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Partitioning of Ce, as a Simulant for Pu, in a Multiphase Ceramic Nuclear Waste Form

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A multi-phase titanate waste form was synthesized using high temperature sintering in air or Ar with the composition of Ca0.71Y0.088Ce0.52Hf0.22Ti2Ox. The phase assemblage was investigated as a function of the sintering atmosphere. X-ray diffraction measurements indicated that oxidizing sintering atmosphere (air) favors the formation of pyrochlore, whereas neutral environment (Ar) promotes the formation of perovskite, although both sintering atmospheres produced samples containing three phases (pyrochlore, perovskite and rutile under air or zirconolite, perovskite and rutile under Ar). Scanning electron microscope (SEM) analysis revealed that Ce is present in perovskite and pyrochlore (or zirconolite), but not in rutile. Micro-X-ray-Absorption-Near-Edge-Structure (micro-XANES) measurements conducted at the Ce L-edge, in combination with electron energy loss spectroscopy (EELS), showed that Ce4+ prefers to inhabit the pyrochlore (or zirconolite) phase and Ce3+ the perovskite phase. This knowledge is valuable for the further development of titanate waste forms to immobilize Pu.

Summary

Primary author(s) : Dr ZHANG, Zhaoming (ANSTO)

Co-author(s) : Dr VANCE, Eric R. (ANSTO); Dr LUMPKIN, Gregory (ANSTO); Mr DAVIS, Joel (ANSTO); Dr SPIERS, Kathryn (Australian Synchrotron)

Presenter(s) : Dr ZHANG, Zhaoming (ANSTO)

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