A new methodology for an improved description of radionuclide retardation in safety assessments

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Conceptual model

In safety assessments usually the K_d concept with temporally constant values is applied to describe the radionuclide retardation in the far field of a repository.

In this study, the existing transport program r³t [1] used for large model areas and very long time scales has been improved. Implementing the smart K_d concept based on surface complexation allows to consider the impact of varying geochemical conditions.

The new methodology describes the sorption of radionuclides as a function of selected, important environmental parameters E_i such as pH, pCO_2, ionic strength, concentration of the cations Ca^{2+} and Al^{3+} and presence of characteristic mineral phases. The Gorleben site as a potential repository site in Germany has been selected as an exemplary case of application for a proof-of-concept.

Most of the individual parameters E_i were not available in r³t so far. Thus the transport of the respective substances as well as equations describing pH and concentrations of ions as a function of accessible mineral phases had to be implemented. Then the reactive transport model r³t can call pre-calculated K_d values for selected sediments for each time-space point. They are stored in a multidimensional matrix depending on the real geochemical conditions. Figure 1 shows the frequency distribution of the pre-calculated smart K_d values for Am^{3+} and UO_2^{2+}.

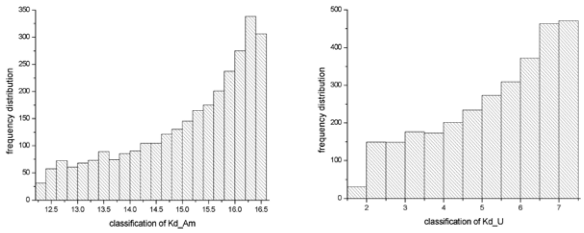

Figure 1: Frequency distribution of the pre-calculated K_d values for Am^{3+} and UO_2^{2+} (in m³/kg).

[1] Fein (2004) *Report GRS-192*.

Combined NanoSIMS and TEM *in situ* analysis of pristine matrix material of ALHA 77307 and Acfer 094

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Only a few structural studies on presolar silicates and oxides exist [1]. Commonly the NanoSIMS mapping technique is used to identify presolar grains in the meteoritic matrix from a polished thin section. To extract these presolar grains for further TEM analysis, a complex and risky lift out procedure by FIB is needed [1], reducing the availability of such grains for structural studies. In contrast, Argon Ion Slicing (ArIS), a recently introduced TEM sample preparation technique in geosciences, yields super large electron transparent areas of up to 40, 000 μm^2 [2]. The availability of extremely large thin foils makes a substantial change in the sequence of procedure steps possible, i.e. preparation of a large thin foil first, followed by the NanoSIMS scan on the previously obtained thin foil (Fig.1). No additional sample preparation for TEM is required once the grains are identified by NanoSIMS.

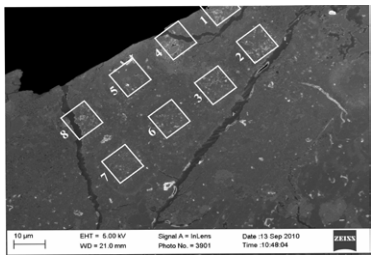

Figure 1: Thin foil section of ALHA 77307 with NanoSIMS scan windows indicated

Pieces of Acfer 094 and ALHA 77307 were ArIS treated until large electron transparent areas were obtained. These TEM thin foils were scanned for oxygen isotope ratios with a Cameca NanoSIMS 50. Based on large O-isotopic anomalies we detected 2 presolar grains (both silicates) out of 8 ion images (10 x 10 μm^2) in ALHA 77307, and 7 presolar grains (6 silicates and one oxide) out of 22 ion images (same size) from Acfer 094.

[1] Vollmer *et al*. (2009) *ApJ* **700**, 774–782. [2] Stojic & Brenker (2010) *Eur. J. Mineral*. **22**, 17–21.