1, G-2, G-3, H-4, and H-5), the corresponding alteration aureole contains three suites of alteration minerals. These are: (1) higher-grade montmorillonitic clays (allevardite-kalkberg clays and discrete illite), Figure 7-12; (2) predominantly sodic-potassic clinoptilolites, Figure 7-14; and (3) higher-grade minerals from the alkali zeolitic series (analcime and albite), Figure 7-11. From Figures 7-25 and 7-28, it may be observed that for these alteration minerals, the K/Ar ages range from 11×10^6 to about 9.5×10^6 years B.P. Importantly, within the northwestern alteration aureole, alteration temperatures diminish with increasing distance from the southern margin of the Timber Mountain Caldera, Figure 7-20. At a constant depth of ~3000 feet, these temperatures keep dropping from a maximum of 130° (borehole USW G-2), through 90-110° (boreholes USW G-1 and H-6), to 80° Celsius in the southernmost borehole USW G-3. Both the southward diminishing alteration temperatures and the K/Ar ages of the corresponding alteration minerals indicate that the northwestern alteration aureole expresses and records the prolonged activity of a hydrothermal system driven by the Timber Mountain Caldera heat source, Section 2.5. For the southeastern segment of Yucca Mountain (boreholes J-13, UE-25p#1,

UE-25a#1 and 25b#1, and USW G-4), the corresponding alteration aureole also contains three suites of alteration minerals. These are: (1) predominantly calcic clinoptilolites, Figure 7-14a; (2) higher-grade zeolites from the calcic series (heulandite and laumontite), Figure 7-14a; and possibly (3) alleverdite clays, Figure 7-12. From Figure 7-28, it may be observed that for these minerals the K/Ar ages range from about 8.5×10^6 to as little as 2.0×10^6 years B.P. This fairly long alteration period, if considered together with the absence of pervasive alteration and the fairly low-grade of this alteration, indicates that the causative hydrothermal process did not operate continuously. Rather, this process appears to have been active intermittently, or ephemerally. Within the southeastern alteration aureole, alteration temperatures seem to be diminishing with northwestwardly increasing distance away from the trace of the Stage Coach Road fault, Figure 7-20. In the vicinity of this fault, the alteration suite indicates a temperature range from 125° (borehole UE-25p#1) to 140° Celsius (borehole J-13). Some 1-2 km westward of this fault, however, the corresponding temperatures are within a 80-100° Celsius range. The observed planar distribution of the inferred alteration temperatures suggests that the southeastern alteration aureole is (1) fault-based, and (2) rooted in the Stage Coach Road fault zone. Is it possible that this aureole expresses and records the intermittent fault-based activity of a hydrothermal system, driven by a conductively replenished heat source? Both the inferred prolonged yet intermittent character of this activity and the strongly metasomatic character of the corresponding alteration minerals are consistent

with this possibility.

Eighth, for the Yucca Mountain alteration minerals, the observed spatial distribution of both the K/Ar ages (Figures 7-25 and 7-27) and the concentrations of alkali earth elements (Figures 7-14a and 7-14b) are directly attributable to the space-differential overprinting of the northwestern alteration aureole by the subsequent southeastern alteration aureole. The envisaged alteration overprinting is schematically depicted in Figure 7-29. This figure has been constructed recognizing that, relative to a magmatic center-based hydrothermal system, a fault-based hydrothermal system is fundamentally different. For the former system, the prolonged circulation of fluids is controlled by a fairly uniformly-distributed intrinsic hydraulic conductivity. Consequently, for the magmatic center-based alteration aureole, the isogradal surfaces are fairly uniform, and reflect both proximity to the topographic surface and proximity to the causative heat source, Figure 7-29a. The corresponding upward fluid flux is spatially extensive, and is fairly uniformly distributed about the causative magmatic heat source. For the latter system, however, the circumstances are markedly different, Figure 7-29b. Two features are particularly important, Figure 7-30. On the one hand, for a fault-based hydrothermal system, the lateral diffusion of ascending fluids, away from the parent fault, is controlled by the spatial distribution of extrinsic hydraulic conductivity. In contrast to intrinsic conductivity, the latter is controlled by the in-situ stress field developed around a yielding, and soon to be

unstable, fault. Szymanski (1989) pointed out that, prior to fault rupture and prior to the subsequent ascent of deep-seated fluids, the corresponding in-situ stress field, and with it the extrinsic hydraulic conductivity structure, is characteristically space-variant. Near the soon-to-be ruptured fault, in-situ stress enhancement (extrinsic) of hydraulic conductivity is relatively deep and large. Away from this fault, however, both the depth extent and amount of conductivity enhancement diminish gradually. In Figure 7-30a, this effect is represented by the space-variant depth $Z_{(x,y,t)}$. On the other hand, for a fault-based hydrothermal system, it is totally inappropriate to envisage the corresponding upwelling of fluids as a spatially extensive and uniform increase in the upward fluid flux. Szymanski (1989) has pointed out that, at Yucca Mountain, the expected fluid flux increase must necessarily be restricted to a *"ruptured fault and a few adjacent fractures."* In other words, at a depth of, say, 2.0 km, the fluid flux increase is narrowly focused around the parent fault, and forms an upwelling center, elongated along the fault strike, Figure 7-30b. Perpendicular to the fault trace, the upward fluid flux increase ($\Delta q_{(x)}$) is strongly dependent upon the distance away from the parent fault. Keeping in mind both of the above inferred features, it is possible to construct a logical explanation for the observed spatially-selective development of the alkali earth metasomatism, Figures 7-14 and 7-29a. Such an explanation is provided in Figure 7-30c. In this figure, two reasonable points are emphasized. These are (1) the calcium-magnesium fluid phases responsible for the observed metasomatic zeolitization were ejected in the southeastern segment of Yucca

Mountain, and (2) the fluid diffusion paths were controlled by the deformation-induced gradients of extrinsic hydraulic conductivity. By overprinting both (1) the earlier alteration aureole (produced by the Timber Mountain Caldera hydrothermal system, Figure 7-29a), and (2) the subsequent alteration aureole (produced by the fault-based hydrothermal system, Figure 7-29b), it is possible to construct a satisfactory explanation for the field relationships actually observed. Such an explanation is depicted in Figure 7-29c. By comparing this figure with Figures 7-11, 7-12, 7-14, 7-16, and 7-28, it may be observed that the proposed explanation does account for: (1) the observed distribution of epigenetic alteration grades, (2) the observed chemical composition of the epigenetic alteration products (clinoptilolites and whole-rock ignimbrites); (3) the corresponding K/Ar ages; and (4) the independently evident hydrothermal character of the local metamorphism. Further testing of the proposed explanation will be performed in Section 8.4. In that regard, particularly helpful is the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio vs. depth gradient, as constructed by Peterman et al. (1991) based on the results of isotopic analyses of samples of the Yucca Mountain calcitic veins.

Ninth, at Yucca Mountain, the K/Ar history of hypogene zeolitic and montmorillonitic alterations fits harmoniously with the independently known history of local magmatism, Figure 7-31. Both montmorillonitic clays and the older alkalic clinoptilolites are temporally correlative with the late Timber Mountain magmatic stage, Noble et al. (1991). The younger calcic clinoptilolites

are also temporally correlative with subsequent local magmatic episodes. Such episodes are expressed and recorded through (1) the "older rift basalts" of Jackass Flat and possibly the Shoshone Mountain rhyolites, and (2) the "younger rift basalts" of Crater Flat, Frizzell and Shulters (1990). The fact that both of the independently known histories are temporally correlative is important. It provides additional assurances that (1) the K/Ar ages of the epigenetic clinoptilolites do adequately reflect timing of the main episodes of hypogene alteration, and (2) the low-grade metasomatic zeolitization expresses and records episodically-continuous, or intermittent, hydrothermal metamorphism.

7.5 Summary and Conclusions

Integrated analysis of the extensive set of data for the Yucca Mountain alteration minerals leads to the establishment of secure views regarding the origin and age of these minerals. For the purposes of this report, these views may be summarized by the following six points. First, the currently available mineralogical and radiometric data do not provide any factual basis to support the belief that, at Yucca Mountain, the observed zeolitic and montmorillonitic alteration minerals are largely the result of deuteritic processes, i.e., processes that were operative during the cooling, syn-depositional devitrification and solidification of the local ignimbrites. To the contrary, both the spatial

distribution of the alteration minerals (relative to the stratigraphic boundaries) and the K/Ar ages of these minerals are in clear and direct conflict with this belief. Furthermore, the metasomatic character of some of the local alteration products requires total rejection of this belief.

Second, the currently available geochemical data do not provide any factual support for the belief that, in the Yucca Mountain vadose zone, low-grade zeolitization is the result of supergene-diagenetic alteration. To the contrary, both the chemical affinity of the vadose zone clinoptilolites with the "saturated" zone clinoptilolites and the metasomatic character of some of the vadose zone zeolites are in direct conflict with this belief.

Third, the currently available radiometric data do not provide any factual basis to support the belief that, in the Yucca Mountain vadose zone, the epigenetic (hypogene) clinoptilolites were all formed in the distant past. To the contrary, the presently known spectrum of K/Ar ages, from 10.5×10^6 to as little as 2×10^6 years B.P., is in direct conflict with this belief.

Fourth, for the Yucca Mountain alteration minerals, all of the presently available geochemical, mineralogical, and radiometric data are satisfactorily reconciled within the context of traditional geologic practice. In accordance with such practice, the pro-grade zeolitic and/or montmorillonitic alteration is a hypogene

process resulting from either burial or hydrothermal metamorphism (see, for example, Steiner, 1955; Turner and Verhoogen, 1960; Coombs, 1970; Meyer and Hemley, 1967; and Iijima, 1975, 1978, and 1980). In the case of Yucca Mountain, however, the burial metamorphism option is not appropriate. Consequently, the observed pro-grade alteration facies must be attributed to epigenetic hydrothermal metamorphism.

Fifth, within the context of suitability considerations of Yucca Mountain to accommodate a high-level nuclear waste repository, the observed low-grade hypogene zeolitization is clearly of two-fold significance. On the one hand, the fact that the corresponding alteration minerals are observed above the contemporary water table provides clear and unequivocal testimony that, at Yucca Mountain, the vadose zone ignimbrites have been affected by epigenetic hydrothermal metamorphism. By standards employed in ordinary geologic practice, this conclusion is regarded as secure beyond a reasonable doubt. This is so because the same conclusion is indicated by paragenetic analysis of both the vadose zone interstitial fluids (Section 5.0) and the surficial "mosaic" breccias (Section 6.0). On the other hand, the fact that the metasomatic clinoptilolites yield a wide spectrum of the K/Ar ages, from 8.5×10^6 to as little as 2.0×10^6 years B.P., indicates that the causative hydrothermal metamorphism was not restricted to a single, yet remote, episode. To the contrary, the currently available K/Ar data indicate that, in the Yucca Mountain vadose zone, the metasomatic formation of epigenetic

clinoptilolites has been an episodically-continuous, or intermittent, process.

Because of the inferred radiometric age overprinting and the limited number of K/Ar age determinations of clinoptilolites, it is not possible to reliably specify an upper age limit for the causative hydrothermal metamorphism. Both the fission-track ages of allogenic zircons (incorporated in "mosaic" breccia cement) and K/Ar ages of calcic clinoptilolites, however, indicate that this metamorphism was extended into Plio-Quaternary time.

Sixth, at Yucca Mountain, the anticipated expressions of both kinds of expected hydrothermal systems have been identified. From Section 2.5, it may be recalled that these expectations were developed completely independently, based on consideration of the local geodynamic configuration. In the upper 2 km of the local stratigraphic section, the combined influence of two thermodynamically different hydrothermal systems is expressed by the observed juxtaposition of the chemico-temporally distinct alteration aureoles produced in association with at least two spatially distinct hydrothermal centers. The northwestern alteration center is older. The respective aureole is associated with three suites of alteration minerals, including: (1) higher-grade montmorillonitic clays, such as alleverdite-kalkberg clays and discrete illite; (2) higher-grade minerals from the alkali zeolitic series, specifically analcime and albite; and (3) predominantly sodic-potassic clinoptilolites. Both the K/Ar ages of the corresponding epigenetic minerals (from 11×10^6 to 9.5×10^6 years B.P.) and the observed spatial arrangement

of the corresponding isogradal surfaces indicate that the northwestern alteration center expresses and records the prolonged activity of a hydrothermal system driven by the Timber Mountain Caldera heat source. The southeastern alteration center is younger. The respective aureole is also associated with three suites of alteration minerals, including: (1) higher-grade zeolites from the calcic series, such as heulandite, laumontite, and analcime; (2) predominantly calcic clinoptilolites; and possibly (3) higher-grade montmorillonitic clays, specifically the alleverdite clays. Both the broad range of K/Ar ages of the corresponding minerals (from 8.5×10^6 to 2.0×10^6 years B.P.) and the observed spatial arrangement of these minerals indicate that the southeastern alteration center expresses and records the ephemeral, or intermittent, activity of a fault-based hydrothermal system driven by conductively replenished heat sources. The fact that the activity of this hydrothermal system is expressed in the local ignimbrites, including those comprising the vadose zone, entails profound implications. Specifically, this fact demands that a determination be made whether or not, at Yucca Mountain, the late Quaternary formation of both calcretes and compositionally and texturally equivalent calcitic veins constitutes a pedogenic expression of fundamental hypogene processes. After all, the result of considering the contemporary geodynamic configuration of the Yucca Mountain region, as set forth in Section 2.5, does not provide a single reason to believe that, during the Quaternary time span, such processes became dormant. If anything, this result suggests that, relative to Pliocene time, augmentation of conductive heat flow by

convective heat flow may have been enhanced. To further explore the possibility that, at Yucca Mountain, the late Quaternary calcretes and the equivalent bedrock veins express and record an intermittently active hydrothermal system, it is necessary to consider these deposits in some further detail. The following Section 8.0 is devoted to this end. The analysis is based on a variety of geochemical data, including: (1) uranium, strontium, oxygen, and carbon isotopic data; (2) uranium-thorium radiometric data; and (3) fluid-inclusion data.

8.0 The Age and Origin of the Yucca Mountain Carbonatization

8.1 General

With regard to the suitability of Yucca Mountain (either for site characterization or for the development of a high-level nuclear waste repository) the origin of the local calcites is clearly of critical importance. This is true because, in contrast to the previously considered alteration minerals, some of the Yucca Mountain calcites are known to have been precipitated subsequent to the local deposition of the Bishop Ash, some 7×10^5 years B.P. These calcites are major components of four distinct depositional phases. These are: (1) calcretes and sinters; (2) compositionally and texturally equivalent surficial bedrock veins; (3) acknowledged spring deposits; and (4) subsurface calcites, developed as infillings of lithophysal cavities, interstitial impregnations, and discrete veins. From Section 3.0 it may be recalled that, for the local calcretes, noteworthy characteristics are: (1) occurrence in the form of five distinct textural varieties; (2) polygenetic, or multistage development; and (3) compositional and textural affinity with bedrock veins and with authigenic cements of the "mosaic" breccias. The surficial calcitic veins occur in both unconsolidated aeolian and colluvial deposits as well as in the underlying ignimbrites. They range in thickness from a few millimeters to a maximum of about 1 m. In addition to calcite, the surficial calcitic veins also contain opal CT, opal A, sepiolite, and smectite, Vaniman et al.

(1988). The calcite-bearing spring facies have been identified in the southern parts of Crater Flat, Hoover et al. (1981) and Taylor (1991). From this area, three morphologically and compositionally distinct deposits have been reported. The so-called "Crater Flat deposit" consists of calcareous clays and silts, and includes several mounds of concretionary carbonate. The so-called "root cast deposit" consists of gypsum cemented silts and clays. It contains abundant glass shards (replaced by authigenic carbonate) and calcareous root casts. The so-called "horse-tooth deposit" consists of silts and clays interbedded with layers of tephra, freshwater limestones, and diatomites. It contains vertebrate fossils, including *Mammathus* sp., *Equus* sp., *Lepus* sp., and a large camelid. The diatoms identified in three samples suggest ". . . *alkaline, at times slightly saline, shallow water. Many of the species identified are found in alkaline, somewhat saline spring systems . . .*" Hoover et al. (1981). The subsurface calcites occur as two textural varieties, namely sparry calcites and micritic calcites. Commonly, calcite is observed to be associated with opaline silica, minor quartz, and trace amounts of manganese and iron oxides. In a few instances, however, calcite is found to be associated with fluorite and barite. In the "saturated" zone, examples of this association were encountered in boreholes UE-25a#1 (depth ranging from 3200 to 3600 feet), USW G-2 (depth ranging from 4200 to 5700 feet), and USW G-3 (depth of about 4700 feet). In the vadose zone, an example of calcite and fluorite association was found in borehole USW GU-3, at a depth as shallow as 318-362 m. Based on the microscopically observed cross-cutting relationships, calcite was

found to be the latest phase of the Yucca Mountain paragenetic sequence, Carlos et al. (1990). Deposition of calcite, however, appears to be a polygenetic, or multistage process. In borehole USW G-2, for example, calcite occurs as at least two generations separated by the deposition of heulandite, Carlos et al. (1990). Two temporally and texturally distinct generations of epigenetic calcite were also observed in borehole USW GU-3, at a depth of 482 feet. In this borehole, sparry calcite was observed to be "*coated by a later growth of fine-grained euohedral calcite*," Vaniman et al. (1984).

To account for the seemingly ubiquitous carbonatization, two sharply contrasting viewpoints may be put forth. On the one hand, considering the manifestly magmatically active character of the region, and following elementary tectono physical principles, one may reasonably speculate that at Yucca Mountain all of the observed textural and paragenetic varieties of calcite are hypogene in origin. These epigenetic minerals may be presumed to represent carbonatization that is typically associated with propylitic alteration. The local presence of the latter, however, is an independently established fact, Section 7.0. The older sparry calcites, particularly those associated with barite and fluorite, may be presumed to express the prolonged activity of a hydrothermal system driven by the Timber Mountain heat source. The subsequent micritic calcites, however, may be presumed to express the intermittent activity of a fault-based hydrothermal system driven by conductively replenished heat sources. After all, it is not

difficult to envisage that the metasomatic or allogenic calcium-magnesium fluids (those that are presently observed in the vadose zone pore space and that were once involved in both the calcic zeolitization and the formation of the "mosaic" breccia cements) were also involved in the formation of veins composed of calcite, opaline silica, and sepiolite. It is also not extraordinarily difficult to envisage that, upon reaching the topographic surface, these fluids would stimulate a variety of pedogenic processes, including the development of the compositionally, texturally, and isotopically equivalent calcretes, sinters, and veins.

On the other hand, it has also been proposed that the overall carbonatization represents a combination of temporally distinct processes, ranging in origin from hypogene to supergene. For obvious reasons, this unorthodox and completely novel viewpoint is being aggressively advocated by some investigators associated with the Yucca Mountain Site Characterization Project. For the "saturated zone," the calcite, barite, and fluorite mineral assemblage is considered to represent the epigenetic mineralization resulting from the hydrothermal stage of activity of the Timber Mountain Caldera. For the vadose zone, however, the calcite and fluorite assemblage is regarded as formed *"during the initial deposition or cooling of the tuffs,"* Bish (1989). Both calcretes and micritic veins (including those encountered below the water table) are regarded as supergene deposits; in other words, they were precipitated from infiltrating rainwater. For rainwater, the source of dissolved calcium is thought to be windblown dust; the source of dissolved carbon

is the local biomass; and the source of radiogenic strontium-87 occurs in the previously metasomatically altered ignimbrites. Finally, the acknowledged spring deposits are considered to be most likely related to so-called perched waters "*from shallow basin-fill aquifer, non-existent today*," Taylor (1991).

The a priori assumption of a supergene origin for the Yucca Mountain micritic calcites is being advocated particularly aggressively by some members of the USGS staff. During the last three years, numerous papers have been published in prestigious scientific journals in support of this viewpoint, see, for example, Szabo and Kyser (1990), Quade and Cerling (1990), Stuckless et al. (1991 and 1992), Peterman et al. (1991 and 1992), and Whelan and Stuckless (1991 and 1992). Routinely, the same authors advocate their viewpoints at various periodic meetings of the American geoscience community (GSA, AGU, etc.). The U.S. Congress, various governmental agencies, and the public are being kept informed about the newly discovered scientific facts. The viewpoints advocated represent the results of more than a decade of research on Yucca Mountain. According to the USGS investigators, a supergene origin of micritic calcites is indicated by the isotopic characters of uranium, strontium, oxygen, and carbon incorporated in these calcites.

Four lines of evidence have been put forth in support of this interpretation. First, the results of isotopic investigations revealed that for bedrock veins the isotopic

characteristics, both stable and radiogenic, are similar to those incorporated in the compositionally and texturally equivalent calcretes and sinters, Stuckless et al. (1991). This undisputed and expected isotopic affinity, in turn, was taken as indicating a supergene origin of the bedrock veins considered. To assess the scientific value of this conclusion, it is useful to examine the underlying syllogism. For this syllogism, the major premise consists of the statement, "the Yucca Mountain calcretes and sinters are of supergene origin," the minor premise consists of the statement, "for a given area, all supergene deposits carry equivalent isotopic signatures;" and the resulting conclusion may be represented by the statement "because the calcretes and the bedrock veins do carry equivalent isotopic signatures, both of these deposits are of supergene (pedogenic) origin." Unacceptably, the unproven statement, "the calcretes and sinters are of supergene origin" occurs in both the major premise and the resulting conclusion. Clearly, the latter has been derived via circular logic and, therefore, can not possibly be regarded as the result of valid scientific reasoning.

Second, the results of isotopic investigations revealed that, at Yucca Mountain, the isotopic character of uranium and strontium incorporated in micritic calcites differ from their counterparts dissolved in contemporary fluids immediately below the water table (mean fluid sampling depth is ~1.0 km), Stuckless et al. (1991). These undisputed and expected isotopic discords, in turn, were taken as completely negating the hypogene origin of both calcretes and isotopically equivalent veins.

Again, to recognize the falsehood of this conclusion, it is necessary to examine the underlying syllogism. For this syllogism, the major premise may be represented by the statement "during the last, say, 2×10^6 years B.P., the isotopic characteristics of uranium and strontium dissolved in the local fluids were both time- and depth-invariant and, consequently, the parent fluids for any hypogene deposits must have been isotopically similar to those currently observed within 0.5-1.5 km of the topographic surface;" the minor premise consists of the statement "both calcretes and isotopically equivalent veins must be either of supergene origin or of hypogene origin;" and the resulting conclusion may be represented by the statement "because the isotopic characteristics of the radiogenic elements dissolved in the subsurface fluids are dissimilar to those incorporated in calcites, the calcites cannot possibly be of hypogene origin and, therefore, must be of supergene origin." In contrast to the previous case, this syllogism does not involve unacceptable circular logic. Instead, it is based on the major premise which (1) in itself is the subject of the isotopic investigations, and (2) is false, if the calcites are of hypogene origin. For the major premise to be true, factors that control the isotopic characteristics of shallow fluids (i.e., fluid circulation depth, fluid fluxes, conditions of isotopic exchange reactions, etc.) must remain time- and space-invariant, for time spans of $\sim 2 \times 10^6$ years B.P. Such ubiquitous tranquillity, however, may only be associated with a hydrologic system for which both the hydraulic conductivity structure and the boundary conditions are time-invariant. In other words, this hypothetical hydrological system behaves

as an unperturbed equilibrium or linear dynamic system. For the Yucca Mountain hydrologic system, however, the principal issue that is being addressed using the isotopic data is whether or not the above attributes are indeed present, Szymanski (1989). In view of this, the assumption that the major premise is true is equivalent to the assumption that both the hydraulic conductivity structure and the boundary conditions remain time-invariant and, consequently, the corresponding position of the water table also remains time-invariant. Because it does not make a bit of sense to investigate water table change by assuming that it remains time-invariant, the major premise, and with it the Stuckless et al. (1991) syllogism, must be rejected as devoid of any rational basis.

Third, the results of isotopic analyses revealed that, at Yucca Mountain, the isotopic character of carbon incorporated in calcites displays a clear bi-modal distribution, Whelan and Stuckless (1991 and 1992). Specifically, calcites collected from within the upper 1 km of the stratigraphic section were shown to be associated with values of the $\delta^{13}\text{C}$ ratio ranging from -3.0 to about -9.5 per mil_{PDB}; calcites sampled from within the 0.9-1.8 km depth interval, however, were shown to be associated with "heavier" values of the $\delta^{13}\text{C}$ ratio, from -2.0 to about +3.0 per mil_{PDB}. The observed and undisputed bi-modality of the $\delta^{13}\text{C}$ ratios, in turn, was taken as unequivocal evidence for supergene-pedogenic origin of the carbon-13 depleted calcites. To recognize the misleading character of this conclusion, it is necessary to examine the underlying syllogism. For this

syllogism, the major premise may be represented by the statement, "at Yucca Mountain, the locally circulating carbon originates exclusively from two sources, namely 1) a biogenic source, providing isotopically 'light' carbon, and 2) an inorganic source (Paleozoic carbonates), providing isotopically 'heavy' carbon;" the minor premise may be represented by the statement, "at Yucca Mountain, hypogene calcites contain exclusively isotopically 'heavy' carbon (derived from the inorganic source) and supergene calcites contain both isotopically 'light' carbon (derived from the biogenic source) and isotopically 'heavy' carbon ('light' biogenic carbon dioxide modified via diffusional carbon-13 enrichment);" and the resulting conclusion may be represented by the statement, "because the calcites in question contain isotopically 'light' carbon these calcites cannot possibly be of hypogene origin and, therefore, must be of supergene origin." Prior to accepting the stated conclusion as valid, it is prudent to examine first the major premise and then the minor premise. With this end result in mind, the following two lines of evidence are considered. On the one hand, it is a well established fact that, in magmatically active regions, the circulating carbon originates from three different sources, instead of the presumed two sources, see, for example, Faure (1987) and Hoefs (1987). These are: (1) an inorganic source, at Yucca Mountain this source is represented by the Paleozoic carbonates; (2) a biogenic source, at Yucca Mountain this source is represented by a biomass containing ~85 percent of plants with the C-3 metabolic pathway; and (3) a deep-seated igneous source. Depending upon CO₂ dissolution depth, or temperature, igneous sources of carbon may yield

hydrothermal fluids for which the isotopic character of dissolved carbon ranges from "light," through "intermediate," to "heavy." On the other hand, it is also a well established fact that, in magmatically active regions, unequivocal hypogene carbonates commonly contain isotopically "light" carbon, in addition to the presumed "heavy" carbon. For travertines from the Long Valley Caldera, for example, White et al. (1990) reported values of the $\delta^{13}\text{C}$ ratio ranging from -7.0 to +3.5 per mil_{PDB}. Furthermore, for hydrothermal carbonates, the actually known variability of the $\delta^{13}\text{C}$ ratios is even wider, from -12.0 to about +12.0 per mil_{PDB}, see, for example, Hoefs (1987) and Turi (1986). Clearly, for the manifestly magmatically active region of Yucca Mountain, both of the stated premises are not appropriate and most likely are false. The resulting USGS conclusions, therefore, can not possibly be regarded as either uniquely valid or based on solid scientific reasoning.

Fourth, the results of strontium isotopic analyses revealed that in the north-western segment of Yucca Mountain, there is a clearly expressed $^{87}\text{Sr}/^{86}\text{Sr}$ vs. depth gradient, Peterman et al. (1991 and 1992). This gradient is evident for both whole-rock ignimbrites and the spatially corresponding calcites. Specifically, for both of these solid phases, values of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio diminish depthward, from the 0.7112-0.7130 range near the topographic surface to the 0.7085-0.7090 range at a depth of ~1.5 km. The observed and undisputed strontium isotopic gradients were taken, in turn, as consistent with and supportive of a supergene origin for

the late micritic calcites. To assess the reliability of this conclusion, it is again helpful to examine the underlying syllogism. For this syllogism, the major premise may be represented by the statement "for the whole-rock ignimbrites, the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio vs. depth gradient was established prior to and independently of the corresponding gradient for the calcites;" the minor premise consists of the statement, "during descent, supergene solutions acquire their dissolved strontium through ionic exchange reactions with the whole-rock ignimbrites and, at the same time, are involved in (1) the development of authigenic veins, and (2) the resulting transfer of the strontium isotopic signal from the ignimbrites to the authigenic calcites"; the resulting conclusion may be represented by the statement, "because the late micritic calcites yield a $^{87}\text{Sr}/^{86}\text{Sr}$ ratio vs. depth gradient which is equivalent to that obtained from the whole-sample ignimbrites, these calcites are of supergene origin." In contrast to the previous cases, the strontium-isotope syllogism can not be rejected solely on epistemological grounds. Its correctness, however, is entirely predicated on the validity of first the major premise and then the minor premise. Particularly strong objections may be raised against the major premise. There are two aspects to these objections. On the one hand, there is no factual basis to support the assumption of sequential development of the strontium isotopic gradients, i.e., first for the whole-rock ignimbrites and then for the corresponding calcites. On the other hand, a combined consideration of the distributions of both the chemical and temporal characteristics of alteration products (whole-rock ignimbrites and clinoptilolites) and the isotopic

characteristics of the corresponding calcites, as described subsequently, reveals that the alteration overprinting is spatially correlative with both of the strontium isotopic gradients. This observation clearly and unequivocally indicates that, rather than being established sequentially, the strontium isotopic gradients were developed concurrently instead. With this information at hand, one is left with little choice but to reject the major premise, and with it the entire strontium isotope syllogism constructed by Peterman et al. (1991 and 1992).

Having rejected all four of the USGS syllogisms as demonstrably false, it is necessary to undertake in-depth and independent re-examination of the isotopic data on which these syllogisms are based. The main purpose of this re-examination is to determine whether or not these data are either consistent with or require a hypogene origin for both the Yucca Mountain calcretes and the compositionally, texturally, and isotopically equivalent veins. The re-examination is presented in five parts. Section 8.2 is concerned with the uranium, oxygen, and carbon isotopic data. The origin of the Yucca Mountain calcites is examined based on both comparative isotopic analyses and paleo-geothermal reconstructions. For the local calcites, interpretation of paragenetic relationships is discussed in Section 8.3. These relationships are interpreted on the basis of combined consideration of five different sets of data. These are: (1) spatial distribution of the alteration minerals; (2) K/Ar ages of clinoptilolites; (3) structural settings of the calcites in question; (4) isotopic characteristics of oxygen and carbon

incorporated in these calcites; and (5) homogenization temperatures of fluid inclusions contained in spatially associated calcites. Section 8.4 deals with the strontium isotopic data. These data are used to further test both the independently inferred origin of the Yucca Mountain calcites and the interpreted paragenetic relationships. The uranium-thorium ages of the late micritic calcites are presented and considered in Section 8.5. These data, together with K/Ar and $^{40}\text{Ar}/^{39}\text{Ar}$ laser fusion ages of the youngest local volcanics, are used to verify the independently inferred origin of carbon incorporated in the late micritic calcites. The overall conclusions are summarized in Section 8.6.

8.2 The Origin of the Yucca Mountain Calcites, Based on Comparative Isotopic Analyses and Paleo-geothermal Reconstructions

For the Yucca Mountain calcites, the currently available uranium, oxygen, and carbon isotopic data are presented in Figures 8-1 through 8-4b. The uranium concentrations and values of $^{234}\text{U}/^{238}\text{U}$, $^{230}\text{Th}/^{232}\text{Th}$, and $^{230}\text{Th}/^{234}\text{U}$ activity ratios, from samples of calcretes and equivalent surficial veins, are summarized in Figures 8-1a through 8-1c. For the vadose zone veins, the corresponding U-series data are presented in Figures 8-2a through 8-2c. The relative concentrations of carbon-13 and oxygen-18 (values of the $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ ratios) from samples of calcretes and equivalent surficial veins are summarized in Figure 8-3. For both the vadose zone calcites and the "saturated" zone calcites, the corresponding stable

isotope data are presented in Figures 8-4a and 8-4b. These data represent calcites occurring in three different structural settings, namely: (1) in-fillings of lithophysal cavities; (2) interstitial impregnations and replacements of phenocrysts; and (3) discrete veins and veinlets.

For deposits elsewhere with known hypogene origin, the matching isotopic data set is presented in Figures 8-5a through 8-9. This set will be used for the purposes of performing comparative isotopic analyses of the origin of the Yucca Mountain calcites.

Figures 8-5a and 8-5b present the U-series data for the Amargosa Basin-Furnace Creek Wash travertine veins. These veins are considered to be appropriate U-series analogs because, similar to the late Yucca Mountain calcites, they occur above the water table. In this setting, calcitic veins are not armored by the ongoing deposition of calcium carbonate, and are exposed to the leaching influence of rainwater. Intermittent flushings by infiltrating rainwater may lead to preferential removal of ^{234}U atoms, resulting in the lowering of the actual value of the $^{234}\text{U}/^{238}\text{U}$ activity ratio. This may occur because, relative to the parent ^{238}U atoms, the radiogenic ^{234}U atoms are more vulnerable to leaching. This relative vulnerability is a consequence of the $^{238}\text{U} \rightarrow ^{234}\text{U}$ nuclear fission, which involves alpha particle emission. The ejection of alpha particles has the effect of damaging the crystal lattice around the former ^{238}U atom. The resulting daughter nuclide

$^{234}\text{Th} \rightarrow ^{234}\text{U}$ occupies the radiation-damaged site which, consequently, is more accessible to rainwater. Both the travertine veins and the Yucca Mountain calcites, however, are equally vulnerable to the preferential mobilization of ^{234}U atoms by infiltrating rainwater. For this reason, the Amargosa Basin-Furnace Creek Wash travertine veins were selected for U-series comparative analysis.

Figures 8-6 and 8-7 present values of the $\delta^{18}\text{O}$ ratio for the Amargosa Basin spring-marsh deposits and carbonate gangue associated with the Carlin and Cortez gold deposits of Nevada, respectively. For the purposes of oxygen-18 comparative analysis, both of these deposits are regarded as appropriate analogs for the following three reasons. First, all three occur in analogous orographic settings. Consequently, it may be presumed that, at a time of atmospheric condensation, the parent fluids for these deposits were carrying somewhat analogous relative concentrations of both deuterium and oxygen-18. In other words, relative to the meteoric water line, the respective fluids could have occupied somewhat equivalent positions. Second, all three deposits are associated with geothermal systems that have (1) low to moderate temperatures, and (2) fairly high values of the fluid/rock ratio. Under such conditions, the magnitude of the so-called "oxygen isotopic shift" is fairly small, see, for example, Clayton and Steiner (1975) and Lambert and Epstein (1980). Consequently, it may be presumed that, for all of the deposits considered, isotopic modifications of the initial compositions (resulting from rock $\leftrightarrow$ fluid exchange reactions) were small

and somewhat analogous. Third, for both the Amargosa Basin - Yucca Mountain surficial deposits and the oxygen-18 enriched Carlin-Cortez gangue deposits, it may be presumed that the respective calcite depositional environments were somewhat equivalent. Under these conditions, the magnitudes of both oxygen isotopic fractionation effects and oxygen-18 evaporative enrichments could have been somewhat analogous.

Figures 8-8 and 8-9 present values of the $\delta^{13}\text{C}$ ratio for carbonate gangue associated with various hydrothermal ore deposits. For the purpose of the carbon isotopic comparative analysis, both of these data sets are regarded as representing appropriate analogs for the Yucca Mountain calcites. This is so because in terms of the relative concentrations of carbon-13, the parent fluids for these analogs are equivalent to contemporary chemically mature fluids from the Nevada Test Site area. For the latter fluids, values of the $\delta^{13}\text{C}$ ratio range from -2.3 to about -9.0 per mil_{PDB}, see, for example, Winograd and Pearson (1976), Claassen (1985), Benson and McKinley (1985), and White and Chuma (1987). However, relict fluids, entrapped in fluid inclusions from hydrothermal ore deposits, carry values of the $\delta^{13}\text{C}$ ratio that commonly range from -3.0 to about -10.0 per mil_{PDB}, Figure 8-8. Similarly equivalent are values of the $\delta^{13}\text{C}$ ratio for geothermal fluids from the Long Valley Caldera area. As shown in Figure 8-9, for cooled and CO_2 degassed fluids, these values range from -1.5 to -9.7 per mil_{PDB}; for deeper hydrothermal fluids, these values are about -4.9 per mil_{PDB}.

Auxiliary carbon isotopic data are presented in Figures 8-10a through 8-10c. These auxiliary data are analyzed to derive best estimates of the isotopic character of carbon which may reasonably be expected to be dissolved in fluids that, at Yucca Mountain, could be involved in hypothetical accumulations of supergene carbonates. These estimates, in turn, are used to evaluate whether or not the isotopic character of carbon incorporated in the Yucca Mountain calcites is consistent with the proposed supergene origin of these calcites. For hypothetical supergene solutions (those that could be involved in a primary deposition of supergene carbonates), however, reliable estimates of values of the $\delta^{13}\text{C}$ ratio are difficult to obtain. This is so because, at Yucca Mountain, neither these fluids nor their solid derivatives may be identified with confidence. To obtain the desired estimate, therefore, there is no choice but to rely on indirect inferences. In Figures 8-10a through 8-10c, five such inferences are considered.

The first inference is based on the assumption that the parent fluids for hypothetical supergene carbonates acquire their carbon solely through dissolution of the locally produced biogenic CO_2 , Figure 8-10a. For this gas, the estimated value of the $\delta^{13}\text{C}$ ratio is based on the results of a plant survey (i.e., observed proportion of plants with the C-3 metabolic pathway), as performed and reported for the Yucca Mountain area. For the dissolved carbonate, the estimated value of the $\delta^{13}\text{C}$ ratio was calculated assuming that (1) the biogenic CO_2 dissolves at an ambient temperature of $\sim 15^\circ$ Celsius, and (2) during residence in the soil horizon,

the biogenic CO₂ does not undergo any isotopic modification.

The second inference is based on the assumption that the parent fluids for hypothetical supergene carbonates acquire their carbon through dissolution of CO₂ gas that is known to be residing in the vadose zone, Figure 8-10b. For this gas, the estimated range of values of the $\delta^{13}\text{C}$ ratio represents means of values observed during three different CO₂ surveys. One of these surveys was performed at Yucca Mountain. For the dissolved carbonate, the estimated value of the $\delta^{13}\text{C}$ ratio was calculated assuming that the vadose zone CO₂ dissolves at a temperature of 15° Celsius.

The third inference is based on the assumption that, in terms of the isotopic character of dissolved carbon, the parent fluids for hypothetical supergene carbonates are similar to local contemporary fluids occurring immediately below the topographic surface, Figure 8-10c. Such fluids were sampled in the Amargosa Narrows area and in the Yucca Mountain well UE-29a#2, at a depth ranging from 1 to 29 m. In both of these instances, the observed proportions of carbon-14 (PMC) are very high, in excess of 60 percent of the modern carbon standard. It is likely, therefore, that the $\delta^{13}\text{C}$ ratios from these evidently young and presumably immature fluids are conservatively representative of the corresponding ratios for local infiltrating rainwater.

The fourth inference is based on the assumption that, in terms of the isotopic character of dissolved carbon, the parent fluids for hypothetical supergene carbonates are similar to interstitial fluids residing in the Yucca Mountain vadose zone, Figure 8-10c. Samples of such fluids were centrifuged from rock cores extracted in boreholes UE-25 UZ-4 and UZ-5, Figure 5-2b. From Section 5.0 it may be recalled that, at Yucca Mountain, the vadose zone interstitial fluids are being contaminated with fairly young meteoric fluids. The strongly negative values of the $\delta^{13}\text{C}$ ratio observed for the composite solutions uniquely indicate that CO_2 dissolved in the contaminating fluids originated exclusively from biogenic sources. It is possible, therefore, that the $\delta^{13}\text{C}$ ratios for the latter fluids are the most representative of the isotopic character of carbon dissolved in the local infiltrating rainwater.

The fifth and last inference is based on the assumption that the parent fluids for hypothetical supergene carbonate veins acquire their carbon through dissolution of pre-existing soil carbonates, whether supergene or hypogene, Figure 8-10c. For these fluids, the estimated range of values of the $\delta^{13}\text{C}$ ratio is based on the results of laboratory soil leaching experiments. Soil samples used in these experiments were obtained from a number of sites situated within the Nevada Test Site proper.

For the Yucca Mountain vadose zone, the results of paleo-geothermal

reconstructions are presented in Figures 8-11a through 8-11d. These reconstructions are based on the $\delta^{18}\text{O}$ vs. depth gradient, constructed from the results of isotopic analyses of samples of late micritic calcites. They were performed selectively, considering only samples that, locally, are the most enriched in oxygen-18. Presumably, for these samples, the oxygen isotopic fractionation effects attained their relative maximum, and the corresponding calcite precipitation temperatures were at their relative minimum. It is likely, therefore, that the reconstructed geothermal gradients represent the mildest geothermal conditions prevailing during the formation of the Yucca Mountain veins. A reliable reconstruction of these minimum conditions is an important element in resolving the dilemma that surrounds the origin of these veins. Specifically, a reliable demonstration that, in the vadose zone, the geothermal gradients undergo fluctuations, with amplitudes of say 10-20° Celsius per 1 km of depth, amounts to a fairly definitive demonstration of two important points. These are: (1) intermittently, warm hypogene fluids ascend into the vadose zone, and (2) the epigenetic calcites, together with the metasomatic zeolites (Section 7.0) and the young thermally reset zircons (Section 6.0), record such ascents.

Based on the $\delta^{18}\text{O}$ vs. depth gradient alone, however, the desired paleo-geothermal reconstructions are difficult to perform. Although these reconstructions may be made in relative terms, thus eliminating the need for assuming the oxygen-18 composition of the parent fluids, they do require two

additional assumptions. The first assumption is that, for the spatially distinct calcite specimens, the parent fluids were carrying $\delta^{18}\text{O}$ ratios which either were the same or the $\delta^{18}\text{O}$ variability is both known and constant. The second assumption is that, during formation of the specimens considered, non-equilibrium fractionation effects were either absent or, relative to the equilibrium fractionation curve, have maintained a known relationship. However, for the parent fluids of the specimens considered, on the basis of currently available data alone it is not possible to know exactly what was the corresponding variability of the oxygen-18 content. At least three observations complicate this undertaking. First, at Yucca Mountain, contemporary subsurface fluids exhibit a $\delta^{18}\text{O}$ spatial variability of about 1.5 per mil_{SMOW}, Benson and McKinley (1985). This variability is known on the basis of bulk fluid samples, pumped from fairly large segments of boreholes. Because of possible fluid mixing and the resulting sampling "smoothing," the actual variability may be larger than reported. Second, for observation periods of a few years, and for a given observation station, $\delta^{18}\text{O}$ fluctuations are known to be as large as 5.0 per mil_{SMOW}, Lyles et al. (1990). Third, for observation periods of a few hundred thousands of years, the actual known $\delta^{18}\text{O}$ fluctuations range from 2.7 to about 4.0 per mil_{SMOW}, Winograd et al. (1988). Similarly uncertain are the isotopic fractionation effects. In view of substantial errors that potentially may be involved, the paleo-geothermal reconstructions were made considering three scenarios.

The first scenario is based on the assumption that non-equilibrium fractionation effects were either absent or depth-invariant. For a constant depth, the observed $\delta^{18}\text{O}$ variability of about 3.0 per mil_{SMOW} (Figures 8-11a and 8-11b) was regarded as reflecting the $\delta^{18}\text{O}$ fluctuations in the parent solutions. Both of these assumptions yield low values of the paleo-geothermal gradient, and the resulting reconstruction is considered the most conservative and reliable.

The second scenario is based on the assumption that, for the shallow specimens, the combined (non-equilibrium plus equilibrium) fractionation effects exceeded the equilibrium effects and the difference was depth-variant, Figure 8-11c. For a constant depth, the observed $\delta^{18}\text{O}$ variability was regarded as expressing (1) $\delta^{18}\text{O}$ fluctuations in the parent solutions, together with (2) variability of the combined fractionation effects. Because it is impossible to separate both of these factors, the resulting reconstruction may be in error.

The third scenario is based on the assumption that, for shallow specimens, the combined fractionation effects were smaller than the equilibrium effects and the difference was depth-variant, Figure 8-11d. Also for this scenario, it is not possible to interpret properly the observed $\delta^{18}\text{O}$ variability, at a given depth. Consequently, the reliability of the corresponding reconstruction may be low, particularly for shallow parts of the vadose zone.

All of the isotopic data that are considered in this section are summarized in Figures 8-12 through 8-18. For the Yucca Mountain subsurface calcites, the depth distributions of values of $^{234}\text{U}/^{238}\text{U}$, $\delta^{13}\text{C}$, and $\delta^{18}\text{O}$ ratios are presented in Figures 8-12, 8-13, and 8-14, respectively. Additionally, these figures show observed ranges of the corresponding isotopic ratios for local calcretes and sinters, as well as for the analogs whose hypogene origin is known unequivocally. Figure 8-15 presents a comparison of the isotopic characters of carbon interpreted and/or inferred to have been dissolved in (1) the parent fluids of the Yucca Mountain calcretes and veins, and (2) the hypothetical Yucca Mountain supergene fluids. The results of paleo-geothermal reconstructions are shown in Figures 8-16 and 8-17. In these figures, two different reconstructions are represented. Figure 8-16 presents a range of values for the local paleo-geothermal gradient. This range has been computed by the USGS investigators, based on the entire set of presently available $\delta^{18}\text{O}$ data. Figure 8-17 shows the paleo-geothermal gradients that were reconstructed for the vadose zone, using only the most oxygen-18 enriched samples. This figure also shows the contemporary geothermal gradient, as measured in borehole USW G-3. Finally, Figure 8-18 presents a relationship between the CO_2 dissolution temperature and the $\text{HCO}_3^- \leftrightarrow \text{CO}_{2(\text{gas})}$ fractionation factor. This relationship is important for understanding the origin of carbon that: (1) typically, is dissolved in hydrothermal fluids, and (2) was dissolved in the parent fluids of the late micritic calcites.

Examination of Figures 8-12 through 8-18 leads to the formulation of the following five observations/interpretations. First, the isotopic characters of uranium, carbon, and oxygen incorporated in samples of the Yucca Mountain calcretes and surficial veins are equivalent to those incorporated in the subsurface calcites. As shown in Figures 8-12 through 8-14, isotopically equivalent calcites are present in three settings, including: (1) the topographic surface; (2) within the vadose zone; and (3) below the contemporary water table. Clearly, in all three settings, the calcites are fairly young and owe their existence to the same carbonatization processes. In addition, there is a population of calcites which is not represented at the topographic surface. For these deeper and evidently older calcites, the relative concentrations of carbon-13 are fairly high; the corresponding values of the $\delta^{13}\text{C}$ ratio range from -2.0 to about +5.0 per mil_{PDB}, Figure 8-13. The observed bi-modality of the carbon isotopic characteristics has led Whelan and Stuckless (1991 and 1992) to believe that, at Yucca Mountain, two genetically distinct generations of calcites are present. The isotopically "heavy" calcites (high values of the $\delta^{13}\text{C}$ ratio) were regarded as representing hypogene carbonatization related to the hydrothermal stage of activity of the Timber Mountain Caldera. The isotopically "light" calcites (low values of the $\delta^{13}\text{C}$ ratio), however, were regarded as representing subsequent supergene-pedogenic carbonatization. Based on the carbon isotopic data alone, however, this interpretation is not unique, and may or may not be correct. An alternative interpretation is that the two identified generations of calcite are expressing two different stages of hypogene

carbonatization. Specifically, it may be presumed that the isotopic bi-modality reflects the independently known overprinting of spatially, chemically, and temporally distinct alteration aureoles, as ascertained independently in Section 7.0.

Second, for all of the Yucca Mountain calcites, the isotopic characters of incorporated uranium, carbon, and oxygen are analogous to those incorporated in the hypogene analogs. For the calcretes and the vadose zone veins, the isotopic character of incorporated uranium is identical to that incorporated in the Amargosa Basin-Furnace Creek Wash travertine veins, Figure 8-12. For all of the locally known calcites, including those occurring at the topographic surface and within the vadose zone, the isotopic character of carbon is identical to that incorporated in: (1) carbonate gangue associated with various hydrothermal ore deposits, and (2) travertines and hydrothermal veins from the Long Valley Caldera, Figure 8-13. Similarly, for all of the locally known calcites, including calcretes and the vadose zone veins, the isotopic character of incorporated oxygen is identical to that incorporated in (1) the Amargosa Basin spring deposits, and (2) the carbonate gangue associated with the Carlin-Cortez disseminated gold deposits, Figure 8-14. The observed close multi-element isotopic affinity indicates that even for the most carbon-13 depleted calcites (values of the $\delta^{13}\text{C}$ ratio ranging from -5.0 to -10.0 per mil_{PDB}, Figure 8-13), a hypogene origin is clearly a permissible interpretation option. As shown in Figures 8-8 and 8-9, hydrothermal

fluids typically carry fairly wide ranges of the $\delta^{13}\text{C}$ ratio, from -1.5 to about -10.0 per mil_{PDB}. For the resulting carbonate deposits, the corresponding range is the same as that observed at Yucca Mountain, from -10.0 to about +5.0 per mil_{PDB}. Hoefs (1987) has explained the observed wide ranges of the $\delta^{13}\text{C}$ ratio by pointing out that, in magmatically active regions, hydrothermal fluids may acquire dissolved carbon from two different sources. During certain periods of hydrothermal activity, the associated fluids acquire their dissolved carbon from marine limestones. Typically, such fluids are enriched in carbon-13, and carry values of the $\delta^{13}\text{C}$ ratio of $\sim 0 \pm 2.0$ per mil_{PDB}. During other periods of hydrothermal activity, however, a deep-seated igneous source of carbon becomes important. In this case, the associated fluids acquire carbon through dissolution of igneous CO_2 . Typically, such fluids are depleted in carbon-13 and carry values of the $\delta^{13}\text{C}$ ratio ranging from, say, -3.0 to as little as -10.0 per mil_{PDB}, Faure (1976).

Third, relative to the isotopic character of carbon expected to be dissolved in local supergene fluids, the isotopic character of carbon interpreted to have been dissolved in the parent fluids of the Yucca Mountain calcites is strongly discordant. This discordance, if considered together with the carbon isotopic affinity with typical hydrothermal fluids, effectively argues against the proposed supergene origin of the calcites. As shown in Figure 8-15, for local supergene fluids, expected values of the $\delta^{13}\text{C}$ ratio fall somewhere within the -10 to -25 per

mil_{PDB} range. For the parent fluids of the Yucca Mountain calcites, however, the corresponding range is from +2.0 to about -10.0 per mil_{PDB}. It should be recognized that this represents a minimum value for the isotopic discord. Two-fold reasoning may be put forth in support of this statement. On the one hand, for local supergene fluids, expected values of the $\delta^{13}\text{C}$ ratio were estimated assuming climatic conditions similar to contemporary conditions. However, it is entirely possible that at Yucca Mountain deposition of both calcretes and equivalent bedrock veins occurred in part during glacial maxima, when the climate was cooler than the contemporary climate. At the time of deposition, the actual climatic conditions could have been analogous to those presently prevailing at altitudes ~750 m higher, in the lower pinyon-juniper-sagebrush zone. It follows then that, during the deposition of the Yucca Mountain calcretes and veins, the local biomass could have contained more plants with the C-3 metabolic pathway (~100 percent against ~85 percent today). Consequently, relative to contemporary circumstances, local biogenically derived CO_2 could have been "lighter" by about 2-3 per mil_{PDB}. Because biogenic CO_2 is considered to be the main source of dissolved carbon, the corresponding paleo-supergene fluids could have been accordingly "lighter." On the other hand, for the parent fluids of the local calcites, the range of values of the $\delta^{13}\text{C}$ ratio was computed assuming, conservatively, that the appropriate value of the $\text{CaCO}_3 \leftrightarrow \text{HCO}_3^-$ fractionation factor ranges from 1.75 to about 3.0 per mil_{PDB}. These values pertain to equilibrium isotopic fractionation occurring at a temperature ranging from 15 to 130° Celsius,

respectively. They are appropriate only for very low rates of carbonate precipitation. Under fast precipitation conditions, however, the actual fractionation factor is much lower, say less than 1.0 per mil_{PDB}, see, for example, Usdowski et al. (1979) and Turi (1986). Because the inferred rates of precipitation of the local micritic calcites are fairly high (Section 3.0), it is possible that, for the parent fluids of these calcites, the actual values of the $\delta^{13}\text{C}$ ratio were about 2.0 per mil_{PDB} "heavier" than those shown in Figure 8-15.

Fourth, the isotopic character of oxygen incorporated in the Yucca Mountain calcites is not consistent with, or supportive of, the proposed supergene origin of these calcites. Such a state of affairs is clearly indicated by a comparison of the paleo-geothermal gradients, reconstructed based on the observed $\delta^{18}\text{O}$ vs. depth gradient, with the contemporary gradients. For hypothetical supergene calcites, the reconstructed paleo-geothermal gradients ought to be equal to the contemporary gradients. This is so because it is totally unreasonable to associate infiltrating rainwater with a noticeable warming of rock through which this rainwater percolates. For ascending hypogene fluids, however, the opposite is true. Even minor heating of rock volumes measured in km³ requires very large amounts of energy and, for natural circumstances, is conceivable only with the involvement of deep-seated hot or warm fluids. As shown in Figure 8-16, the reconstructed paleo-geothermal gradients range from 34 to as much as 140° Celsius/km. For the vadose zone, and based on the most oxygen-18 enriched

specimens (Figures 8-11a through 8-11d), the reconstructed gradients are: (1) the reconstruction by Szabo and Kyser (1989), dT/dz ranging from 17 near the topographic surface to as much as 50° Celsius/km in deeper parts of the vadose zone; (2) reconstruction A, $dT/dz \sim 35^\circ$ Celsius/km; (3) reconstruction B, $dT/dz \sim 33^\circ$ Celsius/km; and (4) reconstruction C, $dT/dz \sim 58^\circ$ Celsius/km. The contemporary gradients are known from downhole measurements of subsurface temperatures. Such measurements were made and reported by Sass et al. (1987). For wells from which the most oxygen-18 enriched specimens were obtained, the corresponding geothermal gradients are: (1) UE-25a#1, $dT/dz \sim 22^\circ$ Celsius/km; (2) USW G-2, $dT/dz \sim 24^\circ$ Celsius/km; (3) USW G-3, $dT/dz \sim 22^\circ$ Celsius/km; and (4) USW G-4, $dT/dz \sim 20^\circ$ Celsius/km. Clearly, for all of the scenarios considered, the reconstructed paleo-geothermal gradients significantly exceed the contemporary gradient, Figure 8-17. The minimum discrepancy is about 50 percent. By any standard of judgment, this is a significant and meaningful discrepancy.

Fifth, as far as the origin of the Yucca Mountain calcites is concerned, the uranium, carbon, and oxygen isotopic data do not provide any factual basis to support the proposed supergene origin of some of the Yucca Mountain calcites. Quite the contrary, three independent lines of evidence may be put forth as arguing against it, and if taken collectively, as effectively nullifying the a priori assumption of supergene origin. These are: (1) the observed uranium, carbon, and oxygen isotopic affinities between hypogene analogs and the Yucca Mountain

calcites; (2) the inferred carbon isotopic discords between hypothetical supergene fluids and parent fluids of the Yucca Mountain calcites; and (3) the interpreted discrepancy between the present geothermal gradient and the paleo-geothermal gradient. In sharp contrast to the proposed supergene origin, the assumption of a hypogene origin leads to a contradiction-free interpretation. This interpretation accounts for all of the known isotopic, textural, and compositional characteristics of the Yucca Mountain calcites. For these calcites, also, the observed bi-modality of the $\delta^{13}\text{C}$ ratios may be accounted for within the context of hypogene origin. In that regard, particularly relevant are interpretations of the origin of carbon circulating in magmatically active regions as proposed, for example, by Hoefs (1987). In such regions, magmatic and related hydrothermal processes do not operate under perpetual steady-state conditions, but evolve and fluctuate instead. During periods of intense activity, for example, both the sources of dissolved carbon and the carbon acquisition depth (temperature) are not the same as those during periods of waning or more subdued activity. These two factors, in turn, profoundly influence the isotopic character of carbon that is dissolved in the respective fluids. On the one hand, carbon derived from different sources is variably enriched in carbon-13. For example, carbon derived from marine limestones is typically isotopically "heavy" and is associated with mean values of the $\delta^{13}\text{C}$ ratio of 0 ± 2.0 per mil_{PDB}; carbon derived from igneous sources, however, is "lighter" and the corresponding values range from -5.0 to -8.0 per mil_{PDB}, Hoefs (1987). On the other hand, the $\text{HCO}_3^- \leftrightarrow \text{CO}_{2(g)}$ fractionation factor is strongly

dependent upon the CO₂ dissolution temperature, or depth. As shown in Figure 8-18, values of this factor range from ~+2.0 (T = 30° Celsius) to about -7.0 per mil_{PDB} (T = 300° Celsius). These general characteristics may be used to construct a plausible explanation for the bi-modal distribution of carbon isotopic characteristics in the Yucca Mountain calcites. Specifically, the observed range of the $\delta^{13}\text{C}$ ratios (from -10.0 to +4.0 per mil_{PDB}, Figure 8-13) may be explained by introducing four assumptions. These are: (1) the carbon-13 enriched calcites record circulation either during subdued stages of hydrothermal activity or during waning stages of igneous activity; (2) the carbon-13 depleted calcites record deeper circulation, either during youthful stages of igneous activity or during invigorated hydrothermal circulation; (3) the parent fluids of the carbon-13 enriched calcites acquired dissolved carbon from local Paleozoic carbonates; and (4) the parent fluids for the carbon-13 depleted calcites acquired dissolved carbon from a local igneous source (the inferred CO₂ dissolution temperature ranges from 200 to 250° Celsius).

8.3 Interpretations of the Paragenetic Relationships

The interpretation of calcite paragenetic relationships developed in this section is based on a variety of isotopic and geochemical data. Collectively, these data were obtained from boreholes USW G-1, G-2, G-3, GU-3, G-4, and UE-25b#1. Locations of these boreholes, relative to the alteration centers identified previously, are

shown in Figure 8-19. In developing the interpreted paragenetic relationships, four data sets discussed previously were considered. These are: (1) the chemical composition of clinoptilolites, Figures 7-8 and 7-14; (2) the K/Ar ages of clinoptilolites, Figures 7-22 and 7-27; (3) the isotopic characters of carbon and oxygen incorporated in the Yucca Mountain calcites, Figure 8-4; and (4) the structural settings of the calcites, Figure 8-4. The paragenetic interpretations were augmented by the available fluid inclusion data. The results of measurements of the homogenization temperatures of fluid inclusions, from calcites encountered in boreholes USW G-2 and G-3/GU-3, are presented in Figure 8-20. For the fluid inclusion sampling sites USW GU3-103 and -429, the observed field relationships are illustrated and summarized in Figures 8-21a through 8-21e. These relationships are helpful in judging the origin of the corresponding calcites.

All of the above data are also presented and summarized in Figures 8-22 through 8-25. Figure 8-22 shows the $\delta^{13}\text{C}$ vs. $\delta^{18}\text{O}$ fields for calcites representing three different settings. These are: (1) in-fillings of lithophysal cavities; (2) interstitial impregnations and replacements of phenocrysts; and (3) discrete veins. For calcites from the northwestern segment of Yucca Mountain (boreholes USW G-2, G-3, and GU-3, Figure 8-19), a comparison of the depth distributions of the fluid inclusion homogenization temperatures with the spatially corresponding values of the $\delta^{13}\text{C}$ ratio is shown in Figure 8-23. This comparison is used to further verify the independently inferred hypogene origin of the carbon-13 depleted calcites.

Figure 8-24 presents a comparison of the depth distributions of: (1) values of the $\delta^{13}\text{C}$ ratio from calcites; (2) mol percentage of Ca + Mg cations from clinoptilolites; and (3) K/Ar ages of clinoptilolites. The data representing the southeastern segment of Yucca Mountain (boreholes USW G-4, UE-25a#1, and -25b#1, Figure 8-19) are shown separately from those representing the northwestern segment (boreholes USW G-1, G-2, and G-3, Figure 8-19). Both of these comparisons are used to determine whether or not the independently evident overprinting of the northwestern alteration aureole, by the subsequent southeastern alteration aureole (Section 7.4, and Figures 7-29 and 7-30) is also expressed through the spatial distributions of the $\delta^{13}\text{C}$ ratios from the epigenetic calcites. Finally, Figure 8-25 summarizes the interpreted paragenetic relationships.

Examination of Figures 8-22 through 8-25 leads to the formulation of the following four observations/interpretations. First, at Yucca Mountain, calcite occurs in three different structural settings, Figure 8-4. These are: (1) in-fillings of lithophysal cavities; (2) interstitial impregnations and replacements of phenocrysts; and (3) discrete veins. Is it possible that, at Yucca Mountain, these different settings represent three different generations of calcite? The in-fillings of lithophysal cavities, for example, may be presumed to represent deuteric carbonatization, developed contemporaneously with the cooling, syn-depositional devitrification and solidification of the welded members of the Paintbrush Tuff. Similarly, the

interstitial impregnations and replacements of phenocrysts may be presumed to record prolonged hydrothermal exposure of the calcite-bearing rocks. The corresponding carbonatization, therefore, may be presumed to record prolonged activity of a hydrothermal system driven by the Timber Mountain Caldera heat source. It may further be presumed that some of the discrete veins are also related to the Timber Mountain Caldera carbonatization. This may be true particularly for the early veins (those that: (1) are associated with barite, fluorite, and pyrite, and (2) are sparry in texture). Slow yet continuous mineral crystallization is a feature to be expected in association with a magmatic center-based system. Such conditions of calcite crystallization promote the development of sparry crystals and, hence, account for the observed sparry textures. One may further speculate that the remaining discrete veins (those that: (1) are associated with opal-CT and sepiolite, and (2) are micritic in texture) record the intermittent activity of a fault-based hydrothermal system driven by conductively replenished heat sources. In association with such systems, the expected rates of mineral crystallization are large and, consequently, the resulting minerals are typically micritic in texture. Based on the combined oxygen and carbon isotopic characteristics of the respective calcites, it should be feasible to either strengthen or weaken the stated paragenetic speculations. Specifically, if these speculations do accurately reflect the actual paragenetic relationships, then one may expect four predictions to materialize. These are: (1) for calcites representing different settings, the corresponding $\delta^{13}\text{C}$ vs. $\delta^{18}\text{O}$ fields ought to

be somewhat distinct; (2) for each setting, the corresponding $\delta^{13}\text{C}$ vs. $\delta^{18}\text{O}$ field ought to be explicable within the context of the presumed paragenesis; (3) the early epigenetic calcites (those that were presumed to record the hydrothermal stage of activity of the Timber Mountain Caldera) ought to be spatially correlative with the Timber Mountain alteration minerals; and (4) the late epigenetic calcites (those that were presumed to be recording intermittent activity of a fault-based hydrothermal system driven by conductively replenished heat sources) ought to be spatially correlative with the late metasomatic (calcic) zeolites.

Second, the combined carbon and oxygen isotopic data indicate that, at Yucca Mountain, calcites represent three clearly distinct isotopic facies, Figure 8-22. The first facies includes the presumed deuteritic in-fillings of lithophysal cavities. The corresponding values of the $\delta^{18}\text{O}$ ratio range narrowly from 14.0 to about 17.0 per mil_{SMOW}; for the $\delta^{13}\text{C}$ ratio, the corresponding values range widely from -8.0 to about +5.0 per mil_{PDB}. Importantly, the observed $\delta^{13}\text{C}$ vs. $\delta^{18}\text{O}$ field is explicable within the context of the presumed deuteritic origin. Two-fold reasoning may be put forth in support of this statement. On the one hand, the observed range of the $\delta^{18}\text{O}$ ratio (from 14.0 to 17.0 per mil_{SMOW}) may be satisfactorily explained by postulating that (1) parent fluids of the deuteritic calcites were isotopically similar to typical magmatic fluids, and (2) calcite precipitation temperatures ranged from 200 to 250° Celsius. For magmatic fluids, the $\delta^{18}\text{O}$ ratio is about +7.0 per mil_{SMOW}, Hoefs (1987). In the 200-250° Celsius temperature

range, the CaCO_3 $\leftrightarrow$ H_2O fractionation factor for oxygen ranges from 9.5 to 7.0 per mil_{SMOW}, respectively. Under these conditions, deuteritic fluids could have precipitated calcites whose $\delta^{18}\text{O}$ values range from 14.0 to 16.5 per mil_{SMOW}. On the other hand, the observed range of the $\delta^{13}\text{C}$ ratio (from -8.0 to +5.0 per mil_{PDB}) may also be satisfactorily explained by postulating that the parent fluids of the deuteritic calcites were associated with two expected characteristics. Specifically, these fluids (1) were carrying igneous carbon with values of the $\delta^{13}\text{C}$ ratio ranging from -8.0 to about -5.0 per mil_{PDB}, Hoefs (1987), and (2) were variably enriched in carbon-13 via the diffusional carbon-13 enrichment mechanism (during CO_2 degassing, the "light" $^{12}\text{CO}_2$ molecules escape more often than the "heavy" $^{13}\text{CO}_2$ molecules and, consequently, the remaining CO_2 is progressively enriched in carbon-13). The second isotopic facies includes the epigenetic calcites which were presumed to be recording prolonged activity of a hydrothermal system driven by the Timber Mountain heat source. These calcites occur in two settings, namely: (1) interstitial impregnations and replacements of phenocrysts, and (2) discrete veins. As shown in Figure 8-22, corresponding values of the $\delta^{18}\text{O}$ ratio range from 4.0 to about 12.0 per mil_{SMOW}; for the $\delta^{13}\text{C}$ ratio, the corresponding values range from -2.0 to about 4.5 per mil_{PDB}. The observed $\delta^{13}\text{C}$ vs. $\delta^{18}\text{O}$ field is again explicable within the context of the presumed association with the Timber Mountain magmatic center. The prolonged thermal exposure (expected near a major magmatic center) may be presumed to have resulted in calcite precipitation temperatures that were generally high, say 100-200° Celsius. For these

temperatures, the CaCO_3 $\leftrightarrow$ H_2O fractionation factor for oxygen ranges from 17.0 to about 8.0 per mil_{SMOW}, respectively. Under these conditions, local meteoric fluids with values of the $\delta^{18}\text{O}$ ratio of about -4.0 per mil_{SMOW} could have precipitated the observed epigenetic calcites. The observed trend toward isotopically "heavy" carbon (with evolution of the Timber Mountain magmatic center, from 13 to 9.5×10^6 years B.P.) may be readily explained by postulating that, during waning stages of activity of the caldera-forming magmatism, the earlier igneous source of carbon was replaced by an inorganic source of carbon, specifically the Paleozoic carbonates. The postulated replacement of the igneous source of carbon by an inorganic source does account for the observed ranges of the $\delta^{13}\text{C}$ ratio (~ 8.0 per mil_{PDB} for the deuteric calcites, $\sim 0 \pm 2.0$ per mil_{PDB} for the Timber Mountain calcites, Figure 8-22). The third and last isotopic facies includes epigenetic calcites which were presumed to be recording intermittent activity of a hydrothermal system driven by conductively replenished heat sources. These calcites occur as discrete veins and, typically, are micritic in texture. As shown in Figure 8-22, the corresponding values of the $\delta^{18}\text{O}$ ratio range from 13.0 to about 20.0 per mil_{SMOW}; for the $\delta^{13}\text{C}$ ratio, the corresponding values range from -3.0 to about -9.5 per mil_{PDB}. For the two sets of epigenetic calcites, differing $\delta^{13}\text{C}$ vs. $\delta^{18}\text{O}$ fields are readily explicable by recognizing the intrinsic characteristics of the respective hydrothermal systems. Relative to the prior magmatic center-based system, the subsequent fault-based system was operating under different circumstances. Specifically, the latter system: (1) was active for

shorter periods of time; (2) is expressed at a shallower depth west of the Stage Coach Road fault; and (3) was associated with greater depth of fluid circulation. The first two circumstances would have meant that, relative to the earlier epigenetic calcites, the later epigenetic calcites were precipitated in a cooler environment. In this case, values of the $\text{CaCO}_3 \leftrightarrow \text{H}_2\text{O}$ fractionation factor for oxygen are higher and, consequently, the corresponding calcites are relatively enriched in oxygen-18 (higher values of the $\delta^{18}\text{O}$ ratio). The third circumstance would have meant that, rather than acquiring dissolved carbon from the Paleozoic carbonates, the corresponding fluids were acquiring carbon through dissolution of deep-seated igneous CO_2 instead. At a temperature of $\sim 200^\circ$ Celsius, dissolution of igneous CO_2 ($\delta^{13}\text{C} \sim -6.0$ per mil_{PDB}) yields fluids with values of the $\delta^{13}\text{C}$ ratio of about -9.0 per mil_{PDB}. In a low pressure environment (near the topographic surface), rapid escape of CO_2 from ascending solutions is typically accompanied by diffusional carbon-13 enrichment and a resulting progressive increase in the $\delta^{13}\text{C}$ ratio. The epigenetic calcites, formed under such non-equilibrium conditions, would carry the observed range of the $\delta^{13}\text{C}$ ratio from -9.0 (low degree of diffusional carbon-13 enrichment) to -3.0 per mil_{PDB} (high degree of the diffusional carbon-13 enrichment). From Section 6.0, it may be recalled that an identical mechanism was devised to explain the observed carbon-13 and oxygen-18 enrichment of the calcretes, relative to both the equivalent veins and the authigenic breccia cement, Figure 6-4. For the authigenic breccia cement, the isotopic character of incorporated carbon is identical to that incorporated in the

least diffusionally enriched calcites. Both of these phases seem to have been precipitated from fluids with the highest values of CO₂ partial pressure and, thus, with the lowest degree of carbon-13 diffusional enrichment.

Third, at Yucca Mountain, both the carbon-13 enriched calcites and the carbon-13 depleted calcites appear to be spatially associated with calcites that yield elevated homogenization temperatures of fluid inclusions. Such a state of affairs is indicated by a comparison of the depth distributions of fluid inclusion homogenization temperatures and the $\delta^{13}\text{C}$ ratios from spatially equivalent samples, Figure 8-23. The currently available data allow for constructing such a comparison only for the northwestern segment of Yucca Mountain (boreholes USW G-2, G-3, and GU-3, Figure 8-19). As shown in Figure 8-23, the carbon-13 enriched calcites may be associated with the fluid inclusion homogenization temperatures ranging from 94 to about 240° Celsius. Importantly, these temperatures (presumably reflecting the calcite precipitation temperatures) are similar to those inferred from the spatially corresponding occurrences of the higher-grade alteration minerals. As shown in Figures 7-18 and 7-19, for borehole USW G-2 the corresponding alteration temperature is about 250° Celsius; for borehole USW G-3, however, the corresponding alteration temperature is 100-130° Celsius. The evident close correspondence of the fluid inclusion homogenization temperatures and the independently known alteration temperatures is of two-fold significance. On the one hand, this correspondence indicates that at Yucca

Mountain fluid inclusion homogenization temperatures do provide an adequate representation of the paleo-geothermal environment. On the other hand, this correspondence indicates that, in the northwestern segment of Yucca Mountain, carbon-13 enriched calcites were formed contemporaneously with the higher-grade alteration minerals, and thus confirms the conclusion drawn previously that these calcites are related to the Timber Mountain hydrothermal metamorphism. As further shown in Figure 8-23, some of the shallower and carbon-13 depleted calcites may also be associated with fluid inclusion homogenization temperatures ranging from 101 to as much as 227° Celsius. This association seems to confirm the independently-inferred hypogene origin of the carbon-13 depleted calcites. Bish (1989) speculated that "*the higher temperature inclusions in the upper part of USW GU-3 probably formed during the initial deposition or cooling of the tuff.*" It should be recognized, however, that there is no factual basis to support this speculation. Quite the contrary, at the fluid inclusion sampling sites USW GU3-103 and -429, the observed field relationships indicate that the corresponding minerals are clearly epigenetic, i.e., formed after "*the initial deposition or cooling of the tuff.*" The following reasoning is put forth in support of this statement. As shown in Figures 8-21a through 8-21d, both of the sampling sites are associated with a hydrothermally altered fault zone. The observed alteration aureole is centered and narrowly restricted to the fault zone itself. Importantly, both the alteration aureole and the fluid inclusion sampling sites occur within three different stratigraphic members of the Paintbrush Tuff. These are: (1) the Tiva

Canyon Member, hosting the fluid inclusion sample USW GU3-103, Figure 8-21a; (2) the Pah Canyon Member, comprising the oxidized hanging wall of the host fault for the sample USW GU3-429, Figure 8-21b; and (3) the Topopah Spring Member, comprising the bleached footwall of this fault, Figure 8-21c. Both the sharply-expressed alteration gradient and multiple transgressions of the stratigraphic boundaries are not features that are attributable to syn-depositional deuteric processes. Instead, these features are commonly regarded as indicative of subsequent epigenetic processes. Bish (1989) further rejected an epigenetic origin of the shallow USW GU-3 samples by pointing out the local absence of higher-grade alteration clays. This, however, is hardly a convincing argument. Two-fold reasoning is put forth in support of this statement. On the one hand, it may be presumed that the parent fluids for both of the shallow USW GU-3 samples have carried high relative concentrations of dissolved alkali earth elements. Such fluids tend to inhibit the smectite --> illite transformations, Section 7.2. On the other hand, development of the higher-grade alteration minerals requires both temperatures above 100° Celsius and a prolonged period of hydrothermal exposure. Near the topographic surface, however, hot ascending fluids may not maintain above-boiling temperatures for the periods of time required for development of the higher-grade alteration minerals.

Fourth, as expected, the independently-known overprinting of the northwestern alteration aureole, by the subsequent southeastern alteration aureole, is clearly

reflected through the observed spatial distribution of the isotopic characters of carbon incorporated in the epigenetic calcites. Such a state of affairs is clearly indicated by comparison of the depth distributions of: (1) $\delta^{13}\text{C}$ values from the epigenetic calcites; (2) chemical composition of clinoptilolites; and (3) K/Ar ages of clinoptilolites. For the northwestern sector of Yucca Mountain (boreholes USW G-1, G-2, G-3, and GU-3, Figure 8-19), this comparison is shown in Figure 8-24a; for the southeastern sector (boreholes USW G-4, UE-25a#1, and -25b#1, Figure 8-19), this comparison is shown in Figure 8-24b. From both of these figures it may be observed that the older and predominantly sodic-potassic clinoptilolites are spatially correlative with the carbon-13 enriched calcites. The K/Ar ages of the associated clinoptilolites further confirm the conclusion drawn earlier that these calcites represent epigenetic carbonatization related to the prolonged activity of a hydrothermal system driven by the Timber Mountain heat source. The subsequent and strongly metasomatic clinoptilolites (fairly large mol percentage of Ca + Mg cations), however, are spatially correlative with the carbon-13 depleted calcites. The K/Ar ages of the associated clinoptilolites support the presumption that these calcites represent epigenetic carbonatization related to intermittent activity of a fault-based hydrothermal system driven by conductively replenished heat sources. From Figures 8-24a and 8-24b, two additional results may be discerned. On the one hand, across Yucca Mountain, the depth of occurrence of carbon-13 depleted calcites appears to display important trends. For the northwestern segment (boreholes USW G-2, G-3, and GU-3, Figures 8-4 and 8-19),

these calcites are known to occur within the vadose zone, some 80 to 400 meters above the contemporary water table. For the southeastern segment (borehole USW G-4, Figures 8-4 and 8-19), however, these calcites are known to be present throughout the 520 m thick vadose zone, as well as throughout the uppermost 320 m of the "saturated" zone. For the carbon-13 depleted calcites, the easterly increasing depth of occurrence (in the direction of the Stage Coach Road fault, Figure 7-20) is important. It indicates that (1) the development of carbon-13 depleted calcites is not restricted to the vadose zone, and (2) the development of these calcites appears to be conformable with the independently-known distribution of the calcic zeolitization (Figures 7-14a and 7-14b). These observations may be taken as confirming both the independently inferred hypogene origin of the carbon-13 depleted calcites and the expected concurrent development of metasomatic zeolitization with associated hypogene carbonatization. On the other hand, at Yucca Mountain, there is a set of calcitic veins whose age remains uncertain. In Figure 8-24b, this set is designated as the "vein set?" The absence of chemical and radiometric data for spatially associated alteration minerals does not allow unequivocal resolution of this issue. Considering that the corresponding calcites (depth 942-1216 m in borehole UE-25b#1, Figure 8-4) are strongly enriched in carbon-13, one is tempted to associate these calcites with the Timber Mountain carbonatization. Considering that the corresponding calcites are accompanied by strongly metasomatic clinoptilolites (Figure 7-14a), however, one is also tempted to associate these

calcites with the subsequent carbonatization. In either case, it is certain that the parent fluids for the carbon-13 enriched calcites have acquired their dissolved carbon from the nearby Paleozoic carbonates. The precipitation of these veins could have occurred during early stages of activity of a fault-based hydrothermal system, when circulation of fluids was confined to the Paleozoic carbonates. It is also possible, however, that this precipitation occurred later, during periods of subdued activity of the fault-based hydrothermal system. During periods of thermal source depletion (subdued activity), carbon derived from a local igneous source is partially trapped in a deep substratum and, consequently, relatively small amounts of it are dissolved in shallower fluids. Temporally, the Paleozoic carbonates become the dominating source of carbon dissolved in these fluids. During subsequent periods of invigorated activity, however, the dominance of the igneous source of carbon is re-established. Greatly enhanced upward flux of fluids from the deep substratum is responsible for that.

All four of the above observations/interpretations are mutually reinforcing. Collectively, they support the speculations stated earlier regarding paragenetic relationships of the Yucca Mountain calcites. The resulting comprehensive and paradox-free interpretation of these relationships is summarized in Figure 8-25. This figure emphasizes that during the last 13.5×10^6 years B.P., Yucca Mountain ignimbrites have been carbonatized in association with three different hypogene processes. For the purposes of this report, the earlier stages of carbonatization

(both the deuteritic carbonatization and the Timber Mountain carbonatization) are of relatively minor importance. In that regard, much more important is the subsequent carbonatization. Based on the K/Ar ages of spatially and chemically corresponding clinoptilolites, it is clearly evident that this carbonatization was initiated about 8.5×10^6 years B.P. Intermittently, this carbonatization has continued until at least 2.0×10^6 years B.P. Both the metasomatic or calcic zeolitization and the carbon-13 depleted carbonatization are directly attributable to the intermittent activity of a fault-based hydrothermal system driven by conductively replenished heat sources. Further verification of this already secure interpretation is provided by the fact that both the carbon-13 depleted calcites and the calcic alteration minerals (borehole UE-25a#1 at a depth of 1200-1400 feet, Figures 8-27b and 8-31) contain abnormally high relative concentrations of radiogenic strontium-87, Section 8.4. For fluids associated with the intermittently active hydrothermal system, the inferred chemical and isotopic characteristics (which are: (1) alkali earth element bulk composition; (2) carbon-13 depletion; and (3) strontium-87 enrichment) could have been acquired only in a deep substratum that includes the local Pre-cambrian basement. From Section 5.0 it may be recalled that, although isotopically modified, strongly allogenic fluids reside in the pore space of the Yucca Mountain vadose zone. Remarkable characteristics associated with these fluids are: (1) alkali earth element bulk composition; (2) elevated concentrations of base and noble metals; and (3) relative HREE enrichment. Collectively, these three characteristics are explicable by assuming

that the fluids evolved deep within the crust in a CO₂-rich environment. During ascent, such fluids could have brought in the thermally reset zircons and could have caused the intermittent explosive fragmentation of bedrock, Section 6.0. During intermittent ascents, these fluids could have also caused: (1) metasomatic zeolitization; (2) alkali earth metasomatism; and (3) late carbonatization, including the surficial or pedogenic aspects of it.

8.4 Combined Carbon and Strontium Isotopes as a Test of the Interpreted Paragenetic Relationships

There are three essential elements of the paragenetic relationships that were interpreted to account for the observed characteristics of the Yucca Mountain hydrothermal metamorphic facies (whole-rock alteration, zeolites, and epigenetic calcites). First, the spatial distribution of the whole-rock alteration and the corresponding zeolites is the result of overprinting of the Timber Mountain alteration aureole by a subsequent fault-based, strongly metasomatic aureole. Most probably, the latter is rooted in the Stage Coach Road fault zone. Second, the spatial distribution of the isotopic character of carbon incorporated in the epigenetic calcites does reflect the independently-known alteration overprinting. Third, for both of the identified isotopic and textural varieties of epigenetic calcite, differences in relative concentrations of carbon-13 are directly attributable to differences in depth of fluid circulation, with resulting substitutions of carbon

sources. All three of these elements will be tested using the combined carbon-strontium isotopic data set. Jointly, this set represents three different substances, specifically: (1) contemporary fluids; (2) epigenetic calcites; and (3) whole-rock altered ignimbrites.

For the purpose of the envisaged test, the strontium-carbon isotopic combination is particularly suitable. This is true because of the following three reasons.

First, the respective isotopic data base is sufficiently broad. It allows for estimating the isotopic characters of carbon and strontium that: (1) are dissolved in local contemporary fluids, with differing degrees of chemical and isotopic maturity and with different bulk compositions (both calcium-magnesium and sodium-potassium); (2) are incorporated in the whole-rock altered ignimbrites (approximate ages of alteration are known based on the K/Ar data); (3) are incorporated in both of the identified isotopic and textural varieties of epigenetic calcite (approximate ages of crystallization may be inferred from their spatial association with K/Ar-dated alteration minerals); (4) are provided by local sources of carbon; and (5) are incorporated in local reservoirs of strontium.

Second, based on the isotopic characters of carbon and strontium incorporated in a precipitate, it is possible to estimate both the $\delta^{13}\text{C}$ ratio and the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio for the parent fluid. This opportunity is the result of two isotopic fractionation

effects. On the one hand, for isotopes of carbon, the $\text{CaCO}_3 \leftrightarrow \text{HCO}_3^-$ fractionation effects are fairly small. For example, in the case of equilibrium fractionation at a temperature of less than 100° Celsius, these effects are lower than 3.0 per mil_{PDB}, Hoefs (1987). For disequilibrium fractionation, however, these effects are almost non-existent. Usdowski et al. (1979), for example, have demonstrated that the kinetic fractionation effects (associated with high rates of CaCO_3 precipitation) cause carbon-13 enrichment in solid CaCO_3 (relative to HCO_3^- in solution) to be very small, less than 1.0 per mil_{PDB}. On the other hand, during common geochemical reactions, strontium isotopes do not fractionate. Consequently, the isotopic character of strontium initially incorporated in a solid compound is identical to that dissolved in the corresponding parent fluid. Furthermore, for most carbonates, the in-situ production of radiogenic strontium-87 is negligible. Thus, the actual value of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio adequately represents the corresponding initial value. This is so because rubidium-87 atoms, which are parent atoms for strontium-87, are too large to be incorporated in the calcite lattice. The oversized ^{87}Rb atoms are excluded from this lattice and, consequently, the in-situ $^{87}\text{Rb} \rightarrow ^{87}\text{Sr}$ production is small or nil, Raukama (1954).

Third, under favorable circumstances, depth of circulation may be inferred from the isotopic character of strontium dissolved in a fluid. Such circumstances may reasonably be expected to be present around Yucca Mountain. The following reasoning is put forth in support of this statement. In the Yucca Mountain area,

there are three litho-stratigraphic complexes that could function as primary reservoirs of strontium dissolved in local hypogene fluids. These are: (1) the Tertiary ignimbritic complex; (2) the Paleozoic carbonate complex; and (3) the Pre-cambrian basement. Importantly, the isotopic character of strontium incorporated in the former two complexes may be markedly different from that incorporated in the underlying Pre-cambrian basement. For the ignimbrites altered during the Timber Mountain hydrothermal metamorphism (bulk of the Tertiary aquifer), present-day values of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio range from 0.7083 to 0.7117 and the corresponding initial values range from 0.7082 to 0.7111, Peterman (1990). Furthermore, it is unlikely that, in the pristine or unaltered state, each of the local ignimbrites contained values of the $^{87}\text{Sr}/^{86}\text{Sr}$ that are significantly higher than 0.7100. Such low relative concentrations of radiogenic strontium-87 are commonly found in association with young rhyolites and compositionally equivalent granites, Faure (1986). For rhyolitic magma from the Yellowstone Park area, for example, Leeman et al. (1977) estimated the value of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio to be 0.7094. Similarly low values of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio are evident in the results of isotopic analyses of Quaternary rhyolites from Long Valley Caldera. Goff et al. (1990), for example, have reported that these values range from 0.7060 to about 0.7074. Interestingly, almost identical values were reported for the Mesozoic batholiths of California, see, for example, Kiestler and Peterman (1973). For southeastern California (near the Nevada border), a representative value of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio is about 0.7070, right in the middle of the range from the much

younger rhyolites. In terms of bulk composition, the crust of the Yucca Mountain region is analogous to that of southeastern California. There is no reason to expect that, in terms of strontium isotopic characteristics, silicic magmas generated at the two locations are significantly different. Typically, for marine limestones (such as those comprising the local Paleozoic carbonate complex) representative values of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio are quite low. This is partly because rubidium concentrations in sea water are very low. Also, during deposition of limestones, complete separation of Sr atoms from Rb atoms takes place. Strontium commonly replaces calcium; rubidium is not incorporated at all. For limestones of Silurian age, for example, Faure (1986) gives a value of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of 0.7084 ± 0.004 . Similarly low values are evident in the results of isotopic analyses of samples of Paleozoic carbonates in the Yucca Mountain region. For these carbonates, the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio ranges from 0.7082 to no more than 0.7099, Peterman (1990). For the Pre-cambrian basement, however, the expected isotopic character of strontium is markedly different from that incorporated in the overlying complexes. Typically, these rocks are associated with both very long ($\sim 10^9$ years) intracrustal residence times and elevated concentrations of rubidium. Both of these factors cause the rate of in-situ $^{87}\text{Rb} \rightarrow ^{87}\text{Sr}$ production as well as ^{87}Sr accumulation times to be correspondingly high. For various Pre-cambrian rocks, the observed values of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio are typically in excess 0.7120 and may be as high as 0.7360, or more, Faure (1986). For the Nevada Test Site Pre-cambrian rocks, the value expected for the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio may be inferred from

the results of a few analyses that are available from southeastern California. Here, the Pre-cambrian rocks and their sedimentary derivatives are associated with values of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio ranging from 0.7112 to as much as 0.7220, Peterman et al. (1970) and Goff et al. (1990). Based on four specimens, the mean value is 0.7168. Strong enhancement of the strontium isotopic signal, which is inferred to be associated with the Nevada Test Site Pre-cambrian basement, is important. This is so because this enhancement allows for inferences to be drawn with regard to depth of circulation of fluids, both past and present. Specifically, fluids involved in circulation restricted to the upper litho-stratigraphic complexes (both the Tertiary ignimbrites and the underlying Paleozoic carbonates) may be expected to carry low values of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio, say, ranging narrowly from 0.7082 to about 0.7110. For fluids involved in deeper circulation, however, the corresponding isotopic signal is decisively stronger, say, in excess of 0.7120.

In view of the above general considerations, the paragenetic relationships interpreted for the Yucca Mountain epigenetic calcites assume a testable form. Specifically, if the interpreted paragenetic relationships do accurately reflect the actual relationships, then the following four expectations ought to materialize.

First, for allogenic fluids associated with the Timber Mountain hydrothermal metamorphism, the isotopic characters of dissolved carbon and strontium ought to be distinct from those dissolved in allogenic fluids associated with the subsequent

hydrothermal metamorphism. This is so because, within the context of the tested interpretation, the respective fluids are regarded as having acquired dissolved carbon and strontium from isotopically different sources and reservoirs.

Specifically, relative to the parent fluids of the Timber Mountain minerals, the parent fluids of the subsequent minerals underwent deeper circulation; these fluids were tapping (1) a more carbon-13 depleted igneous source, and (2) a more radiogenic reservoir of strontium in the Pre-cambrian basement. Consequently, for the late allogenic fluids, the observed carbon-13 depletion ought to be accompanied by strontium-87 enrichment. For the Timber Mountain fluids, however, the observed carbon-13 enrichment ought to be accompanied by strontium-87 depletion.

Second, for contemporary mature calcium-magnesium fluids, the isotopic characters of dissolved carbon and strontium ought to be distinct from those incorporated in the Paleozoic carbonates. Specifically, relative to these carbonates, contemporary calcium-magnesium fluids ought to be both enriched in strontium-87 and depleted in carbon-13. This is so because, within the context of the tested interpretation, contemporary calcium-magnesium fluids are not regarded as having been acquiring dissolved carbon and strontium solely through ionic and isotopic exchange reactions with the host carbonates. The inferred increased depth of circulations had the result that, during Plio-Quaternary time (age of the alteration minerals and associated epigenetic calcites), the calcium-magnesium

fluids were and still are being contaminated by yet deeper allogenic fluids which carry dissolved strontium and carbon from the Pre-cambrian basement and local igneous sources, respectively.

Third, in contrast to contemporary calcium-magnesium fluids, early allogenic paleo-fluids (those that were involved in the Timber Mountain hydrothermal metamorphism) ought to display clear isotopic affinity with the Paleozoic carbonates. This is so because, within the context of the tested interpretation, fluids involved in the Timber Mountain hydrothermal metamorphism are regarded as have experienced shallower circulation, i.e., above the Pre-cambrian basement. Both the shallower depth of circulation and the steadily decreasing influence of the caldera-forming magmatism had the result that, at the end of Miocene time ($9.5-10.5 \times 10^6$ years B.P.), the fluids were acquiring dissolved carbon and strontium solely from the Paleozoic carbonates.

Fourth, for the subsequent allogenic fluids (those that were involved in both the calcic zeolitization and the corresponding carbon-13 depleted carbonatization), the isotopic characters of dissolved carbon and strontium ought to be analogous to those for the contemporary calcium-magnesium fluids. This is so because within the context of the tested interpretation, both of these fluids are regarded as having the same sources and reservoirs of carbon and strontium, respectively.

The available isotopic data to be used for the purpose of the envisaged test are presented in Figures 8-26 through 8-30. The isotopic character of indigenous strontium that has been and remains incorporated in locally important reservoirs may be estimated from the isotopic data presented in Figure 8-26. Figures 8-27a through 8-27c present the results of strontium analyses of whole-rock samples of the Yucca Mountain ignimbrites. For the whole-rock altered ignimbrites, these data will be used to illustrate depth distribution of both the total concentration of strontium and the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio. For the same stratigraphic ignimbrites, these data will also be used to evaluate lateral changes in the total concentration of strontium. Figures 8-28a and 8-28b present data on the isotopic character of strontium incorporated in the Yucca Mountain epigenetic calcites. For both the carbon-13 enriched calcites and the carbon-13 depleted calcites, these data will be used to establish corresponding values of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio. In combination with the observed relative concentrations of carbon-13, these ratios will be used to infer the $\delta^{13}\text{C}$ vs. $^{87}\text{Sr}/^{86}\text{Sr}$ field for the parent fluids. Figures 8-29a through 8-29d present the results of carbon and strontium isotopic analyses of samples of contemporary fluids from the Yucca Mountain area. Four different categories of local fluids are represented. These are: (1) isotopically and chemically immature fluids from the Paleozoic carbonates; (2) the corresponding mature fluids; (3) isotopically and chemically immature fluids from the Tertiary ignimbrites; and (4) the corresponding more mature fluids. The fluid isotopic data will be used for several purposes. Among these are: (1) definition of evolutionary paths in the

del¹³C vs. ⁸⁷Sr/⁸⁶Sr field with increasing intraformation residence time; (2) evaluation of the compatibility of del¹³C vs. ⁸⁷Sr/⁸⁶Sr fields for both the contemporary calcium-magnesium fluids and the parent fluids of the late epigenetic calcites; (3) evaluation of the compatibility of del¹³C vs. ⁸⁷Sr/⁸⁶Sr fields for both the contemporary calcium-magnesium fluids and the corresponding host rock; and (4) evaluation of the compatibility of del¹³C vs. ⁸⁷Sr/⁸⁶Sr fields for both the parent fluids of the earlier epigenetic calcites and the inferred corresponding host rock.

The joint strontium and carbon test of the interpreted paragenetic relationships is carried out in Figures 8-31 through 8-37. For five samples of the altered lower vitrophyre of the Topopah Spring Member, normalized concentrations of K, Ca, Ti, Rb, Sr, Y, Zr, Nb, Ba, La, and Ce are shown in Figure 8-31. In addition, this figure also shows the total concentrations of Ca, Ba, Sr, Ti, Rb, Zr, La, and Ce, plotted as a function of depth. Both the normalized concentrations and the total concentrations, shown in Figure 8-31, are based on samples obtained from borehole UE-25a#1 at depths ranging from 200 to 1400 feet. For the northern and southern ends of Yucca Mountain, the total concentrations of strontium in the same stratigraphic ignimbrites are compared in Figure 8-32. For the northern sector of Yucca Mountain, Figures 8-33a and 8-33b present a comparison of the depth distributions of: (1) the present-day values of the ⁸⁷Sr/⁸⁶Sr ratio for whole-rock ignimbrites; (2) the corresponding total concentrations of strontium;

and (3) the percentage of Ca + Mg cations in the corresponding clinoptilolites and whole-rock ignimbrites. Two separate comparisons are shown. The southeastern segment of Yucca Mountain (boreholes USW G-1 and G-2, Figure 8-19) is represented in Figure 8-33a, and the northwestern segment (borehole UE-25a#1, Figure 8-19) in Figure 8-33b. For both the Timber Mountain calcites and the late epigenetic calcites, a comparison of values of the $\delta^{13}\text{C}$ ratio with corresponding values of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio is shown in Figure 8-34. This comparison has been constructed from the isotopic data for boreholes USW G-1, G-2, G-3, G-4, and UE-25b#1, Figure 8-19. For the parent fluids of both the Timber Mountain calcites and the late epigenetic calcites, the evolutionary path for the $\delta^{13}\text{C}$ vs. $^{87}\text{Sr}/^{86}\text{Sr}$ field is depicted in Figure 8-35. For contemporary fluids from both the Tertiary ignimbrites and the Paleozoic carbonates, carbon and strontium isotopic characteristics are summarized in Figure 8-36. For both the local calcium-magnesium fluids and the local sodium-potassium fluids, this figure also shows the evolutionary paths for $\delta^{13}\text{C}$ vs. $^{87}\text{Sr}/^{86}\text{Sr}$ fields. These paths illustrate the influence of the intraformation residence time on the respective $\delta^{13}\text{C}$ vs. $^{87}\text{Sr}/^{86}\text{Sr}$ fields. Finally, for both the contemporary calcium-magnesium fluids and the parent fluids of the late epigenetic calcites, Figure 8-37 illustrates the isotopic compatibility of the respective $\delta^{13}\text{C}$ vs. $^{87}\text{Sr}/^{86}\text{Sr}$ fields. This figure also illustrates that, for parent fluids of the late epigenetic calcites, the reconstructed $\delta^{13}\text{C}$ vs. $^{87}\text{Sr}/^{86}\text{Sr}$ field is compatible with the independently-inferred intraformation history of these fluids (i.e., long intraformation residence time and

great depth of circulation).

Examination of Figures 8-31 through 8-37 leads to the formulation of the following five observations/interpretations. First, in addition to the calcium and magnesium metasomatism, the Yucca Mountain ignimbrites display clear indications of having experienced a pervasive strontium metasomatism. Such a state of affairs is indicated by two lines of evidence. On the one hand, for unaltered alkali ignimbrites, the expected total abundance of strontium is uniform and low, typically much less than 100 ppm, Faure (1986). This general expectation has been confirmed by the results of chemical analyses of samples of Yucca Mountain glass. Specifically, Peterman et al. (1991) have reported that, for the unaltered lower vitrophyre of the Topopah Spring Member, the representative concentration of strontium is as low as 19 ± 6 ppm. This is a mean value derived from six samples of fresh vitrophyre from three different boreholes. From Figure 8-32 it may be observed that, in the present-day altered state, the Yucca Mountain ignimbrites contain surprisingly large concentrations of strontium. These concentrations, however, are highly erratic and range from 10 to as much as 700 ppm. Clearly, an epigenetic and space-differential enrichment of total strontium is apparent. This enrichment is best expressed through a normalized plot of the results of trace element analyses of five samples from the altered lower vitrophyre of the Topopah Spring Member, Figure 8-31. It may be observed that, relative to the devitrified interior of the Topopah Spring ash-flow sheet, the epigenetically

altered vitrophyre exhibits substantial gains in both calcium and strontium. On the other hand, for a given stratigraphic unit, the average concentrations of strontium do not remain constant, but vary laterally instead. The lateral variability of the average concentration of strontium may be illustrated by examining these concentrations for the Topopah Spring Member, at three different locations. In the southern end of Yucca Mountain (near Highway 95), the average concentrations are uniformly low, say about 25 ppm, Figure 8-32. In the northern end, however, the corresponding concentrations are decisively higher. These concentrations range from 50 ppm for the northwestern segment (boreholes USW G-1 and G-2), to 190 ppm for the southeastern segment (borehole UE-25a#1), Figures 8-27a and 8-27b, respectively. The occurrence of the highest values of average concentration of strontium in the vadose zone of the southeastern segment is particularly important. It indicates that, for both the calcic zeolitization and the late carbon-13 depleted carbonatization, parent fluids were carrying fairly elevated concentrations of total dissolved strontium. From Section 5.0 it may be recalled that, for the vadose zone interstitial fluids, concentrations of dissolved strontium are extraordinarily high. As shown in Figure 5-2a, these concentrations range from 1020 to 1180 ppm. They are a factor of 30-40 higher than those for contemporary fluids from below the water table, Figure 5-3a. Is it possible that, at Yucca Mountain, both the vadose zone interstitial fluids and the spatially differential strontium metasomatism, as well as both the calcic zeolitization and the carbon-13 depleted carbonatization, are related to the same hydrothermal

processes?

Second, at Yucca Mountain, the observed space-differential strontium metasomatism was developed sequentially in response to two distinct allogenic fluids, each having distinct strontium isotopic characteristics. Such a state of affairs is indicated by the depth distributions of values of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio obtained for whole-rock ignimbrites, Figures 8-33a and 8-33b. For the ignimbrites altered during the Timber Mountain hydrothermal metamorphism, the present-day values of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio are low, generally slightly less than 0.7100, Figure 8-33b. The corresponding initial ratios range from 0.7083 to no more than 0.7101, Figure 8-27a. These initial ratios were calculated assuming that present-day values of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio reflect both the relative concentration of strontium-87 acquired from solution and the ^{87}Sr produced in-situ through decay of ^{87}Rb . It has been further assumed that the ^{87}Sr produced in-situ has been accumulating for about 13×10^6 years (the age of emplacement of the host rock). Because the actual timing of the whole-rock alteration ($11-9.5 \times 10^6$ years B.P., Figure 7-25) does not differ appreciably from the assumed age of introduction of strontium, the calculated initial values of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio, from 0.7081 to 0.7101, may be regarded as accurately reflecting the isotopic character of dissolved strontium in the Timber Mountain hydrothermal fluids. For the ignimbrites altered during the subsequent intermittent hydrothermal metamorphism, however, the present-day values of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio are decisively higher. They range from 0.7095 to as

much as 0.7202, Figures 8-33a and 8-33b. The corresponding initial ratios range from 0.7104 to 0.7129, Figures 8-27a and 8-27b. These ratios were calculated assuming erroneously that the ^{87}Sr produced in-situ was accumulating for about 13×10^6 years and therefore are too low. For fluids responsible for the alteration of the lower vitrophyre of the Topopah Spring Member in borehole UE-25a#1, an estimate of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio was reported by Peterman et al. (1991). The reported value is 0.7119 ± 0.0002 , and was said to be identical to that from samples of purified alteration minerals from the altered zone in borehole UE-25a#1. Although too low (because of the erroneous age assumed for the introduction of strontium), the 0.7119 value may be regarded as conservatively representative of relative concentrations of dissolved strontium-87 in the allogenic fluids associated with a fault-based hydrothermal system. The inferred bi-modality of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio indicates that, as far as the proposed paragenetic relationships are concerned, one aspect of the first step of the devised test yields an affirmative result. As expected, relative to fluids associated with the prior magmatic center-based hydrothermal system, fluids associated with the subsequent fault-based hydrothermal system have carried higher relative concentrations of dissolved strontium-87. Locally, the appropriately radiogenic strontium is incorporated in only one primary reservoir, which is the Pre-cambrian basement. The postulated increasing depth of fluid circulation, therefore, is consistent with the clearly evident bi-modality of the strontium isotopic characteristics of the respective allogenic fluids.

Third, for the Yucca Mountain epigenetic calcites, the bi-modality of the $\delta^{13}\text{C}$ ratios noted earlier is clearly reflected through spatially corresponding bi-modality of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios. As shown in Figure 8-34, the carbon-13 enriched calcites are spatially correlative with the strontium-87 depleted calcites. Similarly, the carbon-13 depleted calcites are spatially correlative with the strontium-87 enriched calcites. As shown in Figure 8-35, the $\delta^{13}\text{C}$ vs. $^{87}\text{Sr}/^{86}\text{Sr}$ field reconstructed for the Timber Mountain calcites is clearly distinct from the field reconstructed for the subsequent epigenetic calcites. As far as the test of the paragenetic relationships is concerned, the double isotopic discords indicate that the remaining aspect of the first step in the devised testing scheme also yields an affirmative result. As expected, relative to the Timber Mountain calcites, the subsequent epigenetic calcites were precipitated from solutions that were bearing both low relative concentrations of carbon-13 and high relative concentrations of strontium-87. For the $\delta^{13}\text{C}$ vs. $^{87}\text{Sr}/^{86}\text{Sr}$ field, the observed evolutionary path is consistent with the postulated increased depth of circulation, and the resulting substitution of both the reservoirs of strontium (Paleozoic carbonates --> Pre-cambrian basement) and the sources of carbon (Paleozoic carbonates --> igneous sources). Importantly, at Yucca Mountain, the parent fluids for both the strontium-87 enriched calcites and the carbon-13 depleted calcites were also involved in (1) whole-rock strontium metasomatism, and (2) calcic zeolitization. It follows then that by proposing a supergene-pedogenic origin for the carbon-13 depleted and strontium-87 enriched calcites, the USGS investigators are in fact

proposing that the supergene-pedogenic processes, in addition to being capable of (1) producing veins some 300 meters below the water table, and (2) causing a factor of 1.5 increase in the geothermal gradient, are also fully capable of causing the observed space-differential calcium, magnesium, and strontium metasomatism in large volumes of initially homogenous ignimbrites. Such a proposition, however, is in accord neither with established geological principles nor with common sense. It is doubtful that even a handful of particularly stout proponents of "pedogenic" processes would venture as far as to challenge this statement.

Fourth, at and around Yucca Mountain, the isotopic characters of carbon and strontium dissolved in contemporary calcium-magnesium fluids are different from those incorporated in the host Paleozoic carbonates. These important isotopic discords are illustrated in Figure 8-36. It may be observed that, relative to hypothetical fluids equilibrated with marine limestones of Early Paleozoic age, the actual contemporary Paleozoic carbonate-based fluids are strongly enriched in both radiogenic strontium-87 and isotopically "light" carbon. For such fluids, an expected representative value of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio is, say, 0.7088. The actual observed values, however, are decisively higher, say from 0.7120 to as much as 0.7190, Figure 8-29b. For fluids acquiring dissolved carbon from marine limestones, an expected representative value of the $\delta^{13}\text{C}$ ratio is 0 ± 2.0 per mil_{PDB}. The actual observed value, however, is significantly lower, say -5.0 per mil_{PDB}. Clearly, these isotopic discords are important. As far as the test of the

paragenetic relationships is concerned, these discords indicate that the second step in the devised testing scheme also yields an affirmative result. As expected, contemporary calcium-magnesium fluids are being contaminated (intermixed) with deeper fluids. The latter fluids acquire dissolved strontium from a highly radiogenic reservoir and dissolved carbon from an isotopically "light" source. Considered together, the observed strontium-carbon isotopic characteristics of the contaminating fluids can only be acquired in a deep substratum, a conclusion consistent with both the postulated increased depth of circulation and the postulated reservoir of strontium and source of carbon. Importantly, for contemporary fluids (residing in both the Paleozoic carbonates and the Tertiary ignimbrites), the observed $\delta^{13}\text{C}$ vs. $^{87}\text{Sr}/^{86}\text{Sr}$ fields exhibit important evolutionary trends, Figure 8-36. From this figure it may be seen that, with increasing chemical maturity (increasing fluid intraformation residence time), the $\delta^{13}\text{C}$ vs. $^{87}\text{Sr}/^{86}\text{Sr}$ fields seem to be evolving toward the field reconstructed for parent fluids of the late epigenetic calcites. This observation is in direct conflict with the conclusions drawn by the USGS investigators regarding the isotopic affinity between the parent fluids of carbon-13 depleted calcites and local fluids from below the water table. Based on the results of appropriate isotopic analyses, it is clearly evident that, rather than the calcites being isotopically incompatible with the local deep-seated fluids, these calcites are incompatible with local infiltrating, chemically immature fluids.

Fifth, for parent fluids of both the Timber Mountain calcites and the late epigenetic calcites, the expected affinities of the respective $\delta^{13}\text{C}$ vs. $^{87}\text{Sr}/^{86}\text{Sr}$ fields are clearly evident. These affinities are illustrated in Figure 8-37. From this figure it may be observed that, for the parent fluids of the Timber Mountain calcites (vein set I in Figure 8-37), the reconstructed $\delta^{13}\text{C}$ vs. $^{87}\text{Sr}/^{86}\text{Sr}$ field is compatible with that expected for fluids isotopically equilibrated with marine limestones of Early Paleozoic age. The observed slight difference in values of the $\delta^{13}\text{C}$ ratio is directly attributable to diffusional carbon-13 enrichment. Typically, precipitation of calcite is accompanied by a progressive escape of CO_2 from the parent solution. During CO_2 degassing, the lighter $^{12}\text{CO}_2$ molecules escape preferentially and consequently the affected solution becomes progressively enriched in the heavier $^{13}\text{CO}_2$ molecules. It is not surprising that, for some of the calcites, the $\delta^{13}\text{C}$ ratios are "heavier" than for their carbon source. As far as the proposed paragenetic relationships are concerned, the isotopic affinity for vein set I illustrated in Figure 8-37 indicates that the third step in the devised test yields an affirmative result. From Figure 8-37 it may be further observed that, for the parent fluids of the late epigenetic calcites (vein set II in Figure 8-37), the reconstructed $\delta^{13}\text{C}$ vs. $^{87}\text{Sr}/^{86}\text{Sr}$ field is compatible with that observed in association with contemporary calcium-magnesium fluids. Small differences in values of the $\delta^{13}\text{C}$ ratio are again attributable to diffusional carbon-13 enrichment. Relative to the contemporary fluids, parent fluids for the late epigenetic calcites may be presumed to have been ascending with much higher

velocities. For the latter fluids, CO₂ partial pressures were higher and consequently the degree of diffusional carbon-13 enrichment was lower than that observed in the near-surface aquifer. As far as the proposed paragenetic relationships are concerned, the isotopic affinity for vein set II in Figure 8-37 indicates that the last step in the devised testing scheme also yields an affirmative result.

In light of the four affirmative results, the tested paragenetic relationships seem to be unassailable. These relationships account for all of the isotopic data as well as the field observations (Section 3.0) and, in sharp contrast to the USGS interpretations, do not lead to either paradoxical or nonsensical conclusions. Furthermore, these relationships fit harmoniously with the results of three independent considerations. These considerations pertain to: (1) origin of the vadose zone interstitial fluids, Section 5.0; (2) origin of the "mosaic" breccias, Section 6.0; and (3) origin and age of the alteration minerals, Section 7.0. There is no reason, therefore, to reject the main theme of these combined considerations, namely the intermittent ascent of hypogene fluids into the Yucca Mountain vadose zone.

8.5 Uranium-Thorium Ages of the Carbon-13 Depleted Calcites

A critically important supposition that underlies the hypogene interpretation of

the origin of the carbon-13 depleted calcites is that, at Yucca Mountain, carbon incorporated in these calcites originated from local igneous sources. For this supposition, however, the factual basis is limited and may be summarized using two quotations from Faure (1986). These are: (1) for hydrothermal calcites, "*the $\delta^{13}\text{C}$ values of CO_2 in fluid inclusions range from -4 to -12 per mil with respect to PDB,*" and (2) "*the low $\delta^{13}\text{C}$ values of CO_2 gas and carbonate minerals in hydrothermal ore deposits generally suggest a deep-seated source of carbon.*" For the magmatically active Yucca Mountain region, therefore, the supposition of igneous origin for some of the locally circulating carbon is clearly permissible. Nevertheless, this supposition remains unproven and may be regarded by some as jeopardizing the interpretations advanced herein. This is true particularly in view of the well-known fact that the isotopic character of carbon, either dissolved in a fluid or incorporated in a solid, is not an unequivocal indicator of its origin. With specific reference to hydrothermal fluids, for example, Hoefs (1987) has stated that the equivalent " *$\delta^{13}\text{C}$ values can also be generated by simple mixing between carbonate-derived and organically-derived CO_2 .*" To make matters worse, for many supergene carbonates the isotopic character of carbon is similar to that incorporated in unequivocally hypogene carbonates. For supergene-pedogenic carbonates, for example, Talma and Netterberg (1983) give values of the $\delta^{13}\text{C}$ ratio ranging from +4.0 to about -8.5 per mil_{PDB}. For hypogene-pedogenic carbonates, the same authors note that the corresponding values range from 0 to -5.5 per mil_{PDB}. Furthermore, for travertine deposits, the observed range of the

del¹³C ratio is inclusive of both of the foregoing pedogenic ranges. Turi (1986), for example, notes that the range is from -10.0 to as much as +12.0 per mil_{PDB}.

Clearly, based solely on the isotopic character of carbon, interpreting the origin of carbonates is equivalent to looking "*for a black cat in a coal bin.*"

To gain further confidence in the validity of the supposition that underlies the interpretations advanced herein, it is clearly necessary to devise an independent testing scheme. Fortunately, for the Yucca Mountain area, the desired testing scheme may be constructed from the uranium-thorium ages of calcretes and equivalent veins. Specifically, if the presumed igneous origin of carbon is correct, then one may expect that local igneous events are contemporaneous with local carbonate depositional events. For the former, the corresponding ages are known independently, based on K/Ar and ⁴⁰Ar/³⁹Ar laser fusion analyses of representative volcanic rocks, see, for example, Crow et al. (1982), Wells et al. (1990), and Turi and Champion (1990).

The available U/Th data used for the purpose of the envisaged test are summarized in Figures 8-38 and 8-39. Without exception, these data represent the late epigenetic calcites, those that are enriched in oxygen-18 and, at the same time, are depleted in carbon-13. Two depositional phases of calcite are represented, namely calcretes and associated surficial veins (Figure 8-38) and equivalent veins in the vadose zone (Figure 8-39). The samples analyzed for U/Th

were first ground into a fine powder and then, to convert CaCO_3 to CaO , were heated at 900° Celsius. The acid differential dissolution technique was used to separate the acid-soluble fraction (CaO) from the acid-insoluble residuum. Both of the separate fractions were then spiked with ^{236}U , ^{238}Th , and ^{229}Th standard solutions. Uranium and thorium isotopes were isolated and purified employing the anion exchange procedure, Szabo et al. (1981). The alpha spectrometry counting method was then used to determine concentrations of the isolated isotopes. For pure calcites and opals, the corresponding U/Th ages were calculated from measured values of the $^{230}\text{Th}/^{234}\text{U}$ activity ratio, using the standard radioactive growth and decay equations. These calculations were made assuming that since their formation, the dated phases have behaved as an ideal closed system. For impure specimens, U/Th ages were calculated using measured activity ratios from both acid-soluble and acid-insoluble fractions. The age calculations were made following the pseudo-isochron plot method, Szabo et al. (1981) and Szabo and Rosholt (1982).

The U/Th data for the late epigenetic calcites are also plotted and analyzed in Figures 8-40 through 8-43. Figures 8-40a and 8-40b present a comparison of the $^{230}\text{Th}/^{234}\text{U}$ activity ratios with the corresponding $^{234}\text{U}/^{238}\text{U}$ activity ratios. In these figures, calcites from the vadose zone (travertine veins, calcretes, and micritic calcitic veins) are considered along with calcites from below the water table (Devil's Hole travertine vein). For the former calcites, radiogenic ^{234}U atoms are

exposed to leaching by infiltrating rainwater. For the latter calcites, however, the vulnerable ^{234}U atoms are shielded by continuous precipitation of CaCO_3 . By comparing the activity ratios from the two settings, it is possible to evaluate the appropriateness of assuming ideal closed systems. For coexisting samples of surficial carbonates from the Eagle Mountain area of California, Figure 8-41 shows a comparison of U/Th ages with corresponding ^{14}C ages. This comparison may be used to judge the magnitude of dating errors brought about by deviation from the assumed ideal closed system behavior. For the Yucca Mountain calcretes and vadose zone veins, Figure 8-42 presents U/Th ages plotted against depth of sample acquisition. Finally, Figure 8-43 presents a comparison of the mean U/Th ages of epigenetic calcites with mean radiometric ages of recent nearby volcanic eruptions.

Examination of Figures 8-40 through 8-43 leads to the formulation of the following four observations/interpretations. First, with respect to isotopes of uranium and thorium, the Yucca Mountain calcites seem to exhibit open system behavior. Such a state of affairs is suggested by two lines of evidence. On the one hand, for a few samples, measured values of the $^{230}\text{Th}/^{234}\text{U}$ activity ratio exceed asymptotic secular equilibrium values, Figure 8-40a. These abnormally high values indicate that some vulnerable ^{234}U atoms were selectively mobilized. On the other hand, for the vadose zone calcites, measured values of the $^{234}\text{U}/^{238}\text{U}$ activity ratio are consistently lower than those for the submerged DH-2 vein, Figures 8-40a and

8-40b. This discrepancy in the relative concentrations of vulnerable ^{234}U atoms is attributable to leaching by rainwater and consequent open-system behavior. In light of this observation, the isotopic incompatibility noted by Stuckless et al. (1991) between the isotopic characters of uranium incorporated in the DH-2 vein and in the Yucca Mountain calcites assumes a completely different meaning. Rather than indicating a pedogenic-supergene origin of the carbon-13 depleted and strontium-87 enriched calcites, this incompatibility is clearly indicative of open system behavior of these calcites.

Second, given the inferred open system behavior of the vadose zone calcites, one is tempted to reject the U/Th ages of these calcites as completely erroneous and/or irrelevant. Such, however, is not necessarily the case. The following reasoning is put forth in support of this statement. The selective mobilization of ^{234}U atoms has the effect of diminishing the $^{234}\text{U}/^{238}\text{U}$ activity ratio while, at the same time, enhancing the $^{230}\text{Th}/^{234}\text{U}$ activity ratio. In other words, the selective mobilization of ^{234}U atoms renders U/Th ages positively biased, or too old. This effect is clearly illustrated by the results of ^{14}C and U/Th geochronological studies performed by Schlesinger (1985). As shown in Figure 8-41, for coexisting carbonates from the Eagle Mountain area of Mojave Desert, U/Th ages are consistently older than the corresponding ^{14}C ages. The observed discrepancies, however, are not alarmingly great. For the $\sim 2 \times 10^4$ year-old carbonates, they amount to about 30 percent. Given that equally substantial error bars accompany the Yucca Mountain U/Th

ages, potential errors of even a few tens of percent do not justify complete rejection of these ages.

Third, at Yucca Mountain, both the results of direct field observations and the U/Th ages yield somewhat consistent histories of the formation of local calcretes. Two lines of evidence may be put forth in support of this statement. On the one hand, at Busted Butte, three discrete depositional sequences of calcretes have been documented. These sequences occur below, within, and above aeolian sands comprising the local sand ramps. Although the upper age limit of the formation of these sand ramps remains unknown, one may be reasonably certain that they are younger than the 7×10^5 years B.P. age of the underlying Bishop Ash. It seems that calcretes were formed in association with three temporally distinct depositional episodes. Importantly, all three of these episodes are younger than 7×10^5 years B.P., and are expressed at the topographic surface. On the other hand, U/Th ages of the late epigenetic calcites indicate that, subsequent to the deposition of the Bishop Ash, four generations of these calcites have been formed. As shown in Figure 8-42, the mean U/Th ages of these generations are 3×10^4 , 7.5×10^4 , 1.7×10^5 , and 2.8×10^5 years B.P. Importantly, at and near the topographic surface, only three generations of the epigenetic calcites are evident. The emplacement of the fourth generation of calcites (mean U/Th age of 1.7×10^5 years B.P., Figure 8-42) appears to have been restricted to the lower region of the vadose zone. It seems that, in the time elapsed since the deposition of the Bishop

Ash, both direct field observations and U/Th ages yield the same number of depositional episodes of local calcretes. This observation may be taken as providing some degree of confidence in the U/Th ages.

Fourth, for the Yucca Mountain epigenetic calcites, the depositional history is correlative with the independently-known chronology of local magmatic activity. As shown in Figure 8-43, average K/Ar and $^{40}\text{Ar}/^{39}\text{Ar}$ laser fusion ages for samples of local basaltic flows and cinders are in fairly satisfactory agreement with average U/Th ages of late epigenetic calcites. All five of the recognized igneous episodes of the past 10^6 years seem to have been accompanied by contemporaneous episodes of carbonatization. The fact that these independently determined chronologies are correlative is of two-fold significance. On the one hand, it provides additional assurances that (1) U/Th ages do adequately reflect the timing of deposition of the epigenetic calcites, and (2) a hypogene origin of these calcites has been inferred correctly. On the other hand, as far as the chronological test is concerned, the conformity of radiometric ages indicates that this test yields an affirmative result. Igneous origin of carbon incorporated in the carbon-13 depleted calcites, beyond being a valid option, also is supported independently by circumstantial evidence.

8.6 Summary and Conclusions

The foregoing analysis of the extensive isotopic data set leads to firm conclusions regarding the age and origin of the Yucca Mountain calcites. For the purposes of this report, these views may be summarized by the following four points. First, the currently available isotopic and geochemical data do not provide any factual support for supergene-pedogenic interpretations of the origin of the strontium-87 enriched and carbon-13 depleted calcites as is, ad nauseam, advocated by some members of the USGS staff. Quite the contrary, six independent lines of evidence may be put forth as effectively nullifying this interpretation. These are: (1) the isotopic character of incorporated carbon is not in accord with the proposed supergene origin, Figure 8-15; (2) the depth distribution of the isotopic character of incorporated oxygen is not in accord with the proposed supergene origin and instead indicates crystallization at elevated geothermal conditions, Figures 8-16 and 8-17; (3) the isotopic character of incorporated strontium is not in accord with crystallization from chemically and isotopically immature fluids, Figure 8-36; (4) spatially, the calcites in question are associated with calcites containing fluid inclusions with homogenization temperatures that are substantially greater than ambient temperature, Figure 8-23; (5) spatially, the calcites in question are associated with strongly metasomatic and clearly hypogene alteration minerals, Figure 8-24 and Section 7.0; and (6) temporally, the calcites in question are contemporaneous with periods of local magmatic activity, Figure 8-43.

Furthermore, acceptance of the USGS supergene interpretations leads to three nonsensical conclusions. These are: (1) hypothetical supergene pedogenic processes are activated by local magmatism; (2) these processes, if operating in chemically homogeneous vitric ignimbrites, produce space-differential yet extensive calcium, magnesium, and strontium metasomatism; and (3) during their descent, infiltrating rainwaters cause a factor of 1.5 increase in the local geothermal gradient. With all of these shortcomings, how can anyone regard the USGS interpretation as a suitable foundation for activities that relate to the disposal of high-level nuclear wastes?

Second, in sharp contrast to the USGS interpretation, the inferred hypogene origin of the strontium-87 enriched and carbon-13 depleted calcites not only is supported, but also is demanded, by the multitude of data sets considered above. In that regard, particularly convincing and persuasive are the lines of evidence that involve a number of independent elements. Among these are: (1) the isotopic characters of incorporated uranium, strontium, oxygen, and carbon are readily explicable within the context of the postulated hypogene origin; (2) the isotopic characters of incorporated uranium, strontium, oxygen, and carbon are clearly equivalent to those incorporated in unequivocally hypogene local analogs; and (3) calcite crystallization is contemporaneous with local magmatism. Furthermore, for the Yucca Mountain area, expectations regarding intermittent ascent of hypogene fluids (with chemical and isotopic characteristics broadly analogous to

those inferred for the parent fluids of the calcites considered) were derived entirely independently, based on the consideration of three different sets of data.

Specifically, these data sets pertain to: (1) the contemporary geodynamic configuration of the Yucca Mountain region, including the evolution thereof, Section 2.0; (2) the character and durability of the local hydraulic structure, Szymanski (1989); and (3) the character and durability of the boundary conditions for the local heat-fluid flow system, Szymanski (1989). In consideration of the above attributes, the postulated hypogene origin of the strontium-87 enriched and carbon-13 depleted calcites must be regarded as secure beyond a reasonable doubt.

Third, at Yucca Mountain, the independently inferred overprinting of the Timber Mountain alteration aureole, by the subsequent southeastern alteration aureole, is also reflected in the spatial distribution of the combined strontium and carbon isotopic characteristics of epigenetic calcites. The spatial distribution of the Timber Mountain alteration minerals is correlative with that of the strontium-87 depleted and carbon-13 enriched calcites. Clearly, the crystallization of these calcites is directly attributable to a hydrothermal system driven by the Timber Mountain heat source. The spatial distribution of the subsequent and strongly metasomatic alteration minerals, however, is correlative with that of the strontium-87 enriched and carbon-13 depleted calcites. This spatial conformity, together with the fact that both the alteration minerals and the corresponding calcites were formed in association with fluids carrying remarkably similar

relative concentrations of radiogenic strontium-87, indicate that these calcites are directly attributable to the intermittent activity of a fault-based hydrothermal system driven by conductively replenished heat sources. At Yucca Mountain, therefore, both the propylitic alteration and the associated carbonatization provide clear and convincing testimony that, as expected, convective heat flow has remained operational during the last 10^7 years. Again, as expected, responding to the decreasing degree of partial melting of a rising asthenospheric diapir, the operational forms of convective circulations have exchanged their relative importance. For the immediate vicinity of Yucca Mountain, the earlier magmatic center-based circulations were replaced by intermittent fault-based circulations driven by conductively replenished heat sources.

Fourth, within the context of considering the suitability of Yucca Mountain for either site characterization or for the development of a high-level nuclear waste repository, the locally observed carbonatization is of two-fold significance. On the one hand, the fact that this carbonatization may be identified at the topographic surface provides clear and unequivocal resolution of the dilemma involving the expected magnitudes of co-seismic or co-magmatic hydrologic effects. In contrast to assurances given by some members of the USGS staff, rather than being measured in terms of feet and meters, these effects are measured in terms of hundreds of meters instead. The results of trace-element analyses of the vadose zone interstitial fluids, the results of fission-track analyses of allogenic zircons, the

results of chemical and radiometric analyses of vadose zone clinoptilolites, the results of isotopic analyses of epigenetic calcites, and the results of direct field observations, are all unanimous in that regard. On the other hand, the fact that, collectively, the late epigenetic calcites and the metasomatic zeolites do carry a broad spectrum of the U/Th and K/Ar ages, from 8.5×10^6 to as little as 3×10^4 years B.P., provides an opportunity for judging temporal aspects of the activity of underlying hydrothermal processes. Rather than being restricted to an isolated yet temporally distant episode, these processes operate on an intermittent basis instead. The directly observed field relationships, the K/Ar ages of metasomatic clinoptilolites, and the U/Th ages of epigenetic calcites, are all unanimous in that regard. Furthermore, the intermittent character of local hydrothermal activity has been inferred completely independently, based on consideration of the local geodynamic configuration, Section 2.0. With all these currently known and well documented shortcomings, how can anyone insist that Yucca Mountain is a suitable place for either the forthcoming site characterization or for the development of a high-level nuclear waste repository?

9.0 Overall Summary and Conclusions

The discussion presented in Sections 1.0 through 8.0 may be summarized by the following six concluding remarks. **First and foremost, long-term behavior of the Yucca Mountain coupled fluid-heat flow system is, and always was, profoundly influenced by fundamental geodynamic processes operating deep in the Earth's interior.** Specifically, during the last 13×10^6 years, asthenospheric diapirism, and the resulting corrosion of the lithosphere, is the ultimate cause of local geodynamic developments, including hydrothermal activity and the resulting epigenetic metamorphism and carbonatization. The presence of active mantle diapirism is clearly and unequivocally indicated by the results of combined consideration of the P-wave velocity structure and history of local magmatism. Following well-established tectono physical principles concerning the origin of mafic magmas, it may be inferred that during adiabatic ascent, asthenospheric matter (presumably composed of primitive pyrolite) contained in a gravitationally unstable and rising diapir undergoes depth (pressure)-variant fractional melting.

During the last 10^7 years, the depth of magma source region and, with it the degree of partial melting, have been steadily decreasing. The latter decrease reflects the diminishing availability of water in the melting system. During and before the Timber Mountain magmatism, say 10 to 15×10^6 years B.P., the magma

source region was situated at a depth in excess of 60 km. At the corresponding pressure (20-25 kb), hydrous amphibole is unstable and transforms into eclogite plus water. At this pressure and at greater pressures, therefore, the availability of indigenous mantle water is relatively high, about 0.1 percent. Under these circumstances, the corresponding solidus temperature for pyrolite is relatively low, about 1200° Celsius; and for melting slightly above the solidus temperature, the degree of partial melting is relatively high. With decreasing depth of the magma source region, however, amphibole becomes a stable mineral phase. The previously available water now becomes sequestered in the refractory residuum, which contains stable amphibole. This has the result that, relative to the prior deeper magma source regions, the solidus temperature is higher and, the degree of partial melting for a given adiabat is lower. The increased solidus temperature and the decreased degree of partial melting implies that, relative to the prior temperature of the partial melt, the subsequent temperature increases steadily.

For the Yucca Mountain region, the evolving thermodynamic state of the upper mantle (i.e., (1) diminishing depth of the magma source region, and (2) increasing temperature of the source region) had the result that, during the last $\sim 10^7$ years, the mantle --> crust heat input has undergone a fundamental transformation. Early in the pyrolite melting process, when the magma source region was at a depth greater than 60 km, the advective component of heat input (heat transported via segregated magma bodies) was relatively high and the conductive

component was relatively low. The relatively high degree of partial melting was responsible for the former, and the lower melting temperature and the greater depth of the magma source region was responsible for the latter. During more advanced stages of the melting process, however, the relative and absolute importance of the two components (of the mantle --> crust heat input) have changed. Specifically, with diminishing depth of the magma source region, the conductive component steadily became more prominent at the expense of the advective component. Again, the diminishing degree of partial melting caused the latter to occur, and both the increasing temperature of the source region and the decreasing depth of this region are responsible for the former.

As far as hydrothermal circulation of intracrustal fluids is concerned, the evolving mantle --> crust heat input has profound implications. There are two important aspects of these implications. On the one hand, in the crust of Yucca Mountain, there is, and always was, the need for augmentation of conductive heat flow by convective flow. In other words, at and around Yucca Mountain, there is a chronic potential for convective circulation of intracrustal fluids. This is so because, regardless of whether it is advective or conductive, the elevated mantle --> crust heat input causes the crustal temperatures to have a tendency to increase. Ultimately, the increasing crustal temperature leads to the development of unstable fluid density configurations and, at that time, convective heat flow mechanism becomes operational. The resulting fluid circulations enhance the

crust --> atmosphere heat transfer and, through that, assure that crustal temperatures are not excessively high. On the other hand, the increasing absolute and relative importance of the conductive component of the mantle --> crust heat input, at the expense of the advective component, has caused the forms of convective circulations of intracrustal fluids to change.

During periods when the advective component dominated, the corresponding circulations were driven by "strong" heat sources, consisting of fairly large bodies of molten rock. For such heat sources, the convectively enhanced heat losses are being compensated for readily, via convective circulation within the respective magma body itself. Magmatic center-based hydrothermal systems, therefore, are spatially stationary and temporally prolonged. For such systems, the associated fluid circulations may last uninterruptedly for a fairly long period of time, say $1-2 \times 10^6$ years. Typically, the resulting alteration-mineralization aureoles are spatially extensive and are uniformly distributed around the causative magma body. This spatial uniformity would suggest that, around intracrustal magma bodies, the combined intrinsic and extrinsic hydraulic conductivity is fairly uniformly distributed. In other words, the deformation-induced gradients of extrinsic hydraulic conductivity are broadly distributed, or "flat."

During periods when the conductive component predominates, however, the circumstances are markedly different, as the previous extensive intracrustal

bodies of molten rock are either absent or rare. For the corresponding fluid circulations, therefore, the available heat sources are "weak," in the sense that they may be readily depleted or exhausted. This is so because in contrast to magmatic center-based hydrothermal systems, the enhanced heat loss by fluid convection is not being adequately compensated for by slow conductive heat flow from the far field. Being deprived of adequate replenishment of thermal energy, convective circulations of fluids cease. Such cessations, however, are only temporary. They are followed by periods of thermal recovery, during which heat conduction from the far field slowly restores the original unstable state.

Hydrothermal systems driven by conductively replenished heat sources, therefore, typically remain intermittently active for very long periods of time. In contrast to magmatic center-based hydrothermal systems, however, the individual convective bursts, associated with conductively replenished hydrothermal systems, are of relatively short duration.

Another interesting feature of hydrothermal systems driven by conductively replenished heat source is their "opportunistic" character. Such systems tend to be developed around fault zones, taking advantage of "steep" deformationally induced gradients of extrinsic hydraulic conductivity. In association with a deforming fault zone, both the gradually increasing temperature (which, for fluid circulations, is the driving force) and the enlarging hydraulic conductivity (which, for fluid circulations, is the resisting force) act in concert with each other in

promoting the onset of convective circulations of intracrustal fluids. Conductively replenished hydrothermal systems, therefore, tend to be developed around intermittently active fault zones. The respective alteration-mineralization aureoles are typically fault-based. Importantly, the spatial dimensions of these aureoles are controlled by the depth-variant field of extrinsic hydraulic conductivity. Deep below the topographic surface (below the so-called $Z_{(x,y,t)}$ surface), both the field of extrinsic hydraulic conductivity and the corresponding alteration aureole are narrowly centered on the deforming parent fault. Near the topographic surface, however, the extrinsic hydraulic conductivity field becomes wider and the corresponding aureole widens accordingly. In cross-section, the fault-based alteration aureoles tend to assume a mushroom-like shape, with the root developed around the plane of the parent fault.

In summary, based on the deduced evolution of the mantle --> crust heat input, it is logical to anticipate that both a magmatic center-based hydrothermal system and intermittently active fault-based hydrothermal systems were operational during the last 10^7 years. Initially, the Timber Mountain heat source could have supported prolonged activity of the magmatic center-based hydrothermal system. This system could have remained operational, say, up to $1-2 \times 10^6$ years after the eruption of the Ammonia Tanks Member of the Timber Mountain Tuff. Because the mantle --> crust heat input has remained high (although the advective component has steadily diminished), there is no reason to believe that, after

depletion of the Timber Mountain heat source, convective circulations of intracrustal fluids have ceased completely. Quite the opposite, it is only logical to presume that these circulations have continued intermittently at Yucca Mountain, assuming the form of a fault-based hydrothermal system driven by conductively replenished heat sources.

Second, at Yucca Mountain, the anticipated long-term behavior of the coupled fluid-heat flow system is clearly and unequivocally expressed and recorded through chemical, mineralogical, and radiometric characteristics of the local ignimbrites. In that regard, whole-rock alteration features are particularly relevant. The results of extensive chemical and mineralogical analyses of samples of the local ignimbrites reveal that, locally, at least two such features are present.

On the one hand, the Yucca Mountain ignimbrites display clear and unequivocal evidence of having experienced epigenetic alkali earth metasomatism. Such conditions are apparent through a comparison of the major and trace-element composition of the devitrified ignimbrites with that of the stratigraphically equivalent glasses. Relative to the latter, the former are enriched in a number of major and trace elements, especially including calcium, magnesium, and strontium. Importantly, the alkali earth metasomatism is apparent in both the vadose zone and in the "saturated" zone. The overall metasomatism appears to be

the combined result of two isotopically distinct hydrothermal processes, each involving allogenic calcium-magnesium fluid phases.

Deeper in the stratigraphic section (some 2000 feet below the topographic surface), the alkali earth metasomatism is pervasive and ubiquitous. Spatially, however, there are important differences in the degree of alkali earth element enrichment. For the northwestern segment of Yucca Mountain, the representative mol percentage of Ca + Mg cations is lower, about 10-15 percent. Typically, the stratigraphically equivalent glasses contain less than, say, 3 percent of the corresponding cations. For the southeastern sector, however, the representative mol percentage of Ca + Mg cations is decisively higher, say 25-30 percent. The results of whole-rock strontium isotopic analyses reveal that, in deeper parts of the northwestern sector, the observed alkali earth metasomatism was caused by fluids carrying low relative concentrations of dissolved radiogenic strontium-87. In terms of the isotopic character of dissolved strontium, these fluids were analogous to hypothetical fluids equilibrated with marine limestones of Early Paleozoic age. This observation may be taken as indicating that for deeper parts of the northwestern segment, the observed alkali earth metasomatism was caused by fluids that, in part, have acquired their isotopic and chemical characteristics through ionic and isotopic exchange reactions with the local underlying Paleozoic carbonates.

Higher in the stratigraphic section, however, alkali earth metasomatism is much less pervasive and ubiquitous. Interestingly, this metasomatism appears to be confined to alteration aureoles of varying thickness which, typically, are fault- and fracture-based. Such aureoles are particularly common and/or visible in both vitrophyres of the Topopah Spring Member of the Paintbrush Tuff. For the corresponding altered whole-rock ignimbrites, the representative mol percentage of Ca + Mg cations is sharply increased, and may be as high as 50-65 percent. Furthermore, in contrast to the pervasively altered ignimbrites from deeper parts of the northwestern sector, the strongly alkali earth enriched ignimbrites contain decisively higher relative concentrations of radiogenic strontium-87. It is clear, therefore, that fluids involved in the fracture-based alkali earth metasomatism must have acquired their isotopic and chemical characteristics, in part, through isotopic and ionic exchange reactions with a highly radiogenic reservoir of strontium. At and around Yucca Mountain, however, there is only one primary reservoir of appropriately radiogenic strontium. This reservoir underlies both the Tertiary ignimbrites and the Paleozoic carbonates and is the Pre-cambrian basement. Relative to fluids involved in the pervasive metasomatism, fluids involved in the fracture-based alkali earth metasomatism are clearly distinct and must have ascended from a greater depth.

On the other hand, the Yucca Mountain ignimbrites also display clear and unequivocal evidence of having experienced epigenetic hydrothermal

metamorphism. The observed spatial distribution of chemical, mineralogical, and radiometric characteristics of the resulting alteration minerals indicate that this metamorphism expresses and records hydrothermal activities associated with two chemically, isotopically, and temporally distinct alteration centers.

The northwestern center is older. The respective alteration aureole is associated with three groups of alteration minerals, namely: (1) higher-grade montmorillonitic clays, such as allevardite-kalkberg clays and discrete illites; (2) higher-grade alteration minerals from the alkali zeolitic series, specifically, analcime along with albite; and (3) predominantly sodic-potassic clinoptilolites. Both the K/Ar ages of the corresponding alteration minerals and the observed spatial arrangement of the corresponding isogradal surfaces indicate that the northwestern alteration aureole expresses and records the prolonged activity of a hydrothermal system driven by the Timber Mountain heat source. Importantly, the isotopic character of strontium dissolved in the respective fluids was identical to that incorporated in the Paleozoic carbonates. Evidently, the corresponding depth of fluid circulation was relatively shallow and insufficient to reach the Pre-cambrian basement.

The southeastern alteration center is younger. The respective aureole is also associated with three suites of alteration minerals, namely: (1) higher-grade zeolites from the calcic series, such as heulandite, laumontite, and analcime; (2)

predominantly calcic clinoptilolites; and possibly (3) higher-grade montmorillonitic clays, specifically the alleverdite clays. Both the broad range of the K/Ar ages of the corresponding clinoptilolites (from 8.5×10^6 to 2×10^6 years B.P.) and the observed spatial arrangement of these minerals indicate that the southeastern alteration aureole expresses and records the intermittent activity of a fault-based hydrothermal system driven by conductively replenished heat sources.

Importantly, the isotopic character of strontium dissolved in the respective hydrothermal fluids was significantly different than that incorporated in the Paleozoic carbonates. Specifically, these fluids were carrying high relative concentrations of dissolved radiogenic strontium-87. Evidently, the allogenic calcium-magnesium fluids associated with the fault-based hydrothermal system acquired at least some of their dissolved strontium from the Pre-cambrian basement.

In summary, the observed spatial distribution of characteristics of the alteration products (both the whole-rock ignimbrites and the separated alteration minerals) indicate that, at Yucca Mountain, the Timber Mountain alteration aureole has been overprinted by a subsequent alteration aureole. The latter aureole represents the intermittent activity of a fault-based hydrothermal system driven by conductively replenished heat sources. This overprinting aureole is rooted somewhere in the southeastern segment of Yucca Mountain, possibly in the Stage Coach Road fault zone. For shallow bedrock, the diffusion paths of the ejected

fluids (away from the parent fault) seem to have been controlled by deformation-induced gradients of extrinsic hydraulic conductivity, very much like those observed at the present time, Szymanski (1989). The resulting upwardly branching and spreading mushroom-shaped alteration aureola, documented only in the northwestern segment of Yucca Mountain, consists of a southeasterly thickening wedge, and appears to be rootless. Such seemingly restricted spatial development of this alteration aureole is illusory, and most certainly is an artifice of the spatially limited data base. It is predicted that by following the southeasterly thickening alteration wedge, the USGS investigators will eventually encounter the presently missing root of the overprinting alteration aureole.

Third, at Yucca Mountain, the anticipated long-term behavior of the coupled fluid-heat flow system is also clearly and unequivocally expressed and recorded through the spatial distribution of isotopic characteristics of the local epigenetic carbonatization. Importantly, this carbonatization is apparent throughout the entire 2 km of the explored section as well as at the topographic surface. Comprehensive analysis of the uranium, strontium, and carbon isotopic data reveals that, at Yucca Mountain, epigenetic calcites were produced in response to two isotopically distinct hydrothermal (hypogene) processes. On the one hand, in deeper parts of the stratigraphic section, epigenetic calcites occur in the form of sparry veins, interstitial impregnations, and replacements of phenocrysts; sometimes they are associated

with barite, fluorite, and pyrite. Typically, these calcites exhibit the following isotopic characteristics: (1) the lowest degree of oxygen-18 enrichment, with values of the $\delta^{18}\text{O}$ ratio ranging from +4.0 to about +12.0 per mil_{SMOW}; (2) the highest degree of carbon-13 enrichment, with values of the $\delta^{13}\text{C}$ ratio ranging from +4.5 to about -2.0 per mil_{PDB}; and (3) the lowest degree of strontium-87 enrichment, with values of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio ranging from 0.7086 to say 0.7090. Spatially, the corresponding calcites are correlative with the Timber Mountain alteration aureole, and clearly are of the same age. The isotopic characters of strontium and carbon incorporated in the Timber Mountain generation of calcites indicate that their parent fluids acquired their isotopic signatures from the underlying Paleozoic carbonates. Both the homogenization temperatures of fluid inclusions and the observed oxygen-18 depletion indicate that the Timber Mountain calcites were precipitated at temperatures in excess of 100° Celsius. In the northwestern segment of Yucca Mountain, near the southern margin of the Timber Mountain Caldera, these temperatures were as high as 250° Celsius, at a depth of about 1500 m. With increasing distance from the Timber Mountain Caldera, however, the crystallization temperatures become lower. In the vicinity of borehole USW G-3, these temperatures are about 100° Celsius.

On the other hand, in shallower parts of the stratigraphic section (a wedge-like section with depth increasing in a southeasterly direction, from 450 to about 900 meters), epigenetic calcites occur as micritic veins, commonly associated with

opal-CT, opal-A, sepiolite, and smectite. Typically, these calcites exhibit the following isotopic characteristics: (1) the highest degree of oxygen-18 enrichment, with values of the $\delta^{18}\text{O}$ ratio ranging from +13.0 to about +20.0 per mil_{SMOW}; (2) the lowest degree of carbon-13 enrichment, with values of the $\delta^{13}\text{C}$ ratio ranging from -3.0 to about -9.5 per mil_{PDB}; (3) the highest degree of strontium-87 enrichment, with values of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio ranging from 0.7108 to as much as 0.7130; and (4) values of the $^{234}\text{U}/^{238}\text{U}$ activity ratio near the secular equilibrium value of 1.0. Spatially, the corresponding calcites are correlative with the alteration aureole of the fault-based hydrothermal system. This spatial correspondence indicates that: (1) alkali earth and strontium-87 metasomatism, (2) calcic zeolitization, and (3) carbon-13 depleted and strontium-87 enriched carbonatization, were all developed concurrently, in response to activity of the same hydrothermal system. The combined K/Ar and U/Th ages further indicate that this system remained intermittently active for a very long period of time, from about 8.5×10^6 to as late as 3×10^4 years B.P. Importantly, the isotopic characters of strontium and carbon incorporated in the late epigenetic calcites also indicate that, for the fault-based hydrothermal system, the corresponding fluids must have acquired their isotopic characteristic deep in the crust. The reservoir of dissolved strontium is the Pre-cambrian basement; for dissolved carbon, however, the source appears to be deep-seated igneous carbon dioxide. The contrasting isotopic characters of dissolved strontium and carbon indicate that, relative to fluids involved in the Timber Mountain carbonatization, fluids involved in the late

carbonatization have evolved much deeper in the crust.

As far as geothermal circumstances of formation of the late epigenetic calcites are concerned, the depth distribution of the oxygen isotopic ratio indicates that these calcites were crystallized under temperatures that were decisively higher than those observed at the present time. Although these crystallization temperatures remain unknown, it is clear that during formation of the late epigenetic calcites, the local geothermal gradient was at least 50 percent higher than the contemporary gradient. The homogenization temperatures of fluid inclusions from shallow calcites (which in borehole USW GU-3 are spatially associated with the carbon-13 depleted calcites) suggest that some of the late epigenetic calcites also may have crystallized at temperatures above boiling. The presently known homogenization temperatures of fluid inclusions from the shallow calcites range from 101° to as much as 227° Celsius. In a near surface environment, however, such above-boiling temperatures may not exist for a prolonged period of time. This is so because heat contained in ascending fluids dissipates very rapidly into both the cool surrounding rock and the atmosphere. Furthermore, relatively short-lived circulation of fluids that are expected to be associated with conductively replenished hydrothermal systems do not promote nor allow for a spatially extensive and prolonged heating of a shallow bedrock. Consequently, it may reasonably be expected that, near the topographic surface, the late epigenetic calcites with crystallization temperatures significantly higher than ambient

temperatures exist only locally, along and adjacent to the primary fluid diffusion paths. The majority of the late calcites may be expected to have been crystallized slightly above ambient temperatures, just enough to allow for recognition of their hypogene origin.

Fourth, at Yucca Mountain, the local activity of a conductively replenished hydrothermal system is independently indicated by the results of considering the origin of two additional kinds of specimens, one fluid and the other solid. These are the vadose zone interstitial fluids and the "mosaic" breccias, respectively.

On the one hand, the vadose zone interstitial fluids display geochemical characteristics indicating that, partly, they are remnant and strongly allogenic hydrothermal fluids. In terms of stable and radiogenic isotopes (carbon and tritium, respectively), they appear as fairly young meteoric fluids, which have acquired dissolved carbon from a local biogenic source. In terms of both the base and noble metal abundances and the REE abundance pattern, however, the vadose zone interstitial fluids appear as fluids which have evolved outside of the Tertiary ignimbrites in a CO₂-rich environment. Furthermore, in terms of the relative and absolute abundances of the major dissolved constituents, these fluids are strikingly similar to the mature calcium-magnesium fluids which, at and around Yucca Mountain, are indigenous to the Paleozoic carbonates. The conflicting

appearances of the vadose zone interstitial fluids are satisfactorily reconciled by assuming that these fluids are two-component mixtures. One component represents infiltrating meteoric fluids, and the other component represents much older hydrothermal fluids. The intermixing of these two components and the resulting acquisition of both the man-made tritium and the biogenic carbon occurs in-situ and involves the vapor phase as well as the liquid phase. In part, the intermixing and the resulting fluid exchanges appear to be facilitated by the observed circulations of vadose zone gases. During no-recharge periods, these circulations cause a fracture-based "drying" of the vadose zone rocks. This "drying" process involves vapor phase and, consequently, only pure (distilled) water is removed. The dissolved solids, however, remain behind. During episodic recharge events, the evaporative losses are compensated for by an influx of tritium- and biogenic carbon-carrying rainwater. The result of such back and forth fluid exchanges is two-fold, namely: (1) the vadose zone interstitial fluids evolve isotopically, with time, becoming very much like infiltrating rainwater in some respects, and (2) being strongly held by capillary forces, these fluids remain immobile and, while exchanging vapor phases, retain their initial dissolved solids. As far as the origin of the hydrothermal component of the interstitial fluids is concerned, the strongly alkali earth bulk composition indicates that this component consists of fluids remarkably similar to those that concurrently were involved in: (1) alkali earth metasomatism; (2) calcic zeolitization; and (3) late carbonatization. It is permissible and reasonable, therefore, to assume that, at

Yucca Mountain, the vadose zone interstitial fluids are remnants of fluids associated with the intermittent activity of a conductively replenished hydrothermal system.

On the other hand, the Yucca Mountain "mosaic" breccias display geochemical, isotopic, and radiometric characteristics indicating that these breccias were also formed in association with the intermittent activity of a conductively replenished hydrothermal system. This association is indicated by the isotopic characters of carbon and oxygen incorporated in the authigenic cements of the "mosaic" breccias. These characteristics (being analogous to those incorporated in the carbon-13 depleted and strontium-87 enriched calcites) indicate that the allogenic calcium-magnesium fluids associated with the conductively replenished hydrothermal system, in addition to being involved in the metasomatic alteration and carbonatization, were also involved in the intermittent explosive fragmentation of local bedrock. This paragenetic relationship is confirmed by the fission-track ages of allogenic zircons incorporated in the "mosaic" breccia cements. These ages indicate that the explosive fragmentation of bedrock occurred sometime during the last 4.8×10^6 years B.P. Importantly, the hydrothermal and explosive mode of formation of the Yucca Mountain "mosaic" breccias, as independently inferred herein, is in accord with conclusions drawn by Hanson et al. (1986). Based on the results of direct field observations, these authors have diagnosed the Trench 14 "mosaic" breccias as "hydrothermal eruption breccias."

Fifth, at Yucca Mountain, a critically important element associated the currently active hydrothermal processes is the involvement of carbon dioxide from a deep-seated igneous source. Recognition of a possible role this gas may play in the development of the carbon-13 depleted and strontium-87 enriched carbonatization has led one member of the U.S. Department of Energy staff to conclude that "*Szymanski may have seriously, if not grossly, underestimated the hazardous conditions at Yucca Mountain in his 1989 report.*"

Concerns regarding the possible involvement of gaseous carbon dioxide were also voiced by Archambeau and Price (1991). The following discussion is intended to summarize the factual bases that underlie these concerns. One of the most intriguing isotopic characteristics of the late epigenetic calcites, including the authigenic cements of the "mosaic" breccias, is the fact that these calcites contain isotopically "light" carbon. Based on the corresponding values of the $\delta^{13}\text{C}$ ratio, it may be inferred that, in a pristine state (i.e., prior to the near-surface CO_2 degassing, and the resulting carbon-13 diffusional enrichment), the parent fluids for the late epigenetic calcites were carrying values of the $\delta^{13}\text{C}$ ratio ranging from -9.0 to about -11.0 per mil_{PDB}. Typically but not uniquely, such low relative concentrations of dissolved carbon-13 are characteristic of hydrothermal fluids which contain CO_2 from igneous sources. These low concentrations may be explained by assuming conservatively that igneous $\text{CO}_{2(\text{gas})}$ (with $\delta^{13}\text{C}$ ratio ranging from -5.0 to about -8.0 per mil_{PDB}) was dissolved at a temperature of about 200° Celsius. For this temperature, the value of the $\text{HCO}_3^- \leftarrow \text{CO}_{2(\text{gas})}$

fractionation factor is about -4.0 per mil_{PDB}. The fact that the isotopically "light" carbon is incorporated in the authigenic cements of the "mosaic" breccias is of extraordinary importance. Specifically, this fact suggests that the explosive mode of formation of the "mosaic" breccias, as independently inferred by a number of experts, is the result of so-called gas-assisted hydrothermal eruption and/or seismic pumping. The isotopic characters of incorporated carbon and oxygen, the fission-track ages of allogenic zircons, and the multistage development, all unanimously indicate that, at Yucca Mountain, the "mosaic" breccias express and record the intermittent activity of a conductively replenished hydrothermal system. In association with such a system, however, intermittent and possibly explosive degassings of CO₂ accumulated at depth are not surprising and are to be expected providing, of course, that a deep-seated source of CO₂ is present locally. Both the great depth of circulation and the intrinsically transient behavior have the result that, for a fault-based and intermittently active hydrothermal system, rapid CO₂ degassings are feasible, particularly in magmatically active regions. This is true because (1) presence of fluids in a deep substratum, where CO₂ solubility in water is greatly enhanced, makes it possible for igneous CO₂ to accumulate, and (2) transient circulations influence CO₂ solubility and may lead to catastrophic positive feedback releases of the accumulated gas.

In light of these remarks, it is entirely possible that, at Yucca Mountain, intermittent and rapid CO₂ degassing is the ultimate cause that underlies both

the observed dilatory fragmentation of bedrock and the subsequent and seemingly very fast cementation of this bedrock. While exploring this speculation further, it is critically important to be absolutely certain that, indeed, some of the locally circulating isotopically "light" carbon is being provided by a deep-seated igneous source. In that regard, two observations seem to be particularly noteworthy. On the one hand, the radiometric ages of both the late epigenetic calcites and the mafic volcanic rocks indicate that, at Yucca Mountain, episodes of carbon-13 depleted carbonatization were contemporaneous with episodes of mafic magmatism. Such a state of affairs is indicated by the fact that, for the last 10^6 years, all five of the locally recognized magmatic episodes were accompanied by temporally equivalent episodes of carbon-13 depleted carbonatization. As far as the origin of isotopically "light" carbon is concerned, this remarkable correspondence indicates that, rather than being derived from local biogenic sources, this carbon was derived from a local igneous source instead. On the other hand, the isotopic character of carbon dissolved in the contemporary calcium-magnesium fluids is significantly different from that incorporated in marine limestones in which these fluids reside. For fluids indigenous to the Paleozoic carbonates, observed values of the $\delta^{13}\text{C}$ ratio range from -4.2 to -5.2 per mil_{PDB}. For hypothetical fluids equilibrated with marine limestones, however, the expected values fall within a limited range, from +2.0 to no less than -2.0 per mil_{PDB}. This well expressed and important isotopic discord clearly and unequivocally indicates that, at and around Yucca Mountain, the mature

calcium-magnesium fluids (i.e., those that are chemically and isotopically equivalent to the parent fluids for the concurrent alkali earth metasomatism, calcic zeolitization, and carbon-13 depleted carbonatization) are being intermixed and contaminated with fluids that acquire dissolved carbon from an isotopically "light" source. There are only two conceivable sources of such carbon, namely a biogenic source and a deep-seated igneous source. With respect to the Nevada Test Site calcium-magnesium mature fluids, however, a biogenic source for carbon dissolved in the contaminating fluids does not appear a reasonable proposition. This is so because of the following three reasons: (1) the carbon-14 activity is very low, typically less than 3 percent of the modern carbon standard; (2) the tritium activity is also very low, typically less than 3TU; and (3) the abnormal carbon-13 depletion is accompanied by a likewise abnormal enrichment in radiogenic strontium-87. The latter clearly and unequivocally indicates that, in addition to tapping a carbon-13 depleted source, the contaminating fluids are also tapping a highly radiogenic reservoir of strontium. Around Yucca Mountain, however, appropriately radiogenic strontium is incorporated in only one primary reservoir, which is the Pre-cambrian basement. In view of this observation, a deep-seated origin of some of the carbon dissolved in the mature calcium-magnesium fluids is the only logical explanation remaining. The above two observations may be collectively taken as indicating that an igneous origin of carbon incorporated in the carbon-13 depleted authigenic cements of the "mosaic" breccias, in addition to being a permissible option, also appears to be an independently verified fact.

Sixth, by even the most stringent standards of geologic judgment, the above five concluding remarks are well supported by the presently known facts, and thus must be regarded as reasonably secure. It is clear that a number of independent lines of evidence lead to the same conclusion, namely, at Yucca Mountain, and for time spans measured in 10^4 years, hydrothermal and auxiliary gas-assisted processes pose a significant hazard. With regard to the long-term performance of a high-level nuclear waste repository, such a hazard cannot be mitigated and reduced to prudent and acceptable levels. In view of the evidence presented herein, full implementation of the site characterization program at Yucca Mountain must be regarded as futile and wasteful. Further, in the context of the evidence presented herein, one may remain hopeful that continued efforts by some members of the USGS staff to deliberately discredit and nullify the recognition of the manifestly prohibitive site conditions will be rightly regarded as self-serving but otherwise pointless.

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