Dissolution of single-phase (U, Pu)O_2 mixed oxide powders

The main objective of this study is to acquire data on the dissolution of (U, Pu)O_2 compounds to support the understanding of the phenomena occurring during the dissolution steps of MOX fuels irradiated in light water or sodium fast reactors. Previous studies, in particular on unirradiated MOX fuel, have highlighted the complexity of understanding the dissolution mechanisms through a direct approach. Indeed, the dissolution depends on a large number of parameters, which are mainly chemical dissolution parameters (acidity, temperature…). But it also depends on the physico-chemical characteristics of the fuel pellets (plutonium content, homogeneity of the plutonium content, microstructure, geometry...), a majority of which being highly dependent on the manufacturing process used. To avoid getting averaged responses due to the presence of heterogeneity in the Pu distribution in pellets, it is proposed to carry out a study on single-phase compounds in the shape of powders characterized by a well-defined stoichiometry (U and Pu) and a perfectly determined morphology. A step approach will allow understanding individually each elementary process to really grasp the dissolution mechanisms and improve the control of the dissolution process. A detailed understanding of the influence of chemical parameters on the dissolution thermodynamics and kinetics is expected at the end of this research. Besides, the acquisition of these data is an essential support for experimental studies conducted on irradiated MOX fuels.

The synthesis of materials for the study constitutes a significant part of the thesis to obtain single-phase mixed oxides at different Pu contents (Pu/U+Pu = 8, 20, 30, 45, 80, 100 %), with all comparable morphologies (crystallites size and form, equivalent agglomerates size) on the one hand, and mixed oxides with the same Pu contents but different morphologies on the other hand.

A bibliographic research will be conducted to determine the physico-chemical parameters of the dissolution environment that will be studied. A non-exhaustive list can already be drawn:

- temperature,
- acidity,
- nitrate concentration,
- nitrous acid concentration,
- NOX pressure,
- S/V ratio (powder total specific surface on acid volume),
- [U] + [Pu] or/and $[UO_2(NO_3)_2]$ + $[Pu(NO_3)_n^{(4-n)+}]$ (need a bibliographic work on complex speciation)