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Preparation of $\text{Ln}_{1-2x}\text{CaxTh}_x\text{PO}_4 \cdot n\text{H}_2\text{O}$ rhabdophane-type as low precursors to monazite-Cheralite ceramics

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Monazite, i.e. LnPO_4 ($\text{Ln} = \text{La-Gd}$), has been widely studied as specific ceramic for the conditioning of trivalent and tetravalent actinides.¹ Owing to its chemical and structural flexibility, the monazite allows a variety of possible substitutions following different mechanisms.² Among them, the so-called cheralite-family, i.e. $\text{Ln}_{1-2x}\text{CaxAnIV}_x\text{PO}_4$. In this frame, the major part of protocols reported in the literature was based on solid-state chemistry routes. However, this method requires the use of repetitive grinding steps and re-heating of actinide bearing powders and usually forms heterogeneous compounds in term of cationic distribution.

In this work, we present an alternative method based on wet chemistry route to prepare thorium-based cheralites. It is based on the initial precipitation of $\text{Ln}_{1-2x}\text{CaxTh}_x\text{PO}_4 \cdot n\text{H}_2\text{O}$ low temperature precursors crystallizing in the rhabdophane type structure (monoclinic, $C2$ space group).⁴ A multiparametric study allowed the determination of the appropriate conditions to form homogeneous and single phase powders. Then these hydrated precursors were converted to monazite-cheralite ceramics through thermal treatment. A coupled in situ PXRD/TGA allowed the identification of the different dehydration steps before the irreversible stabilization of the cheralite anhydrous compounds.⁵ Moreover, since such hydrated phases could also control the release of radionuclides from the ceramics in the field of underground repository, dissolution and solubility experiments have been finally carried out.

References

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