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Experimental approach to study the alteration of MOX MIMAS fuels in an underwater storage situation

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After irradiation in a nuclear reactor, the irradiated Mimas® MOX (Mixed Oxyde) fuel assemblies are stored in a pool before reprocessing or geological disposal. This long-term storage requires considering an incidental scenario corresponding to the presence of a through defect in the zircaloy cladding, responsible for a containment breach and potential degradation of the defective rod [1].

This defect leads to the direct interaction between the spent fuel and the pool water. The pool water is pure aerated water that can contain boric acid at a concentration of 2g/L used as neutron absorber with a pH ranging from 4 to 6.5. It is also submitted to an intense gamma radiation field (around 1 k Gray/h) due to the presence of many fuel assemblies. The temperature of the pool ranges from 40 to 80°C because of the residual power released by the radioactive decay of the fuel assemblies.

These conditions are favorable to the oxidizing dissolution of the fuel under α and $\beta\gamma$ radiolysis of water which produces hydrogen peroxide H_2O_2 , the main oxidizing species. This alteration can lead to the release of radionuclides in solution as well as to the formation of $(U,Pu)O_{2+x}$ oxidized layers and to the precipitation of secondary phases such as studtite or schoepite. These phases having a density lower than that of the fuel can induce a worsening of the defect and thus impact the mechanical strength of the rods. It is therefore important to study these degradation mechanisms with the aim of recovering the rods after several years and decades of storage.

In order to assess this problem, an experimental approach coupling the study of simulating materials and highly radioactive materials is developed. It aims to describe the alteration mechanisms of fuels in contact with water as well as the reaction kinetics of dissolution/precipitation. This approach consists in implementing leaching experiments on materials ranging from model compounds $(U, Th)O_2$, to MOX Mimas® fuel $(U, Pu)O_2$ irradiated or not in reactor.

The model materials are developed at the ICSM by using thorium as a redox free plutonium surrogate. Thorium has the advantage of being easily manipulated but also of having a stable +IV valence, moreover like PuO_2 , ThO_2 crystallizes in a fluorite structure Fm-3m. Homogeneous materials $(U,Th)O_2$ are synthesized by hydroxide route in a first step [2]. They are used as a precursor for the synthesis of materials with a heterogeneous microstructure produced by powder mixing in order to mimic the heterogeneous character of the MOX Mimas®.

Moreover, after irradiation in a reactor, the chemical composition of MOX is modified by the production of fission products that are classified into four families ((I) volatile fission products, (II) metallic precipitates, (III) oxide precipitates and (IV) FPs in solid solution in the fuel [3]. Two types of FPs are also simulated in the $(U,Th)O_2$ model materials by addition of lanthanides (category IV) and platinum group metal elements (category II)) during the synthesis of [4].

Leaching experiments for these model or plutonium-containing materials are performed using two approaches:

- Under dynamic conditions at the ICSM on $(U,Th)O_2$ simulant/surrogate materials. These multiparametric experiments allow to study the effect of fuel heterogeneity and chemistry as well as water chemistry (boric acid, $[H_2O_2]$, pH) on the oxidative dissolution of the fuel.

- Under static conditions in a shielded cell in the Atalante facility at Marcoule on highly radioactive materials. The gamma dose rate within an assembly will be reproduced experimentally using a ^{60}Co source given the small amount of fuel involved for the leaching experiment.

These alteration experiments will be monitored over time by analyzing the solutions chemically and radiometrically and using surface characterization tools, the main one being Raman spectroscopy. This technique allows monitoring the evolution of many key parameters such as the chemical composition of the solid/solution interface, uranium oxidation state and the nature of the precipitated phases. It is also an isotopically sensitive technique for studying oxidation processes at the atomic scale [5, 6].

All this work will improve our understanding of the mechanisms of fuel oxidation and secondary phase precipitation in order to develop mechanistic model coupling chemistry to transport of chemical species in a defective rod. It will also contribute to the analysis of data acquired on defective rods extracted from storage pools.

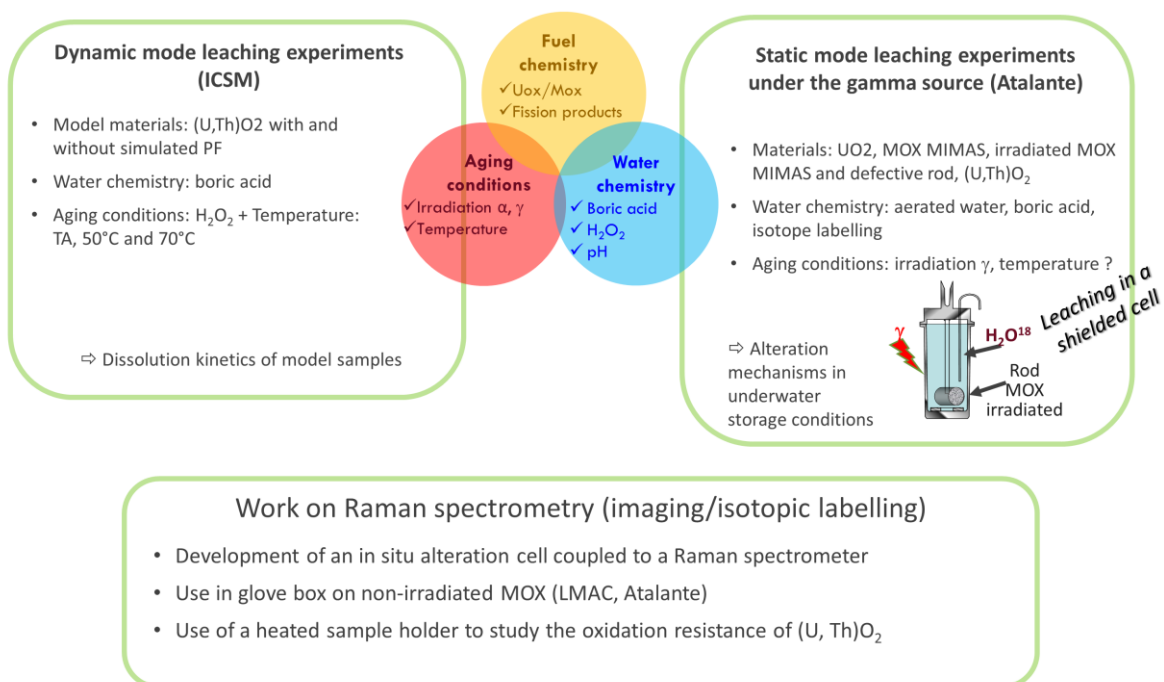


Figure 1 : Experimental approach

- [1] IRSN. Report DSR N° 316, (2008).
- [2] J.Martinez et al., J. Nucl. Mater. 462 (2015) 173–181.
- [3] Kleykamp, H., J. Nucl. Mater. 131(2–3) (1985) 221–246.
- [4] Théo Cordara et al., J. Nucl. Mater 528 (2020) 151836.
- [5] C. Jegou et al., J. Nucl. Mater. 458 (2015) 343–349.
- [6] L. Sarrasin et al., J. Phys. Chem. C 125 (2021) 19209–19218.