

Actinide pyrochlores: Fabrication and new structural insights

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Besides already established waste forms such as spent fuel or glasses, ceramics are considered as tailor-made potential nuclear waste forms for e.g. the minor actinides (MA = Am, Cm, Np) and Pu. Specifically zirconia based pyrochlores ($A_2Zr_2O_7$) have favourable properties as potential nuclear waste forms with respect to their radiation resistance and their high aqueous durability [1] [2].

To predict the long-term behaviour of these ceramics it is crucial to gain insight into the structural uptake and environment of the actinides. Luminescence spectroscopy of Cm and Eu doped pyrochlore and defect fluorite ceramics identified the actinide and surrogate to adopt the A site of the pyrochlore crystal structure. Moreover the species in the defect fluorite structure are different from the pyrochlore species due to the increase of disorder when the defect fluorite structure is realised.

In order to get closer to a realistic waste form Pu-pyrochlore ceramics with 5 and 10 mol% Pu content were fabricated via a wet chemical synthesis route. The pyrochlore crystal structure was verified by X-ray diffraction and a homogeneous Pu-239 distribution was proven by SEM and EDX. Besides these bulk characterisation techniques XAS was applied to probe the short range order of these Pu-pyrochlores. EXAFS spectra at the Pu L3-edge show the coordination of Pu by eight oxygen which is typical for the A-site of the pyrochlore crystal structure. Complementary spectra of the Zr K-edge were recorded for a complete characterisation of the synthesised ceramics.

[1] Ewing (2011) *C.R. Geosci.* **343**, 219-229. [2] Finkeldei et al. (2014) *Appl. Geochem.* **49**, 31-41.