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Reply to "Commentary: Assessment of past infiltration fluxes through Yucca Mountain on the basis of the secondary mineral record – is it a viable methodology?", by Y.V. Dublyansky and S.Z. Smirnov

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Abstract. Xu et al. (2003) presented results of a reaction-transport model for calcite deposition in the unsaturated zone at Yucca Mountain, and compared the model results to measured abundances in core from a surface-based borehole. Marshall et al. (2003) used the calcite distribution in the Topopah Spring Tuff to estimate past seepage into lithophysal cavities as an analog for seepage into the potential repository waste emplacement drifts at Yucca Mountain in southern Nevada (USA). Dublyansky and Smirnov (2005) wrote a commentary paper to Marshall et al. (2003) and Xu et al. (2003), containing two points: (1) questionable phenomenological model for the secondary mineral deposits and (2) inappropriate thermal boundary conditions. In this reply we address primarily the modeling approach by showing results of a sensitivity simulation regarding the effect of an elevated temperature history that approximates the temperature history inferred from fluid inclusions by Wilson et al. (2003). Modeled calcite abundances using the time-varying temperature history are similar to the results for the steady-state ambient temperature profile (Xu et al., 2003), and are still consistent with the measured abundances at the proposed repository horizon.

Key words: Yucca Mountain, calcite precipitation, thermal boundary conditions, infiltration rate, reactive transport modeling.

1. Introduction

Dublyansky and Smirnov (2005) comments on Marshall et al. (2003) and Xu et al. (2003) contain two major points: (1) a questionable origin of the secondary calcite and opal deposits found within cavities and on fracture surfaces at Yucca Mountain and (2) inappropriate thermal boundary conditions for simulations used by Xu et al. (2003). In Marshall et al. (2003) the calcite distribution in the Topopah Spring Tuff was used to estimate past seepage into cavities as an analog for seepage into the potential repository waste emplacement drifts. The latter paper estimated the water flux required to form calcite and opal using a batch geochemical model calculation that assumed equilibrium of percolating water with calcite and silica gel. Xu et al. (2003) presented results of a dual-continuum (fracture-matrix) reaction-transport model for calcite precipitation in the unsaturated zone at Yucca Mountain, and compared the results to measured abundances from cores taken from a deep surface-based borehole (WT-24). This model considered local equilibrium between calcite, water, and CO_2 in the gas phase.

2. Discussion

The origin of calcite and silica deposits in open fractures and lithophysal cavities at Yucca Mountain (from meteoric water descending along fractures or from upwelling hydrothermal fluids) has been discussed in numerous earlier papers, including Whelan et al. (2002, 2004). Whelan et al. (2004) reaffirmed that secondary minerals in open fractures and lithophysal cavities at Yucca Mountain formed primarily by infiltrating meteoric water. This point has also been discussed briefly in the reply of Marshall et al. (2005).

Here, we address primarily the issue of inappropriate thermal boundary conditions as pertains to the Xu et al. (2003) paper. The reactive transport model presented in Xu et al. (2003) considered several important factors controlling calcite precipitation: (1) infiltration rate, (2) geothermal gradient, (3) gaseous CO_2 diffusive transport and partitioning in liquid and gas phases, (4) fracture-matrix interaction for water flow and chemical constituents, and (5) water-rock interaction. At the time these models were developed, no detailed temperature history of the unsaturated zone was available, and abundant evidence indicated that the majority of the calcite in the unsaturated zone precipitated from downward flowing water of meteoric origin (Paces et al., 2001; Whelan et al., 2002). Therefore, simulations were performed using the present day geothermal gradient and a range of infiltration rates.

More recent fluid inclusion data (Wilson et al., 2003) showed that calcite precipitated under elevated temperatures up to about 5 million years ago, with ambient temperatures and fluid compositions similar to the present day for at least the past 2-3 million years. Temperatures in the Topopah Spring Tuff at the level of the Exploratory Studies Facility (ESF) prior to approximately 6 Ma may have been around 45-60°C, with a maximum value of 83°C. Even with these elevated temperatures, the conclusion of the latter authors is that calcite formed predominantly by precipitation from downward infiltrating meteoric fluids, not hydrothermal circulation. As discussed in Whelan et al. (2004), abundant evidence (morphological and isotopic) presented over several years by numerous researchers at various institutions has clearly demonstrated that calcite in the unsaturated zone at Yucca Mountain was formed primarily by infiltrating meteoric water. In addition to direct evidence from calcite compositions, evidence for a downward flux of Ca and Sr in the unsaturated zone over millions of years at an average infiltration rate of about 5 mm/yr was gained from studies of the compositions of zeolitic rocks in the Calico Hills unit that underlies the Topopah Spring tuff (Vaniman et al. 2001). These results were also consistent with infiltration rates used to model Cl and Sr concentrations in unsaturated zone pore and perched waters reflecting input over only the past 10-20 Ka (Sonnenthal and Bodvarsson, 1999).

We appreciate the comments by Dublyansky and Smirnov (this issue) as an opportunity to test the effect of elevated temperature on calcite precipitation in the unsaturated zone, which has recently been adequately quantified by Wilson et al. (2003). Some of the other points they make may require further evaluation, such as the anomalous temperatures for some fluid inclusions at shallow depths, and the unexpectedly high estimated salinities. Yet, fumarolic activity early in the history of the cooling Topopah Spring tuff, as well as deposition of later tuffs are potential explanations for some of the early-formed high-temperature calcite. Calculated salinities are not derived from directly measured concentrations, and may reflect effects of other chemical components. While some early-formed calcite may show some variable temperatures and compositions, it is still clear that relatively compositions and low temperatures of formation are the rule for the past 2-3 Ma (Wilson et al., 2003).

Based on the recent temperature history inferred from fluid inclusions by Wilson et al. (2003), enough data are now available to better evaluate the effect of changing thermal conditions on calcite precipitation in the unsaturated zone at Yucca Mountain. The following sensitivity study considers a hypothetical change in the thermal gradient over time on calcite precipitation in the unsaturated zone. As mentioned in our original paper, a number of uncertainties and approximations are involved in the numerical simulations, that do not necessarily change the underlying conceptual model for calcite precipitation but can change the conditions and parameters for modeling calcite abundances. For example, time variations in infiltration, CO_2 partial pressure, cation exchange of Ca with Na in zeolites, and other water-rock reactions can affect calcite solubility and abundances.

3. Coupled Thermal-Hydrological-Chemical Model of Calcite Precipitation

A variable temperature lower boundary at the water table was developed (using successive fixed volumes of the lower boundary grid block) such that a rough correspondence to measured temperatures in the Topopah Spring Tuff resulted. The temperature was set initially at 95°C at the base of the 1-D model domain (with the borehole WT-24 stratigraphy) at 10 Ma and allowed to decrease through conductive and advective cooling into the overlying rock and with the atmosphere. An infiltration rate of 5.92 mm/yr and the base-case geochemical system (primarily calcite and silica polymorphs) was used (see Xu et al, 2003). The simulation was carried out using the non-isothermal reactive geochemical transport code TOUGHREACT (Xu et al., 2004).

4. Results

The temperature distribution over the depth of the borehole WT-24 column at different times is presented in Figure 2. Initially, the temperature at the approximate depth where many of the calcite samples were collected in the ESF was about 60°C, similar to the higher temperatures of measured fluid inclusions. Fluid fluxes under these elevated temperatures are still dominated by the infiltration flux, although there is somewhat greater evaporation. Because temperatures are below boiling, and the system is unsaturated, the decreases in density owing to fluid heating does not result in upward fluid flow. Thus hydrothermal circulation is possible only under saturated heated conditions or under boiling conditions in the unsaturated zone where strong vapor flow, condensation, drainage, and further boiling take place (see Spycher et al. 2003).

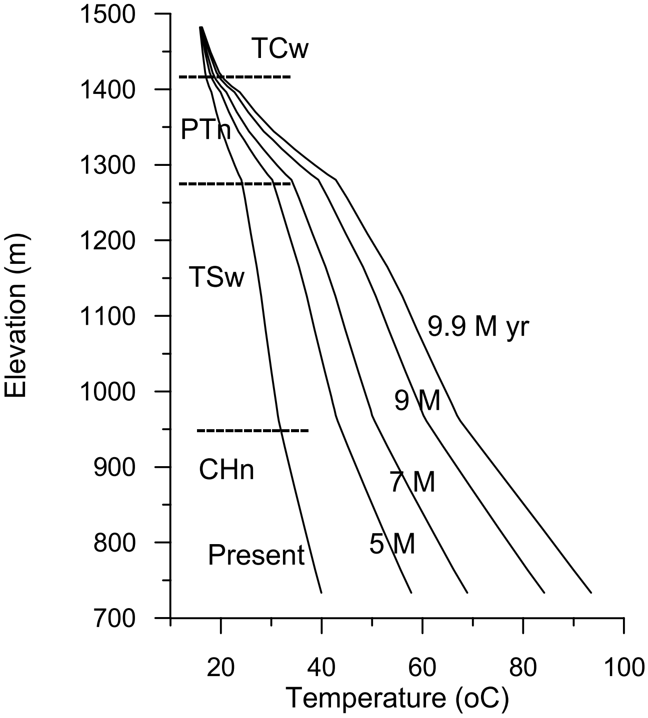

Figure 1. Modeled 1-D temperature profiles (from the surface to the water table for the WT-24 borehole) as a function of time for a ten million year period. Hydrostratigraphic units: TCw = Tiva canyon welded tuff. PTn = Paintbrush nonwelded tuffs. TSw = Topopah Spring welded tuffs. CHn = Calico Hills nonwelded tuffs.

Simulated total calcite abundances for the variable thermal conditions, compared to the Xu et al. (2003) simulated abundances and the measured values (Fabryka-Martin, 2000), are shown in Figure 2. Somewhat greater abundances result from the higher temperature gradients, yet the overall pattern and magnitude is similar to that for the steady-state ambient temperature distribution. Higher temperatures result in lower solubilities and somewhat higher calcite abundances, yet the abundances are dominated by the Ca flux into the unsaturated zone. The variable temperature simulation equally captures the U.S. Geological Survey measured data for the TCw, TSw and CHn hydrostatigraphic units, and equally overestimates abundances in the PTn. Such deviations in the PTn may result from water-rock reaction and cation exchange in the glass-rich and sometimes zeolitized bedded tuff that make up this unit, and not treated in these simulations.

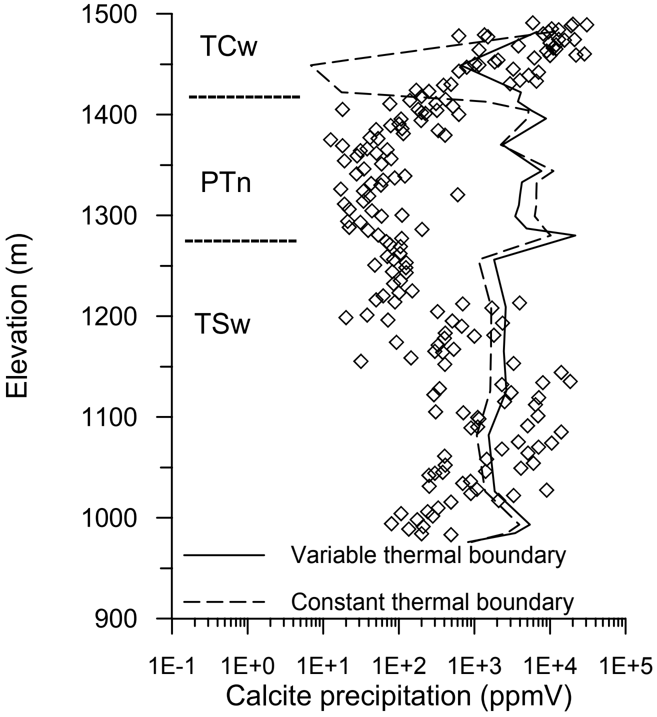

Figure 2. Total (fracture plus matrix) calcite abundances (volume fraction) obtained with two types of thermal conditions applied at the bottom boundary (WT-24 column, after 10 million years). Diamonds represent bulk rock calcite abundances measured by the U.S. Geological Survey (Fabryka-Martin, 2000).

5. Conclusions

Dublyansky and Smirnov (2005) comments on Marshall et al. (2003) and Xu et al. (2003), contain two major points: (1) a questionable phenomenological model for the secondary mineral deposits and (2) use of inappropriate thermal boundary conditions. The first issue regarding the mode of origin of calcite and opal in the unsaturated zone has been addressed by several other authors, and additional supporting geochemical evidence leads to the well-based conclusion that much of the calcite, and certainly all of the calcite formed within the past few Ma, is of meteoric origin and not derived from hydrothermal circulation of upwelling fluids.

Regarding the second point, we acknowledge that a better treatment of the thermal evolution of the unsaturated zone could have been made, but well-constrained data were not available at the time the original work was performed. We have addressed this point with a variable temperature simulation approximating the temperatures measured in fluid inclusions. As in the Xu et al. (2003) paper, the reaction-transport model for calcite precipitation at Yucca Mountain unsaturated zone considers (1) infiltration, (2) the geothermal gradient, (3) gaseous CO_2 diffusive transport and partitioning in liquid and gas phases, (4) fracture-matrix interaction for water flow and chemical constituents, and (5) water-rock interaction. The results of this simulation show somewhat greater abundances of calcite compared to the ambient temperature simulation, yet show similar trends with depth and do not change the conclusions of Xu et al (2003).

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