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## Elaboration of (U,Pu)O<sub>2-x</sub> mixed oxides with high plutonium contents: 60, 65 and 70 mol.% Pu.

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Uranium-Plutonium mixed oxides U<sub>1-y</sub>Pu<sub>y</sub>O<sub>2-x</sub> (MOX) are considered as the reference fuels for Sodium-cooled Fast neutron Reactors (SFRs). The plutonium contents  $y = \text{Pu}/(\text{U}+\text{Pu})$  will range between 20 to 35 mol. % and will exhibit an oxygen hypo-stoichiometry with an Oxygen/Metal ratio (O/M with  $M = \text{U} + \text{Pu}$ ) ranging between  $1.94 \leq \text{O/M} < 2.00$ .

Under irradiation, a thermal gradient along the fuel pellet radius (typically ~4 mm) is observed with a temperature close to 500°C at the periphery while the center can reach more than 2000°C [1]. At the beginning of the irradiation, the thermal gradient induces a redistribution of plutonium and oxygen atoms along the radius. On the one hand, the pellet is enriched in plutonium near the central hole (formed in the center of the full pellet during the first hours and days at full power) with a local Pu content greater than 40% and a lower O/M ratio ( $\sim 1.95$ ). On the other hand, the periphery of the pellet is depleted in plutonium with a local Pu content around 20%, and exhibits an O/M ratio close to the stoichiometry ( $\sim 2.00$ ). This phenomenon leads to heterogeneities in the MOX properties along the pellet. For instance, the local melting point will be modified as it depends upon the O/M ratio and the Pu content. A plutonium content rise involves a decrease in the melting temperature and thus a decrease in the safety margin of the reactor. Then it is essential to study the thermodynamics and structural properties of U-Pu mixed oxides with high plutonium content.

The main difficulty lies in the lack of data (enthalpy, heat capacity, conductivity or melting temperature, ...) for MOX with Pu content greater than 45% mol. and even more for MOX with Pu > 60% mol. [2]–[6]. These data would allow to feed the database to complete the modelling of the U-Pu-O phase diagram by the CALPHAD method and the input data in the GERMINAL performance calculation code. In order to obtain new data for that Pu content range, it is necessary to elaborate pellets. In this way, manufacturing single-phase and dense MOX pellets with Pu contents ranging between 60-70% mol., where the lack of data is the most significant, remains a great challenge. Indeed the difficulties associated with the manufacturing of high plutonium content pellets is to avoid the formation of secondary phases observed in the U-Pu-O phase diagram, constituting the miscibility gap denoted MO<sub>2-x</sub> + MO<sub>2-x1</sub> [7]. The study of this miscibility gap as a function of the temperature, O/M ratio and Pu content, implies the manufacturing of hypo-stoichiometric and stoichiometric pellets for each Pu content studied. First, stoichiometric pellets are elaborated and then, half of them are annealed in order to reduce (i.e. to decrease their O/M ratio) the samples. To obtain

these characteristics, the choice of the process parameters is essential, and in particular the optimization of the oxygen potential during sintering.

In this way, we will present a detailed manufacturing process based on powder metallurgy that is suitable to obtain such MOX pellets with 60, 65 and 70% mol. of Pu. The success of the elaboration will be highlighted by characterization results obtained by X-Ray Diffraction, Raman Spectroscopy, Scanning Electron Microscopy and Electron Probe Micro-Analysis.

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