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Effect of platinoid elements on the dissolution of uranium dioxide

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Dissolution or leaching of the spent nuclear fuels (SNF) is a key step either in the field of their reprocessing or their long-term storage in underground repository. Moreover, SNF contain a wide variety of fission products including platinoid elements (PGM's) either incorporated in the UO2 matrix, or present in various separated phases for which the specific impact on the overall dissolution kinetics has not been yet fully discriminated. In order to answer this question, several samples doped with 0.6 to 3 mol.% of PGM's (55 % Ru ; 9.6 % Rh ; 35.4 % Pd) were prepared from hydroxide based synthesis [1,2]. After conversion then sintering, the prepared pellets were submitted to multiparametric dissolution tests in various media (0.1 to 4 M HNO3) and temperatures (25℃ to 60℃). The macroscopic description of the dissolution showed that the normalized dissolution rates were significantly increased for UO2 doped with PGM's compared to pure UO2 used as reference compound. This effect was strengthened in less acid media. As instance, a factor of 4500 was observed after 175 days of leaching in 0.1M HNO3. Simultaneously, the dissolution of the pellets was followed in operando by ESEM. The combination of macroscopic and microscopic approaches confirmed the modification of the preponderant mechanism occurring at the solid/liquid interface from redox-controlled dissolution in strong nitric acid media to surface-controlled dissolution for less acidic media.

1. Martinez, J. et al., J. Nucl. Mater., 462, 173-181, 2015
2. Martinez, J. et al., J. Europ. Ceram. Soc., 35, 4535-4546, 2015

Summary

Primary author(s) : Prof. DACHEUX, Nicolas (University of Montpellier)

Co-author(s) : Dr LAVALETTE, Claire (AREVA NC/BG aval); Dr CLAPAREDE, Laurent (University of Montpellier); Dr PODOR, Renaud (ICSM); Dr SZENKNECT, Stéphanie (ICSM); Dr CORDARA, Théo (ICSM); Dr MESBAH, adel (ICSM Marcoule, France)

Presenter(s) : Prof. DACHEUX, Nicolas (University of Montpellier)

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