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Experimental simulation of phase evolution in conditions of underground storage: from million years to one day

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Long-living radiotoxic isotopes present in spent nuclear fuel (SNF) requires procedures of complete immobilization of these species. Incorporation of the corresponding elements on atomic level into robust host crystalline matrices is one way to secure SNF during long-term underground storage. Derivatives of zirconia, ZrO_2, are promising materials for these applications since these phases are known to remain structurally stable in geological cycles of up to 10^9 years [1]. The candidate host matrix must provide a sufficient solubility limit for radiotoxic elements, which is studied initially. Afterwards, structural stability of these phases against irradiation and leaching are established in order to asses possible discharge of the incorporated radioactive elements over a long-time scale. In this work we studied systematically structural behaviour of ZrO_2-based materials incorporated with Th^{4+} and Ce^{4+} under extreme conditions of temperature (T) and pressure (P) in order to simulate experimentally possible phase evolution in conditions of underground storage.

In situ synchrotron radiation powder diffraction experiments under ambient and extreme conditions were performed at the HZDR ROBL BM20 beamline at ESRF, Grenoble, France [2]. It was found that the cubic YSZ phases could dissolve 20% more of Th atoms compared to their tetragonal analogues. *In situ* T-dependent diffraction studies on radionuclide surrogate tetragonal and cubic Ce-YSZ series in a *RT*-1150 K range revealed excellent phase stabilities. No discharge of guest Ce^{4+} ions was observed. Nevertheless, application of external pressure on tetragonal Ce-YSZ phase induced transition towards a higher cubic symmetry around the $P \sim 8.5$ GPa. Remarkably, occupancy of Ce^{4+} remains stable throughout the transition. This together with T-dependent data indicates excellent affinity of guest Ce atoms with the YSZ structures. Thus, we suggest *in situ* studies under extreme conditions as a part of standard protocol to validate phases of interest as host matrixes for long-term underground storage of SNF.

References

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