

JÉRÉMIE MANAUD

will present his Ph.D. dissertation

Hydrothermal conversion of tetravalent actinides oxalates from the synthesis to the sintering of oxide powders

The defense will take place on Tuesday, November 11th, 2020 at 10.00 am

in the ICSM Auditorium

In the frame of the development of new nuclear reactors (so-called generation III and IV), innovative methods for oxide fuels preparation have been under study for several years. They are frequently based on wet-chemistry routes, involving the precipitation of crystallized precursors. However, heat treatment remains necessary to ensure the conversion of the precursor into oxide, which can cause the presence of residual carbon traces into the final oxide compound. During this thesis work, an alternative preparation route was investigated, which is based on the oxalic precipitation of actinides (U and Th) and/or lanthanides, and then on the hydrothermal conversion of the precursors into hydrated oxides. Conditions of formation of An(IV) hydrated dioxides (i.e. temperature and duration of the hydrothermal treatment and pH of the initial solution) were first determined for thorium, which constitutes a reference system exempt from redox reactions, then for uranium(IV). In parallel, a complete characterization of the obtained solids was undertaken, from a structural, chemical and morphological point of view. In addition, in the case of uranium compounds, XAS experiments highlighted variations in the O/U stoichiometry within the samples, leading to deformations of the crystal lattice. Generally, the hydrothermal conversion of tetravalent actinide oxalates was achieved after heating at 250°C and pH = 5 for 24 hours, and allowed the formation of nearly-anhydrous oxides with residual carbon content lower than 200 ppm. Then, a preliminary study dedicated to sintering revealed the good densification ability of the produced oxides. The sintering map of 3 different uranium oxides presenting different morphologies was then established, and paves the way to the control of the final pellets microstructure. Finally, the hydrothermal conversion of oxalates has been extended to mixed oxides $U_{1-x}Th_xO_2$ and $U_{1-x}Ce_xO_2$, cerium being used herein as a plutonium surrogate.

