MS15-1-19 Evaluation of surrogate-models for the incorporation of tetravalent actinides in monazite phases #MS15-1-19

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Abstract
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 this 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 challenging [3-4], even though natural monazite is known to contain up to 15 wt% UO2 and up to 32 wt% ThO2, respectively [5]. In this context, it has been suggested that the synthesis of single-phase Ca0.5Ce0.5PO4 was impossible [4], given the tendency of Ce4+ to partially reduce to Ce3+ at elevated temperatures. Here, we report on the successful synthesis of the solid solution series LaPO4 - Ca0.5Ce0.5PO4 in a metastable form via a co-precipitation method. This system will be investigated in view of acting as an inactive model system for comprehensive physical and chemical studies of coupled substitutions in monazite, comprising tetravalent metals.
The chemical composition of the samples was determined using EPMA; PXRD measurements show no evidence for a miscibility gap (see images attached; where not visible the error bars are smaller than the symbols used). Further XANES and EPR measurements are scheduled to verify the tetravalent oxidation state of cerium. The properties of the model system will be investigated, as well as their leaching resistance and the radiation stability.
The authors acknowledge support by BMBF under project number 02NUK060B.

References
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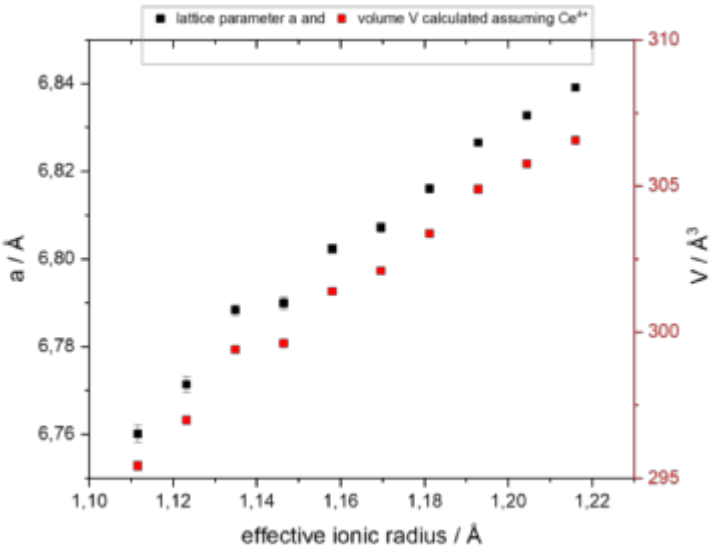
PXRD of LaPO4-Ca0.5Ce0.5PO4 solid solution



EPMA of La0.5Ca0.25Ce0.25PO4