NON-LINEAR PALEOHYDROGEOLOGY AT A PROSPECTIVE HIGH-LEVEL NUCLEAR WASTE DISPOSAL SITE: INTEGRATION OF THE FLUID INCLUSIONS AND THE STABLE ISOTOPE DATA FROM YUCCA MOUNTAIN, NEVADA, USA

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The proposed permanent geological high-level nuclear waste repository would be located within an 800 to 1000 m-thick vadose zone of Yucca Mountain in southern Nevada. The mountain is build up of the Miocene (12.7 Ma) rhyolite as-flow tuffs. The potential repository block is intersected by a C-shaped 7.8 km-long Exploratory Studies Facility tunnel (ESF) as well as a 2.8 km-long East-West Cross-Drift (EWCD), which exposes abundant hydrogenic secondary mineralization in lithophysal cavities and fractures. The secondary mineral paragenesis comprises major calcite, silica miners (quartz, chalcedony) and opals, plus accessory fluorite, zeolites (heulandite), strontianite, and barite. Fluid inclusions and stable isotope studies of these minerals have been carried out by a number of research groups including: Institute of Mineralogy and Petrography SB RAS (IMP; representing State of Nevada), University of Nevada, Las Vegas (UNLV), U.S. Geological Survey (USGS), and Los Alamos National Laboratory (LANL). The ultimate aim of the studies was to gain information on the paleohydrogeology of the site. This paper presents our interpretation of the paleohydrogeological implications of the integrated fluid inclusion-stable isotope dataset that is currently available.

Stable isotope data

Calcite shows prominent unidirectional shift of the $\delta^{13}C$ values from paragenetically early varieties (+8 to +9 ‰) to the latest ones (–8 to –9 ‰ PDB). The $\delta^{13}C$ > ca. +4 ‰ require deposition of calcite from a fluid in which dissolved carbon species (CO_2 and CH_4) have equilibrated in a strongly reducing anoxic environment (log fO_2 <-35). No independent evidence exists regarding the presence of such environment in the vadose zone at Yucca Mountain in geologic past. Importantly, calcite with characteristic "heavy positive" carbon signatures was found in the ESF throughout the repository block.

The δD values of water from fluid inclusions (Wilson et al., 2002) range from –59 to –131 ‰ SMOW as compared to –96 to –110 ‰, characteristic of the modern Yucca Mountain waters. Combined with the $\delta^{18}O$ values of paleowaters (calculated from $\delta^{18}O$ of calcite and homogenization temperatures, T_h's) the data reveal prominent ^{18}O-shift to the right of the "meteoric water line". Such shift is characteristic of the hydrothermal waters. The resulting δD vs. $\delta^{18}O$ data overlap the fields for modern thermal springs and for mineral forming fluids from hydrothermal deposits of Nevada and California.

Chemistry of gases trapped in inclusions

Major CH_4 and subordinate CO_2 contents were determined in calcite from Yucca Mountain by gas chromatographic analysis. Levy et al. (1995) reported Quadrupole MS data indicating that calcite from the ESF contains gases, whose ratios are characteristic of the reducing (dominant CH_4, very little O_2) and a phreatic (H_2O = 99.2 to 99.9 mol %) environment. Both O_2 contents and O_2/N_2 ratios are identical to those of the hydrothermal carbonates (Fig. 1) and dissimilar, by as mush as 1 to 2 orders of magnitude, from the vadose zone pedogenic carbonates found in the southwestern United States. On the basis of strong Raman luminescence, Dublyansky (2001) inferred likely presence of aromatic/cyclic hydrocarbons in gas filling the monophase all-gas inclusions.

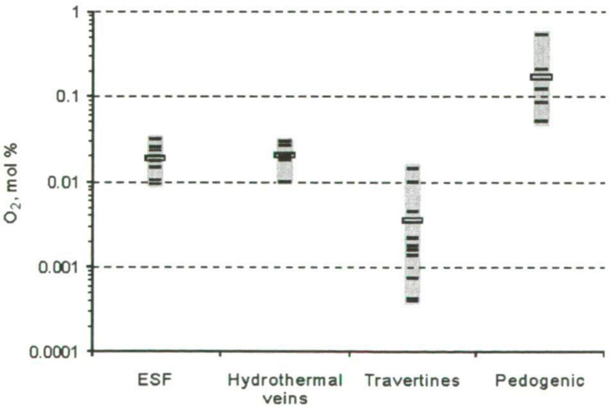

Fig. 1. Contents of O_2 in calcite from Yucca Mountain (ESF) compared with contents in hydrothermal veins, travertines and pedogenic calcites from New Mexico. Data sources: LANL (ESF) and Newman et al. (1996).

Combined fluid inclusions – stable isotope data

Fluid inclusions were studied in calcite, fluorite and quartz. T_h's tend to decrease, in mineral crusts, from early to late members of the paragenesis from 45-90 to <35-50°C (all-liquid inclusions). The two-phase fluid inclusions become scarcer with relative time (i.e., further from the base of crusts), so that all-liquid inclusions are the dominant type of inclusions present in the outer layers. In one sample, two-phase fluid inclusion assemblages (FIAs) have been found in the outermost blocky calcite. Two-phase FIAs were found in calcite with $\delta^{13}C$ ranging from +9 to –6 ‰ PDB.

Calcite yields consistent thermometric information (commonly, 20 to 30 inclusions in a FIA homogenized within a 4-5°C-interval). Inclusions in quartz and fluorite, although scarce, yield T_h's similar to those measured in adjacent calcite.

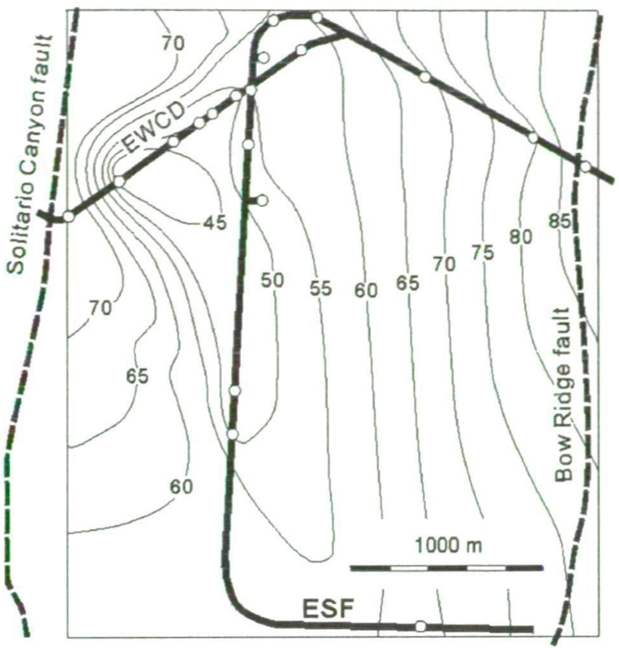

Fig. 2. Reconstructed maximum paleotemperature field in the repository block at the level of the ESF (by max. modal T_h's). Sample locations are shown as white dots. Each sample is characterized by tens to hundreds of the T_h's measurements. Data sources: Dublyansky et al. (2001), Wilson et al. (2002), and Whelan et al. (2001). Graphic interpolation by Mathcad 6 PLUS.

Structure of the paleotemperature field

The paleotemperature field, as represented by the maximum modal T_h's measured in samples from the ESF and EWCD, is non-uniform. It shows negative correlation with the geothermal gradient: the highest T_h's were measured in calcite from the vicinity of the Bow Ridge fault, at a depth of only 25-50 m. The temperatures become cooler as the ESF goes deeper (45-50°C at a depth of ca. 300 m). Further, the maximum modal T_h's exhibit conspicuous east-west gradient (Fig. 2). These temperature/depth relationships translate into paleo heat flows as great as 3.5 to 62 HFU; roughly 3 to 50 times today's average in the western United States.

Salinities of fluids

Temperatures of ice melting measured in calcite inclusions revealed salinities ranging from near 0 to 27,400 ppm NaCl-equiv. The waters, thus, fall in three hydrogeological categories: fresh (<1,000 ppm), brackish (1,000 to 10,000 ppm) and saline (10,000 to 100,000 ppm) waters. The measured salinities are up to 70 times greater that those of the most salty modern groundwaters at Yucca Mountain (pore waters of the non-welded member of the Paintbrush tuff; ca. 400 ppm NaCl).

Discussion and conclusions

It has been proposed that the paleohydrogeology of the prospective nuclear waste repository site was very stable over the last 10-12 Ma, and was characterized by low-intensity flux of meteoric water percolating gravitationally, as thin water films, through thick (800-1000 m) vadose zone (e.g., Wilson et al. 2002; Whelan et al., 2001). Fluctuations of hydrogeological parameters expected in such a system are restricted to cyclic variations of the infiltration flux caused by the climate change.

Meanwhile, a number of lines of evidence presented above reveal traces of the deep-seated hypogene component in the paleohydrogeology of the site. Carbon isotope data for calcite and chemistry of gases trapped in inclusions indicate that, at least at early stages, mineralization was associated with fluids equilibrated with reducing, strictly anoxic environment. Preliminary stable isotope data on paleowaters (Wilson et al., 2002) show $\delta^{18}O$ values that are too "heavy" for the pristine meteoric waters, which suggests substantial water-rock oxygen exchange. The combined δD-$\delta^{18}O$ values are typical of the hydrothermal-meteoric waters. Elevated T_h's (up to 85-90°C) and the pronounced east-west gradient of the paleothermal field reinforce this interpretation and point to the deep-seated fault zone as a major avenue for movement of thermal fluids.

Neither elevated temperatures, nor reducing atmosphere could have persisted within thick aerated vadose zone of Yucca Mountain for geologically substantial times. We propose, therefore, that the relatively "flat" vadose paleohydrogeology was interrupted by a spike (or spikes) of the short-lived invasion(s) of the deep-seated thermal waters. Such invasion(s) would necessarily be of a transient character. Nevertheless, they would introduce into the vadose zone large amounts of thermal, chemically evolved waters, which cooled down, degassed, mixed with oxidized waters and deposited minerals there.

References

DUBLYANSKY, Y. V. (2001): Environment of deposition suggested by all-gas inclusions and stable isotopes in secondary minerals from Yucca Mountain, Nevada, USA. F. Noronha and A. Guedes (Eds.) XVI ECROFI Faculdade de Ciencias do Porto, Departamento de Geologia, Memoria. **7**, 131-134.

DUBLYANSKY, Y., FORD, D., REUTSKI, V. (2001): Chemical Geology, **173**, 125-149.

LEVY, S., CHIPERA, S., NORMAN, D. (1995): Alteration History Studies in the Exploratory Studies Facility, Yucca Mountain, Nevada, USA. Proc. Materials Res. Soc. Symp. Boston, MA.

NEWMAN, B., NORMAN, D., GUNDIMEDA, N., LEVY, S. S. (1996): Understanding the genesis of nonmarine calcite deposits through quadrupole mass spectrometric analysis of fluid inclusion gases. Chemical Geology, **132**, 205-213.

WHELAN, J., ROEDDER, E., PACES, J. (2001): Evidence for an Unsaturated-Zone Origin of Secondary Minerals in Yucca Mountain, Nevada. Proc. Int. Con., Amer. Nucl. Soc. "High-Level Radioactive Waste Management". CD ROM.

WILSON, N., CLINE, J., AMELIN, Y. (2002): Thermochronological Evolution of Calcite Formation at the Potential Yucca Mountain Repository Site, Nevada: Part 2, Fluid Inclusion Analyses and U-Pb dating. HRC Technical Report TR-02-005.2. 52 pp.