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Coprecipitation of the actinides Th(IV), U(IV) and actinide homologues Y(III), La(III), Ce(III) and Yb(III) in secondary minerals from crystalline fracture systems: A natural analogue study from Forsmark, Simpevarp and Laxemar (Sweden)

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As studies on the retention of radionuclides in natural systems over geological times are not feasible, it is common practice to resort to natural analogues. The coprecipitated actinides and homologues of the equivalent oxidation state in secondary mineral phases are a good approximation to estimate the radionuclide retention potential of such minerals.

Fracture fillings formed within crystalline host rock, as e.g. the calcites studied in this work, preserve the element content of the precipitation solution. The specific element composition of single growth zones thus allows conclusions to be drawn about the evolution of the precipitation solution and the precipitation conditions. Fe for example is sensitive to changes in redox conditions and was found in elevated concentrations in connection with paleo-biofilms [1]. Sr contents on the other hand vary between e.g. meteoric, marine, glacial and hydrothermal fluids, hence are specific to the origin.

To obtain highly spatially resolved information at ppm–mg range, we applied 2D element analysis (μ XRF and LA-ICP-MS) to scan for the trace elements Na, Mn, Fe, Sr, Y, La, Ce, Yb, Pb, Th and U accumulated in low-temperature calcite fracture fillings handpicked from exploration drill cores at Forsmark, Simpevarp and Laxemar (Sweden) [2]. Based on the element maps, we found several growth zones and other microstructures (e.g. point accumulations, microfractures, gradual element enrichment) within the fracture fillings. Particular structures from the Laxemar area (ca. 400–430 core meters) indicate paleo-biofilms along which extremely high trace and rare earth element concentrations were observed. Further line scans with an extended element spectrum were performed to illustrate the high retention potential of such biogenic structures. At one sample from the Simpevarp site (ca. 200 core meters) face depending partitioning within a single growth zone could be visualized. It occurred in three clusters with major enrichment in Mn-Fe-Sr, Y-Yb-U and Na-La-Ce-Pb-Th. Furthermore, partition coefficient DTE based on the element assemblage of the most recent overgrowth and connected formation waters were determined as described in equation 1. These ratios were compared to experimentally generated coefficients [3] and values obtained by medium-term precipitation experiment conducted at the Äspö Hard Rock Laboratory [4].

$$D_{\text{TE}} = \frac{\left(\frac{\text{TE}}{\text{Ca}^{2+}}\right)_{\text{solid}}}{\left(\frac{\text{TE}}{\text{Ca}^{2+}}\right)_{\text{liquid}}}$$

Equation 1

References

- [1] Heim, Lausmaa, Sjövall, Toporski, Dieing, Simon et al. (2012), *Geobiology* 10, 280–297.
- [2] Tullborg, Drake & Sandström (2008), *Applied Geochemistry* 23, 1881–1897.
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- [4] Drake, Mathurin, Zack, Schäfer, Roberts, Whitehouse et al. (2018), *Environmental Science & Technology* 52, 493–502.

Primary author: KUSTURICA, Annemie

Co-authors: Dr VANLAATEN, Neele (Friedrich-Schiller University); Prof. SCHÄFER, Thorsten (Friedrich-Schiller University Jena); Prof. DRAKE, Henrik (Linnaeus University)

Presenter: KUSTURICA, Annemie

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