Institut de Chimie Séparative de Marcoule / CEA Marcoule (UMR 5257, CEA, CNRS, Université Montpellier, ENSCM)

THÉO CORDARA

will present his Ph.D. dissertation

Impact of the presence of fission products on the mechanisms of dissolution of uranium dioxide based model compounds

The defense will take place on Monday, November 27, 2017 at 10.00 am

in the ICSM Auditorium

The dissolution of spent nuclear fuels, performed in concentrated nitric acid solution, is a complex process due to some chemical and microstructural heterogeneities. As instance, the presence of Fission Products (FP) in various phases constitutive of spent nuclear fuels can affect the chemical durability of the materials. In this context, three systems of interest representing fresh UO_2 used as reference material, and UO_2 doped with FP products dissolved in the fluorite structure (lanthanide elements) or incorporated as metallic precipitates (platinoids) were prepared by oxalic or hydroxide precipitation. Dense pellets with microstructure representative for spent nuclear fuel were prepared through sintering step at high temperature.

The multiparametric study of the dissolution (macroscopic approach) was developed by varying independently several parameters. While incorporation of lanthanide elements induced a significant increase of the normalized dissolution rates, it remained lower compared to that observed for platinoids. Moreover, increase of nitric acid concentration or temperature led to the decrease of the chemical durability of the prepared materials. Additionally, significant modification of the preponderant mechanism of dissolution occurred for nitric acid concentration higher than 0.5 mol.L⁻¹.

For the three selected systems, the monitoring of the evolving solid/solution interface (microscopic approach) was performed operando by Environmental Scanning Electron Microscopy. Compared to pure UO₂, the presence of FP (lanthanide elements or metallic platinoids) induced preferential dissolution of grain boundaries and triple junctions. Coupling the results obtained from micro- and macro-scopic approaches led to complementary data of strong interest in order to improve the understanding of the role of FP during the dissolution of UO₂.

Keywords: UO₂; fission products; model compounds; dissolution



Deletive







