

Site-selective TRLFS of Eu(III) doped rare earth phosphates for conditioning of radioactive wastes

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Crystalline ceramic materials show promise as potential waste forms for immobilization of high-level radioactive wastes. Rare earth (RE) phosphate ceramics have been found to be extremely stable over geological time scales [1] and they show good tolerance to high radiation doses [2]. These ceramics are able to incorporate radionuclides in well-defined atomic positions within the crystal lattice up to high (~25%) loadings [3], which will reduce the volume of waste in the radionuclide conditioning process. The dehydrated RE phosphates are known to crystallize in two distinct structures, depending on the ionic radius of the cation: the larger lanthanides from La³⁺ to Gd³⁺ crystallize in the nine-fold coordinated monazite structure, while the smaller lanthanides such as Lu³⁺ form eight-fold coordinated xenotime structures.

Structural information on the radionuclide substitution in these RE phosphates can be obtained by site-selective time-resolved laser fluorescence spectroscopy (TRLFS). In our previous work [4] we studied the structural incorporation of Eu(III), taken as an analogue for the long-lived trivalent actinides Pu(III), Am(III) and Cm(III) found in spent nuclear fuel, in the monazite LaPO₄. Eu³⁺ was found to incorporate on the La³⁺ site in LaPO₄ as expected, despite slight discrepancies between the ionic radii of the host and dopant cations.

In the present work we study synthetic Eu(III) doped LaPO₄, GdPO₄, and LuPO₄, as well as mixtures thereof, to investigate the influence of the ionic radius and crystalline structure on Eu(III) substitution in the ceramic material. Results on Eu(III) substitution in the monazite and xenotime materials will be presented and influences of the ionic radii on Eu(III) substitution within the host cation sites will be discussed.

[1] Donald *et al* (1997) *J. Mater. Sci.* **32**, 5851–5887. [2] Luo & Liu (2001) *J. Mater. Res.* **16**, 366–372. [3] Bregiroux *et al* (2007) *Inorg. Chem.* **46**, 10372–10382. [4] Holliday *et al* (2012) *Radiochim. Acta* **100**, 189–195.