

Evaluation of surrogate-models for the incorporation of tetravalent actinides in monazite phases

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Monazite has long been considered as one of the most promising crystalline host materials for long-term storage of radioactive waste, especially actinides. The main reasons for that are its chemical flexibility, its excellent chemical durability and its low recrystallization temperature which allows for rapid repair of radiation induced damage [1,2].

It has been shown that monazites can accommodate large amounts of trivalent actinides as well as various trivalent lanthanides (acting as surrogates for actinides) within its crystal structure [2]. However, the experimental incorporation of tetravalent dopants via a coupled substitution has proven difficult [3-4], though natural monazite is known to contain up to 15 wt% UO₂ and up to 32 wt% ThO₂, respectively [5]. Recently, the substitution of trivalent La³⁺ for Ca²⁺ and Ce⁴⁺, the latter acting as a surrogate for Pu⁴⁺ and U⁴⁺, has been attempted via solid state reaction and co-precipitation.

The evolution of the lattice parameters deviates from that of an ideal mixture (fig. 1) indicating either excess properties or changes in the oxidation state. This will be addressed via various analytical methods. Using XANES and EPR the oxidation state of cerium incorporated into the structure will be determined. The overall chemical composition of the obtained monazite-type phases will be ascertained using EPMA.

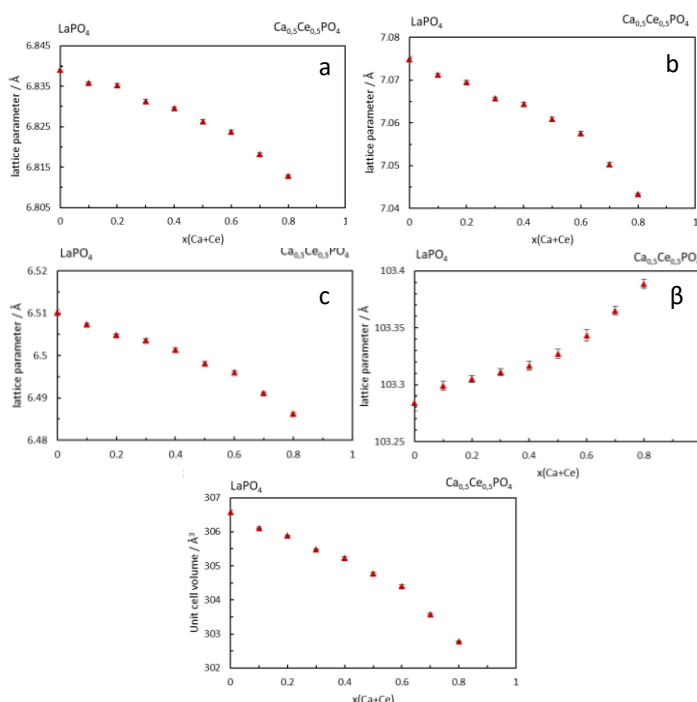


Fig. 1 Evolution of the lattice parameters of the LaPO₄ – Ca_{0.5}Ce_{0.5}PO₄ solid solution

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