



Insights into the origin of secondary calcite at Yucca Mountain (Nevada) from high-resolution stable isotope data coupled with fluid-inclusion microthermometry and δD analyses

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The Miocene rhyolitic tuffs comprising the thick vadose zone of Yucca Mountain host abundant fracture-filling calcite, which has previously been studied to reconstruct the paleohydrogeology at the proposed high-level nuclear waste disposal site. The origin of calcite has been explained by deposition from infiltrating meteoric water (Whelan et al., 2002). Early isotopic studies of this calcite found that its $\delta^{18}O$ values increase with depth below surface, which was interpreted as a reflection of the “normal” geothermal gradient (Szabo and Kyser, 1990). Subsequently, $\delta^{18}O$ was found to increase from early to late stages of calcite growth; this was interpreted as indication of a temperature decrease. $\delta^{13}C$ values showed a concomitant decrease attributed to the evolution of carbon sources from methanogenic bacteria to plants (with a gradually increasing contribution from C4-plants; Whelan et al., 2002).

In this study we performed, for the first time, high-resolution (80 μm) isotopic profiling across two calcite crusts. The two samples turned out to be isotopically heterogeneous, with two and three growth episodes, respectively. Based on fluid-inclusion microthermometry the early calcite generation in both samples was deposited from thermal waters (38 to 48°C). The first sample shows an isotopic pattern in which $\delta^{18}O$ values uniformly increase and $\delta^{13}C$ values decrease with time resulting in a strong negative correlation in both growth zones. In the second sample, O and C isotope values show little variation in the early-stage calcite, followed by several maxima and minima (amplitude 0.5 to 1.5 ‰) in the intermediate-stage calcite, and by a uniform decrease in the late-stage calcite. The $\delta^{18}O$ and $\delta^{13}C$ show positive correlation in two later growth episodes. The δD values of inclusion water in early and intermediate calcite are similar, (-100 to -97 ‰), characteristic of meteoric precipitation in this region younger than ca. 780 ka (Winograd et al., 1985). The $\delta^{18}O$ values of mineral-forming water calculated from the $\delta^{18}O$ values of calcite are -6 to -8 ‰ indicating a significant shift to the right of the Meteoric Water Line. The latter feature strongly suggests the involvement of the deep-seated fluids. The new results suggest that (a) the meteoric infiltration model of calcite is most likely incorrect, and (2) the processes of fluid flow and calcite deposition at Yucca Mountain were more complex than previously thought.

Szabo and Kyser (1990) Geol. Soc. Amer. Bull., 102(12). Whelan et al. (2002) Appl. Geochem., 17. Winograd et al. (1985) Science, 227.