LAURA BONATO

will present her Ph.D. dissertation

Sonochemical reactivity of nanocrystalline actinide oxides

The defense will take place on **Friday, November 13th, 2020** at **10.00 am**

in the ICSM Auditorium

Nanocrystalline materials constitute an important topic because of their specific physico-chemical properties related to the nanometric size of the particles that compose them. However, the nanocrystallinity of actinide oxides and their subsequent reactivity are scarcely reported in the literature. New interest is focused on actinide oxides with nanoscale organisation because of the potential contribution of actinide nanoparticles in the environment (actinide migration) or their potential formation in industrial processes (High Burn-up Structure). In this context, the characterisation of the structural properties of nanocrystalline actinide oxides (Th, Pu) is carried out and the effect of the nanocrystallinity of these oxides on their reactivity is then studied in different media subjected to an ultrasonic irradiation in order to enrich the data on the actinide chemistry. Sonochemistry is used here to improve the reactivity of materials in solution through the physical and chemical effects resulting from the acoustic cavitation phenomenon. As a first step, thorium and plutonium oxides were prepared with nanoscale structuring in order to study the effect of particle size on their corresponding local structure. A local structural disorder driven by a nanoparticle surface effect is highlighted with the decreasing particle size. The reactivity of these nanocrystalline thorium and plutonium oxides was then studied in different aqueous solutions (H_2O, 0.05 M H_2SO_4, 0.5 M H_2SO_4) subjected to an ultrasonic irradiation. A significant dissolution of ThO_2 is observed in sulfuric medium at 20 kHz and leads in particular to the partial conversion of ThO_2 into a thorium peroxo sulfate. This compound, poorly described in the literature, has been the subject of a related study in which its crystal structure is resolved. Finally, the study of the sonochemical reactivity of PuO_2 reveals a reductive dissolution of PuO_2 into Pu(III) followed by its reoxidation into Pu(IV) with ultrasound. More generally, this thesis provides a better understanding of the structural properties of nanocrystalline actinide oxides that can be useful in the understanding of actinide migration in the environment but also in the preparation of an alternative future nuclear fuel. This work also provides new knowledge on the reactivity and the chemistry of actinide nanoparticles in different media under ultrasonic irradiation. These data will be useful in the case of the potential application of ultrasound in the spent nuclear fuel reprocessing process.