

# **The 2011 Frédéric Joliot/Otto Hahn Summer School**

**August 24 – September 2, 2011  
Karlsruhe, Germany**

**TOPIC ④**

**FUELS AND MATERIALS**

**4.2 Fuel behaviour modelling**

**Multi-scale modelling of fuels –  
From the atom to the mesoscopic scale**

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## ***Introduction***

Up to now developing and qualifying nuclear fuel has been a process essentially based on an empirical approach. One of the challenges for the next generation of reactors is to significantly increase the efficiency in designing innovative fuels. One way of doing this is to complement this empirical approach by a physically-based description of fuels to enhance the predictive capability of models. This should enable a reduction of the effort necessary and a better selection of promising fuel systems.

Actinide compounds, however, are very complex and specific materials. Moreover, the fuel element under irradiation is submitted to a wide variety of coupled phenomena involving among others the temperature, the irradiation damage, as well as the chemistry of the material and of the various fission products. The improvement of the understanding of the phenomena under irradiation requires to de-correlate the complex phenomena involved in the material evolution and to conduct fundamental research studies towards the atomistic level.

The modelling methods from the atomic to the mesoscopic scale are now ripe enough to obtain quantitative data on fuel materials and multiscale modelling approaches are being developed to study them. These approaches can in addition be coupled to separate-effect experiments, as well as to post-irradiation examinations (PIE) at the relevant scales. Recent programs and networks dedicated to such an effort have recently been created worldwide, in particular the F-BRIDGE project in Europe.

The lecture will be separated in two parts. First, the development of a multiscale approach from the atomic to the mesoscopic scale will be presented. The methods involved and the links between scales will be particularly discussed. Then, illustrations of the application of these methods, essentially on uranium dioxide, and the information which can be obtained will be presented. The examples chosen will focus on the links between scales, as well as on the coupling of modelling and experiments.

### ***Development of a multiscale approach from the atomic to meso scale and coupling with the experiments to obtain relevant data for the description of the fuel behaviour***

The main modelling methods used for the study of fuels from the atomic to the mesoscopic scale, in particular electronic structure calculations empirical potentials, kinetic Monte Carlo, cluster dynamics, are the same as on structural materials and will be presented in Topic 4.1. Their specificities when applied to fuel materials under irradiation will be shown. In particular the description of strong correlation and of the various oxidation states present in actinide compounds will be discussed. The DFT+U formalism which improves the treatment of the strongly correlated  $f$  electrons will be presented. Then, the specific validation of the atomic scale methods, for the description of nuclear fuel under irradiation will be detailed. This validation is essential since the results of the atomic scale calculations form the foundation of the multiscale approach. Finally, the development of a multiscale approach and coupling with the experimental studies and the analysis of the relevant data to pass from scale to scale will be shown.

## *Application for the investigation of the transport properties in uranium dioxide*

Three recent studies performed in the F-BRIDGE European project will be presented to illustrate the coupling of modelling and experiments and of various scales, as well as the results which can be obtained.

First, a joint electronic structure modelling and experimental study of the oxygen point defect formation and migration mechanisms in  $\text{UO}_2$  will be shown.

Formation and migration energies of oxygen vacancies and interstitials were calculated in the DFT+U formalism using electronic occupancy control. From an experimental standpoint, a combination of tracer diffusion coefficient and electrical conductivity measurements were carried out at different oxygen potentials and temperatures on  $\text{UO}_2$  samples containing different impurity levels. They pointed to oxygen migration being due to isolated oxygen interstitials over a wide range of oxygen potential. In addition, the temperature study provided an estimate of interstitial formation and migration energies.

The results strongly pointed to the fact that DFT + U calculations quantitatively describe transport phenomena in uranium dioxide, at least for oxygen self-diffusion. The coupling of the electronic structure results with the experimental energies indicates the indirect interstitial mechanism as being the relevant oxygen migration mechanism. These results open up the prospect of using first-principles DFT + U calculations as part of a predictive approach to determining transport properties in other actinide oxides.

Second, the simulation of displacement cascades using molecular dynamics and empirical potentials and the comparison with TEM observations to study the formation of bubbles will be presented.

The first stages of defect cluster formation resulting from irradiation in uranium dioxide were investigated by overlapping 10 keV displacement cascades in a small crystalline area. Nanometre size cavities and dislocation loops were shown to appear as a result of the irradiation process. A specifically designed TEM experiment involving He and heavy ions implanted thin foils were carried out to support this modelling work. These results, in conjunction with several other observations taken from the literature of ion implanted or neutron irradiated uranium dioxide, suggest a radiation damage controlled heterogeneous mechanism for insoluble fission product segregation in  $\text{UO}_2$ .

Third, the development of a cluster dynamics model using the results of the atomic scale methods will be presented. This type of models allows the simulation at the mesoscopic scale of the long term evolution of the material microstructure during irradiation or annealing conditions, i.e. the size distribution of the clusters. Simplified versions of this kind of models can be used in fuel performance codes. An existing cluster dynamics code developed for He and other solutes in metals was adapted to the case of xenon in  $\text{UO}_2$ . The main achievement was the coding of a generic thermodynamics of gas bubbles, particularly useful in situations where the bubbles can dissociate and grow. This model was fed thermodynamic parameters, such as Xe incorporation energy and Schottky formation energy determined by electronic structure calculations. Kinetic parameters (migration energies of Xe and Schottky) were taken from empirical potential calculations.

The code was first tested to simulate post-irradiation annealing of  $\text{UO}_2$  fragments. Two thought experiments were compared: in the first one, Xe was supposed quasi insoluble, as is commonly done. In the second one, Xe incorporation energy was set to the much lower value obtained using electronic structure calculations. Different behaviours were obtained, which underlines the importance of having precise input data for the mesoscale models.

## *Bibliography*

### **1) On methods and validation**

- Bibliography given for Topic 4.1.2
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### **2) On illustrations**

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