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October 27, 2003

Dr. Michael L. Corradini, Chairman  
Nuclear Waste Technical Review Board  
2300 Clarendon Blvd., Suite 1300  
Arlington, VA 22201-3367

Dear Dr. Corradini:

Enclosed you will find a copy of a document entitled "Review Of The Report 'Thermochronological Evolution Of Calcite Formation At The Potential Yucca Mountain Repository Site, Nevada'" published under the auspices of the Siberian Branch of the Russian Academy of Sciences United Institute of Geology, Geophysics and Mineralogy, which was authored by two members of the Institute of Mineralogy and Petrography, Dr. Yuri V. Dublyansky and Dr. Sergey Z. Smirnov. The document is a review of a two part report written by UNLV coordinators, Nick Wilson, Jean Cline and Y. Amelin, of the Yucca Mountain Thermochronology Project, a project conducted in response to a suggestion by the Nuclear Waste Technical Review Board in 1998 to resolve differences in the interpretation of certain fluid inclusion and stable isotope data, which had spawned a heated controversy between scientists representing the State of Nevada and those representing the interests of the DOE (primarily the USGS) concerning the origin and ages of secondary minerals in the interior of Yucca Mountain.

Dr. Dublyansky was Nevada's representative for the UNLV Thermochronology Project. Except for DOE representatives' concession that the secondary minerals in question were, indeed, formed from heated aqueous fluids, the disagreement between the scientists, particularly the source of the heat, has persisted to the present day. The DOE aligned interests still maintain that the source of the depositing fluids was meteoric water in the form of infiltrating rainwater passing through a mountain that remained hot for millions of years. Dr. Dublyansky and a group of internationally based scientists working with him, which include many of his colleagues at the Institute, Jerry Szymanski of Las Vegas, Nevada and Dr. Tim Harper of England are convinced, based on many lines of evidence, that the secondary minerals were deposited by hydrothermal fluids driven from deep beneath Yucca Mountain and that episodes of such deposition are recent in geologic time. If hydrothermal fluids were to flood the proposed repository during its 10,000-year lifetime or even an extended period of many tens of thousands of years, steam explosions would undoubtedly result and the canisters would be breached. As the fissile material is rearranged tremendous quantities of radioactivity would be released through a variety of pathways to the biosphere, not the least of which are those created by predictable low yield nuclear explosions and uncontrollable *in situ* criticality processes.

In a letter written to the NWTRB by the Yucca Mountain Project Manager, J. Russell Dyer,

dated January 24, 2002, the lack of a consensus in the lingering rainwater-upwelling controversy was documented. Inexplicably, however, NWTRB Chairman Jared Cohon wrote a letter addressed to Mr. Lake H. Barrett, Acting Director of OCRWM dated March 11, 2002, which stated:

At the Board meeting and in a letter to the Board dated January 24, 2002, the DOE concluded that the hypothesis of hydrothermal upwelling proposed by Mr. Jerry Szymanski had been adequately addressed and may be discounted. These conclusions were based on the DOE's positive response to a Board recommendation that a joint federal-State of Nevada project be conducted to determine the ages of fluid inclusions at Yucca Mountain. A systematic joint study was coordinated by University of Nevada-Las Vegas scientists and can be considered a model for successful resolution of some contentious scientific issues. The Board concurs with the DOE's conclusions and considers this issue resolved.

The important point to recognize with respect to the foregoing communications is that they contain nothing more than political opinion. The decision whether or not the controversy is scientifically resolved is a technical issue related to the safety of the site, which is committed to the jurisdiction of the Nuclear Regulatory Commission's licensing board. The NRC is the sole entity responsible for safety considerations concerning the licensing of the Yucca Mountain site. Furthermore, the decision whether or not the State of Nevada will raise a contention based upon the continuing controversy is a question, which rests solely with the Nevada Attorney General. The bottom line is that the controversy is resolved neither politically nor scientifically.

Other political statements such as the one attributed to you as the consequence of your recent co-authorship of an editorial in a Madison, Wisconsin newspaper that in your opinion nuclear waste can be "stored safely at Yucca Mountain" are counter productive in the effort to provide the world community with a fair and unbiased process. Since that bell cannot be unringed, an appropriate strategy for the mitigation of the effects of the dissemination of misinformation might come in the form of reopening the scientific review of the origin and ages of the secondary minerals at Yucca Mountain before the NWTRB.

An unbiased consideration of reasonable interpretations, which may be attributed to data acquired during the UNLV Thermochronology Project, is warranted. A number of questions, which were raised by Board members, regarding findings by the Thermochronology Project in a meeting of the full Board on May 9, 2001 need to be resolved. Among these were questions raised concerning the source of magnesium found in samples of secondary minerals, the source of hydrocarbons in all gas inclusions, an explanation for the high salinities in the fluids of the inclusions, the use of a constant lead correction for uranium-lead age dating, thermodynamic limitations to the rainwater hypothesis, etc.

The review authored by Dr. Dublyansky and Dr. Smirnov enclosed herein and a second review authored by them: "Commentary on: 'Physical and stable-isotope evidence for formation of secondary calcite and silica in the unsaturated zone, Yucca Mountain, Nevada' by J.F. Whelan, J.B. Paces, and Z.E. Peterman" (submitted for publication in Applied Geochemistry, a peer-reviewed journal) as well as the reports of the USGS and UNLV researchers regarding their interpretations of the data produced by the UNLV Thermochronology Project can provide valuable resources to define the issues. The position of the international group of scientists referred to above will be fully discussed in a book length monograph presently in a draft format pending review, which will contain multiple lines of evidence proving without question that the deposition of the secondary minerals was caused by the upwelling of hydrothermal water.

The NWTRB has the statutory mandate in Section 503 of the NWPA, 42 U.S.C. 10263, to

evaluate the technical and scientific validity of activities undertaken by the Secretary of Energy in relation to, among other things, site characterization activities. This broad grant of authority provides the Board with the power and the duty to oversee the DOE's consideration of potentially disruptive events such as the possible flooding of the proposed repository by upwelling water and to intervene with appropriate admonitions and recommendations to the Department of Energy. It is a dereliction of this duty for the Board to disregard its mandate by leaving contentious issues affecting the performance of the proposed repository left unresolved.

The Board also has the duty to report to the Congress and the Secretary of Energy with regard to findings, conclusions and recommendations as to matters within its purview. See 42 U.S.C. 10268. To the extent the Board has prematurely terminated consideration of the need for a comprehensive risk assessment of potential consequences associated with the controversy discussed herein, it appears that both the Secretary of Energy and the Congress have been misled by previous reports from the Board. Eventually, evidence of the dangerous nature of the site will certainly cause the abandonment of the site. At that time certain individuals and entities will be held accountable for the expenditure of billions of dollars and, more importantly, years of lost time in the resolution of a pressing national environmental problem. There will be plenty of blame to go around. Unless the NWTRB takes steps to rectify its past nonfeasance, it will likely become the scapegoat for the misfeasance of many.

I commend the enclosed review for your careful consideration and appropriate action.

Cordially,

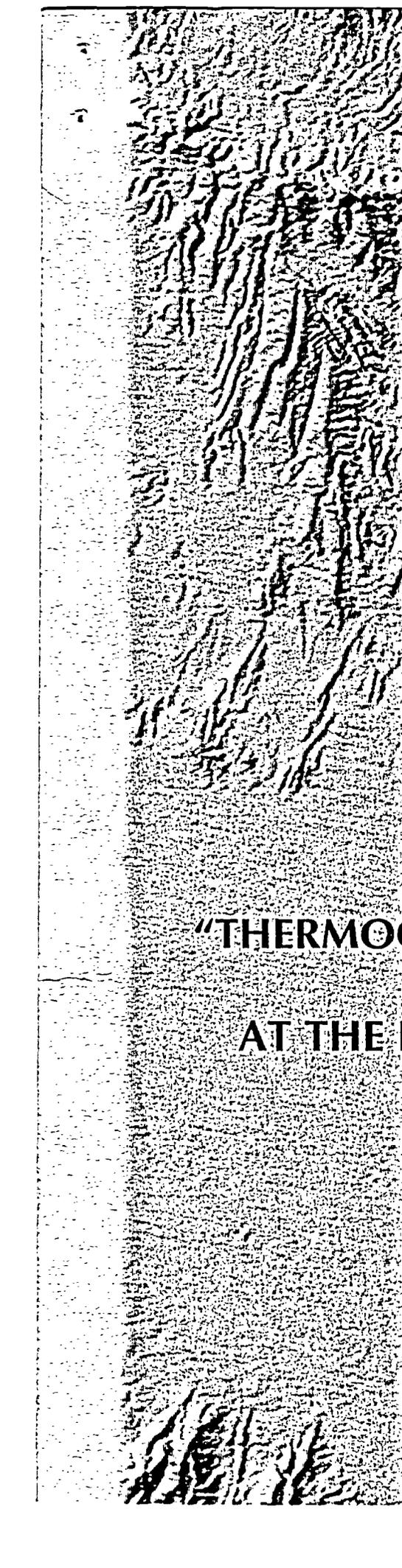
  
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Enclosure

cc:

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The Honorable Kenny Guinn, Nevada Governor, Carson City, NV  
The Honorable Harry Reid, Nevada Senator, Washington DC  
The Honorable John Ensign, Nevada Senator, Washington DC  
The Honorable Jim Gibbons, Nevada Representative, Washington DC  
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William J. Broad, New York Times, New York, NY



**Yuri V. Dublyansky and Sergey Z. Smirnov**

**REVIEW OF THE REPORT  
"THERMOCHRONOLOGICAL EVOLUTION  
OF CALCITE FORMATION  
AT THE POTENTIAL YUCCA MOUNTAIN  
REPOSITORY SITE, NEVADA"**

**NOVOSIBIRSK  
2003**



**SIBERIAN BRANCH OF RUSSIAN ACADEMY OF SCIENCES**

**UNITED INSTITUTE OF GEOLOGY, GEOPHYSICS  
AND MINERALOGY**

**INSTITUTE OF MINERALOGY AND PETROGRAPHY**

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**REVIEW OF THE REPORT: "THERMOCHRONOLOGICAL EVOLUTION  
OF CALCITE FORMATION AT THE POTENTIAL YUCCA MOUNTAIN  
REPOSITORY SITE, NEVADA"**

**Reviewed by:**

**Dr. Yuri V. Dublyansky and Dr. Sergey Z. Smirnov**



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Publishing House of SB RAS**

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**2003**

**Review of the report: "Thermochronological Evolution of Calcite  
Formation at the Potential Yucca Mountain Repository Site,  
Nevada"**

**Part 1. Secondary Mineral Paragenesis and Geochemistry  
Report TR-02-005.1. 2002, 43 p.**

**By N.S.F. Wilson and J. Cline**

**and**

**Part 2. Fluid Inclusion Analyses and U-Pb Dating  
Report TR-02-005.2. 2002, 52 p.**

**By: N.S.F. Wilson, J. Cline, and Y. Amelin**

**Reviewed by:**

**Dr. Yuri V. Dublyansky and Dr. Sergey Z. Smirnov**

**ISBN 5-7692-0588-1**

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## Executive summary

The reviewed UNLV report and, particularly, the technical data files posted at the UCCSN WebPages, contain a large volume of meticulously collected observations and measurements of fluid inclusion homogenization temperatures. Other data, not directly related to fluid inclusions, are presented in an overly abbreviated form. For example, the numeric results of the microprobe analyses are not given in the report and are not tabulated in the technical data files. The results of the LA-ICP-MS are not reported at all, although contents of such elements as U, Th, Sr and Mn have been measured (Wilson and Cline, 2002, p. 17). The results of the cathodoluminescence studies are not documented in the report (except for a single photograph) and documentation is inadequate in the technical data files. These technical shortcomings cause most of the geochemical information to be largely unusable, and does not permit the independent verification of a number of statements that appear in the report.

We found that the mineralogic descriptions given in the report are overly generalized, and those in the technical data files are too sketchy. The presence of at least one mineral identified by the UNLV researchers, strontianite, was not reported either in the report's text or in the technical data files (identification of this mineral was confirmed by personal communication with N. Wilson, 2001). Another mineral, barite, although reported in the technical data files, was not mentioned in the report. The paragenetic relationships between these accessory minerals (as well as accessory zeolite) and other major minerals (calcite and silica) were not described or discussed. Neither the genetic significance of the observed accessory minerals nor how their presence constrains the chemistry of the mineral forming fluids was discussed.

A more serious problem with the UNLV report is the interpretation of the data: No rational models of the processes responsible for the formation of secondary minerals at Yucca Mountain are proposed; their formation in the vadose zone is simply asserted. In many instances, the causal nexus between processes and features discussed by the UNLV researchers cannot be traced (see Section 1.2 of this review, for example). Even more problematic, some speculative models appear to be in conflict with the principles of physics (see Section 1.3.2, for example). Overall, the authors of the UNLV report have failed to develop a coherent and defensible model, explaining the origin of secondary minerals at Yucca Mountain. Rather, they arbitrarily subscribe to the DOE/USGS model of a rainwater origin.



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## Preface

Between April 1999 and August 2001, Yuri Dublyansky served as an official representative of the State of Nevada, overseeing the progress of the Yucca Mountain Thermochronology Project, a U.S. DOE funded project that was being conducted by researchers at the University of Nevada at Las Vegas (UNLV). Early independent studies of fluid inclusions in the Yucca Mountain secondary minerals had been conducted by two research groups, the first from the Institute of Mineralogy and Petrography, Russian Academy of Sciences, Novosibirsk (IMP) represented the State of Nevada, and the second from the U.S. Geological Survey (USGS). These studies yielded conflicting results and interpretations (see e.g., Roedder et al., 1994; Roedder and Whelan, 1998; Dublyansky and Reutski, 1995; Dublyansky, 1998). In response to a suggestion by the Nuclear Waste Technical Review Board (NWTRB) in July 1998, the U.S. DOE made an offer to the Nevada Agency for Nuclear Projects/ Nuclear Waste Projects Office (NWPO) in September 1998 to enter into a joint investigation to resolve the differences in the findings of the two groups. Together, in concert with the UNLV researchers, the NWPO and DOE initiated the UNLV project to provide a third set of data, which would verify the results of either one or the other of the previous fluid inclusion studies. Researchers from the USGS elected to carry out a parallel study; researchers from the IMP were commissioned by the State of Nevada's Agency for Nuclear Projects to pursue separate studies of the subject minerals. Under this arrangement, IMP researcher, Yuri Dublyansky, carried out fieldwork, as well as laboratory fluid inclusion and stable isotopic studies; another researcher Sergey Smirnov, collected samples and conducted mineralogic and geochemical studies of the Yucca Mountain minerals.

Pursuant to the contract with the State of Nevada, Yuri Dublyansky attended quarterly technical meetings of the UNLV and the USGS researchers. He maintained a detailed record of the activities of the group, which is contained in a series of letter reports, memos, and minutes from meetings, submitted to the Agency for Nuclear Projects. Although not stipulated in Dr. Dublyansky's contract, a critical evaluation of the final report(s) detailing the outcome of the UNLV project would have been a logical conclusion to the State's oversight of the project. Since the final reports were not filed by the UNLV researchers until May 2002, and were available for downloading from the UNLV Internet site even much later (i.e., well after the expiration of Dr. Dublyansky's contract with the State of Nevada), this task was not accomplished.

The evaluation report presented below is an unsolicited document. It has been prepared on a pro bono publico basis by the researchers of the Institute of Mineralogy and Petrography – an entity within the United Institute of Geology, Geophysics and Mineralogy, Siberian Branch of Russian Academy of Sciences, Novosibirsk, Russia.

The authors of this evaluation have reviewed the two parts of the UNLV report (TR-02-005.1 and TR-02-005.2) prepared by the UNLV researchers, as well as a collection of the supporting technical data files entitled «Compilation of Task 4 Data» by Dr. N. Wilson, submitted to the Harry Reed Center of the UNLV on December 24<sup>th</sup> 2001. The UNLV report has been downloaded from the University and Community College System of Nevada (UCCSN) WebPages (<http://hrcweb.nevada.edu/qa/Tech.htm>). Technical data files have been downloaded from the «non-qualified data» section of the same WebPages (<http://hrcweb.nevada.edu/data/tda/>).

We focus our review on what we see as major problems with the UNLV reports that affect, quite severely in our judgment, the soundness of the conclusions reached by the UNLV researchers. Numerous minor technical lapses and inaccuracies in presentation and interpretation of the data are not addressed.

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## Commentary: Handling dissent in science

In his letter to Dr. Jared L. Cohon, Chairman of the Nuclear Waste Technical Review Board (NWTRB) dated January 24, 2002 the Yucca Mountain Project Manager, Russel Dyer, outlined the scope and organization of the UNLV project as follows:

As the Board suggested, the DOE funded a joint research program coordinated by Dr. Jean Cline, University of Nevada, Las Vegas (UNLV) in which scientists from the State of Nevada, the U.S. Geological Survey (USGS) and UNLV conducted detailed analyses of the fluid inclusions found in mineral deposits. Participants met on a regular basis between March 1999 and March 2001 to establish a common methodology for sample collection and handling and share the results of their investigations.

The scientific outcome of the project was summarized in Mr. Dyer's letter by the following three paragraphs:

*Paces et al state in the abstract of the 2001 USGS report «Ages and Origins of Calcite and Opal in the Exploratory Studies Facility Tunnel, Yucca Mountain, Nevada»: The physical and isotopic data from calcite and opal indicate they formed from solutions of meteoric origin percolating through a limited network of connected fracture pathways in the unsaturated zone rather than by inundation from ascending groundwater originating in the saturated zone.*

*Wilson, Cline, and Amelin state in the abstract for Part 2 of their report: Results from this study are consistent with a model of descending meteoric water that infiltrated the cooling tuff sequence, became heated, and precipitated secondary minerals within the vadose zone. And further, This study demonstrates that the hypothesis of geologically recent upwelling hydrothermal fluids is untenable and should not disqualify the Yucca Mountain as a potential nuclear waste storage site.*

*The position on this issue by scientists representing the State of Nevada seems unchanged. In a pre-publication excerpt from the «Scientific status of the lingering "upwelling water" controversy in light of the joint UNLV/USGS/State of Nevada research project» by Jerry S. Szymanski and Dr. Yuri V. Dublyansky, May 2001, pp. 19, «The proposed conceptual model implies that vadose zone is occasionally subjected to an upward flux of heat and gas-charged fluid, in addition to being subjected to a small flux of infiltrating rainwater.»*

It is apparent from the excerpts of Mr. Dyer's letter above that the parties to the Project have failed to reach a consensus as to the origin of secondary minerals at Yucca Mountain. What is particularly unusual and alarming, however, is that diametrically opposite interpretations emerged from parallel studies of the same collection of samples by virtually identical methods. Under such circumstances, we believe, the responsible course of action for the U.S. DOE would be to arrange for a thorough evaluation of both the quality of the data produced by the parties, and the soundness of the interpretations that have lead the scientists to their conclusions.

This procedure, known as peer review, is a hallmark of academic science. Such an approach is dictated by common sense. If two groups of qualified researchers, studying the same samples by the same methods, have produced substantially different results, one of the groups must have done something wrong. Until the cause is found and the discrepancy is explained, the results reached by both groups should be viewed with suspicion. If, however, the technical results are similar but the conclusions based on these results are different, the soundness of the reasoning and interpretations must be evaluated.

It seriously concerns us that, instead of pursuing a proper course of evaluation and verification, the DOE hastily embraced the conclusions of the UNLV and USGS groups and bluntly dismissed the dissenting opinion of scientists representing the State of Nevada. Russell Dyer, DOE's Yucca Mountain Project Manager, in a letter dated January 24, 2002 addressed to Jared L. Cohon, Chairman of the NWTRB stated: *«The data collected by both DOE and UNLV researchers confirm that the conceptual model of descending percolation is correct. The DOE further concludes that the «upwelling waters» or «seismic pumping» hypotheses for the origin of secondary mineralization at the Yucca Mountain site have been adequately addresses and may be discounted.»* (Dyer to Cohon, 01.24.2002). We find this attitude both arrogant and inappropriate.

It is scientifically unacceptable to read only the conclusions of the reports and to judge which of the proffered hypotheses is correct on the basis of the number of «votes cast in support» of the competing models. Back in 17<sup>th</sup> century, a famous philosopher and mathematician, Rene Descartes, warned against the futility of such an approach: *«It would be no use to total up the testimonies in favour of each, meaning to follow that opinion which was supported by the greater number of authors; for if it is a question of difficulty that is in dispute, it is more likely that the truth would have been discovered by few than by many.»* (Descartes, Rules for the Direction of the Mind).

It is particularly disturbing to us that the NWTRB, an entity charged with a high public calling and, as a consequence of their duty, needs to be respected for integrity and open-mindedness, seems to have adopted the same mode of perfunctory inquiry. A letter from Jared L. Cohon, Chairman of the NWTRB, addressed to Mr. Lake H. Barrett, Acting Director Office of Civilian Radioactive Waste Management of the U.S. Department of Energy dated March 11, 2002 reads: *«At the Board meeting and in a letter to the Board dated January 24, 2002, the DOE concluded that the hypotheses of hydrothermal upwelling proposed by Mr. Jerry Szymanski had been adequately addressed and may be discounted. These conclusions were based on the DOE's positive response to a Board recommendation that a joint federal-State of Nevada project be conducted to determine the ages of fluid inclusions at Yucca Mountain. A systematic joint study was coordinated by University of Nevada-Las Vegas scientists and can be considered a model for successful resolution of some contentious scientific issues. The Board concurs with the DOE's conclusions and considers this issue resolved.»*

With due respect, we disagree. As far as science is concerned, the issue is far from being resolved by the UNLV study. The validity of the USGS-UNLV depositional model was not proved. DOE's rush to judgment was self-serving. The suggested invalidity of the hydrothermal upwelling hypothesis was not demonstrated by the study. The DOE's conclusion, in this regard, was equally self-serving, and on a larger scale, potentially dangerous. We believe that a critical evaluation of the UNLV report and the technical data presented below will provide ample justification for our opinions.

### **Hydrothermal upwelling concept and phreatic environment**

It appears to us that many of the contentions that will be treated in this review stem from a misunderstanding, by the authors of the UNLV report, of the hydrothermal upwelling concept in addition to their arbitrary treatment of the terminology: phreatic environment. In order to clarify the sub-

ject matter, we precede our review with a brief explanation of the hydrothermal upwelling concept. The subject is treated exhaustively in Szymanski *et al.* (2002); we refer the interested reader to that source for a more complete explication of the concept.

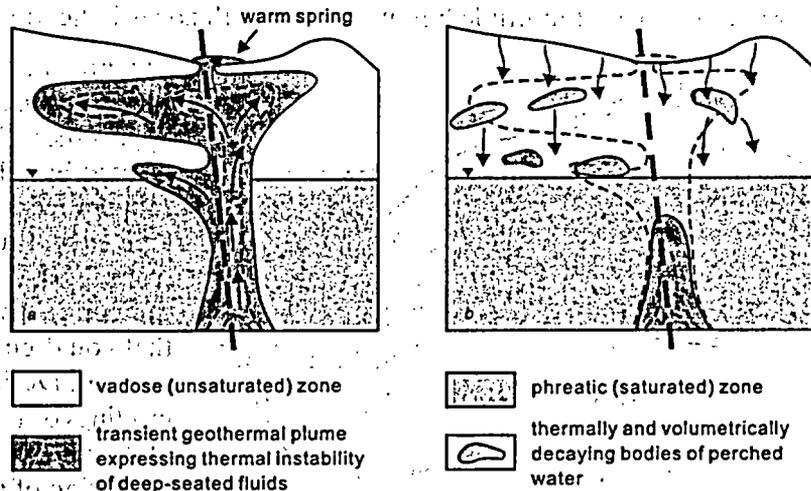
The hydrothermal upwelling concept does not envisage a more or less uniform rise of the water table, so that the whole of the Yucca Mountain tectonic block (horst) becomes flooded. Instead, the concept envisages relatively short-term invasion(s) of deep-seated fluids into the vadose zone that follows a large («characteristic») earthquake (Figure 1). The invasion is induced by the establishment within a fault-based conductivity channel of a so-called Rayleigh-Bernard instability (convective cell) and is enhanced by a mechanism known as seismic pumping. The hydraulic «mound», formed in this way, is necessarily a transient feature, so that the dissipation of the mound begins almost simultaneously with its establishment. During the decay of the mound, the waters are generally controlled by gravitation and move downward, toward the water table.

The deep-seated fluids, which are injected into the vadose zone along the fault-based enhanced conductivity channel move laterally away from the channel. As they move they evolve; they cool, degas, oxidize, mix with other waters and deposit minerals. Their path is primarily controlled by the conductivity structure of the host rock mass; therefore, not all cavities in the rock are necessarily accessed by the fluids; and fluids do not deposit minerals in all cavities that are accessed.

In the context of the model, the term, phreatic environment, indicates a situation whereby the cavities in which minerals grow are filled with water, so that the growth occurs in a submerged state. This may happen within transient water bodies located well above the static water table. This approach parallels the terminology employed in karst science, where two broad categories of cave formations, subaerial and subaqueous, are distinguished based on their formation above or below the surface of the water. Deposits acquire characteristic features of subaqueous mineral growth even if they form in a small cave pond, located thousands of meters above the regional water table.

Figure 1. Schematic diagram illustrating changing hydrology of vadose zone at a time following the occurrence of characteristic earthquake, «hydrothermal upwelling concept» (from Szymanski *et al.*, 2002).

*a* – Seismic pumping-induced and Rayleigh-Bernard instability assisted invasion of waters into the vadose zone along the fault-based conductivity channel. *b* – Decay of the mound, associated with formation of perched bodies of water, gravitation-driven filtration, and interaction with meteoric (rain) waters.



Characteristic features of the model: 1. Upwellings of thermal fluids are restricted to deep-seated horst-bounding faults, such as Solitario Canyon and Paintbrush. 2. Both upward and downward movements of geothermal fluids occur in the vadose zone. 3. Asymmetry of the diffusing plume is a result of the near-surface enhancement of the conductivity, mostly affecting the hanging wall block. 4. The local temperature gradient in the vadose zone expresses the transient nature of the instability. 5. At and around the fault-based conductivity channel, cavities in the rock are completely lined with minerals, whereas away from this channel the mineral distribution in cavities may be an expression of the dissipation of perched bodies of water.

# 1. REVIEW OF THE REPORT: THERMOCHRONOLOGICAL EVOLUTION OF CALCITE FORMATION AT THE POTENTIAL YUCCA MOUNTAIN REPOSITORY SITE, NEVADA: PART 1, SECONDARY MINERAL PARAGENESIS AND GEOCHEMISTRY

By N.S.F. Wilson and J. Cline. Community College and University System of Southern Nevada. Report TR-02-005.1. 2002

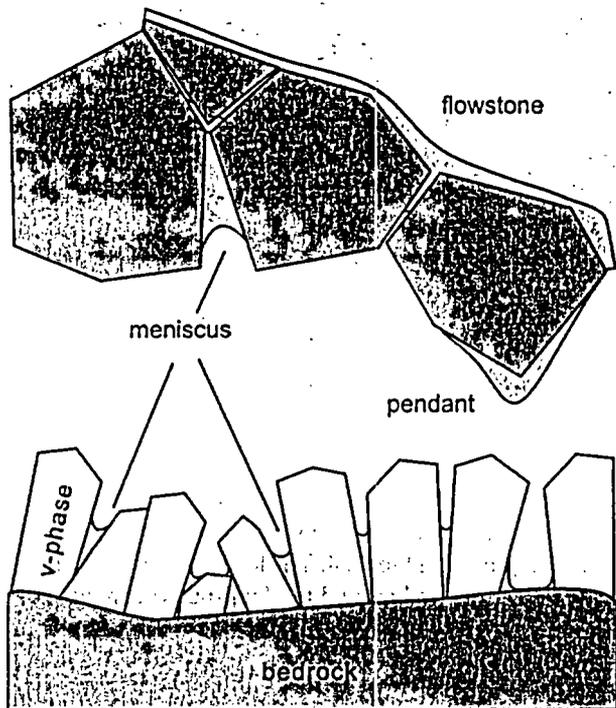
## 1.1. Observations «not consistent» with the phreatic environment of mineral growth

In this section of the review we first address, on a point-by-point basis, the list of observations that, as Wilson and Cline (2002) believe, «... are not consistent with the formation of secondary minerals in the phreatic zone». The list of arguments appears in the section 8.5. «Formation of secondary minerals in the Vadose Zone» (p. 23–24).

**Argument 1.** The majority of primary and secondary porosity contains no evidence of secondary minerals. If secondary minerals precipitated in a phreatic environment by upwelling hydrothermal fluids or rising groundwaters, a more extensive record of secondary minerals would be expected in the available open space.

As we explained in the section, «Hydrothermal upwelling concept and phreatic environment,» above, the absence of secondary minerals in part of the open cavities is not inconsistent with and is, in fact, to be expected within the hydrothermal upwelling model. The meaning of “precipitated in a phreatic environment by the upwelling of hydrothermal fluids” into the vadose environment must be adjusted to reflect the transient nature of hydrothermal upwelling.

**Argument 2.** Some pores in lithophysal cavities, fractures and breccias are rimmed by thin layers of calcite, suggestive of meniscus textures that indicate precipitation in a vadose environment. Some pore fillings exhibit multiple luminescent growth zones indicating multiple fluid events.



The presence of the telltale vadose zone textures, such as meniscus and pendant textures, or flowstone appearance of calcite (Figure 2), has not been documented by other researchers working on splits of samples from the UNLV collection. We have examined several tens of fluid inclusion sections, both from UNLV project and from our independent sampling activities, and have not observed textures that could be interpreted as meniscus or pendant textures. During the quarterly UNLV-USGS-IMP meetings of 1999–2000, which meetings had, as one of the major goals, joint work with samples and the discussion of petrographic observations and interpretations, neither meniscus nor pendant nor other gravity asymmetric textures were observed or discussed. So, we would be hesitant to accept the in-

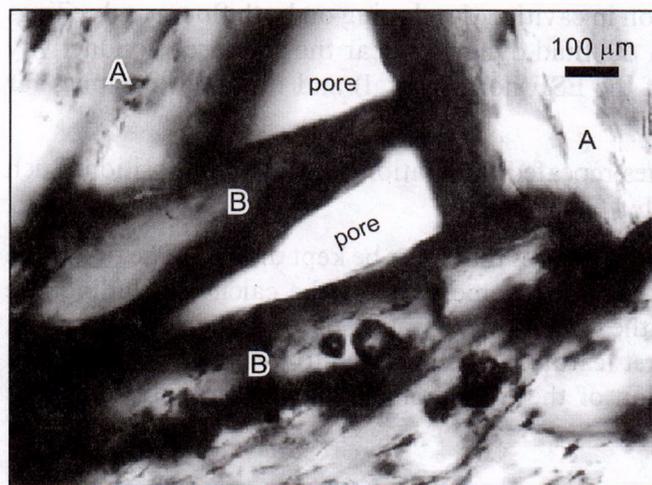
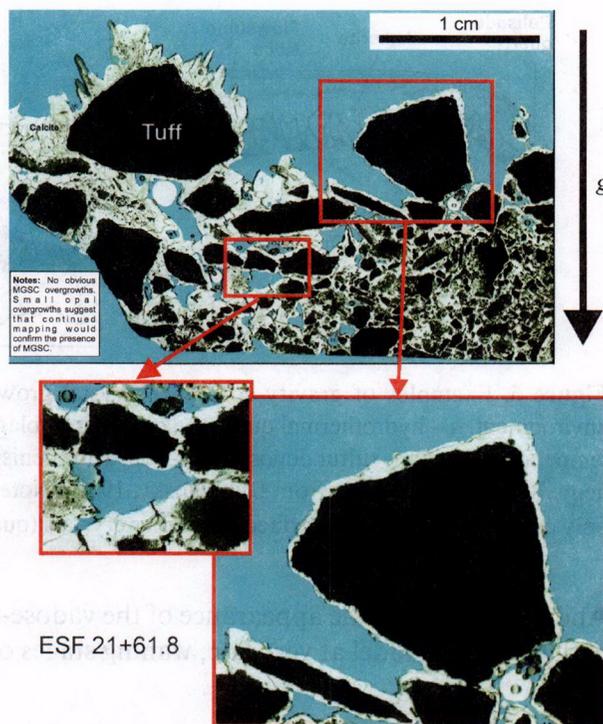
**Figure 2.** Schematic examples of the characteristic vadose-zone textures expressing surface tension and gravity asymmetry. None of these textures have been reported from the Yucca Mountain samples.

**Figure 3.** Textures of the sample ESF 21+61.8. Note complete coating of the tuff clasts with white calcite, showing no meniscus textures. For many tuff clasts the whole coating is represented by optically continuous calcite. *g* – is the direction of gravity force. Photograph is from the technical data files posted at the UCCSN WebPages. Blue is epoxy resin.

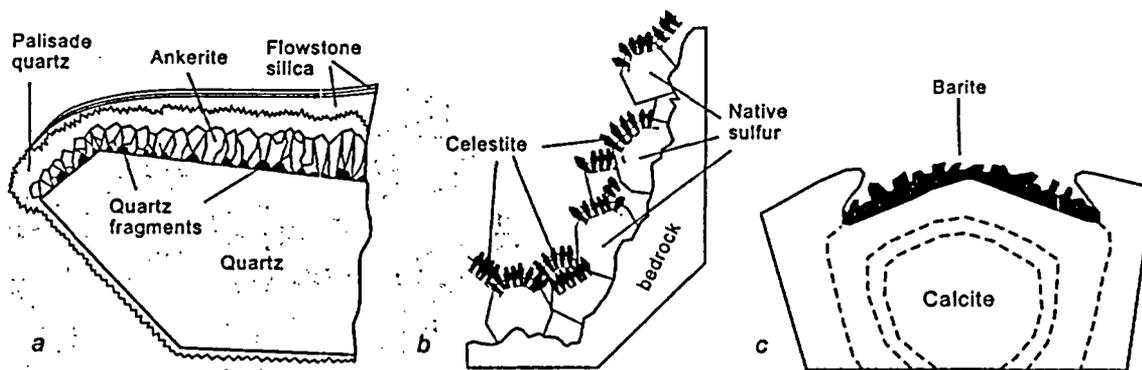
terpretation given in the UNLV report («suggestive of meniscus textures») without a documented demonstration of the presence of such textures.

We have examined photographs of the two samples (ESF 21+61.8 and ESF 74+19), presented by Wilson and Cline as an example of the meniscus texture, and also examined actual sections prepared from these samples. In both cases we failed to observe the meniscus texture or other textures suggestive of a vadose zone environment. Sample ESF 21+61.8 is shown in Figure 3. It is clearly seen that tuff fragments are completely coated with crystalline calcite. On many fragments, coatings have a fairly uniform thickness (<1 mm). No concave surfaces (meniscus textures) are observed in abundant voids. Examination under polarized light reveals that the small tuff fragments are typically embedded in optically continuous calcite crystals, while the meniscus texture is characteristic of polycrystalline aggregates. Uniform thickness of coatings over several loci (within the sample) argues against gravity-controlled deposition. Summarizing, the textures of the samples cited by Wilson and Cline (2002) as supporting the vadose depositional setting is perfectly compatible with, if not indicative of, the phreatic environment of mineral deposition.

One additional comment is in order. The appearance of the meniscus textures has a simple physical cause, which is the surface tension at the interface between liquid and vapor phases. If a water film wets a «rugged» angular substratum, menisci inevitably form in the reentrants (see Figure 2). At Yucca Mountain, the inner surface of the lithophysal cavities and some fractures is remarkably rugged due to the presence of a «palisade» of the vapor-phase crystals. Similarly, aggregates of bladed calcite crystals possess abundant narrow reentrants. A water film placed over such a surface would inevitably form menisci in large numbers. If secondary minerals indeed were deposited from films of water, as the UNLV researchers believe, menisci would be expected to be the most common texture of the secondary minerals at Yucca Mountain. It is revealing, therefore, that such textures have not been observed in the Yucca Mountain samples. By contrast, the conspicuous absence of such textures is commonly noted (Figure 4).



**Figure 4.** Micro-photograph showing early bladed calcite crystals (A; formed at ~50 °C) and late bladed crystals (B) formed at T ~40 °C in interstices between the early crystals. Note angular pores without any indication of the meniscus textures. IMP sample #1291, ESF station 28+81.



**Figure 5.** Examples of gravity-controlled crystal growth in the phreatic hydrothermal and ambient-temperature environment. *a* – hydrothermal quartz-ankerite assemblage, Urals, Russia (from Grigoriev and Zhabin, 1975), *b* – low temperature gypsum-sulfur deposit Shor-Sou, Turkmenistan (Yushkin, 1966), and *c* – hydrothermal karst cave Mátiás-hegy, Budapest, Hungary (from Dublyansky, 1995). Note that crystals of ankerite (*a*), celestite (*b*) and barite (*c*) grow only on the upward-facing surfaces of earlier crystals (quartz, native sulfur and calcite, respectively).

And finally, a sporadic appearance of the vadose-zone textures may be expected within the hydrothermal upwelling model at very late, waning stages of the dissipation of the hydraulic mound (see Figure 1-*b*).

**Argument 3.** The lack of isopachous textures is not consistent with formation in the phreatic zone. Secondary minerals occur predominantly on the footwalls and bases of lithophysal cavities, fractures and breccias.

Isopachous textures (i.e., thin linings of inner pore surfaces with mineral layers of nearly equal thickness) are characteristic textures of the phreatic, particularly marine cements, but they are, by far, not the only textures characteristic of the phreatic environment. They are not *sine qua non* features. By contrast, large euhedral (three-dimensional) crystals of calcite simply do not form from water films. They require, instead, submerged conditions of growth (e.g., Kendall and Broughton, 1978). This subject will be discussed in more detail below.

As for the occurrence of minerals at the floors of cavities, two observations are in order. First, although not very common, gravitation-controlled distribution of minerals (nucleation and growth on the upward-facing surfaces), is reported from both hydrothermal and ambient-temperature phreatic environments (Figure 5). This feature, therefore, is not incompatible with the phreatic setting. Second, it is important that near the north portal of the ESF both silica minerals and calcite commonly fail to show gravitation control in their distribution in cavities, developing on both floors and ceilings of cavities. This may emphasize the difference in the fluid dynamics near the enhanced conductivity channel (Paintbrush fault zone in the vicinity of the ESF north portal) and the fluid dynamics far removed from it.

**Argument 4.** Growth zonation of MGSC indicates repeated fluctuations in fluid composition. Such fluctuations are difficult to reconcile with saturated environment.

The authors of the report seem to lump together two features that must be kept distinct. The first is the generally increased (up to ~1.3 wt.%) contents of Mg in some parts of the calcite (including late oscillatory-zoned calcite, MGSC). The second is the oscillatory character of the Mg growth zonation exhibited by MGSC. We would agree that the first feature is likely related to the generally increased Mg/Ca ratio of the fluid at the time of formation of the Mg-enriched calcite (this subject will be discussed in more detail in Section 1.2).

However, explaining the oscillatory Mg zonation by external causes (i.e., by cyclic fluctuations of the fluid's Mg/Ca ratio), as Wilson and Cline do<sup>1</sup>, represents a clearly non-unique interpretation. Oscillatory mineral zonation is usually associated with crystal growth in a system, which may be driven sufficiently far from thermodynamic equilibrium to produce autonomous patterns by geochemical self-organization (Holten *et al.*, 2000). The coupling of the chemical reaction with diffusive transport through a boundary layer leads to a feedback mechanism that can produce oscillatory zonation. Researchers Wang and Merino (1992) studied oscillatory zonation in calcite. They developed a model based on growth inhibition by divalent cations such as Mn<sup>2+</sup>, Fe<sup>2+</sup> and Zn<sup>2+</sup>. Although they did not consider Mg<sup>2+</sup>, it is well known that the latter is also an inhibitor for calcite growth (Zhang and Dawe, 2000), and the Wang-Merino model, thus, should apply to Mg zonation in calcite. Holten *et al.* (2000) studied mathematically the effect of external disturbances (noise) on the calcite zonation produced by the Wang-Merino model and concluded that such zonation is not sensitive to the noise.

The bottom line of these studies is simply that oscillatory zonation in calcite does not require fluctuations of the Mg/Ca ratio of the mineral forming water. It may develop in a growing crystal even if the Mg/Ca ratio of the bulk mineral forming fluid remains constant or undergoes non-periodic fluctuations. Mathematical analysis of the zonation pattern may be helpful in determining the causes of the zonation (e.g., Wang and Merino, 1992; Halden and Hawthorne, 1993; Halden, 1996; Bryxina and Sheplev, 1997), but this analysis has not been done for the Yucca Mountain MGSC. Thus, unless the external cause of the Mg zonation in MGSC is persuasively demonstrated, interpreting it as a feature expressing a climatic signal and thus irreconcilable with the hydrothermal upwelling model would certainly be in conflict with the universally accepted understanding in modern mineralogy concerning the origin of mineral zonation: «*Oscillatory mineral zonation is usually associated with crystal growth in an open system, either a hydrothermal system or a melt ...*» (Holten *et al.*, 2000, p. 1893).

**Argument 5.** If upwelling hydrothermal fluids periodically invaded Yucca Mountain, the fluids would be cooled by contact with colder rocks, and silica minerals, particularly quartz, would precipitate. Although silica minerals are relatively abundant in some samples in the NPR and SPR, these minerals are part of the early to intermediate assemblages, and silica minerals are sparse in younger assemblages and in other parts of the site.

This argument is ambiguous and a *non sequitur*. First, Wilson and Cline postulate that upwelling hydrothermal fluids must deposit silica minerals. Then they confirm that such minerals are particularly abundant in the vicinity of the north and south portals of the ESF. It is to be noted that these are the parts of the ESF tunnel closest to the horst-bounding fault zones, which provide the major avenues for fluid upwelling. Near the portals the highest fluid inclusion temperatures have been measured (see Figures 12 through 14 below). Thus, the ESF portal areas are exactly the places to look for silica minerals, if their hydrothermal origin is presumed. Observations of the mineral assemblages are perfectly compatible with, and even suggestive of the hydrothermal upwelling model.

Wilson and Cline bluntly dismiss the significance of the observations, arguing that silica minerals «*are part of early to intermediate assemblages*». Should their argument be taken to mean that they believe that the early and intermediate parts of the assemblages formed from hydrothermal upwelling fluids, and only the younger minerals of the assemblage did not? How else can their reasoning be understood?

**Argument 6.** No single location or sample records the entire history of secondary mineralization, suggesting that whatever process was responsible for deposition did not occur uniformly through the repository horizon. Such a record is difficult to reconcile with a phreatic environment.

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<sup>1</sup> «*The fine oscillatory growth zoning shows that the Mg/Ca ratio in the fluid fluctuated repeatedly and in a fairly regular, cyclical manner.*» (Wilson and Cline, 2002, p. 21).

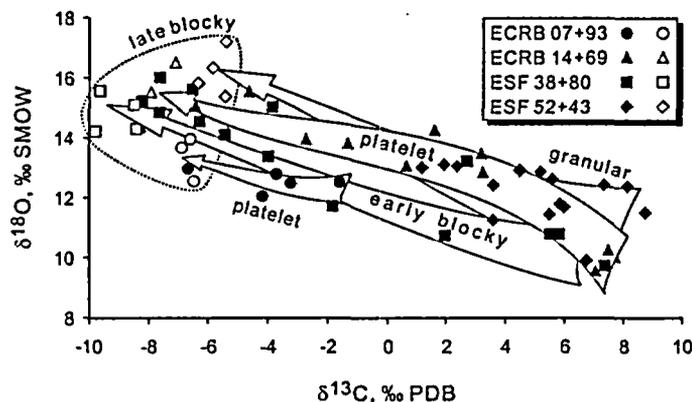


Figure 6. Summary cross-plot for four samples with different morphologies. Overall direction of growth is shown by arrows. The latest morphologic variety in all four samples is blocky calcite with opal (circles). Note that in sample ECRB 07+93 early part of the record is missing.

The claim that no single location records the entire history of secondary mineralization is debatable. Our studies show that in different parts of the repository block, minerals deposited from geochemically (and temporally) equivalent fluids may have different

morphological appearances. Isotopic studies show, for example, that  $\delta^{13}\text{C}$  in the fluid evolved from strongly positive (ca. +8–9 ‰ PDB) to negative values (ca. –10 ‰; Figure 6). The trend is always unidirectional (i.e., no reversals have been documented in samples with readily interpretable textures). Importantly, in different parts of the repository, similar trends have been measured in crystals with different morphologies (e.g., combinations of granular, bladed, and blocky crystals) and in crusts of different thickness (1 mm to 2–3 cm). This indicates to us that notwithstanding the differences in the appearance, mineral crusts in many locations do record a consistent depositional history.

It is not uncommon also that, at some locations, parts of the record are missing (e.g., sample ECRB 07+93 in Figure 6). This is to be expected in a dynamic system, in which fluids evolve in time and space. As the fluids move away from the fault zone, along which they were injected, they cool down, degas, oxidize, mix with other waters and deposit minerals. Their path is primarily controlled by the permeability structure of the host rock; therefore, these fluids may not access some cavities. Fluid inclusion temperatures record this spatially complex picture as shown in Figure 14 below. The spatial structure is also emphasized by the decreasing abundance of silica minerals away from the horst-bounding fault (see discussion in the preceding sub-section).

**Argument 7.** The presence of glass in the host tuff suggests that these tuffs have not been below the static water level. These data strongly indicate that the volcanic rocks have not been in contact with fluids in a phreatic environment for any length of time during their history.

Although it is true that within the horst of Yucca Mountain the glass-bearing tuffs are generally confined to the present-day vadose zone, but in the adjacent tectonic depressions, Jackass Flats and Crater Flat, it is not so. In western Jackass Flats the preserved glass has been found deep below the present-day water table at a depth of about 230 m in drill holes J-12 and J-13. In central Crater Flat the glass has been found in drill hole USW VH-2 at a depth up to 920 m below the present-day water table. Closer to the repository footprint, glass was found below the water table in drill holes USW G-3, UE25 p#1 and a#1 (Carlos and Chipera, 1989). This glass was preserved, in the phreatic zone for a time comparable to the lifetime of these tectonic depressions, that is, for millions of years. The argument that the volcanic rocks could not have been «in contact with fluids in a phreatic environment for any length of time» must therefore be discarded as demonstrably invalid.

In addition, as was stated in the section «Hydrothermal upwelling concept and phreatic environment» above, the hydrothermal upwelling concept does not envisage a geologically extended state of saturation within what is now the vadose zone of Yucca Mountain. Instead, it envisages relatively short-term injections of the deep-seated fluid into the vadose zone with the formation of a transient hydraulic mound followed by dissipation of the mound. The time of the exposure of volcanic glass to aqueous fluid in such a system would be much shorter than the time of exposure of glass found below the water table in boreholes.

### 1.1.1. Summary on section «Formation of Secondary Minerals in the Vadose Zone»

Above we addressed arguments and observations that were deemed by Wilson and Cline to be inconsistent with the formation of the Yucca Mountain minerals in the phreatic environment. We conclude that none of these arguments and observations invalidates the hydrothermal upwelling model of formation of the Yucca Mountain secondary minerals.

### 1.2. Relations between the Mg-enriched calcite and the climate (an example of ambiguous interpretation)

The subject of the discussion by Wilson and Cline that appears on p. 22 of their report deals with the cause for the appearance of the Mg-enriched calcite that exhibits oscillatory growth zonation. The authors postulate that the latter feature (Mg oscillatory zonation) is related to climatic changes that occurred from around 2.9 Ma to the present day. The style of argumentation by Wilson and Cline, characteristic of their scientific analysis, deserves scrutiny and evaluation. They postulate the following contentions and we respond:

**Contention 1.** «*This time period corresponds with a major change in Northern Hemisphere glaciation which occurred around 2.67 Ma (Prueher and Rea, 1998). An abrupt change in deep-sea sediment character related to a change from non-glacial to a glacial environment occurred across the North Pacific at this time. This change occurred too rapidly to be a direct response to tectonic or orbital forcing (Prueher and Rea, 1998) and terrestrial changes that corresponded with deep sea-related changes would be expected.*»

The onset of major glaciation in the Northern Hemisphere, as Prueher and Rea (2001) argued, was triggered by explosive volcanism in the Kamchatka-Kurile and Aleutian arcs. What bearing, if any, this fact has on the sources of Mg in the Yucca Mountain calcite remains unclear. Since no specific mechanism relating the transition from inter-glacial to glacial climate and the increase in Mg content of the mineral forming waters is proposed, the whole passage seems to be irrelevant.

**Contention 2.** «*The gradual uplift of the Sierra Nevada, west of Yucca Mountain, caused a change in water chemistry during the last 3 m.y. (Smith et al., 1983).*»

Wilson and Cline do not specify what waters they are talking about. The uplift of the Sierra Nevada, by some 950 m over the last 3 million years could have influenced the chemistry of some waters, particularly the chemistry of the runoff from the Sierra; the chemistry of desert lakes (related to increased evaporation due to rain-shadow effect); the chemistry of groundwaters (related to changing proportions of watersheds with different lithologies), and the isotopic geochemistry of atmospheric precipitation (related to orographic depletion of deuterium in inland bound Pacific storms). How the uplift could cause an increase in the Mg/Ca ratio in meteoric waters that fall as atmospheric precipitation on the surface of Yucca Mountain and percolated down fracture pathways (a model, advocated by the UNLV researchers) remains unexplained.

**Contention 3.** «*This change in chemistry led to deposition of considerable dolomites and Mn-enriched clays in playas and lakes in the Amargosa desert during the Pliocene (3.2-2.1 Ma) (Hay et al., 1986). These minerals may have contributed Mg to fluids that percolated into Yucca Mountain during climate-related cycles, forming MGSC. This process may have been accelerated by the earlier Pliocene nonglacial environment, when the climate was substantially wetter and springs in the Amargosa Desert were more widespread and had greater discharge (Hay et al., 1986).*»

The description above does not provide any explanation of how the dolomites and Mg clays deposited in the Amargosa Desert at the discharge sites of moderately thermal springs (Hay *et al.*, 1986) would contribute to the «fluids that percolated into Yucca Mountain». It should be noted that Yucca Mountain is located some 30–60 km to the north (up-gradient) and some 800 m higher relative to the Amargosa discharge sites. As a consequence, it remains equally unclear how this (unspecified) process could be accelerated by a wetter climate and a greater discharge in the Amargosa springs.

**Contention 5.** «*Alternatively, it is possible that the increase in Mg could be related to atmospheric dust related to erosion of Paleozoic dolomites in the southwest (J. Stuckless, personal communication, 2000).*»

Outcrops of Paleozoic dolomites and limestones are present not only generally in the southwest, but also in the immediate vicinity of Yucca Mountain (Bare Mountain ~15 km to the west and in Calico Hills ~10 km to the northeast). Wind erosion of these rugged terrains should produce atmospheric dusts similar to the one that Wilson and Cline, following the lead of Stuckless, invoked as a source-term for Mg in Mg-enriched calcite.

Guthrie *et al.* (1995) studied the mineral composition of modern dusts collected in the vicinity of Yucca Mountain. The dusts contain 45–47 wt. % feldspar, 18–22 % quartz, 6–8 % smectite, 3–4 % zeolite (clinoptilolite), 1–3 % mica, as well as minor amounts of tridymite, cristobalite, chlorite, amphibole, and hematite. Calcite was found in trace amounts or, in some samples, was not determined at all. Dolomite was not found. From this example we can see that this speculative model does not withstand scrutiny when confronted with the hard data.

**Summary.** The discussion by Wilson and Cline, evaluated above, may create a perception for the lay reader that a reasonable explanation of the appearance of Mg-enriched calcite has been offered. In fact, however, the discussion provides no more than a set of assertions. The possible genetic links between the processes discussed by Wilson and Cline and features described by them are not shown, and scientifically defensible models of the development of the Mg enrichment are not proposed.

### 1.2.1. Mg-enriched calcite and the hydrothermal upwelling model

When considering possible sources for the Mg-enriched calcite at and around Yucca Mountain, it is important to recognize that Mg enrichment is a geochemical feature that is typically associated with minerals deposited at ancient discharge sites, commonly from fluids with elevated temperatures. Vaniman *et al.* (1995) reported that Mg-enriched calcite commonly occurs at contemporary and fossil discharge sites of thermal waters in Death Valley (Travertine Point, Grapevine Spring, and Nevares Spring) and at the southern end of Yucca Mountain (USGS site #199). The Mg content varies between 1.53 and 2.63 wt. % MgO, for the Death Valley locations, and between 3.0 and 5.8 wt. % MgO, for site #199. Further, the Nye County well NC-EWPD-1S revealed that the USGS site #199 is underlain, at a depth of about 20 meters, by a ~100 meters tall hydraulic «mound», which is composed of thermal water with a Ca-Mg bulk composition (Farnham *et al.*, 2002).

The calcite of the surficial calcretes and bedrock veins (a potential source-term of the vadose-zone calcite in the USGS-UNLV model) contains low abundances of Mg (<1 wt.% MgO; Vaniman *et al.*, 1995). The distribution coefficients for Mg in the solution-calcite system are very low ( $K_d \sim 0.02\text{--}0.03$  at  $T = 20\text{--}40\text{ }^\circ\text{C}$ ; Rimstidt *et al.*, 1998). Thus, calcite crystallizing from solution that acquired its Mg through dissolution of surface calcretes is expected to contain substantially (orders of magnitude) less Mg than the «parent» calcretes. The actual contents of Mg in the MGSC, however, are greater than those in the surface calcretes (up to 1.3 %), which makes the latter an unsuitable source of Mg for the subsurface calcite. We conclude that neither the rhyolite tuffs nor the surface calcretes could have served as an adequate source of Mg for the subsurface minerals at Yucca Mountain.

The Paleozoic dolomite of the Roberts Mountain Formation is widely recognized as a source of Mg for the Mg-rich carbonates found in spring deposits of the region. At Yucca Mountain, the Roberts Mountain dolomite underlies the tuffs and occurs at a depth ranging between 1.2 and >3.5 km. Upwelling thermal waters would pass through the dolomite; it is therefore perfectly compatible with the hydrothermal upwelling model that this dolomite served as a source of Mg in secondary calcite.

### 1.3. Questions not asked

We found that a number of important questions have not been discussed by the UNLV researchers, notwithstanding the fact that the information, which they obtained in the course of their study clearly permitted such a discussion, or even begged for it. These questions, in our opinion, have a profound bearing on the issue of the origin of secondary minerals at Yucca Mountain.

#### 1.3.1. Mineralogy of secondary deposits

Wilson and Cline (2002) report the presence of calcite ( $\text{CaCO}_3$ ), opal ( $\text{SiO}_2 \cdot n\text{H}_2\text{O}$ ), chalcedony ( $\text{SiO}_2$ ), quartz ( $\text{SiO}_2$ ), fluorite ( $\text{CaF}_2$ ) and zeolite (likely heulandite,  $\text{Ca}_4[\text{Al}_8\text{Si}_{28}\text{O}_{72}]\cdot 24\text{H}_2\text{O}$ ). Although it is not stated in the report, they also observed barite ( $\text{BaSO}_4$ ; reported in technical data files posted at the UCCSN WebPages) and strontianite ( $\text{SrCO}_3$ ; Wilson, 2001, pers. com.). Although Wilson and Cline describe the major secondary minerals (calcite and silica minerals) in considerable detail, they do not describe paragenetic relationships between major and accessory minerals and fail to discuss the genetic significance of the mineral assemblage as a whole. The minerals are present; they are co-genetic; so what does this mean?

In order to deposit minerals, an aqueous solution must first react with the rock to dissolve the requisite components (in the case of Yucca Mountain these are: Ca, Si, Ba, Sr,  $\text{HCO}_3^-$ ,  $\text{F}^-$ ,  $\text{SO}_4^{2-}$ ) and then become supersaturated with respect to these components. Wilson and Cline do not discuss how the fluids acquire their solutes. According to the genetic model they advocate (along with the USGS researchers), these minerals were formed, essentially, from rainwater that fell on the ground, percolated through soils and infiltrated along open fractures toward the precipitation sites. The travel distance for such fluids would be relatively short (roughly, equal to the depth from the surface, 30 to 300 m). The only rocks available for dissolution along this path are (a) surface soils (that could be calcareous) and (b) rhyolitic tuffs. Thermodynamic modeling by Palyanova *et al.* (2003) has demonstrated that natural water reacting with these rocks within a temperature range of 25 to 100 °C, at  $p\text{CO}_2$  ranging from «atmospheric» (0.03) to «soil» (0.1) values and rock/water ratio, R/W ranging from  $10^{-8}$  to  $10^2$  cannot reach supersaturation relative to some of these minerals, particularly fluorite, under any geologically reasonable set of conditions.

The deposition of strontianite does not seem to be possible within the model envisaging the derivation of the dissolved components from surficial calcareous deposits. The Sr/Ca ratio in the solution resulting from dissolution of surficial carbonates in rainwater would be equal to that in the dissolving calcite (assuming congruent dissolution). Surficial carbonate deposits studied in Trench 14 on Exile Hill (east slope of Yucca Mountain above north ramp of the ESF) contain, on an average, 2100 ppm of Sr (Vaniman *et al.*, 1995), which gives a Sr/Ca ratio of about 0.0024. Fairchild *et al.* (2000) have demonstrated that, due to the selective leaching, the Sr/Ca values of leachates of weathered bedrocks show up to 5-fold enrichment relative to the respective values of the bedrock carbonates. Taking into account this effect, the reasonable estimate of the Sr/Ca value of the solution equilibrated with the Yucca Mountain slope calcretes would be Sr/Ca @ 0.01. By contrast, solutions in equilibrium with both calcite and strontianite in the 25 to 100 °C-range must have  $\text{Sr}^{2+}/\text{Ca}^{2+}$  ratios of 0.1 to 0.5 (Helz and Holland, 1969; Kinsman, 1969). This simple calculation shows that the hypothetical rainwater dissolving slope carbonates would be strongly undersaturated with respect to strontianite.

**Summary.** To be acceptable, the model of formation of secondary minerals must rationally explain where and how the fluids acquired their solutes. The model adopted by Wilson and Cline (2002) does not provide a plausible explanation.

### 1.3.2. Euhedral character of minerals

All researchers of the secondary minerals at Yucca Mountain, Wilson and Cline included, report that in many locations cavities host large, up to 3 cm, freestanding calcite crystals developed roughly perpendicularly to the substratum. The morphology of such crystals is termed «euhedral».

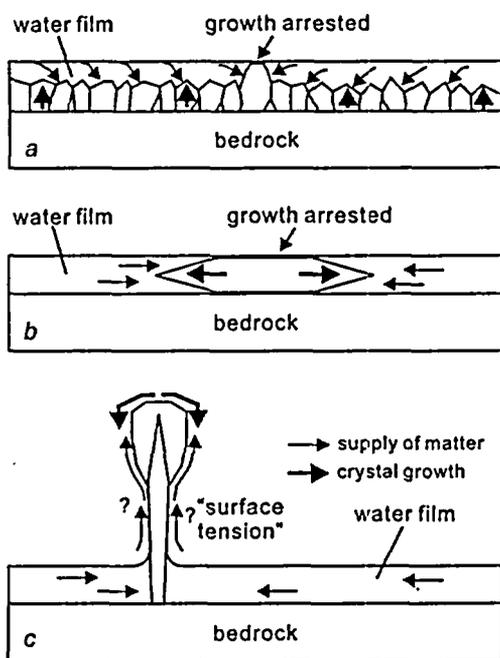
The concept suggested by the USGS researchers and endorsed by UNLV researchers envisages the formation of secondary minerals at Yucca Mountain via the growth of crystals from thin water films (Figure 7-c). From the standpoint of the theory of crystal growth this does not seem to be possible. For minerals with relatively low solubilities, such as calcite and quartz, the growth of large euhedral crystals may only occur in a submerged state from a slightly supersaturated fluid, because molecules that build the crystal lattice must be supplied uniformly to all crystal faces at a steady low rate (e.g., Sunagawa, 1982).

Two important questions do not seem to have satisfactory answers within the «film water» growth model. First, how and why did the large euhedral crystal start to acquire their shape? And second, how was the matter supplied to the tips of large crystals, where the growth preferentially occurred at advanced stages of growth?

Describing the characteristic textures of calcite deposits formed from water films (speleothems), Kendall and Broughton (1978) provided a simple and logical explanation why the crystals formed through this mechanism always have very small sizes: «...distinctive fabrics of palisade calcite are formed because precipitation usually occurs from thin water films that flow over the growing speleothem surfaces. Large crystal terminations do not form on the speleothem surface because they form projections that disturb the water flow away from the projections which, as a consequence, are gradually eliminated.» (p. 519). The situation is schematically shown in Figure 7-a. This rationale is universally used by petrologists to discriminate between vadose and phreatic environments of formation of mineral deposits. For example,

describing a case-study of calcite spar from Lake Valley Formation, New Mexico, Goldstein and Reynolds (1994) write: «Some growth bands define crystal terminations of well-formed euhedra with as much as 5 mm of relief on the termination, suggesting growth within a water-saturated pore, such as that which would be present below a perched or permanent water table ...» (p. 159).

If an individual crystal, rather than a multitude of micro crystals, develops within a water film (requiring a very low



**Figure 7.** Examples of crystal growth from water films (thickness exaggerated).

*a* – growth of palisade aggregate (typical of speleothems); growth is arrested as soon as the crystal’s length reaches the thickness of water film; *b* – growth of a single euhedral crystal; *c* – hypothetical growth of the Yucca Mountain scepter calcite from water films. Drawing *c* combines the observed (inferred from physical evidence) directions of crystal growth with postulated supply of matter by water films (Paces *et al.*, 2001; Whelan *et al.*, 2002; Wilson *et al.*, 2002).

nucleation rate and low supersaturation) the preferential flux of the matter to such crystal will be directed parallel to the film surface (Figure 7-b). In accordance with Curie's principle of symmetry, the direction of the fastest growth will also be oriented parallel to the film surface (i.e., parallel to the cavity surface; Shafranovski, 1968). In striking contrast, bladed crystals at Yucca Mountain are almost invariably oriented roughly perpendicularly to the cavity floors (Figure 7-c). It is unclear, therefore, how and why large freestanding euhedral crystals would form from water films, instead of the «normal» palisade aggregates of micro crystals typical for such a setting (see Figure 7-a).

The next important question is: what process would be capable of taking a hypothetical mineral-laden water sitting as a film on the cavity floor and transporting it all the way up to the top of a 2–3 cm-tall crystal? The USGS researchers proposed a model in which water films are drawn up the crystal faces by surface tension (Paces *et al.*, 2001; Whelan *et al.*, 2002). Wilson and Cline seem to accept this model<sup>2</sup>, which is schematically presented in Figure 7-c.

The model, however, seems to be in conflict with basic principles of physics. Capillary forces can only be effective in lifting up liquids if they operate in thin channels or pores. In geological objects, the size range in which capillary forces play a discernible role is from 0.0002 to 0.5 mm for cylindrical channels and from 0.0001 to 0.25 mm for slot-shaped channels (Chukhrov, 1955). In wider channels capillary forces are insignificant and do not produce any substantial lifting of water. It is easy to demonstrate that under the most conservative assumptions, the water film rise along a flat surface of a free standing crystal cannot be greater than ~4 mm. Simple experiments with actual Yucca Mountain crystals partly immersed in waters with different salinities have shown that the water meniscus «climbs» up the crystal surface for approximately 1.5 mm. Thus, the proposed mechanism of supply of the mineral-forming solutions to the tips of 2–3 cm-tall crystals by surface tension represents a physical impossibility.

**Summary.** We contend that the growth of large euhedral crystals and aggregates similar to those found at Yucca Mountain is only possible in the submerged (phreatic) state and cannot occur from films of water. We dispute, thus, the statement by Wilson and Cline: «*The question of whether the secondary minerals formed in a vadose or phreatic environment cannot be directly answered, because no single texture observed in the rocks unequivocally identifies the environment of precipitation.*» (Wilson and Cline, 2002, p. 23). The euhedral character of large crystals provides an unequivocal indication for a phreatic setting during their growth.

### 1.3.3. Significance of the «heavy positive» $\delta^{13}\text{C}$

The  $\delta^{13}\text{C}$  values as «heavy» as +5 to +10 ‰ PDB are common for the early parts of the Yucca Mountain calcite. Wilson and Cline (2002) do not discuss the genetic significance of these unusually heavy  $\delta^{13}\text{C}$  values but, nevertheless, claim that their stable isotope data «... indicate that calcite precipitated from meteoric fluids ...» (p. 21). We challenge the validity of this claim.

The  $\delta^{13}\text{C}$  in calcite is controlled by isotopic properties of the sources of carbon. All such potentially available sources at Yucca Mountain (within the UNLV-USGS genetic model) have an isotopically light composition of carbon (e.g., +1 to -1 ‰ for marine carbonate dust; -7 ‰ for atmospheric  $\text{CO}_2$ ; -10 to -12 ‰ for surface carbonates; -9 to -17 ‰ for soil  $\text{CO}_2$ ; -14 to -16 ‰ for  $\text{CO}_2$  in the underground air). Calcite with  $\delta^{13}\text{C}$  heavier than approximately +4 ‰ **cannot be formed** in equilibrium with an aqueous fluid acquiring its carbon from these sources.

The formation of calcite with a «heavy positive» isotopic composition of carbon requires a preferential enrichment of dissolved oxidized carbon species that take part in the precipitation of calcite with

<sup>2</sup> «Water was drawn up the faces of growing crystals by surface tension ...» (Wilson and Cline, 2002, p. 24).

the «heavy» carbon isotope ( $^{13}\text{C}$ ) at the expense of the reduced species (notably,  $\text{CH}_4$ ). Effective partitioning requires a strictly anoxic environment and is favored by a long length of time available for reaction and elevated temperatures (Ohmoto, 1986). This suggests that at least the early parts of the mineral forming fluids at Yucca Mountain have been equilibrated with a reducing, strictly anoxic environment (see Section 2.7.1 of this review for additional independent corroborating evidence). From general geologic and hydrogeological considerations, the persistence of an anoxic reducing environment within the thick vadose zone of Yucca Mountain in the past is highly improbable (see Section 2.7.1. for discussion).

**Summary.** If the UNLV researchers intend to defend their model of deposition of secondary calcite at Yucca Mountain by downward migrating meteoric water, they need to provide a reasonable and verifiable explanation of how calcite with a  $\delta^{13}\text{C}$  of +4 to +10 ‰ PDB was formed.

## 2. REVIEW OF THE REPORT: THERMOCHRONOLOGICAL EVOLUTION OF CALCITE FORMATION AT THE POTENTIAL YUCCA MOUNTAIN REPOSITORY SITE, NEVADA: PART 2, FLUID INCLUSION ANALYSES AND U-Pb DATING

By N.S.F. Wilson, J. Cline, and Y. Amelin. Community College and University System of Southern Nevada. Report TR-02-005.2. 2002

### 2.1. Conclusions by Wilson *et al.* (2002)

In the conclusion of their report Wilson and others state: «Results from this study are not consistent with models requiring formation of secondary minerals in a saturated environment at Yucca Mountain. Results, furthermore, provide no evidence for the former presence of upwelling hydrothermal fluids. Alternatively, results are consistent with infiltration of a cooling tuff sequence by descending meteoric water. This study demonstrates that the hypothesis of geologically recent upwelling hydrothermal fluids is untenable and should not disqualify Yucca Mountain as a potential nuclear waste storage site.» (p. 26, emphasis added).

After studying the report and accompanying technical data posted at the UCCSN WebPages, we have failed to find any data that could be deemed inconsistent with the hydrothermal upwelling model. The report does not contain a summary of the results and arguments that, as Wilson with co-authors assert, negate the possibility of a hydrothermal upwelling mode of formation for secondary minerals, and indicate formation from descending meteoric fluids instead. We attempted to compile a list of arguments presented in the report, which could be interpreted by a reader as supporting the *per descensum* model. These arguments are:

- 1) Fluid inclusions trap a low-salinity water (p. 16);
- 2) Fluid inclusion water has  $\delta\text{D}$  values characteristic of the meteoric waters (pp. 24, 25);
- 3) The fluid inclusion temperatures are uniformly distributed across the repository block; they do not show significant lateral gradients and do not exhibit a central hot plume. (p. 22).

In subsequent sections we critically evaluate these three positions.

### 2.2. What does «low salinity fluid» mean?

Summarizing their Section 4.4, «Fluid Inclusion Ice Melting Temperatures», Wilson and others state: «Results indicate that the 2-phase FIAs [fluid inclusion assemblages] consistently trapped a low-salinity

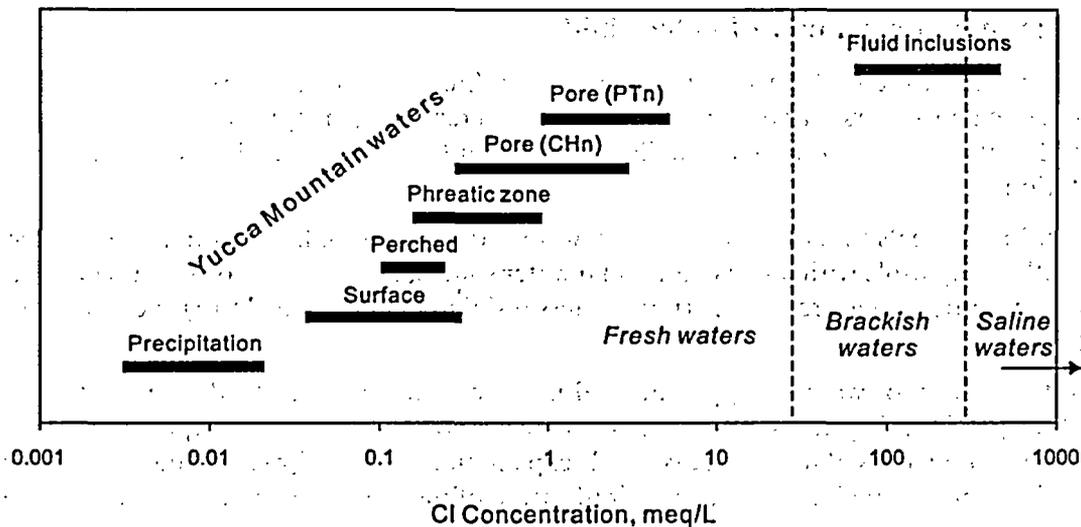


Figure 8. Chloride concentrations in water samples from Yucca Mountain (plotted from Meijer, 2002) compared with the concentrations measured in fluid inclusions, as reported by Wilson et al. (2002). Boundaries between fresh, brackish, and saline waters are according to the classification of Davis and De Wiest (1966).

fluid.» (p. 16). The quoted statement employs a loose term, «low-salinity fluid», which is not defined in the text. The use of such a term might be misleading to a lay reader. In hydrogeological terms, waters analyzed by Wilson and others from fluid inclusions (salinities ranging from 0.35 to 2.74 wt. % = 3,500 to 27,400 ppm; see p. 16) would be classed as brackish (1,000 to 10,000 ppm) and saline (10,000 to 100,000 ppm; classification by Davis and De Wiest, 1966).

A comparison of the fluid inclusion salinities with salinities of different types of Yucca Mountain waters is shown in Figure 8. The most saline is the pore water of the PTn non-welded tuff. The maximum contents of Cl for this water range up to about 7 Meq/l (245 ppm), or the equivalent of 405 ppm of NaCl. Thus, the «low-salinity fluid» of Wilson and others, inferred from fluid inclusions data, appears to be 8 to 70 times more salty than the most saline of the naturally occurring waters found at Yucca Mountain, and up to 7000 (!) more salty than the surface runoff waters.

Finally, does the alleged «low salinity» of the water in the fluid inclusions make it incompatible with the hydrothermal origin? Of course not! For example, Criss and Taylor (1986) state: «*Geothermal waters commonly contain appreciable amounts of solutes. Total concentrations are typically several thousand ppm, but range from essentially fresh water (few ppm) to concentrated brines (25 wt. % solute). The most important are neutral to alkaline chloride waters, which have a predominance of alkali and alkaline earth chlorides (NaCl, KCl, CaCl<sub>2</sub>), are often close to saturation with calcite and amorphous silica, and contain a wide variety of other constituents (Ellis and Mahon, 1964, White et al., 1971).*» (p. 390).

The general characteristic of the geothermal waters given in the citation above bears a striking resemblance to the characteristics of the Yucca Mountain paleo waters inferred from mineralogic and fluid inclusion studies. Total concentrations ranged from near 0 to 27,400 ppm NaCl equivalent (Dublyansky et al. 2001; Wilson et al. 2002). Since the waters were depositing calcite, silica (opal, chalcedony, quartz), fluorite, heulandite, strontianite, and barite, they should have reached saturation levels with these minerals; hence, substantial amounts of dissolved silica, bicarbonate, fluorine and sulfate must be inferred.

### 2.2.1. Summary on salinity of paleo fluids

1. A loose usage of the undefined term, «low-salinity fluid,» by the authors of the UNLV report to characterize paleo waters trapped in fluid inclusions creates a perception that such waters are not compatible with the hydrothermal upwelling model and that they have likely derived from meteoric precipitation.

2. A closer look reveals that the fluid inclusion waters: (a) range in salinity from brackish to saline; (b) are perfectly compatible with the hydrothermal water source; and (c) are 8 to 70 times more saline than the most concentrated natural waters (pore waters residing in non-welded tuff) encountered at Yucca Mountain.

3. Thus, the salinity of waters trapped in fluid inclusions in calcite, instead of indicating a surficial origin of the precipitating fluids, provides, in our view, a strong argument in support of the hydrothermal upwelling model. As of now, no reasonable explanation for these brackish and saline waters in the context of the model involving descending infiltration of meteoric precipitation waters has been proposed.

### 2.3. The $\delta D$ of waters trapped in fluid inclusions

Wilson and others reported the data on  $\delta D$  values of waters trapped in fluid inclusions. Ten  $\delta D$  measurements were obtained from three calcite samples. We are not satisfied with the way either the methodology of the analyses or the results are described in the report. For example, the data in the main report (reproduced as Table 1) do not correspond to the results presented in technical data files posted at the UCCSN WebPages (Table 2). Only one method of analysis was briefly described in Section 3.3, whereas from Table 1 it is apparent that two different methods were employed (an online continuous He flow-laser ablation technique and «conventional» offline Zn reduction method). Further, when describing the method of the  $\delta D$  analysis in the continuous He flow, Wilson and others refer the reader to a paper by Sharp *et al.* (2001). The latter citation, however, does not contain a description of the method in which water from fluid inclusions is released by «decrepitation» (?) of samples by a  $CO_2$  laser.

Table 1.  $\delta D$  compositions of fluid inclusion fluids (reproduced from Wilson *et al.*, 2002)

Sample Number	$\delta D$ (‰)	Mean $\delta D$ (‰)	Description	Th (°C)
AL#5 00+28.5	-120, -90	-105	Outer part of mineral crust – intermediate calcite	35–45
ESF 27+84	-110, -115*	-112,5	MGSC	<35
ESF 60+52.5	-131*	-131	MGSC	<35

\* Duplicate analyses were performed by the conventional technique of heating the sample in vacuum with an external furnace.  $H_2O$  is collected in a 6 mm diameter Pyrex tube with "magic" Indiana zinc. The tube is sealed off, heated to 550 °C and the Zn reacts with  $H_2O$  to make  $ZnO + H_2$ . The  $H_2$  is cracked directly into the mass spectrometer and analyzed using dual inlet–bellows system.

Table 2. Summary of  $\delta D$  isotopic composition of fluid inclusion fluids (as presented at the UCCSN WebPages)

Sample Number	$\delta D$	Comments
AL#5 00+28.5	-120, -90	Sample was from the outer part of sample above the 8.24 Ma chalcidony layer that contained inclusions with Th's of 35–45 °C.
AL#5 00+28.5	-61, -59	Sample was from the inner part of sample older than 6,24 Ma chalcidony layer that contained inclusions with Th's of 35–45 °C and greater.
ESF 60+52	-131	Sample of MGSC
ESF 27+84	-90, -87, -69	Sample from the basal calcite layer containing 2–phase FIAs with Th's of >35–45 °C.
ESF 27+84	-110 (-115 conventional analyses)	Outer MGSC layer.

Two replicate analyses for sample Al#5 00+28.5, by this method, showed a discrepancy in the measured  $\delta D$  values as large as 30 ‰ (see Tables 1 and 2). Instead of trying to address the reason for this large difference, Wilson and others simply report the mean for these two analyses, which is clearly inappropriate. The difference between the replicate samples may be real; alternatively, it might be an artifact indicating analytical problems. Before the reasons of the discrepancy are understood, it would be prudent to assume that actual errors associated with the data could be much greater than the declared analytical precision of  $\pm 2$  ‰.

The  $\delta D$  data from fluid inclusions are novel and have not been available for Yucca Mountain samples before. We expected to see a comprehensive discussion of them. Unfortunately, the discussion is reduced to one assertive statement repeated several times in different parts of the report: «*These data indicate that the inclusions trapped meteoric fluids.*» (p. 16), «*The low  $\delta D$  signatures of fluid inclusion fluids (e.g.  $\leq -105$  ‰; Table 1) indicate that intermediate calcite and MSGC could only have formed from meteoric fluids.*» (p. 24); and « *$\delta D$  compositions of inclusion fluids indicate formation from meteoric fluids.*» (p. 25).

Since no discussion is provided, it is unclear what Wilson and other mean by «*meteoric fluids*». If they use the term to define a broad category of waters antithetic to «*juvenile*» (i.e., derived from the upper mantle of the Earth), or magmatic waters, the statement is justifiable<sup>3</sup> (although water from early calcite from Alcove #5 is compatible with both these sources<sup>4</sup>). It must be noted, however, that a determination that waters, which deposited calcite are of meteoric origin, cannot be used as an argument in support of the formation of secondary calcite from surficial waters percolating through the rocks. It has long been established that «*... essentially all geothermal waters on continents and islands are dominantly of meteoric origin, although a small (~5%) component of magmatic waters cannot be excluded.*» (Criss and Taylor, 1986; p. 390). Thus, the definition of «*meteoric fluids*» does not exclude other possibilities such as hydrothermal upwelling water as the origin of water, which formed the secondary calcite.

We were astonished, also, by the fact that the UNLV researchers based their interpretation on only one isotope, hydrogen. In modern hydrogeologic isotope studies,  $\delta D$  properties are never used in isolation. As Sheppard (1986) argued: «*The combined H- and O-isotope approach has overwhelming advantages because potentially it can give information concerning both the source and history of the water...*» (p. 165). In meteoric waters (e.g., precipitation), H- and D-isotope compositions vary in a very systematic way, described by linear equation of the so called meteoric water line:

$$\delta D = 8\delta^{18}O + 10 \text{ (in ‰)}.$$

Most meteoric waters that have not undergone extensive evaporation plot within a band up to  $\pm 1$  ‰  $\delta^{18}O$  of this line (Sheppard, 1986). Multiple studies have demonstrated that chloride-type waters from hydrothermal systems normally show  $\delta D$  values identical or close to the values of local meteoric precipitation waters. Meanwhile, the  $\delta^{18}O$  in such waters exhibits a so-called  $^{18}O$  shift toward «*heavier*» values, which is caused by an oxygen isotopic exchange between heated meteoric waters and  $^{18}O$ -rich rocks.

A determination of  $\delta^{18}O$  for waters derived from fluid inclusions is technically possible but not feasible for oxygen-bearing minerals formed at elevated temperatures (e.g., calcite or quartz) since these waters could have exchanged oxygen isotopes with the host mineral. Therefore, the  $\delta^{18}O$  of mineral-forming water is normally calculated from  $\delta^{18}O$  values of the hydrogenic minerals. To make such calculations

<sup>3</sup> «*Juvenile*» waters are generally believed to have  $\delta D = -48 \pm 20$  ‰, and waters of magmatic origin are characterized by  $\delta D = -40$  to  $-90$  ‰ (Sheppard and Epstein 1970; Hoefs, 1976; Rollinson, 1993), so the values of  $\delta D < -90$  ‰ are not compatible with these sources.

<sup>4</sup> The data for this sample are not shown in the report (see Table 1) and appear only in technical data files posted at the UCCSN WebPages (see Table 2).

Table 3: Temperatures of formation (by fluid inclusions),  $\delta^{18}\text{O}$  of calcite (measured),  $\delta^{18}\text{O}$  of mineral forming water (calculated), and measured  $\delta\text{D}$  for selected Yucca Mountain samples

Station, description	T indicated by Wilson <i>et al.</i> (2002), °C	T determined from $T_h$ distributions, °C*	Measured $\delta^{18}\text{O}_{\text{cat}}$ , ‰ SMOW	Calculated range of $\delta^{18}\text{O}_{\text{water}}$ , ‰ SMOW	$\delta\text{D}$ , ‰ SMOW
Al#5 00+28.5, Inner part, 2-phase FIAs	35–45 and more	51–55	12.9; 13.7	–9.3 to –10.7	–59; –61
Al#5 00+28.5 Outer part, 2-phase FIAs	35–54	37–43	13.4; 14.4	–10.3 to –12.6	–90; –120
ESF 27+84, Basal, 2-phase FIAs	>35–40	40–50**	13.3; 14.8	–9.0 to –12.1	–87; –89; –90
ESF 27+84, Outer part, MGSC, all-liquid FIAs	25–35***	n/a	16.2; 17.1	–9.3 to –12.2	–110; –115
ESF 60+52.5, MGSC, all-liquid FIAs	25–35***	n/a	18.2; 18.6	–7.8 to –10.2	–131

\* – Temperatures obtained from  $T_h$  histograms reported as technical data at the UCCSN WebPages and include 90% of data in the distribution. \*\* – the  $T_h$  distribution for this sample is bimodal; the analyzed water most likely represents a mixture of waters trapped at two different temperatures. \*\*\* – the temperature estimates accepted for calculations.

possible, the temperature of the fluid must be established independently, for example from fluid inclusions (Sheppard, 1986; Rollinson, 1993). It is assumed that isotopic equilibrium was nearly complete between a given mineral and the mineral-depositing solution.

All necessary data for these calculations are available from the UNLV report. The temperature of formation for Yucca Mountain samples has either been measured from fluid inclusions, or inferred from the absence of two-phase inclusions and the presence of all-liquid inclusions. We recalculated  $\delta^{18}\text{O}$  values measured in those calcite samples for which  $\delta\text{D}$  values and  $T_h$ 's were obtained from fluid inclusions (Table 3) using the equation:

$$\delta^{18}\text{O}_{\text{wat}} = \delta^{18}\text{O}_{\text{cat}} - \left[ 2.78 \cdot \left( \frac{10^6}{(T + 273)^2} \right) \right] - 2.89$$

taken from Faure (1986). The resulting «boxes» are shown in Figures 9 through 11, where the results are placed in a proper context with the isotope geochemistry of local waters and in the broader context of the meteoric precipitation waters and geothermal fluids.

Obviously, any interpretation based on such a scant database must be considered tentative. Although we assume in the analysis below that the ten  $\delta\text{D}$  values of fluid inclusion water reported by Wilson *et al.* (2002) are valid and representative, it must be born in mind that much more data need to be obtained to make interpretations scientifically defensible.

Modern waters sampled from wells in the Yucca Mountain region have  $\delta\text{D}$  values ranging between –96 and –110 ‰ (Paces *et al.*, 2002). It is apparent from Figure 9 that  $\delta\text{D}$  values of waters trapped in fluid inclusions overlap the  $\delta\text{D}$  values of modern waters and extend to both «heavier» and «lighter» values. Waters trapped in the latest calcite, MGSC, have  $\delta\text{D}$  values substantially lower (10–20 ‰) than those of modern waters. The data for most calcites exhibit a prominent shift of  $\delta^{18}\text{O}$  values to the right of the meteoric water line, representing the classic  $^{18}\text{O}$ -shift common for the near-neutral chloride-type geothermal waters. Interestingly, the  $^{18}\text{O}$  shift for late calcite from Yucca Mountain is greater than that observed in the modern hydrothermal system at Steamboat Spring, Nevada (see Figure 10). Importantly, the  $\delta\text{D}$ – $\delta^{18}\text{O}$  data for the Yucca Mountain paleo fluids from late calcite (MGSC in Figure 9) seem to provide strong evidence against the USGS concept, relating deposition of secondary minerals with evaporation occurring underground (Whelan *et al.*, 2002). Evaporation leads to the concomitant increase of both  $\delta\text{D}$  and  $\delta^{18}\text{O}$  in the residual fluid, so that its composition moves to the right and upward on the  $\delta\text{D}$ – $\delta^{18}\text{O}$  cross-plot, remaining at all times below the meteoric water line.

Figure 9. Boxes, enveloping measured  $\delta D$  and calculated  $\delta^{18}O$  values of waters trapped in inclusions in the Yucca Mountain calcite (UNLV data) compared with values for different waters from the Yucca Mountain region (data from Yang *et al.*, 1996). Gray arrows connecting boxes indicate the trend of isotopic change with time.

Actual data reported by Wilson *et al.* (2002) indicate the decrease of  $\delta D$  instead of its increase.

The higher-temperature calcite ( $T_h = 50-55^\circ C$ ) has the heaviest  $\delta D$  values and shows an apparent depletion in  $^{18}O$  so that the data plot to the left of and above the meteoric water line. Such isotopic properties are unusual for most natural waters. Thus far they have been reported only from deep-seated brines, saline waters and brackish groundwaters from crystalline basement rocks in Canada, Scandinavia and Central and Western Europe (Kloppmann *et al.*, 2002; see Figure 10).

In Figure 10 we compare measured and calculated isotopic properties of the Yucca Mountain fluid inclusion waters with those of the modern thermal waters in Nevada and California. It is apparent from the Figure that the isotopic characteristics of most of the fluids trapped in inclusions are similar to those of modern thermal waters. In Figure 11 we compare isotopic properties of the fluid inclusion waters with those from a number of hydrothermal ore deposits in Nevada, as reported by Criss and Taylor (1986). In this case we can again see an almost perfect match between the two sets of data in terms of both hydrogen and oxygen isotope properties.

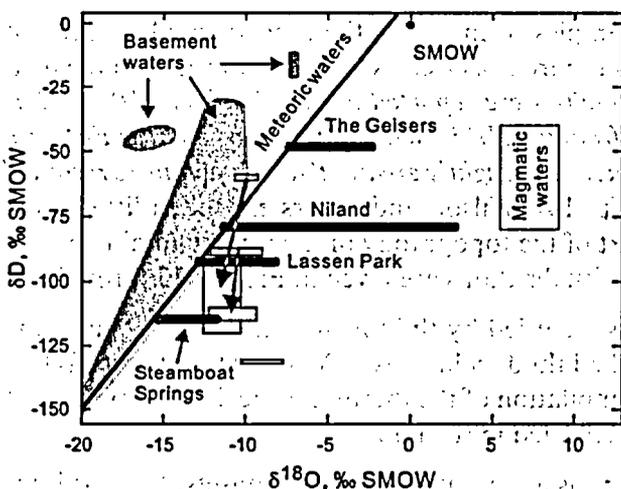
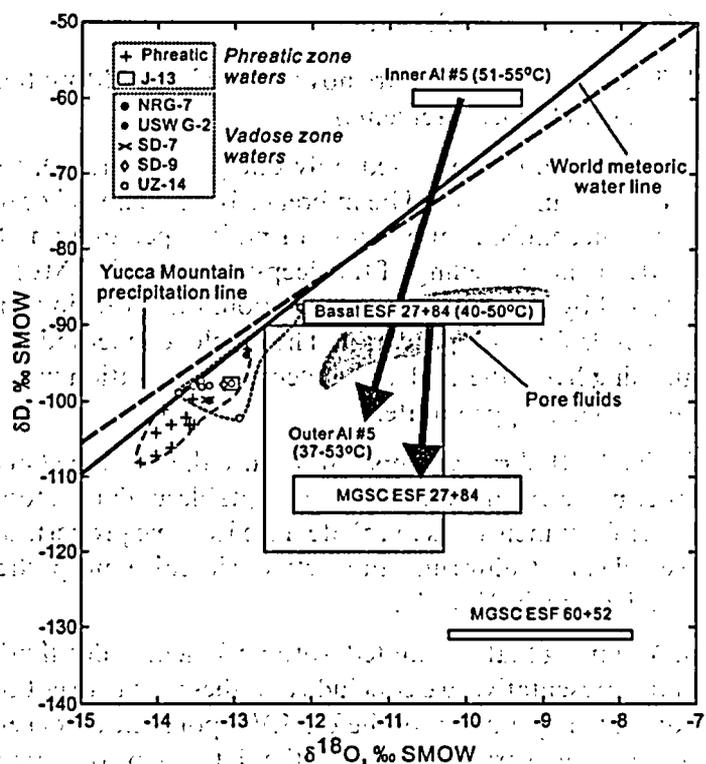


Figure 10. Comparison of the data from inclusions from the Yucca Mountain calcites (boxes) with hydrothermal-meteoric waters.  $\delta D$  and  $\delta^{18}O$  values for hydrothermal systems are from Hoefs (1986); for basement fluids – from Kloppmann *et al.* (2002). Gray arrows connecting boxes indicate trend of isotopic change with time.

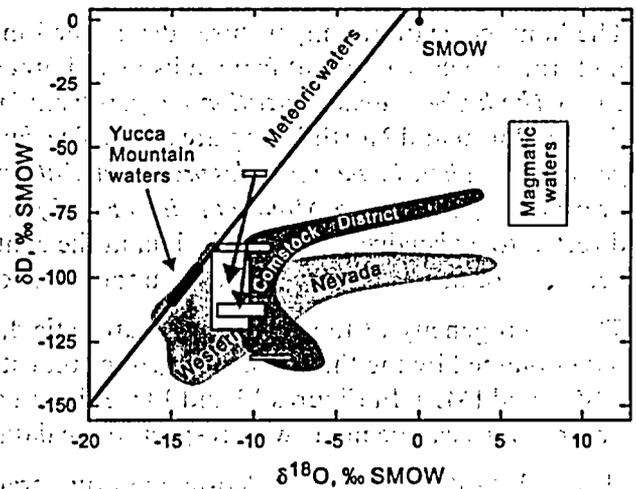


Figure 11: Comparison of the data from inclusions from the Yucca Mountain calcites (boxes) with isotopic compositions of fluids from selected meteoric-hydrothermal ore deposits in Nevada (simplified from Criss and Taylor, 1986). Gray arrows connecting boxes indicate trend of isotopic change with time.

### 2.3.1. Summary on $\delta D$ data

1. The data of this type are novel in the Yucca Mountain secondary mineral studies and the database is meager (10 measurements).
2. Wilson and others used only one parameter,  $\delta D$ , to infer the «meteoric water» origin of waters trapped in inclusions. The inference is ambiguous because it does not discriminate between two proposed mechanisms of mineral formation: percolation of surface waters through heated rock (USGS, UNLV) and upwelling of the deep-seated fluids (IMP). The  $\delta D$  values in near-neutral water are little affected by the water-rock interaction; therefore, in a given region these values are typically indistinguishable for both processes. The employed methodology, thus, is in principle not capable of distinguishing hydrothermal and meteoric precipitation waters. As such, the employed methodology must be deemed inappropriate.
3. The UNLV researchers did not use the combined analysis of H and O isotopes, which provides a more powerful and universally acceptable analytical tool. After applying this approach, we found that water from inclusions in calcite have isotopic properties similar to those of modern thermal springs in Nevada and California, and virtually identical properties to fluids that have formed many hydrothermal deposits in Nevada.
4. Assuming that the limited isotopic data on paleo fluids obtained by UNLV researchers are accurate and representative, we conclude that these data are not compatible with the paleo hydrogeologic model advocated by authors of the UNLV report (the model envisaging percolation of meteoric waters from topographic surface). Instead, the data seem to be perfectly compatible with the hydrothermal upwelling model.
5. A more detailed discussion of the isotopic trends and features that seem to be present in the  $\delta D$ - $\delta^{18}O$  data is not appropriate at this time due to the scarcity of the data and the substantial variations (30 %) obtained from two replicate samples, indicating a potential problem with the data. Further discussion must be preceded by a demonstration of the reliability of the data and enlargement of the database.

### 2.4. Structure of the paleo temperature field

The homogenization temperatures determined in samples from across the repository block are not uniform, which is apparent from Wilson and other's Figure 5, as well as from descriptions in the text, e.g.: «*The highest homogenization temperatures in calcite were reported in samples from NP and NR [North Portal and North Ramp areas]. ... The data from the NR indicate that, where 2-phase FIAs are present, the calcite in these samples precipitated at temperatures slightly greater than temperatures recorded over most of the Yucca Mountain site (Fig. 5)...*» (p. 14). Wilson and others expended little effort directed to a visualization of their data in the context of the topography of Yucca Mountain and the geometry of the repository block. Examples of such visualizations are given in Figures 12 through 14.

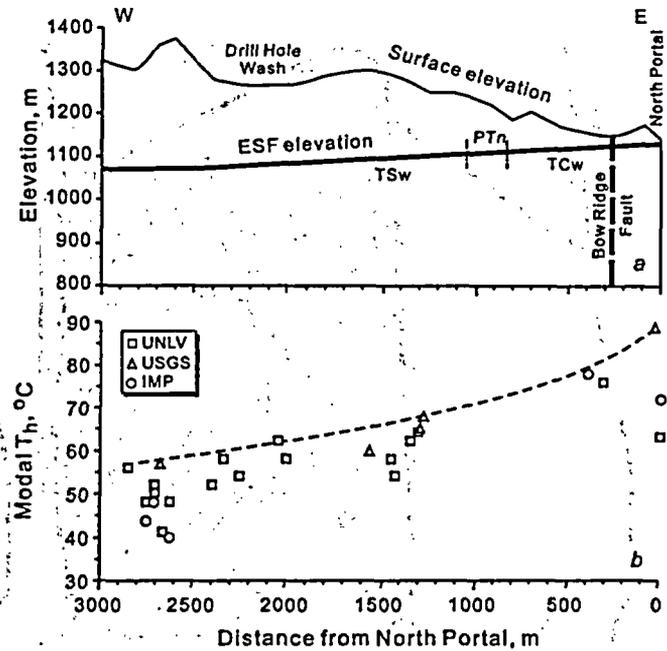
The heterogeneity of the paleo temperature field, as recorded by fluid inclusions, is apparent in the data obtained by the three research groups that studied fluid inclusions at Yucca Mountain (UNLV, USGS and IMP). In Figures 12 and 14 we show a compilation of the thermometric data. A number of observations can be made with respect to the data presented in the figures.

(1) A substantial east-west paleo-temperature gradient is apparent in all three datasets. From this we may safely conclude that the gradient is real. Although it is more prominent in the North ramp, the east-west gradient is also apparent in the South ramp. A prominent minimum is recorded between ESF stations 35+00 and 55+00.

(2) The temperature change seems to be unrelated to the lithology. Although the highest temperatures measured from the ESF are from the Tiva Canyon tuff (TCw), the trend continues into the Topopah

**Figure 12.** Schematic east-west cross-section of Yucca Mountain showing surface and ESF elevations (a) and distribution of the high- $T$  modes of the  $T_h$ 's in samples from this part of the ESF (b). Each mode was calculated on the basis of tens to hundreds of  $T_h$  measurements. Data by UNLV (Wilson *et al.*, 2002; data from the UCCSN WebPages), USGS (Whelan *et al.*, 2001), and IMP (Dublyansky *et al.*, 2001; Dublyansky, 2001-a). All data were statistically treated except for those from Whelan *et al.* (2001), which are reported as modal values.

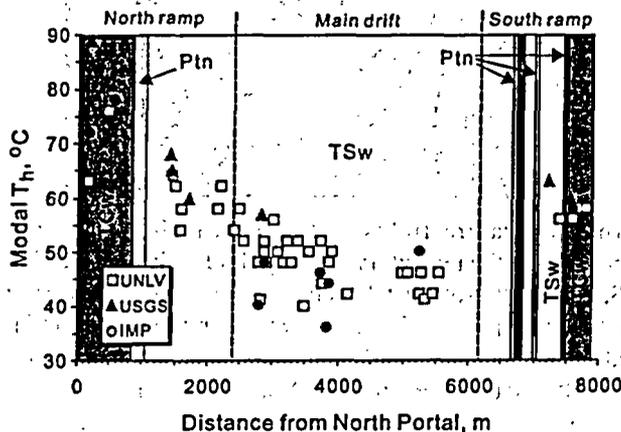
Note: (a) the east-west decline in  $T_h$ 's is recorded by all three datasets; (b) the decline trend seems to persist both between the TCw and TSw tuff units and within the TSw unit, so it does not exhibit direct relationship to the lithology; and (c) the  $T_h$  distribution is unrelated to «normal» geothermal gradient, with temperatures declining with increasing depth.



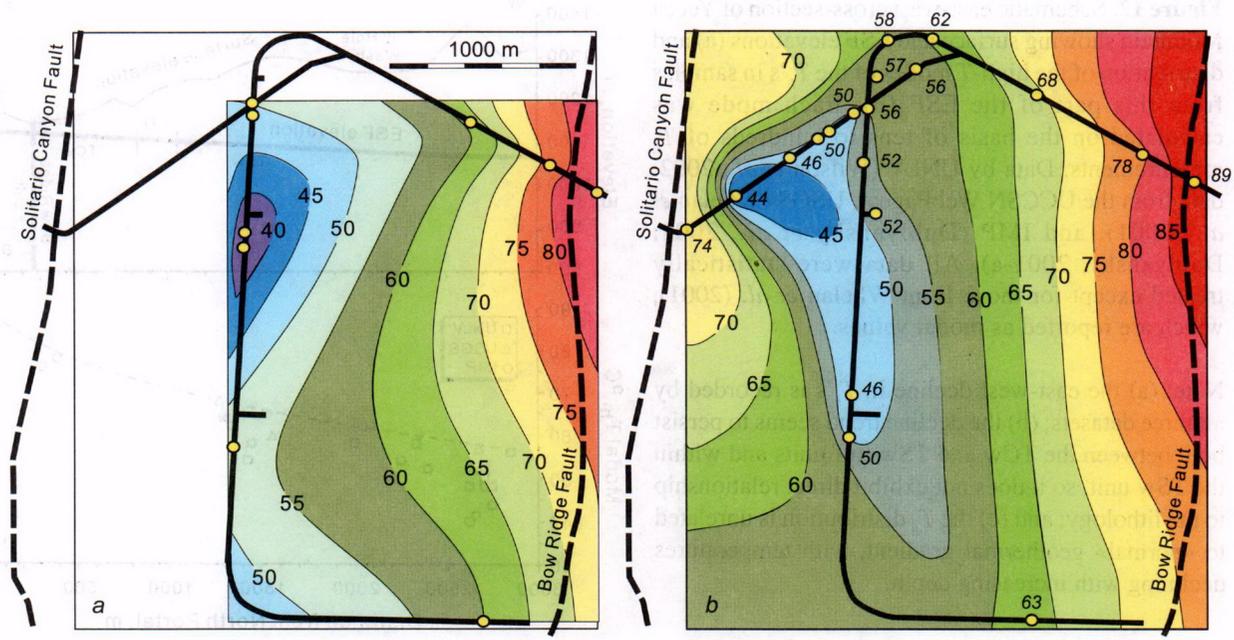
Spring tuff unit (TSw) (see Figure 13). Thus, we have to disagree with the statement of Wilson and others: «*Early, high temperature fluids were restricted to welded Tiva Canyon Tuff and did not extend to deeper areas.*» (p. 22).

(3) The paleo-temperatures are inversely correlated with depth from surface (see Figure 13). The highest temperatures of 75–90°C were measured near the North portal, at a depth of only 30–50 m. The temperatures, thus, are unrelated to the geothermal gradient; in fact, they show an inverse relationship, declining rather than increasing with depth.

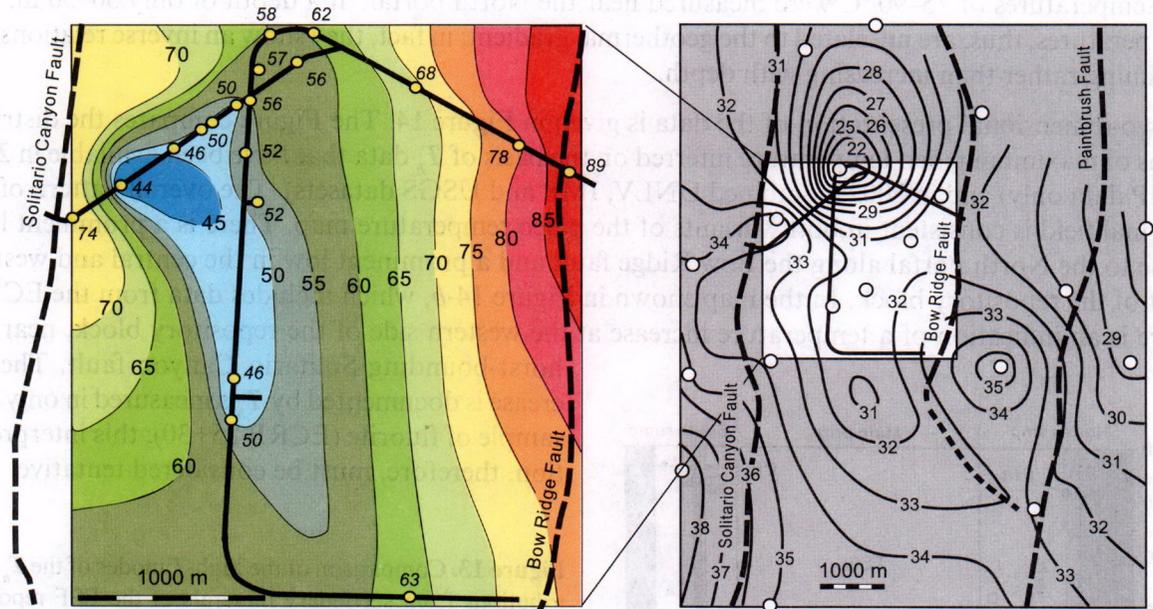
A two-dimensional presentation of the data is given in Figure 14. The Figure compares the distributions of maximum paleo temperature inferred on the basis of  $T_h$  data that have been available in 2001 (IMP data only) and in 2002 (combined UNLV, IMP and USGS datasets). The overall pattern of the thermal field is consistent in both variants of the paleo temperature map. There is a prominent high close to the North portal along the Bow Ridge fault and a prominent low in the central and western part of the repository block. In the map shown in Figure 14-b, which includes data from the ECRB, there is an indication of a temperature increase at the western side of the repository block, near the horst-bounding Solitario Canyon fault. The increase is documented by  $T_h$ 's measured in only one sample of fluorite (ECRB 25+30); this interpretation, therefore, must be considered tentative.



**Figure 13.** Comparison of the high- $T$  modes of the  $T_h$  distributions from secondary minerals of the ESF reported by three research groups. Each mode was calculated on the basis of tens to hundreds of individual  $T_h$  measurements. Data by UNLV (Wilson *et al.*, 2002; UCCSN WebPages), USGS (Whelan *et al.*, 2001), and IMP (Dublyansky *et al.*, 2001; Dublyansky, 2001-a). All data were statistically treated except for those from Whelan *et al.* (2001), which are reported as modal values.



**Figure 14.** Reconstructed field of maximum paleo temperatures (in °C) in the repository area, by fluid inclusions. Yellow circles indicate locations of samples. *a* – Map based on the data of Dublyansky *et al.* (2001) and Dublyansky (2001); *b* – Map based on the integrated data of UNLV (Wilson *et al.*, 2002; UCCSN WebPages), USGS (Whelan *et al.*, 2001), and IMP (Dublyansky *et al.*, 2001; Dublyansky, 2001-a). Black lines show the footprint to the ESF and the ECRB tunnels. Graphic interpolation was done with *Mathcad PLUS 6.0* software. Note that reliability of isotherms is poor at the corners of the map owing to the absence of the data there.



**Figure 15.** Structural similarity in paleo- and modern temperature fields in the Yucca Mountain area. *a* – Reconstructed field of maximum modal paleo temperatures (°C) in the repository area, by fluid inclusions. Yellow circles indicate locations of samples (data from UNLV, USGS, and IMP). Note that the temperatures of fluids in eastern part of the repository block were substantially higher than in its central part. *b* – Present-day temperatures measured in boreholes at the water table (°C) in the vicinity of the proposed repository (based on data of Sass *et al.*, 1987). Boreholes are shown as white circles. Black lines show the footprint to the ESF and the ECRB tunnels. Note two maxima indicating hydrothermal circulation along the horst-bounding fault zones. Graphic interpolation was done with *Mathcad PLUS 6.0* software.

The configuration of the paleo-temperature field, as recorded by fluid inclusions, bears a striking similarity to the structure of the modern temperature field, revealed by measurements in boreholes at the water table. These two fields are compared in Figure 15. Both fields show maxima associated with the horst-bounding fault zones (Solitario Canyon and Paintbrush-Bow Ridge faults) and a minimum associated with the central part of the Yucca Mountain block. From a hydrologist's perspective, the configuration of the modern temperature field can be interpreted as a feature, which reflects the enhanced conductivity of the fault zones, allowing for the convective circulation of heated fluids (Sass *et al.*, 1983; Lehman and Brown, 1995; Bredehoeft, 1997). Similarly, the configuration of the paleo temperature field, inferred from the fluid inclusion data, strongly suggests that these fault zones served as conduits for circulation of heated fluids, from which minerals were deposited.

#### 2.4.1. Interpretation of the paleo temperature field by Wilson and others

In view of the information presented in Figures 12 through 15, it is inconceivable to us how the following statement could have been made by Wilson and others: «*It is noteworthy that temperatures recorded across the Yucca Mountain repository horizon do not exhibit a central hot plume and large lateral thermal gradients that are present in geothermal and epithermal systems (Henley, 1985). The lack of a significant temperature gradient and presence, instead, of relatively uniform temperatures argues against an upwelling hot fluid model.*» (p. 22).

In their statement Wilson and others bluntly deny the presence of lateral thermal gradients that are clearly observed in their own and the other's data. We note that terms such as «significant gradient» and «relatively uniform distribution» are arbitrary. We reiterate, in this regard, our opinion stated in Section 2.1 above, that usage of loose and undefined terms could be seriously misleading and should, therefore, be avoided. We insist, that from the standpoint of both hydrogeology and geothermometry, the characterization of the paleo temperature field shown in Figure 15-a as «relatively uniform» is grossly misleading and, therefore, absolutely inappropriate.

Wilson and others further refer to a «central hot plume» (!), which they do not observe. The data show, however, (e.g., Figure 15-a) that the location of the thermal high, both from a hydrologist's perspective, as well as from the perspective of the hydrothermal upwelling model, is precisely where it should be: along the zone of enhanced permeability accompanying the deep-seated fault. Thermal highs presently exist at the water table in association with these zones (see Figure 15-b).

Wilson and others opined that the temperature variations recorded by fluid inclusions do not reflect a spatial trend but, rather, a temporal trend: «*These temperatures, furthermore, are not related to lateral temperature gradient across the site because the temperature variations occurred at different times.*» (p. 16) or «*The distribution of fluid temperatures is related to the timing of mineral precipitation at various sites.*» (p. 21). Such conclusions, if proven to be correct could be very important; therefore, the observations and arguments supporting them must be carefully documented. We could not find in the Wilson and other's report any convincing evidence in this regard.

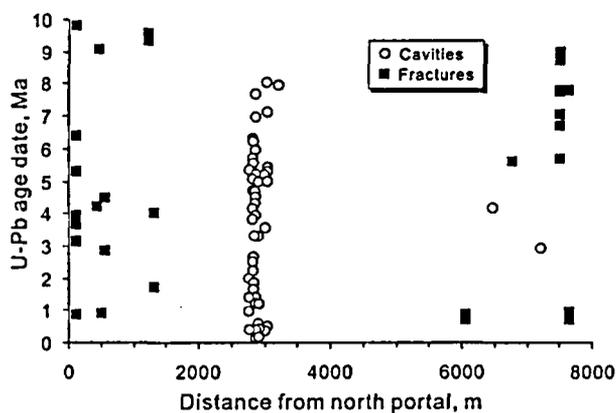
The following is an unsubstantiated argument made by the UNLV researchers: «*The wall rock surface in most fracture and breccia samples consists of broken tuff that lacks vapor-phase minerals. These surfaces suggest that fracture/breccia development occurred after vapor-phase alteration of the host tuffs and, in turn indicate that secondary minerals in fractures and breccias began to precipitate later than secondary minerals in LC [lithophysal cavities]*» (Wilson and Cline, 2002, p. 19–20). Without basis, they stretch this argument further and state: «*The low homogenization temperatures in samples from the IFZ [Intensely Fractured Zone] suggest that fracture- and breccia-related calcite probably had not precipitated when the earliest, higher temperature fluids invaded the site. The lack of vapor phase minerals in these samples is consistent with their later formation. These observations suggest that the various tem-*

perature ranges recorded across the site reflect fluid fluxes that occurred at different times... « (Wilson *et al.*, 2002, p. 14).

The argument that the absence of vapor-phase minerals in fractures and breccias indicates that secondary minerals there «*began to precipitate later than secondary minerals in lithophysal cavities*» is a *non sequitur*. During early stages of the cooling of the tuff, vapor-phase minerals formed on the walls of cavities. Formation of the vapor-phase minerals ended shortly after the deposition of the red-hot ash flow mass and its compaction and conversion into welded tuffs. The tuffs at Yucca Mountain likely compacted and cooled to ambient temperatures within about 100 to 1,000 years after deposition (Riehle, 1973; U.S. DOE, 2001) and the stage at which vapor-phase minerals formed took only a fraction of this time. Thus, the absence of the vapor-phase minerals on the fracture walls tells us no more than that the fracture is younger than 12.7 Ma but, in the context, «younger» could mean millions of years or just tens of years.

The fallacy in Wilson and others argument is best demonstrated by the fact that the highest homogenization temperatures measured at Yucca Mountain (for example, ESF 01+62.3 and ESF 04+73.4) came from fractures devoid of vapor-phase minerals: «*A NR [North Ramp] sample from a fracture occurrence ... contains primary 2-phase FIAs with homogenization temperatures that reach 75 °C.*» (p. 20). This means that the fractures were present during the earliest stages of fluid circulation, but were absent earlier, during the stage of the vapor-phase alteration. In addition, both the U-Pb age dates of Wilson *et al.* (2002) and Neymark *et al.* (2002) show that many fractures contain minerals, which in fact appear to be older than their counterparts from the lithophysal cavities (Figure 16).<sup>5</sup>

The relative ages of minerals can also be assessed on the basis of their stable isotope properties. It has been shown for the Yucca Mountain samples that the «... $\delta^{13}\text{C}$  compositions decrease from positive values in older calcite at the base of crusts to negative values around  $-6.0$  to  $-8.0$  in the youngest calcite.» (Wilson and Cline, 2002, p. 17; see also Figure 6 in this review). Figure 17 shows clearly that the early, « $\delta^{13}\text{C}$ -positive» calcite is present in all temperature zones of the ESF – from the high-temperature portal areas (see Figure 13) to the coolest area between ESF stations 35+00 and 55+00 (Wilson and Cline's IFZ zone). This is consistent with conclusions made by the USGS researchers, e.g.: «*The large range of  $\delta^{13}\text{C}$  values, as plotted against location in the ESF ..., shows that the entire paragenetic sequence is present in mineral coatings throughout the ESF.*» (Whelan *et al.*, 2002, p. 742).



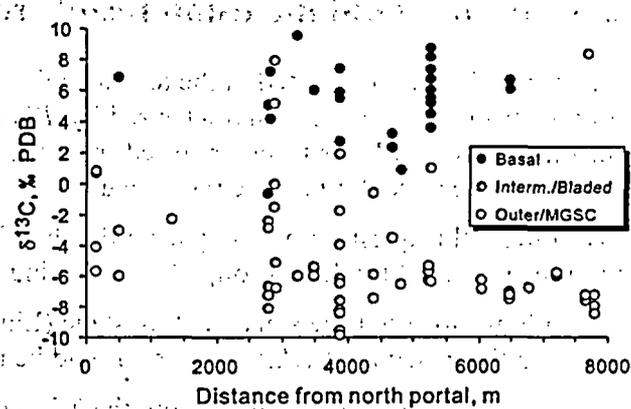
Carbon is a «conservative» component of the fluid, whose isotopic composition reflects the compositions of sources of dissolved bicarbonate, such as deep-seated-, soil-, or atmospheric  $\text{CO}_2$ , for example. Fractionation of carbon isotopes changes little with temperature (for example, the fractionation coefficient between  $\text{HCO}_3^-$  and  $\text{CaCO}_3$ )

Figure 16. U-Pb age dates obtained from opal and chalcedony in fractures and lithophysal cavities of Yucca Mountain. Combined data from Wilson *et al.* (2002) and Neymark *et al.* (2002). Circles – lithophysal cavities; squares – fractures.

<sup>5</sup>We cannot, at this time, accept the validity of the U-Pb ages reported by Wilson *et al.* (2002) and by Neymark *et al.* (2002). We refer to these age data simply to highlight the internal inconsistency of the argument. Specific problems with the U-Pb dating will be discussed in Section 2.6 of this report.

Figure 17.  $\delta^{13}\text{C}$  values of calcite from basal, intermediate and outer parts of mineral crusts. Data from Wilson and Cline (2002) and Dublyansky (2001).

changes by 0.4 ‰ between 20 to 90 °C). Nevertheless, in the Yucca Mountain samples carbon shows a dramatic unidirectional shift, decreasing from ca. +10 at the bases to ca. -10 ‰ in the outer parts of crusts. Reversals or repetitions have not been observed (in samples that have readily interpretable textures). From this, one must infer dramatic and unidirectional changes in the source of  $\text{CO}_2$  dissolved in the fluid.



Wilson and others speculate that waters with different temperatures entered the repository block at different times: earlier in the portal areas and later in the IFZ area. In order to reconcile this model with the  $\delta^{13}\text{C}$  data shown in Figure 17, one must assume that the waters that entered the portal area, say around 10 Ma ago (Wilson *et al.*, 2002), and those that entered the IFZ area a few million years later had the same initial isotopic compositions (positive  $\delta^{13}\text{C}$ ) and subsequently evolved in the same manner (toward negative  $\delta^{13}\text{C}$  values of ca. -8 to -10 ‰). It is difficult to imagine a geological situation in which sources of the dissolved carbon changed so dramatically (by about 20 ‰) more than once.

A straightforward explanation that accounts for all observations is that the waters injected along the Paintbrush-Bow Ridge fault zone (north and south portal areas of the ESF) moved westward and cooled down. The carbon isotope properties of the fluid are not expected to change substantially in such a system, so that the early parts of the fluids deposited calcite with nearly the same «heavy positive»  $\delta^{13}\text{C}$  values (+8 to +10 ‰ PDB). While in the portal areas these minerals were deposited at temperatures as high as 70–85 °C, in the IFZ area located far from the «feeder conduit» the temperature of the fluid was lower. For example, in a number of samples, which possessed  $\delta^{13}\text{C}$  values of +9 ‰ characteristic of the earliest calcite, two-phase fluid inclusions (indicating elevated temperatures) were not found (e.g., sample ESF 52+43 shown in Figure 6). This reflects the progressive cooling of the fluid as it moved away from the conduit.

#### 2.4.2. Summary on the structure of the paleo temperature field

1. The fluid inclusion data indicate that a strongly non-uniform temperature field existed within the repository block during crystallization of the early secondary minerals. Lateral gradients of 15–20 °C/km appear to be characteristic of that stage of mineral deposition. Maximum temperatures were associated with the Paintbrush-Bow Ridge fault zone. The existence of strong lateral gradients is apparent in the data of the three independent research groups (UNLV, USGS, IMP; see Figures 12 through 15).
2. Wilson and others asserted that the observed fluid inclusion temperature distribution is caused by different times of fluid infiltration, so that locations with higher fluid inclusion temperatures were accessed with early hot fluids, whereas those showing lower fluid inclusion temperatures were accessed only later by cooler fluids. Based on this assertion they proposed the «lack of a significant temperature gradient and presence, instead, of relatively uniform temperatures».
3. We have found that the interpretation of Wilson and others is not supported, in the report, by any factual evidence. As a hypothetical mechanism, it would require a geologically unrealistic chain of events. Actual realization of such a chain of events is not supported by available geologic and geochemical data.

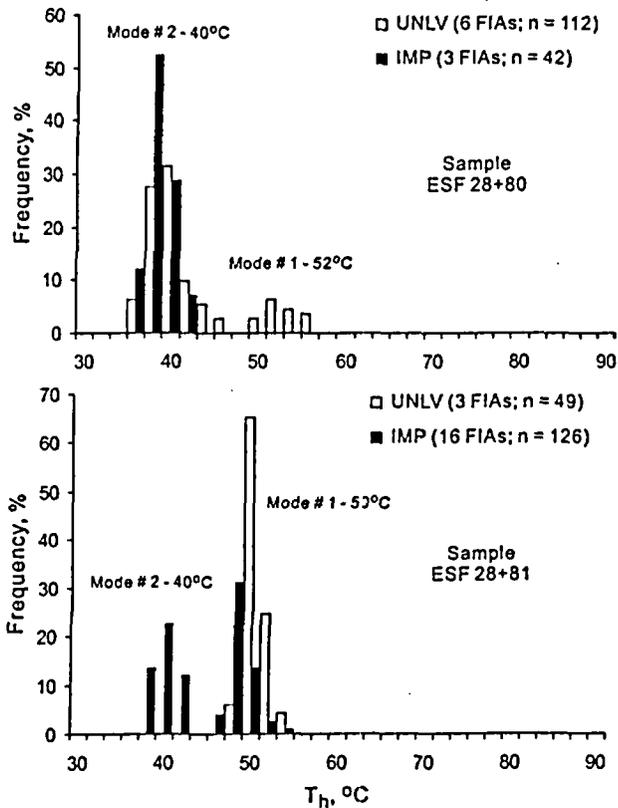
## 2.5. Are the fluid inclusion results representative?

Analyzing the data presented in the technical files posted at the UCCSN WebPages, we observed that a number of samples were characterized by  $T_h$ 's measured from only one to three FIAs. It was not uncommon for all FIAs to be analyzed from the same chip of a polished section. The question thus arises: how representative were these results?

We address this question by comparing the fluid inclusion results reported by the UNLV researchers with the results of the IMP group. The following needs to be noted in this regard:

- 1). Both groups used the same heating stage (Linkam THMSG 600) calibrated with synthetic inclusions, so the instrument-related errors should be minimal.
- 2). In all cases used in this comparison, the  $T_h$ 's measured for individual FIAs are internally consistent (i.e., fall within a relatively narrow temperature interval); and
- 3). Samples for fluid inclusion studies were collected independently and at different times. This means that although the specimens were collected from the same locations (fractures, cavities), they do not necessarily sample immediately adjacent parts of mineral crusts.

By comparing the results, therefore, we may assess both the role of the number of analyses done on the sample, as well as the variability of the  $T_h$ 's within an individual mineralized cavity. Figure 18 shows the results from two adjacent cavities (located within 1 m from one another; ESF 28+80 and 28+81). It is immediately apparent that the number of analyses is quite important. In sample ESF 28+81 two modes are clearly seen in the IMP data (16 FIAs analyzed), while only one mode is apparent in the UNLV data (3 FIAs). Similarly, in sample ESF 28+80, fewer data obtained by IMP (3 FIAs) show only one mode at 40°C, whereas 6 FIAs analyzed by UNLV reveal a small, but statistically distinct mode at 52°C.



treated as showing different  $T_h$  distributions (monomodal and bimodal). If the two datasets are combined, it becomes apparent that both samples show bimodal distributions with virtually identical characteristics.

In the two examples given in Figure 18, the results obtained by the two groups are consistent in the fact that the positions of one of the two  $T_h$  modes coincide. This suggests that the samples, most likely, are adequately characterized by the combined dataset. Figure 19 shows a somewhat different situation, in which a relatively large number of FIAs analyzed by UNLV (9 FIAs) from two chips of the thick section, yielded internally consistent results, but these results are distinctly different from the IMP results obtained on only 3 FIAs from a sample collected in the same fracture.

Regarding this sample Wilson and others stated: «Figure 6 illustrates the consistency of homogeniza-

Figure 18. Comparison of the fluid inclusion results obtained from two samples (ESF 28+80 and 28+81) by UNLV and IMP groups.

Figure 19. Data obtained by UNLV and IMP from sample ESF 01+63.2.

tion temperatures for 181 fluid inclusions in sample ESF 01+62.3 from NP [North Portal]. Most inclusions homogenized from 61-67 °C, a temperature range of 6 °C. Data include all homogenization temperatures from one chip and data from six FIAs identified in a second chip. These results show that a single FIA is representative of all data from this sample.» (p. 13; emphasis added). Our Figure 19 shows, that although these 9 FIAs might be representative of the sample (i.e., thick section or chip); the sample itself, however, is not necessarily representative of the mineral occurrence (i.e., of the assemblage of secondary minerals present in a given mineralized cavity). Therefore, it may not provide complete information regarding the temperatures of fluids that deposited minerals in this particular cavity.

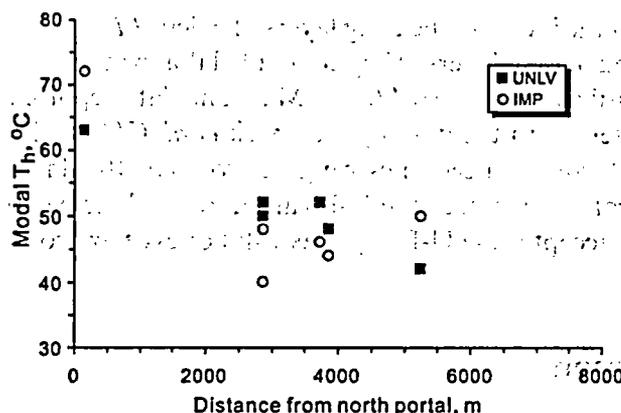
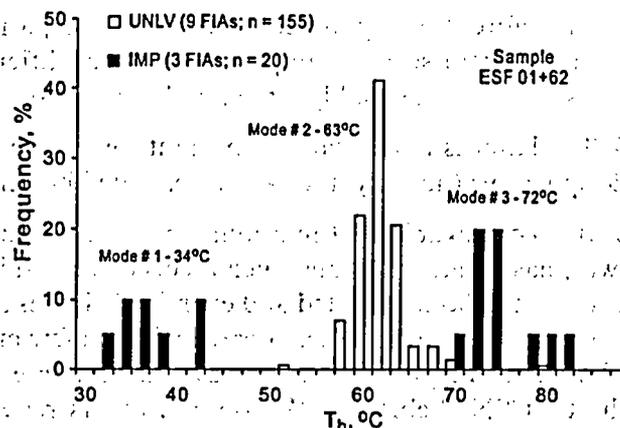


Figure 20. Comparison of the highest  $T_h$  modes for samples collected in the same cavities (but not exactly at the same locations) by UNLV and IMP.

As a consequence, inferences based on such data may be in error. For example, Wilson and others have stated: «*Interestingly no samples from NP and NR [north portal and north ramp] have bimodal distributions...*» (p. 22). Our Figure 18 shows that the sample 01+63.2 from the NP area does show a polymodal distribution. This emphasizes the need of a more spatially distributed sampling within

individual cavities. Samples must be taken and analyzed from different parts of the cavity. This is particularly important for mineral occurrences in which the paragenetic context is complex and not readily decipherable (e.g., breccia cementation).

Fluid inclusion data obtained by UNLV and IMP from the same cavities but from different samples are compared in Figure 20. The Figure shows statistical modes of the  $T_h$  distributions (for each sample, only the highest- $T$  mode is shown). It is apparent that deviations in the modal values may be as great as 10 °C.

### 2.5.1. Summary on the representativeness of the fluid inclusion data

1. Several samples from report by Wilson and others were characterized by  $T_h$ 's measured from one to three FIAs. While these data do reflect real temperatures of mineral forming fluids (provided the FIAs meet the criteria of consistency), they may sample only a fragment of the thermal history of a given mineral occurrence.

2. Nearly all of the data reported by Wilson and others were obtained from single thick sections, prepared from samples collected in the course of the joint UNLV-USGS sampling program. A comparison with the data obtained from different sections (from other parts of the same mineralized cavities) indicates that in some cases individual sections reveal only a part of thermal history of the given mineralized cavity.

3. Therefore, for any mineralized occurrence, a reliable picture of the thermal history can only be obtained from several samples taken from different parts of the cavity and characterized by  $T_h$ 's measured in a reasonably large number of FIAs.

4. The latter is particularly important for cavities in which the style of mineralization is not readily decipherable (notably, the calcite-cemented breccias).

5. It is conceivable that in a number of mineralized occurrences that have been deemed lacking the two-phase fluid inclusions, such inclusions could have been found, if samples from other parts of the cavities had been collected and examined. This is supported by the fact that even in sections prepared from the same sample (i.e., located within a few mm from one another) there are «... variations in the abundance of 2-phase FIAs.» (p. 21). In another example made apparent from the materials posted at the UCCSN WebPages, Wilson and others reported two-phase fluid inclusions from sample ESF 29+79. However, in a sample collected from the same lithophysal cavity earlier, two-phase fluid inclusions were not found (Dublyansky, 1998).

## 2.6. Problems with the U-Pb ages

In their report, Wilson and others assessed the ages of the inclusion bearing layers of calcite ( $T_h = \sim 45\text{--}60\text{ }^\circ\text{C}$ ) by «bracketing» them between the U-Pb-dating of opal layers. They concluded that waters with elevated temperatures accessed Yucca Mountain until as recently as 4.0–5.3 Ma.<sup>6</sup> The data reported by Wilson and others appear to be generally consistent with the data by Neymark *et al.* (2002) who reported U-Pb age dates for Yucca Mountain opals and chalcedonies ranging from *ca.* 10 Ma to several Ka. There seem to exist, however, a serious conflict between the U-Pb age dates and the paleo temperatures, a conflict which does not permit us to accept these U-Pb ages as valid or even approximate.

### 2.6.1. General thermodynamic consideration

It is generally accepted that modern landforms were already established at Yucca Mountain 11.6 million years ago, and since that time, the rates of erosion were very low (<0.1 to 0.5 cm per thousand years; U.S. DOE, 2001). It is further believed that no more than ~100 m of the rocks could have been removed from Yucca Mountain. This means that secondary minerals studied from the Yucca Mountain vadose zone have formed at a depth similar, or just slightly greater ( $\leq 100$  m) than they are located today.

Thus, the acceptance of the two independent data sets: paleo temperatures (based on fluid inclusions) and U-Pb ages would lead to the conclusion that temperatures as high as 45–60 °C have persisted in the shallow vadose zone of Yucca Mountain (at a depth of 30 to 300 m from the paleo surface; see, e.g., Figure 12) during the several million year-long period (between 9–10 Ma or earlier and 4–5 Ma). These temperature/depth relationships translate into paleo heat flows as great as 3.5 to 62 HFU<sup>7</sup>; roughly 2 to 36 times the average in the western United States. These values are extraordinarily high.

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<sup>6</sup> Ma = million years ago; Ka = thousand years ago.

<sup>7</sup> Heat flow,  $q$ , is defined as geothermal gradient  $dT/dz$  [ $^\circ\text{C}\cdot\text{km}^{-1}$ ] multiplied by thermal conductivity of the rock,  $k$ , [ $\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ ] and is expressed in [ $\text{mW}\cdot\text{m}^{-2}$ ] or in Heat Flow Units, abbreviated HFU (1 HFU = 42  $\text{mW}\cdot\text{m}^{-2}$ ). The mean value of heat flow for the Western USA is  $q = 1.8$  HFU ( $\sigma = 0.87$ ,  $n = 190$ , Sass *et al.* 1971).

In northern Nevada, for example, heat flows of ~24 HFU support geothermal power generating plants as well as other non-electrical geothermal applications (Sass, 1999). Such high heat flows are possible only in association with geologically short-lived events (e.g., cooling of the freshly deposited pyroclastic rock) or in active geothermal systems. There is no geologically reasonable source of energy available to generate and discharge, through Yucca Mountain, that much heat over such a long period of time (4-6 million years or more).

### 2.6.2. Thermal history of Yucca Mountain known on the basis of geologic and mineralogic evidence

Two thermal events are considered well established in the Yucca Mountain geological history. The first event was the deposition of the ash-flow tuffs, which built up layers of the mountain. Following its deposition by 12.7 Ma, the tuff "pile" was hot; however, cooling to ambient temperatures took a relatively short time (between 100 and 1000 years; Riehle, 1973; U.S. DOE, 2001). The second was the Timber Mountain Caldera hydrothermal event. A hydrothermal convection system was set off by a granitic magma body, which resided under the Timber Mountain caldera, some 8-10 km to the north of Yucca Mountain. The Timber Mountain event is held responsible for pervasive zeolitic, montmorillonitic and carbonate alteration of the rhyolitic tuffs and the deposition of abundant calcite and silica below ~1.2 km under the surface of Yucca Mountain. The data of Bish and Aronson (1993) on the K-Ar ages of clay minerals and zeolites constrain the age of this alteration to 10.0-10.6 Ma, after which the source of heat was exhausted, and the hydrothermal activity ceased. Mineralogical data indicate that, during the Timber Mountain Caldera event, values of heat flow in the planned repository area did not likely exceed 2.9 to 5.0 HFU and temperatures at the ESF level (reference depth of ~250 m) were less than approximately 35-37 °C (Szymanski *et al.*, 2000).

Summarizing the time and temperature constraints briefly discussed above, it is easily demonstrated that secondary minerals collected at the level of the ESF or higher, with an established temperature of formation >~45°C and/or ages younger than ~12.7 Ma cannot be related to the Timber Mountain Caldera hydrothermal event.

**Summary.** Elevated temperatures (35-85 °C) recorded by fluid inclusions in the vadose zone between ca. 10 Ma and 4.0-5.3 Ma (U-Pb ages by Wilson *et al.*, 2002) cannot be related to either of the known thermal events at Yucca Mountain on the basis of geologic and mineralogical records. An apparently irreconcilable conflict, thus, exists between the U-Pb radiometric dating results and the paleo-thermometric data.

### 2.6.3. What is wrong with the U-Pb dating?

The U-Pb dating is a method, which is typically applied to relatively old geological objects (hundreds of Ma and older). This method has helped earth scientists determine the age of the earth. The amounts of radiogenic <sup>206</sup>Pb and <sup>207</sup>Pb isotopes accumulated due to the *in situ* decay of their parent <sup>238</sup>U and <sup>235</sup>U over relatively short periods of time (e.g., 10 Ma) is very small because of the long half-lives of the parent isotopes ( $T_{1/2} = 4.7$  billion years for <sup>238</sup>U and  $T_{1/2} = 0.7$  billion years for <sup>235</sup>U). Even though these amounts could be measured by modern equipment (particularly if the content of U in dated mineral is high), the results remain highly susceptible to perturbations.

Pashenko and Dublyansky (2002-a and -b) developed a physicochemical model, which demonstrates that the applicability of the U-Pb method could be severely limited when dating minerals that: (a) form in open cavities (>0.1 cm), (b) form from colloidal solutions, and (c) are young (Miocene or younger). The model stipulates that minerals growing in an open cavity are exposed to a flux of addi-

tional radiogenic Pb isotopes, not accounted for by the “common lead” correction. The parent for these isotopes is U, which resides in the surrounding rock. The decay chains of U contain radon, which, being a gas, readily diffuses into the cavity and, after several decays, produces stable isotopes of Pb. Modeling shows that concentrations of this Rn-derived radiogenic Pb in relatively large cavities will be substantially higher than in thin fractures (<0.1 cm).

Opals, particularly uraniferous ones, normally form from colloidal solutions (Zielinski, 1982). The USGS researchers seem to agree that the Yucca Mountain opals also formed by this mechanism. Discussing the geological meaning of the U-Pb ages, they stated: «*If amorphous opal-A forms from maturing silica gel precipitated from water (Ludwig et al. 1980; Zielinski, 1982), its U-Pb age would reflect the time when the water redistribution and migration of dissolved ions terminated within the precipitating solid phase. This is probably very close to the time of the silica gel deposition, assuming closed system behavior from that time until the present. However, subsequent crystallographic ordering and transformations like opal-A→opal-CT→chalcedony complicate the exact meaning of U-Pb ages.*» (Neymark et al. 2002, p. 724).

In colloidal solutions, the accumulation of the Rn-derived Pb isotopes occurs through adsorption on the micelles. Upon coagulation and sedimentation, the micelles become incorporated in the opal. Calculations show that concentrations of  $^{206}\text{Pb}$  and  $^{207}\text{Pb}$  on the order of  $n \cdot 100$  ppb, typically observed in the Yucca Mountain opals, may be acquired by micelles of silica over a period of several days. Controlled primarily by the velocity of water exchange in cavities, the absolute quantities of Pb contained in the Yucca Mountain opals could accumulate within  $n \cdot 100$  to  $n \cdot 1000$  years.

#### 2.6.4. Summary on the U-Pb dating

1. The results of the U-Pb age dating by Wilson et al. (2002), indicating the presence of thermal waters, within the vadose zone at Yucca Mountain over a 4 to 6 million years period of time seem to be problematic. Keeping several cubic kilometers of rock constituting the vadose zone of the mountain this hot (as required by the USGS and UNLV models) requires an extremely potent source of energy. The existence of such a heat source does not seem to be possible from general thermodynamic considerations; it is also not supported by the available geologic and mineralogical data.

2. The model developed by Pashenko and Dublyansky (2002-a, -b) explains why the U-Pb ages of secondary minerals may not be correct. The Yucca Mountain silica minerals could have formed within a much shorter period of time, on the order of thousands of years. Due to the incorporation of the additional radiogenic Pb isotopes produced by emanation and diffusion of radon, its decay to Pb, subsequent adsorption of Pb on silica colloids and coagulation and sedimentation of the latter, the apparent U-Pb ages calculated by employing conventional dating equations would become much greater than the true ages.

3. Our analysis of results of the U-Pb dating of secondary silica minerals, reported in Wilson et al. (2002), shows that the analysis was done «by the book». Nevertheless, we contend that the use of a method in a situation, which stretches the limits of its applicability, must be preceded by a careful evaluation of the possible limitations of the method. The physical mechanism outlined above (involving emanation of Rn and coagulation of colloids) may be one such limitation. The conceptual model supporting hydrothermal upwelling fluids suggests there are others.

4. Unless a reasonable and verifiable explanation reconciling the apparent conflict between the U-Pb age dates and paleo temperature data is offered, the U-Pb ages cannot be accepted.

## 2.7. Questions not asked

The UNLV researchers formulated the goal of their research as follows: «*Since secondary minerals formed from fluids that invaded the repository rocks, the secondary minerals have been examined to determine whether they formed in a vadose or phreatic environment, and from downward percolating meteoric fluids or from upwelling hydrothermal fluids.*» (Wilson and Cline, 2002; p. 6). The UNLV researchers failed to use at least one method, which had the potential for providing answers to the question posed.

### 2.7.1. Compositions of gases trapped in inclusions

One potential means for discriminating between the vadose (above water surface) and phreatic (below water surface) origin of minerals is the study of the composition of gases trapped in fluid inclusions (Newman *et al.*, 1996). Preliminary data reported by Levy *et al.* (1995) suggest that calcite from the ESF contains gases, whose ratios indicate reducing anoxic conditions (dominant CH<sub>4</sub>, very little O<sub>2</sub>) and a phreatic environment of formation (H<sub>2</sub>O = 99.2 to 99.9 mol %). Both O<sub>2</sub> contents and O<sub>2</sub>/N<sub>2</sub> ratios are identical to those of the hydrothermal carbonates and dissimilar, by as much as 1 to 2 orders of magnitude, from the vadose zone pedogenic carbonates studied at different locations in the southwestern United States (Newman *et al.*, 1996).

Peculiar all-gas inclusions have been reported by all researchers studying secondary minerals from Yucca Mountain. Wilson and Cline (2002) and Wilson *et al.* (2002) have also reported them. No effort was made to analyze the chemistry of these inclusions or to evaluate their significance. Based on strong luminescence under Raman studies, Dublyansky (2001-b) suggested the presence of gaseous aromatic (cyclic) hydrocarbons in these inclusions. As the gases trapped in fluid inclusions seem to have chemistries that are not compatible with the chemistry of the underground air, the results of both studies argue against the model in which minerals are deposited in the vadose zone from water films. It is unfortunate that studies were not performed on these inclusions during the UNLV project.

If the overall topography of Yucca Mountain remained unchanged and the 300-700 m-thick vadose zone existed at the mountain at all times over the last 10 million years or so (as the «meteoric water» model of secondary minerals postulates, e.g., U.S. DOE, 2001; Whelan *et al.*, 2002), the physical processes controlling the migration of gases through the mountain may be expected to persist throughout its history. The present-day data, thus, may be used as a baseline for assessing the chemistry of the underground atmosphere. The modern-day underground air contains O<sub>2</sub> and N<sub>2</sub> in «atmospheric» proportions and slightly elevated concentrations of CO<sub>2</sub> (up to 0.13 vol. %). Both underground air and soil gases are depleted in CH<sub>4</sub> (0.05 to 0.2 ppmv and 0.5 to <1.7 ppmv), in comparison to the atmospheric air (1.7 ppmv; Thorstenson *et al.*, 1989). It is apparent that modern day underground gases have no genetic relationship to the gases trapped in the fluid inclusions.

## 2.8. Genetic models for the formation of secondary minerals

In section 6.5. «Genetic Models for the Formation of Secondary Minerals», Wilson and others state that their data are consistent with the models proposed by U.S. Geological Survey geologists. They provide an abbreviated description of how, in their view, the minerals were deposited. The description rests heavily on the publications of USGS scientists, which are extensively referenced. Since it appears to us that Wilson and others have perfunctorily endorsed everything that was proposed by USGS scientists, we address below the most glaring inconsistencies of this joint USGS-UNLV model.

### 2.8.1. Origin of elevated temperatures

One of the crucial questions raised in the interpretation of the fluid inclusion data is: what was the source of heat that is expressed by circulation of waters with elevated temperatures? Wilson and others seem to believe that the tuff sequence at Yucca Mountain was «warm» (>50 °C) for several million years «... following intrusion of the Timber Mountain at around 10 Ma (Marshall and Whelan, 2000).» (p. 25; emphasis added).<sup>8</sup> Furthermore, that thin films of surficial waters infiltrated down into the rock and became heated upon contact with the warm rocks. In order to be compatible with the fluid inclusion data and the U-Pb dating results, the rock must have been heated conductively to temperatures significantly exceeding ambient temperatures (35 to 85 °C, as compared to ca. 22 °C at the ESF level today) for some 4–6 million years (see discussion in Section 2.6 above).

We submit that such a prolonged conductive cooling of the rock mass is not possible. Cooling of a shallow magma chamber, which was invoked by the USGS researchers as a heat source (Marshall and Whelan, 2000) takes about 1–2 million years depending strongly on the volume of melt that remained in the chamber after eruption (e.g., Wohletz and Heiken, 1992). In the case of the Timber Mountain caldera complex, recent work suggests that the magma chambers experienced nearly total evacuation during each eruption (Bindeman and Valley, 2003).

Many additional factors contribute to faster cooling, the major factor being convective removal of heat from surrounding rocks by circulating fluids. Extensive magma chamber-based hydrothermal activity existed around the Timber Mountain caldera complex continuously between ca. 13 and ca. 9 Ma (Weiss *et al.*, 1994). During the Timber Mountain Caldera hydrothermal event between 11.5 and 10.0 Ma, a large south flowing hydrothermal plume existed just beneath Yucca Mountain (Bish and Aronson, 1993). As was stated above in Section 2.6.2, the temperatures at the ESF level during that time did not exceed 35–37 °C, and could only be lower afterwards (Szymanski *et al.*, 2000). So, the model proposed by the USGS researchers and accepted by the UNLV researchers is not supported by what is known regarding the cooling of shallow intrusive bodies, in general, or by the geologic record of the thermal history of Yucca Mountain.

In connection with their description of the model for the formation of secondary minerals, Wilson and others state: «*The NP [North portal] recorded localized elevated temperatures that were not recorded in the underlying tuffs units.*» (p. 25). This statement merely summarizes their results of the fluid inclusions studies and does not explain why the highest temperatures (up to 75 °C by Wilson and others and up to 95 °C by Whelan *et al.*, 2001) were present at a depth of only 30 to 50 m from the land surface and become cooler with increasing depth (see Figure 12). We believe that it would be extremely difficult to explain such a distribution of temperatures by conductive heating of the rock mass by a distant magma chamber. Neither Wilson and others nor the USGS researchers seem to have a reasonable answer to this question, so further discussion of the phenomenon is carefully avoided.

Furthermore, the east-west temperature gradient established by temperatures obtained from the fluid inclusions (see Figure 14 for example) was not explained by the magma-chamber heating model. Such a gradient does not seem to have a rational explanation if the presumed heat source is located some 8–10 km to the north of the ESF (Marshall and Whelan, 2000). One known hydrothermal system related to the Timber Mountain magma chamber did produce, as expected, the north-south geothermal gradient under Yucca Mountain (Bish and Aronson, 1993).

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<sup>8</sup> In addition to sloppy phrasing («intrusion of the mountain») the Wilson and others take undue freedom with numbers: the original publication reads: «...a gradual cooling of the rocks over millions of years, in agreement with thermal modeling of magma beneath the 12-Ma Timber Mountain caldera just north of Yucca Mountain.» (Marshall and Whelan, 2000; emphasis added).

**Summary.** No meaningful model, which explains the origin of the elevated temperatures or their distribution within the repository block has been presented by Wilson with co-authors.

### 3. SUMMARY ON THE PART I AND PART II OF THE UNLV REPORT

The report provides a summary of a large volume of meticulously collected fluid inclusion data along with a lesser amount of other data (petrography, stable isotopes, electron microprobe elemental analysis, cathodoluminescence). The report, however, does not represent a standalone document. Due to the substantial generalizations presented, it can only be evaluated along with the accompanying technical (non-QA) data posted at the UCCSN WebPages.

The report contains quite a number of minor lapses and inaccuracies associated with the presentation of the data (e.g., the data presented in the main report do not match those presented in the technical data files; erroneous entries were found in the Table 1 of the Part I (C and O isotope data), etc.). Self-contradictory statements are common (e.g. statements such as: «*Liquid-only inclusions comprise the only inclusion assemblages in bladed calcite...*» and «*A small number of 2-phase FIAs were identified in the basal part of the bladed calcite...*» which appear in Part II on the same page, page 12). It appears to us that the report has undergone, before submittal, neither technical nor editorial review.

The discussions and interpretation of the results, as they appear in the report, are not satisfactory. In many instances meaningful discussions are either absent altogether or replaced by debatable assertions (examples of which are abundant throughout this review). In most, if not all instances, clearly plausible and straightforward alternatives to the proposed mechanisms are not considered. A number of important questions have not been addressed at all (see Sections 1.3 and 2.7 of this review).

Out of the three conclusions formulated in Part I, only one deals with the inferred origin of the secondary minerals from the Yucca Mountain vadose zone. The conclusion states that textures and features of these minerals are not consistent with saturation of the site with water and formation of minerals in a phreatic environment. Our analysis shows (see Section 1.1 of this review) that the conclusion is not warranted. Factual evidence presented in Part I is susceptible to radically different interpretation. Additional data and discussion presented in Section 1.3 of this review reinforce this opinion.

The conclusion of Part II, stating that the results of the UNLV study are not consistent with the formation of secondary minerals in a saturated environment and with the former presence of hydrothermal upwelling fluids appears to be equally hollow and unsubstantiated by factual evidence.

No meaningful model, which might serve as an alternative to the hydrothermal upwelling model, was formulated in the report. The presentation that appears in Section 6.5. «Genetic Model for the Formation of Secondary Minerals», Part II, is so general and incoherent that it is not amenable to serious evaluation. Some fragments of the model appear to violate principles of physics (e.g., moving water films rising up the flat faces of crystals for 2–3 cm «by surface tension»). The model, as presented, does not seem to be capable of rationally explaining most features of the mineral forming system at Yucca Mountain, inferred from mineralogic, geochemical, and fluid inclusion studies (e.g., complex mineralogy of secondary deposits, euhedral morphology of minerals, strong east-west thermal gradient, high salinity of mineral forming fluids, chemistry of gases trapped in inclusions, etc.).

Unless: (a) unwarranted or otherwise problematic statements and conclusions present in the UNLV report are revised; (b) alternative interpretations and models are discussed and demonstrated to be wrong or irrelevant, and (c) a number of critical but omitted issues are addressed and satisfactory explained by the UNLV genetic model, we cannot accept the UNLV genetic model presented as a viable alternative to the hydrothermal upwelling model.

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**Reviewed by:**

*Dr. Yuri V. Dublyansky and Dr. Sergey Z. Smirnov*

*Дублянский Юрий Викторович, Смирнов Сергей Захарович*

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