New capabilities of analyses with a versatile nuclearized dual beam

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ABSTRACT

In order to replace a 1993 XL30 Scanning Electron Microscope (SEM), the Irradiated Fuel Microstructure Study Laboratory in the CEA-Cadarache LECA-STAR Hot-cell Facility has acquired and installed a new microscope. After more than 4 years of work, this equipment opens new research lines on irradiated fuels.
This ZEISS Auriga SEM FIB (Focused Ion Beam) microscope combined with Electron BackScattering Diffraction (EBSD) and Energy Dispersive X-ray Spectroscopy (EDS) equipments is able to perform a wide range of analyses. This microscope and the glove box that is connected to, had to be installed in the same hot cell as the previous SEM, which was the most difficult point of the design.
Another challenge was to keep the initial technical performances of the machine by cancelling vibrations induced by the ventilation of the facility. Furthermore, the microscope can be disconnected from its glove-box. A translational and rotational movement of the microscope gives access to all detectors installed around the chamber as well as to the glove-box, for basic or servicing hot lab facility operations. A specific anti-contamination shield has been designed to maintain a low level of contamination in the SEM FIB chamber from nuclear materials.
The FIB capabilities while performing observations with the SEM, give access to 3D information. It also allows to prepare micro samples such as TEM (Transmission Electron Microscope) lamellaes, but also, pillars or cantilevers for in-situ mechanical tests, using a specifically designed nano-indentor. Also, a motorized STEM (Scanning Transmission Electron Microscopy) detector can be mounted on the microscope stage.
The paper puts forward the versatility of this nuclearized SEM FIB, the specific developments made and some applications.

1. Introduction

The Hot Lab LECA-STAR (Figure 1), a CEA nuclear facility dedicated to R&D on irradiated fuels has been designed to analyze the behavior of a wide range of nuclear fuels after irradiation. Hot cells allow to carry out tests or transformations on fuel rods, as well as non-destructive or destructive examinations on irradiated fuel from both industrial or research reactors.

In order to characterize phenomenon involved at a lower scale in irradiated fuels, the microanalysis laboratory (Figure 2), situated in the basement of the facility, is equipped with complementary devices:

- An electron microprobe (EPMA),
- A secondary ion mass spectrometer (SIMS),

- An X-ray diffraction ,
- A profilometer (confocal microscopy),
- An electron microscope.

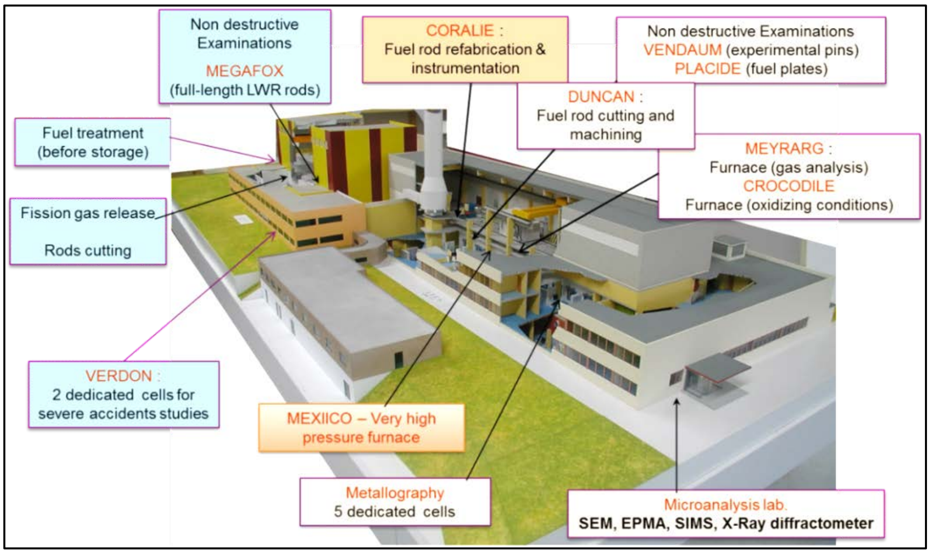


Figure 1: The LECA STAR hotlab facility

In 2016, the old FEI XL 30 SEM of this laboratory has been replaced by an Auriga 40 SEM FIB from Carl Zeiss. This FEG SEM allows classical SEM observations, and opens especially new possibilities with simultaneous material sputtering thanks to the gallium Focused Ion Beam, as TEM specimen's preparations or cantilevers for example.

Almost 5 years were necessary to carry through this project. Six months were first dedicated to project definition, specifications writing of a microscope adapted to the expected performances, and of course nuclearization studies to answer nuclear environmental specific requirements. The commercial procedure took place over one year. Three and a half years were finally necessary to complete successfully all the stages for the installation and the on-site tries, for both the glove box and the new microscope with the different prototypes.

In order to answer to the increasing demand for validation of fuel modeling codes which require continuously experimental information at a lower scale, a FEI's Talos Transmission Electron Microscope will be also installed at the LECA in December 2016.

Figure 2: Panoramic view of the microanalysis laboratory

2. The MEB FIB and its setup

The SEM FIB and the glove box are installed in a small closed room (2,1 m x 2 m x 1,9 m), Figure 3. Setting up was performed by the front side of the room as the shielded wall can be moved thanks to an air cushion. This front shielding is equipped with four bar manipulators and two windows to allow samples movements from the adjacent storage cell, either to be placed on the SEM FIB stage, or to be analyzed by the EPMA installed in the following room, or by the confocal microscope. As required by the environment, control systems are outside the room, in front of the shielded wall to protect the operators from irradiation.

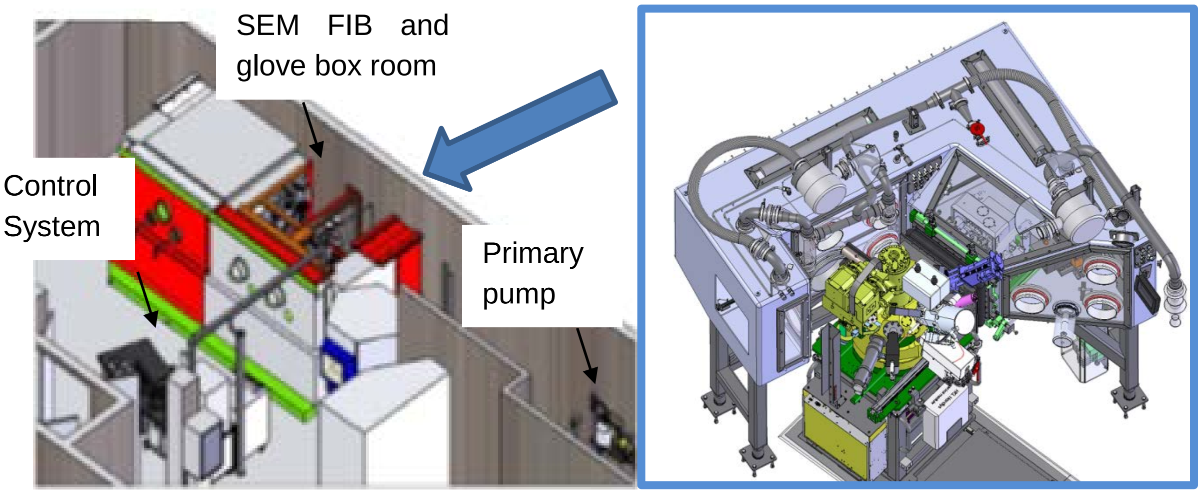


Figure 3: FIB SEM and the glove box in the microanalysis laboratory

The glove box, connected to the microscope for contamination containment, is also used to transfer samples from the storage cell to the EPMA's glove box, thanks to two tunnels each equipped with a proper transfer system. In order to limit the handling of these contamination sources, a system, settled in the SEM glove box, allow to easily move the samples from the exit of the tunnel coming from the storage cell, either to the entry of the tunnel connected with the EPMA glove box, or to the SEM loading system.
The sample, put on a specific sample holder well adapted to the remote working, is guided on the stage with a loading system (Figure 4) mounted on the glove box. This system avoids all risks of sample dropping during the loading stage, as doing this operation with remote handle could lead to problematic situations should the sample drop and be blocked in an unreachable position. Loading off the sample is done same way, the stage moving to predefined positions.

Figure 4: SEM loading system

The SEM is a Carl Zeiss Auriga 40 equipped with a Orsay Physics COBRA FIB column (Figure 5). A Gallium source produce the ion beam, and the angle between the SEM and the FIB beam is 54°. The microscope is equipped with

- two in lens detectors: one for secondary electrons (in lens) and one for backscattered electrons (EsB),
- a SESI Robinson detector for secondary electrons or ions imaging and a removable YAG detector for backscattered electrons, located on the sides of the chamber,
- the EBSD and EDS, for crystallographic and chemical analyses, are both from Oxford, and are also removable and specifically shielded,
- an Orsay Physics Gas Injection System allows Platinium injections on the surface of the sample for paddings or weldings,
- a Kleindick's micromanipulator can be mounted to take off thin foils or micro-samples,
- an Anton-Parr nano-indenter,
- a STEM detector to control the preparation of TEM specimens.

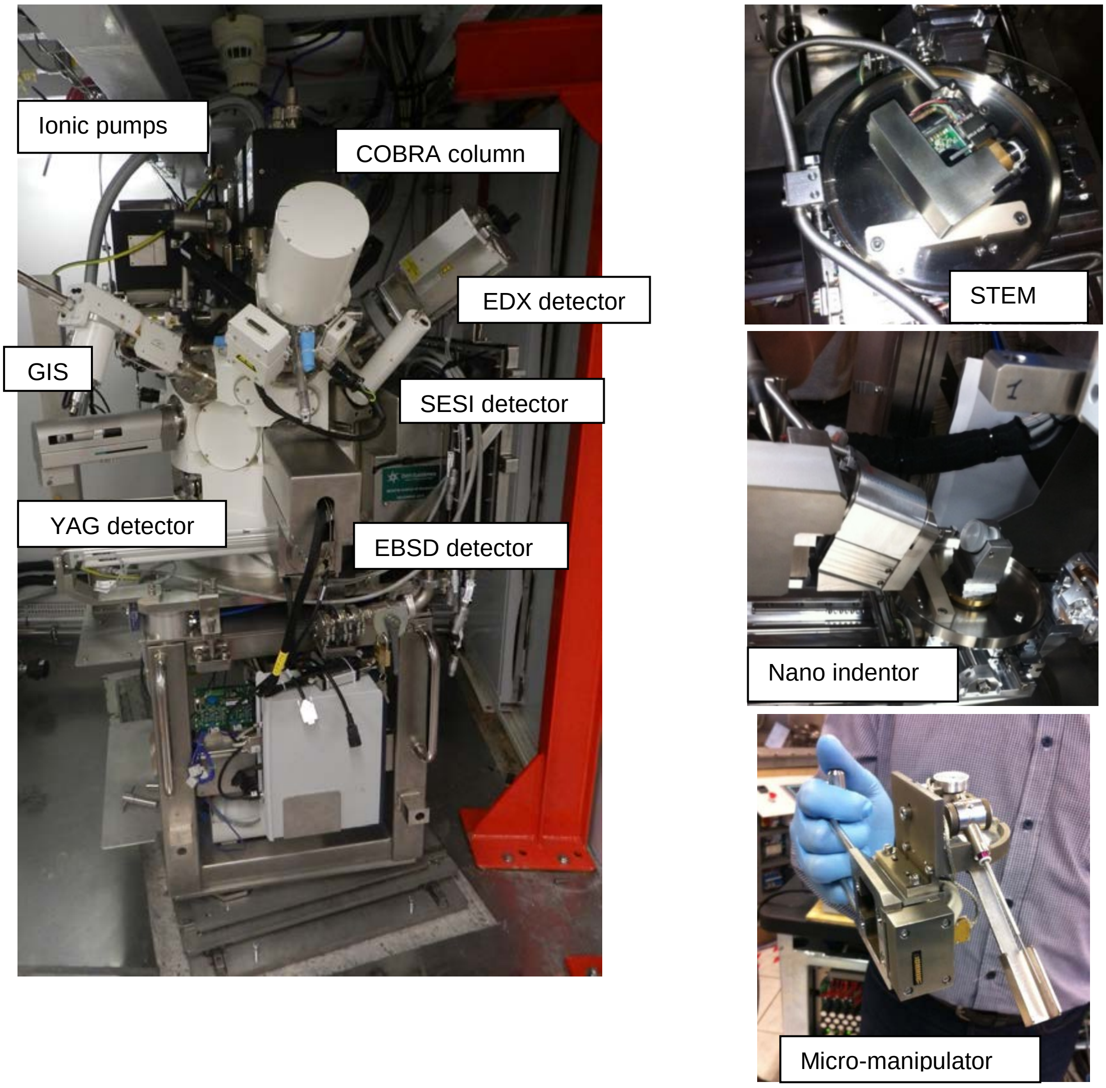


Figure 5: Auriga 40 SEM FIB and accessories

The micro-manipulator and the nano-indentor are loaded on two rails of the door of the chamber and the STEM is installed on the stage. As all these accessories have to be put in place and plugged manually via the gloves of the glove-box, radioactive samples have to be removed of the microscope and the glove box before opening the room.
The EDX and the STEM detector have to be used only on small samples with very low irradiation level to avoid damages on the sensitive parts of the detectors.

3. Specific developments

3.1 Vibrations elimination systems

The ventilation system of LECA hotlab induces vibrations which can seriously decrease the resolution performances of the SEM. To limit such perturbations, a specific setup has been studied and settled.
Beside the pendular system of the pneumatic table on which the SEM FIB is placed, all the different connections between the microscope and the glove box, linked either on adjacent cells, or to the ventilation system, are realized with boots to filter the vibrations. Also, the vacuum primary dry pump is located in the corridor adjacent to the room.

To assess the efficiency of all these anti vibrations settings, SEM images on gold balls were first acquired without the facility ventilation system connected to the glove box (Figure 6). Same areas were afterwards observed after connecting the glove box to the ventilation (Figure 7) and compared to the firsts. Any difference at x300 000 were observed. The resolution tests on site are 1,4 nm at 20 kV and 10kV.

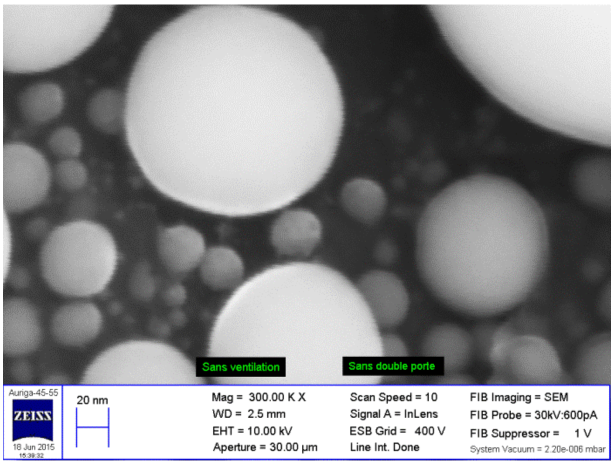


Figure 6: Test without ventilation

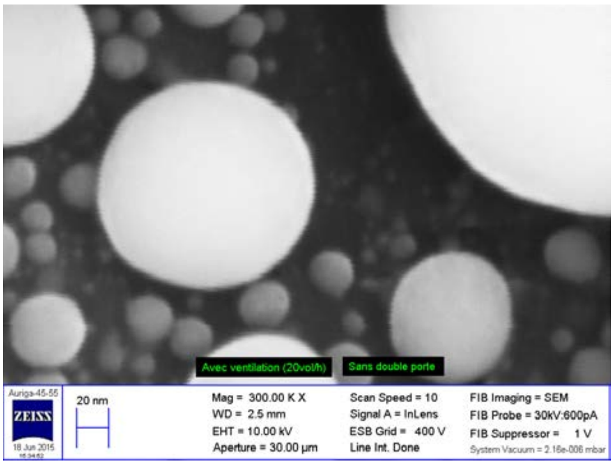


Figure 7: Test with ventilation

3.2 Maintenance of the equipment and its glove box

SEM FIB servicing and glove box operations require an access to the room. After nuclear samples are removed and stored in the adjacent cell, a secondary door gives easily access to the room avoiding moving the front shielded wall on air cushion.
The microscope, connected to the glove box (Figure 8) for samples exams, can be disconnected in order to give access to all parts of the glove box as well as to all detectors or items of the SEM-FIB. When disconnected, the microscope being installed on a specific stage, can be translated or rotated (Figure 9), given so space needed for servicing

The servicing operations of the equipment and glove box working have been tested on site and are now used. They are quite easy and convenient, even if the room is very tight.

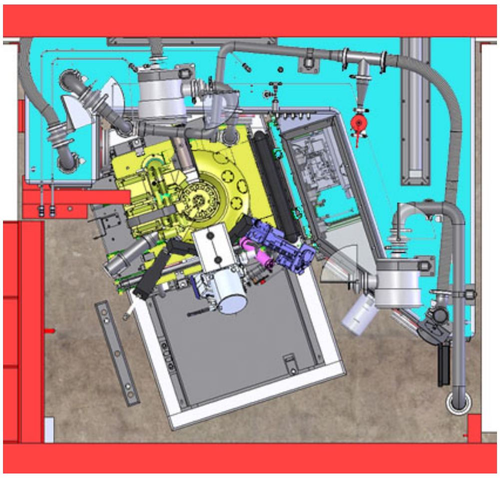


Figure 8: Glove Box and MEBFIB in basic use

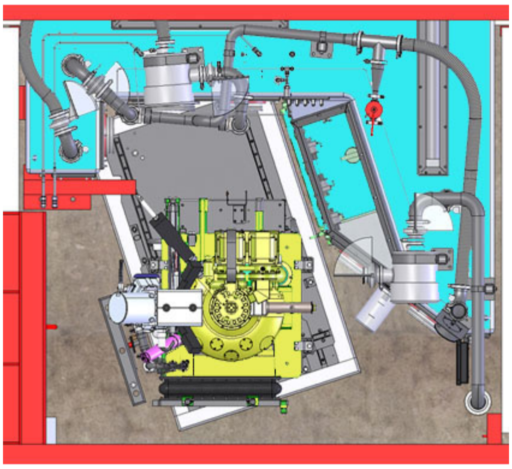


Figure 9: Glove Box and MEBFIB servicing

3.3 Shield against contamination

Sputtering nuclear fuel material by gallium beam leads to contaminate the chamber by successive accumulations. To limit this contamination, a removable collector shield has been studied and installed (Figure 10). The shield will have to be changed periodically and the frequency will be adjusted after 6 months of analysis on irradiated fuels according to contamination level.

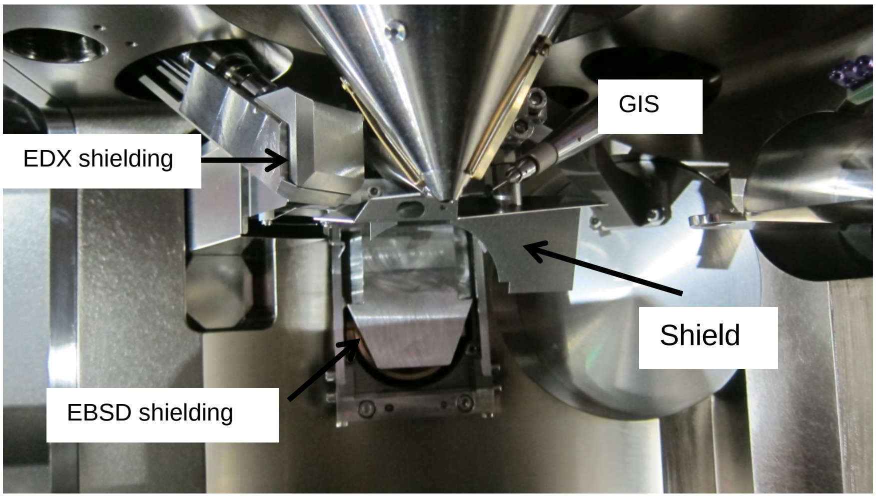


Figure 10: Inside view of the chamber: Shield and detectors

3.3.1 Sputtering study

A study has been made to identify the deposition areas of the sputtered material. For this, a copper grid sample has been sputtered (Figure 11) and the volume of the sputtered area on the grid has been measured using SEM and optical means.

The copper sputtered has then been collected on two stickers (Figure 12):

- the first one, located around the sample. The sticker has a hole on its centre to put the sample on the holder,
- the second one, mounted on stubs at 3 mm above the sample. It has a hole in its centre for the passage of the fib beam.

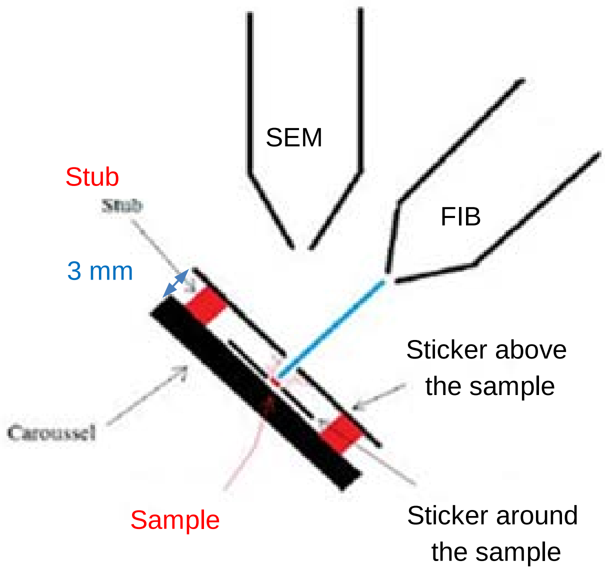


Figure 11: Plan presenting the sample and the two stickers

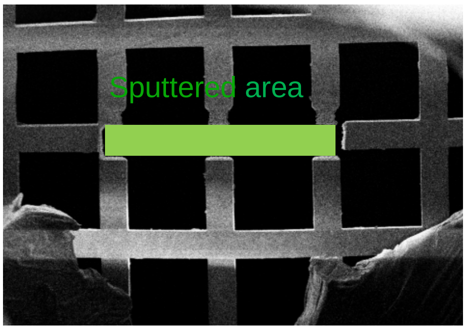


Figure 12: Copper grid after sputtering

After the test, the stickers have been collected and dissolved in nitric acid and the solutions were analysed by ICPMS. The quantities collected on the two stickers have been compared to the quantity sputtered on the grid.

The measures show that 57% of the sputtered copper has been collected on the sticker above the sample and mounted at 3mm. Between 10 and 36% of the sputtered copper has been collected on the sticker around the sample (this measure doesn't take account of the deposition near the sample). This part of sputtered material deposited around the sample doesn't contaminate the chamber as the sample is removed after the analyses.

3.3.2 Efficiency of the shield

The shield has been designed with different holes for electron beam, ion beam and signals for the detectors. The quantity lost in the holes has been calculated between 35% and 40%. As the measured quantity, collected on the shield is 57% (results on scotch at 3mm), the total efficacy of the shield (collected on the shield / total sputtered) should be about 34-37%.

3.3.3 Shield efficiency control

The efficiency of the developed shield has been checked. For this, the shield has been mounted in the chamber under the FIB and SEM columns. A cerina sample has been sputtered in a squared crater (Figure 13). The volume of the crater has been measured by profilometry (Figure 15). To measure how the shield collects the sputtered material, scotches have been put on different parts of the shield, as shown in the Figure 14. After the sputtering step, the shield has been removed from the chamber and the scotches collected. Each scotch has been dissolved in nitric acid and as previously, the solutions analyzed by ICPMS. The shield has collected 35 % of the total sputtered material.

This result is very coherent with the calculated efficiency when the shield has been designed (3.3.2). The scotch 1 on the shield (Figure 14) has collected 98% of the total collected cerium. With the sputtered material collected around the sample between 10 and 36 %, more than 45-71% % from the sputtered material will be collected after the test.

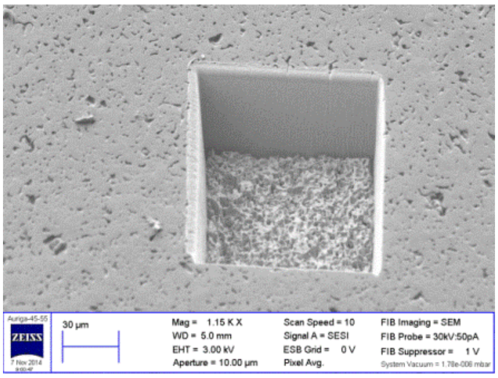


Figure 13: Crater after sputtering

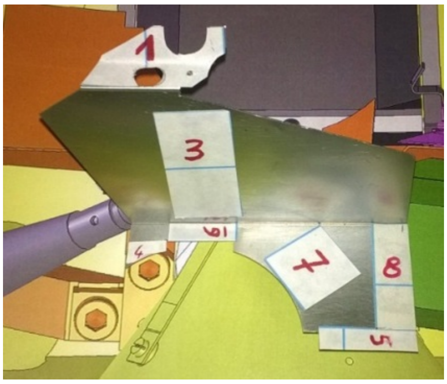


Figure 14: Efficiency test of the shield

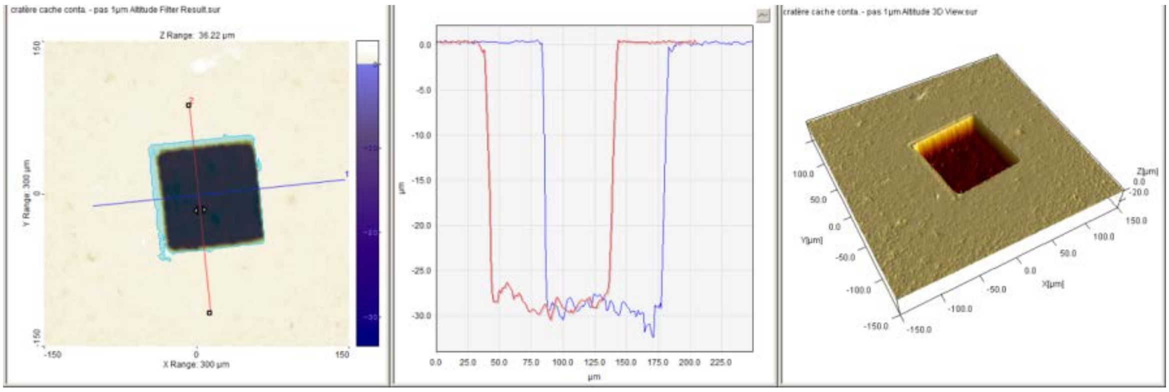


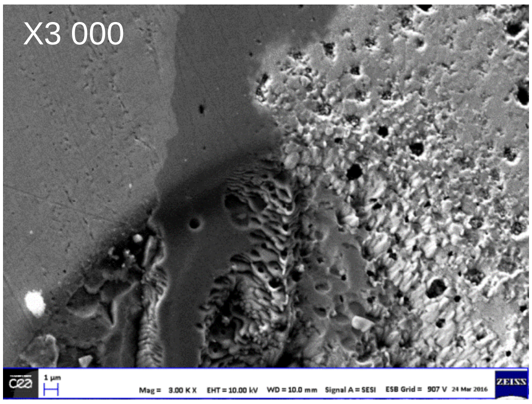
Figure 15: Crater profiles

4. Some applications

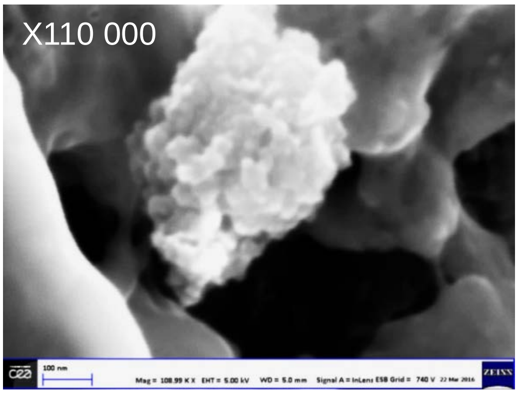
4.1 2D analysis – nuclear fuel

The performances of this new microscope have been tested on irradiated fuels. A few examples of pictures with different magnifications are given in the Figure 16.

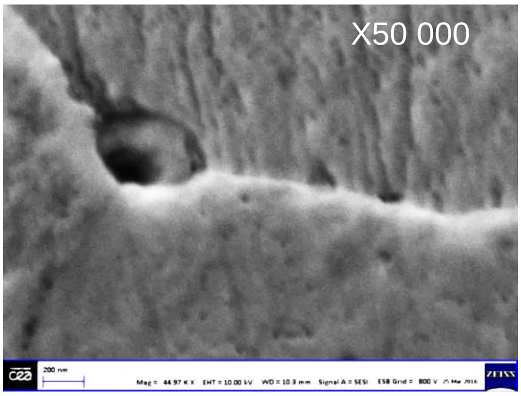
At x210 000, the carbon contamination under the SEM beam is deposited on the surface of the sample (Figure 16d).



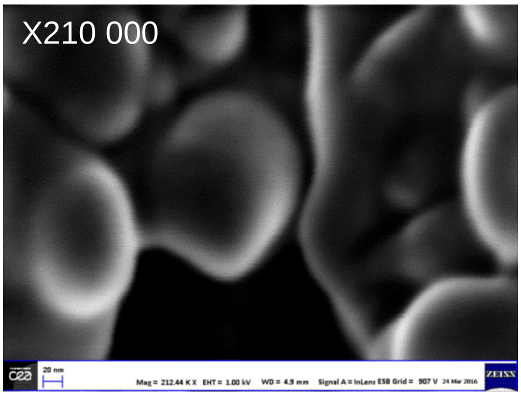
a.Zirconia and rim



b.Irradiated fuels



c.Grain boundary of irradiated fuel

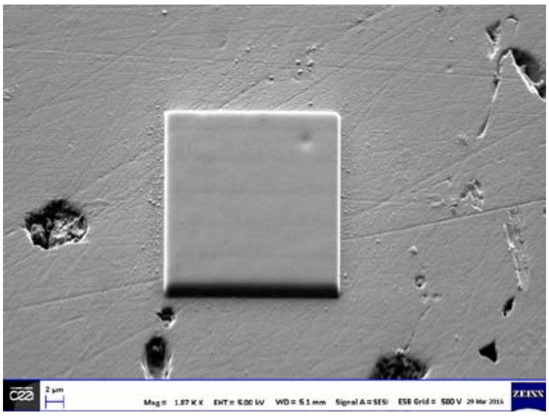


d.Porosity of irradiated fuel

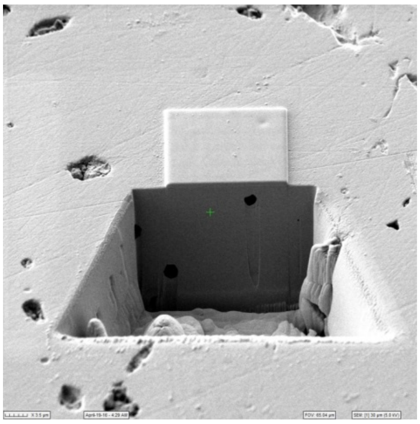
Figure 16: Resolution images

4.2 3D Analysis – Volume of a fission gas bubble

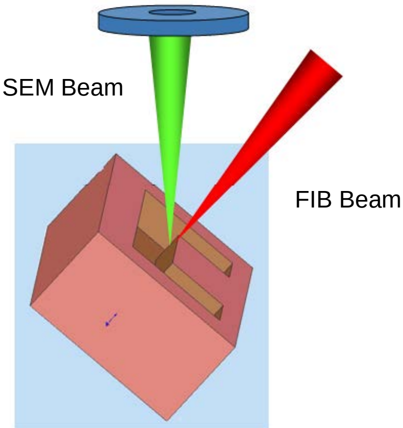
A fuel cube of 10x10x10 µm3 has been sputtered to investigate the real form and the exact volume of one xenon bubble detected previously by EPMA in an irradiated fuel.
Beforehand, a Platinium pad of 1 µm in thickness has been deposited to protect the surface of the cube (Figure 17a) and to avoid the curtain effect when sputtering. A large cavity has then been produced in front of the cube (Figure 17b) to clear SEM view. Afterwards, the cube has been sliced with the ion beam, acquiring a SEM image at each step (Figure 17c). The different slides show that the bubble is not spherical as expected and extended to 10 µm in length.



a.Platinium pad



b.Cavity in front of the cube

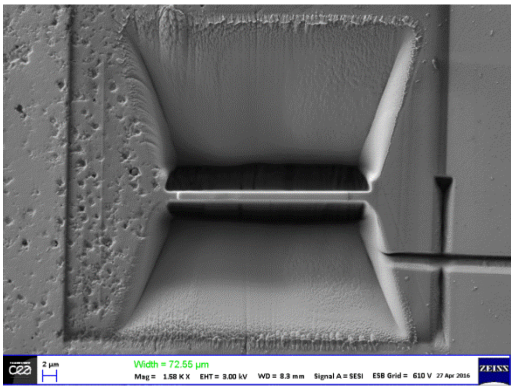


slices

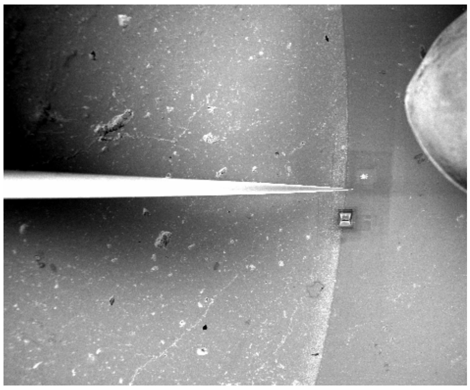
Figure 17: Volume of a fission gaz bubble

4.3 Thin foil

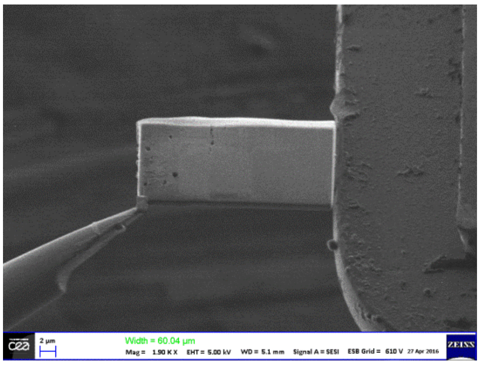
A thin foil has been manufactured by FIB in a zirconia layer created by oxidation between the fuel and the cladding [2]. The first stage involves locating the area of interest and depositing a layer of Platinum to protect the specimen. Then, two trenches are milled on each side of the layer to give space for extraction with the micro-manipulator. The 2 microns' thick specimen is moved to a grid to be weld and freed from the manipulator. The specimen is then gradually thinned to roughly 100 nm of thickness by using beam with lower intensity and lower high tension (1 kV) to reduce most of the defect created by the gallium during the previous stages.



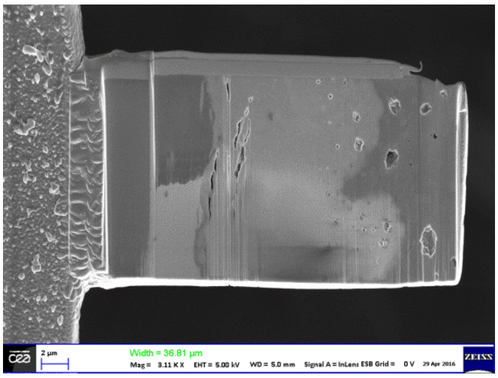
Wall creation



Punction with the micromanipulator



Welding on the grid



Thinning of the foil

Figure 18: Thin foil preparation

5. Conclusion

The MEB FIB exploitation has started the 16th February 2016. Despite the complexity of the microscope and its installation in nuclear environment, the first tests and analysis on irradiated fuels are promising. This equipment will open new research lines on irradiated fuels behaviour. As shown, this machine allows 2D or 3D analyses and creation of TEM lamellae. It will also allow the study of the fragile mechanical properties of the irradiated fuel with the realization of in situ micro-mechanical tests on pillars or cantivelers. At last, grain boundaries of the irradiated fuels could be analysed by EBSD.

6. References

[1] D. Nozais, O.Dugne, V. Basini, M.C. Anselmet – “Keys figures and services at the LECA STAR facility Hotlabs” - Hotlabs Conference - Sept 2015

[2] C. Ciszak - “From cladding oxidation to fuel-clad strong bonding in PWR fuel rods: investigation of the fuel-clad interface evolution by morphological, structural and elementary analyses” - Numat Conference – Nov 2016