Fluid Inclusion Studies of Samples from the Exploratory Study Facility, Yucca Mountain, Nevada

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Main Findings and Recommendations

This report analyzes mineral samples of calcite collected from Yucca Mountain in June 1998 by the author. Calcite (calcium carbonate) is a mineral that often forms veins and incrustations in rock fractures. It is practically always formed by precipitation from water. Calcite can be formed in geologic media by percolation of water from the surface or by upwelling of water from below.
Examination of calcite samples from the Yucca Mountain subsurface discussed in this report leads to two principal conclusions:

- the studied calcite was formed by upwelling of water and not from percolation of surface water; and
- the water that entered the Yucca Mountain repository area in the past from below was at elevated temperatures.

The main evidence for these findings is as follows:

1. Water was found trapped in tiny cavities in the calcite samples. These trapped water bodies are called fluid inclusions. Many fluid inclusions had vapor bubbles formed in them, indicating that the water had shrunk after it became trapped. The shrinkage of water evidences that the water has cooled from its original temperature. This is evidence of the presence of water at elevated temperature in the repository zone in the geologic past that could not have come from surface sources.
2. A few samples showed the presence of hydrocarbons. These are all-gas inclusions in calcite, in which traces of aromatic hydrocarbons were found. Aromatic hydrocarbons are heavy molecules that could not have originated in surface sources. There is evidence of hydrocarbons in the geologic media beneath Yucca Mountain area. Hence, the trapped hydrocarbons provide supplementary, though at present, fragmentary additional evidence of upwelling of water into the repository horizon.
3. Veins and crusts at Yucca Mountain besides calcites contain other minerals such as opal, quartz, and minor fluorite. These minerals are typically precipitate from warm or hot water. In particular, it is extremely rare for quartz and fluorite to be formed from surface water percolation. Hence, the presence of these minerals is strong evidence of past presence of upwelling warm water in the Yucca Mountain area.
4. Minerals formed in unsaturated zone, that is, above the water table, are typically deposited in laminated formations consisting of millions of tiny crystals. For example, stalactites in caves are

created in this way. By contrast, large individual perfectly shaped crystals require a saturated environment to form. The calcite at Yucca Mountain often forms perfectly shaped individual crystals, clearly indicating that the area was, at some time in the past, saturated.

5. The study also addresses the question of the age of the calcites. This is because the only way to estimate the future performance of any site as a geologic repository is to study its past. The timing of the formation of the calcites is important because it provides evidence of when the area was saturated and hence of the probability of its becoming saturated in the future during the period relevant to repository performance.

Peak radiation does from Yucca Mountain are expected to occur in the period between 100,000 years and one million years from now. Saturation in the recent geologic past would have serous negative repercussions for the suitability of Yucca Mountain as a repository. This is because saturation of the Yucca Mountain repository after burial of highly radioactive waste may cause the waste canisters to corrode far more rapidly than if the area remained dry. On the other hand, if water entered Yucca Mountain many millions of years ago, and not since then, this specific issue would be of far less consequence. The findings of the research for the timing of past repository saturation are only tentative and indicative. There are indications that the calcite may have been formed in the recent geologic past (less than one million years). This is a very complex and difficult area of work and considerable further research is needed to clarify this crucial question.

Recommendations

Much more data need to be acquired and analyzed in order to assess the implications of the new findings on the repository suitability. Specifically, three questions need to be addressed:

- What is the age and what was the recurrence period of water upwelling?
- What was the volume of fluids involved at different stages of this activity?
- What was the spatial structure of ancient hydrothermal system?

This may be accomplished through concerted effort of researchers, involving:

- Detailed fluid inclusion studies in calcite and other minerals from Yucca Mountain. Such study may provide important information on the spatial structure of the ancient hydrothermal system;
- Careful dating of calcite samples hosting fluid inclusions indicating elevated entrapment temperatures. Such study would constrain timing of ancient hydrothermal system; and
- Detailed isotopic study of mineral phases may provide important information on the origin of fluids and pattern of fluid migration.

Introduction

This report summarizes results of research on fluid inclusions from the calcite samples gathered in the Exploratory Study Facility (ESF), Yucca Mountain, Nevada in June, 1998.

The purpose of this research was to get insight into the paleo hydrology of Yucca Mountain and specifically address one important question: was the currently unsaturated zone [1] of the mountain unsaturated during the formation of these calcites, or did the deposition occur in a saturated environment?

According to the presently accepted concept by the Yucca Mountain Project, the unsaturated zone at Yucca Mountain was formed 9-10 million years ago and since that time the water table has never risen more than 85-100 m above its present level (e.g., Marshall et al., 1993), which is 300 m below the repository horizon. This would mean that the water table never reached zone where the high-level nuclear waste repository is planned to be constructed. The concept of the Yucca Mountain repository relies on the unsaturated environment as a major barrier that will prevent migration of radionuclides from repository into the accessible environment.

According to current regulations, performance of the repository must be ensured for at least 10,000 years in the future (10CFR60). Moreover, peak doses are expected to occur after 100,000 years or more. Therefore, the viability of the site critically depends on whether or not the hypothesis on the long-term stability of the unsaturated zone is correct.

Calcite-opal veinlets found in drill cores from unsaturated zone on the early stages of Yucca Mountain characterization indicated that water with chemistry “alien” to silicate bedrock tuff moved through the mountain in geological past. In 1995-1997, when a 7.8 km long tunnel (Exploratory Study Facility or ESF) was excavated into Yucca Mountain, many more occurrences of secondary minerals become available for study.

The origin of these secondary minerals, or, strictly speaking, the origin of the waters that deposited them, is of great importance, because it may provide information regarding the long-term stability of the unsaturated zone.

From the onset of the studies on Yucca Mountain calcite and opal, they were presumed to be formed in unsaturated zone from gravitation-driven water films that percolated down along open interconnected fractures (Szabo and Kyser, 1985; 1990; Whelan and Stuckless, 1992; Vaniman and Whelan, 1994, etc.). Calcite and opal were extensively studied in terms of their stable (carbon, oxygen) and radiogenic (strontium, uranium, thorium, lead) isotope compositions, and isotopic ages (U-series disequilibrium, 14C, and U/Pb methods).

The fluid inclusion method, probably the only method capable of unequivocal determination of mineral origin, has never been adequately applied in these studies. To date, the U.S. DOE has published only 7 temperatures measured by the fluid inclusion method (DOE, 1993). The DOE has published no data since the ESF was actually constructed.

These (elevated) temperatures were either attributed to calcite of old, 8-10 million years age, or simply dismissed (Roedder et al., 1994). Recent work by the DOE contractors on calcite samples removed from ESF have failed to discover fluid inclusions suitable for determination of paleo temperatures. The research concluded that calcite in the ESF was formed from low-temperature waters in unsaturated environment (Roedder and Whelan, 1998).

In 1995 I had an opportunity to collect and study samples from the first 200 m of the ESF tunnel excavated by that time. [2] I was able to make 82 measurements of paleo temperatures from 6 samples. Obtained temperatures along with auxiliary data on calcite textures clearly indicated that calcite in question was formed in saturated environment from aqueous fluids with slightly elevated temperatures (Dublyansky and Reutsky, 1995; Dublyansky et al., 1996a,b; Dublyansky, 1998a).

My fluid inclusion data have been evaluated by the U.S. Nuclear Waste Technical Review Board (NWTRB). In the course of this evaluation, I spent one week at Virginia Technical Institute and State University, carrying out a verification study with the Board’s consultant, Dr. Robert Bodnar. [3] In its follow-up letter to the Board, Dr. Bodnar wrote:

> The most important result of the work conducted in the Fluid Research Laboratory during the week of June 15-19, 1998, is that the high temperatures reported earlier by Dublyansky were confirmed to be real and not an artifact of sample preparation or data collection. There is little doubt that the calcite in sample SS#85-86 either formed at or was later exposed to aqueous fluids with temperatures of at least 72o C. The important question, then, that must be answered is “What is the age of the calcite being studied? [4]

In June 1998 I collected more samples from the entire extent of the 8.7 km-long ESF tunnel, and in October 1998 I performed a study of the fluid inclusions in them. This report discusses the results of my study.

Altogether I obtained about 300 measurements of fluid inclusion temperatures. Along with other features of the studied samples, they represent compelling evidence indicating that during the deposition of calcite, a saturated environment existed within Yucca Mountain at the level of planned repository.

This issue has direct and significant bearing on the suitability of the site as a potential host for the high-level nuclear waste repository. The critical questions remaining to be resolved are:

- When did it happen?
- Did it happen as one-stage process, or water was upwelling and receding intermittently?
- If the upwelling occurred in pulses, what was the recurrence period of these pulses and what was the duration of each pulse?
- How much water was involved?
- What was the spatial distribution of this upwelling?
- What was the cause of the upwelling?

Only when all these questions have been satisfactorily answered can we address the ultimate question:

- *Could it happen in the future, on time scales comparable to those during which radiation doses could be significant?*

Without theses answers, any assessment of the site viability or suitability will necessarily be incomplete.

Notes:

1. Unsaturated, or vadose, zone extends from water table upwards to land surface, as opposed to saturated, or phreatic, zone extending from water table downwards. ? Return
2. The study of the 12 samples collected in 1995 was carried out in the Institute of Mineralogy and Petrography in Novosibirsk, Russia. The results are briefly summarized in this report. ? Return
3. Report on this study is attached as Appendix 1. ? Return
4. Letter of July 8, 1998, from Robert J. Bodnar to Dr. Leon Reiter of the Nuclear Waste Technical Review Board. Available at the official NWTRB web site ? Return