

Reconstructing the fluid evolution in an argillaceous aquitard sequence based on isotope geochemistry of veins

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Argillaceous rocks constitute natural barriers to fluid flow, which makes them potential hosts for radioactive waste disposal or seals for gas storage. However, argillaceous rocks can contain veins as relicts of past deformation in the presence of fluids. Thus, investigating these veins regarding the timing of (de)formation, fluid origin and corresponding transport processes provides important insight into the long-term hydrogeological behavior and stability of the host rocks.

This study examines calcite veins in the Opalinus Clay (Middle Jurassic) of northern Switzerland under the above aspects. Calcite veins were collected from drillcores and analyzed for $\delta^{18}\text{O}$, $\delta^{13}\text{C}$, $^{87}\text{Sr}/^{86}\text{Sr}$ and clumped isotope thermometry (Δ_{47}) in combination with U-Pb geochronology (by LA-ICP-MS).

Owing to consistently low U and high common Pb contents, U-Pb geochronology was largely unsuccessful for veins in the Opalinus Clay. However, in the more calcareous units above and below, a substantial number of reliable U-Pb ages in the range of 202.4 – 3.4 Ma could be obtained. Coupled with Δ_{47} data, an evolution from modified seawater prior to Eocene times towards meteoric waters in the Cenozoic is indicated for units above and below the Opalinus Clay. For the latter, (undated) Δ_{47} data are generally in agreement with such an evolution, however, the process(es) by which the meteoric signals migrated across the unit are not yet fully understood. Based on $\delta^{13}\text{C}$ and $^{87}\text{Sr}/^{86}\text{Sr}$ data, a local source of the mineral-forming fluids is conceivable, with $\delta^{18}\text{O}$ values having evolved by diffusive exchange with waters in the units above and below. However, C and Sr isotope signatures could also attest to an efficient rock-buffering potentially obliterating external signals. In any case, Δ_{47} data show that the veins in the Opalinus Clay precipitated from fluids with less negative $\delta^{18}\text{O}$ values compared to the present-day porewater and/or at higher temperatures [1]. Ultimately, this indicates that veining in the Opalinus Clay occurred before 10 – 5 Ma, i.e., before the strata was uplifted and cooled.

[1] Gimmi, Aschwanden, Waber, Gaucher, Ma & Traber (2023), *Applied Geochemistry*, 105846.