One Year of Operation of the Shielded SIMS with Irradiated Materials in the LECA Facility

L.Desgranges, B.Pasquet
CEA Cadarache 13108 Saint Paul lez Durance France

A shielded SIMS, IMS 6fR from CAMECA® company, was installed in CEA hot cells at Cadarache, France [1] and has been operating with irradiated materials since June 2002. The apparatus worked satisfactorily good during this period and brought some new scientific results, especially concerning rare gases. In this paper we shall focus on how the apparatus was used with irradiated materials and we will only briefly present the main scientific issues which have been tackled up to now.

I Operating with irradiated materials

The SIMS is presented on figure 1. It is divided in three main components : the apparatus itself, a glove box and a shielding consisting in a lead box and two additional metallic panels. The sample comes from a shielded box linked to the SIMS with a tunnel. It is entered in the SIMS through an airlock. During the analysis the sample stays inside the vacuum chamber.
It is the first time a CAMECA IMS is used for the analysis of irradiated material. Therefore special care was taken for the introduction of the first samples in the apparatus, especially the nuclear safety of the IMS 6fR was checked for both irradiation and contamination. The influence of radio-active sample on the instrumentation and on the quality of the measurement was also estimated.

1°) Irradiation

From a regulation point of view, the SIMS is located in a room which is considered as a green area, i.e. the rate dose must not exceed 25 µGy/h. Inside the lead box (see figure 1) the area is red (no entrance) when a sample is in the apparatus, it can be downgraded to orange area which allows the maintenance inside the glove box (dose rate less than 100mGy/h). The area between the lead box and the metallic panel is considered as yellow area (dose rate less than 2mGy/h) because it not contaminated but the dose rate can be high. The SIMS is indeed not firmly linked to the lead box because the vibration coming from the building must be avoided. So the metallic panels have been settled to protect the operator from the radiation coming through the connection between the lead box and the IMS. A radioactive leak was considered when the dose rate was higher than to 25µG/h in the green area and 2 mGy/h in the yellow area.

The control of the radiation tightness of the shielding was performed in several steps. Once the shielding of the apparatus was installed in CEA Cadarache, it was checked with a 6 Gy/h caesium source that no major leak exists in the shielding.
Then the apparatus was connected thanks to a tunnel to the shielded box where the radioactive samples are stored. The efficiency of the whole shielding, tunnel and apparatus, was then tested with a well characterised radioactive sample. This sample consisted in a slice of a 6 cycles UO_2 French PWR fuel rod, embedded in cylindrical holder thanks to a low boiling point metallic alloy. This sample was set in four different positions corresponding to the position where the sample can stay for a while in normal operating conditions. These positions were :

- inside the tunnel between the apparatus and the storage box,
- inside the IMS 6fR vacuum chamber at the position where the analysis is performed,

- inside the airlock through which the sample is introduced from the glove box inside the apparatus,
- inside the glove box in special location designed as a parking position, where the sample is settled waiting either to exit the glove box or to enter the airlock.

The rate dose was then measured outside the apparatus in 14 different points. No radioactive leak was detected when the sample was in the vacuum chamber or in the parking position. Weak leaks were measured with the sample in the tunnel and in the parking position.

After these measurements, the shielding was improved by adding some more metallic parts. These modifications were performed by CAMECA company. Some new dose rate measurements were performed after these modifications and no more radioactive leak was detected.

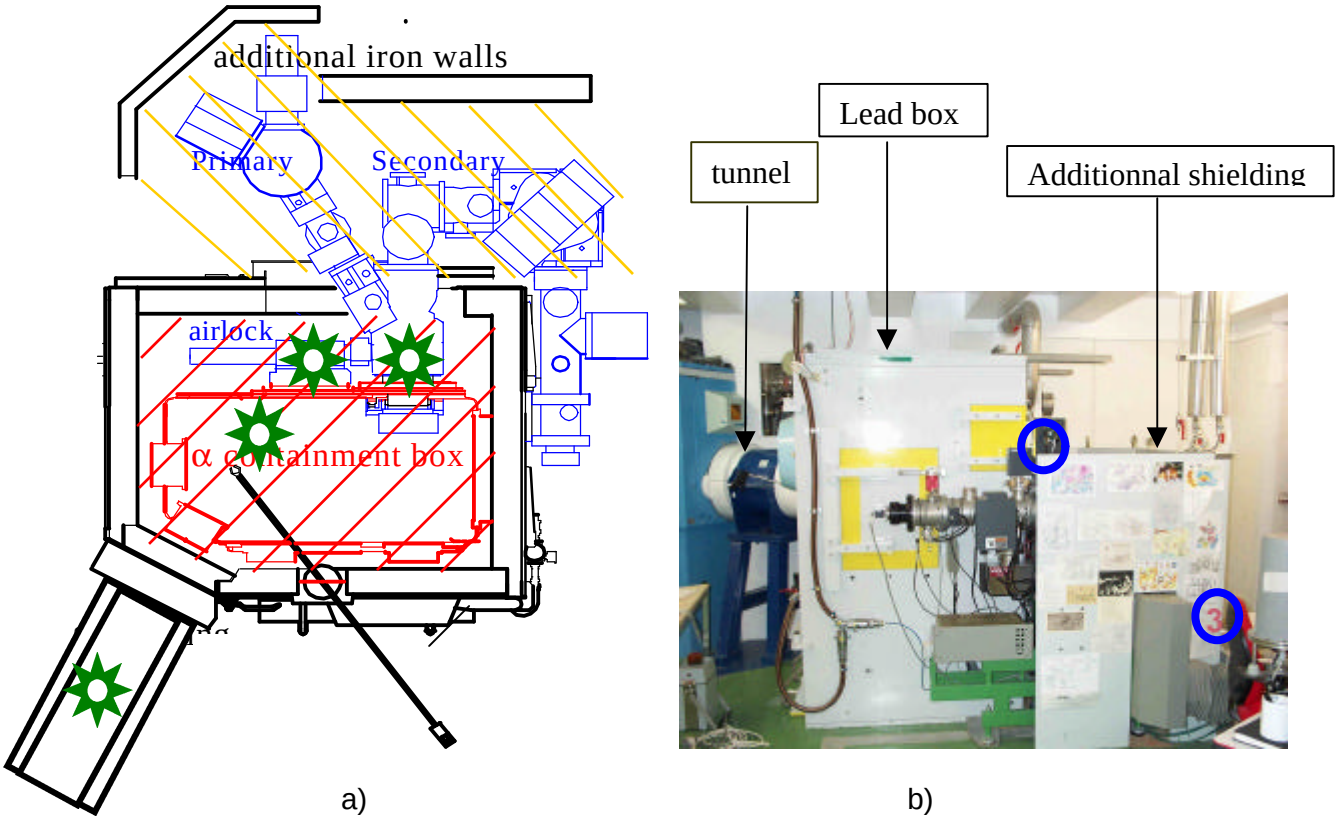

Figure 1 : a) Schematic drawing of the SIMS. The IMS 6f is dawn in blue, the shielding in black and the glove box in red .The red area is symbolised with hatched in red and the yellow area in yellow. The locations where the radioactive sample was put during the radiation measurement are symbolised with green stars.

b) photograph of the SIMS. The points where the main radioactive leaks were detected are symbolised with blue circles.

2°) Contamination

The contamination in the apparatus comes mainly from the measurement during which the sample surface is sputtered. The geometry of the vacuum chamber is presented on figure 2. The primary ion beam hits the sample surface with a 30°angle. The sputtered atoms are ejected in all direction over the sample surface with an approximately cosθ distribution, where is the angle made with a line perpendicular to the surface. As a consequence most of the contamination is deposited on the so called immersion lens, a metallic plate which creates the electrical field necessary to extract the secondary ion beam and which is drilled with holes allowing the passing through of the ions beams. A metallic plate can be removed from the front side of the immersion lens, most of the contamination is expected to be located on this metallic plate.

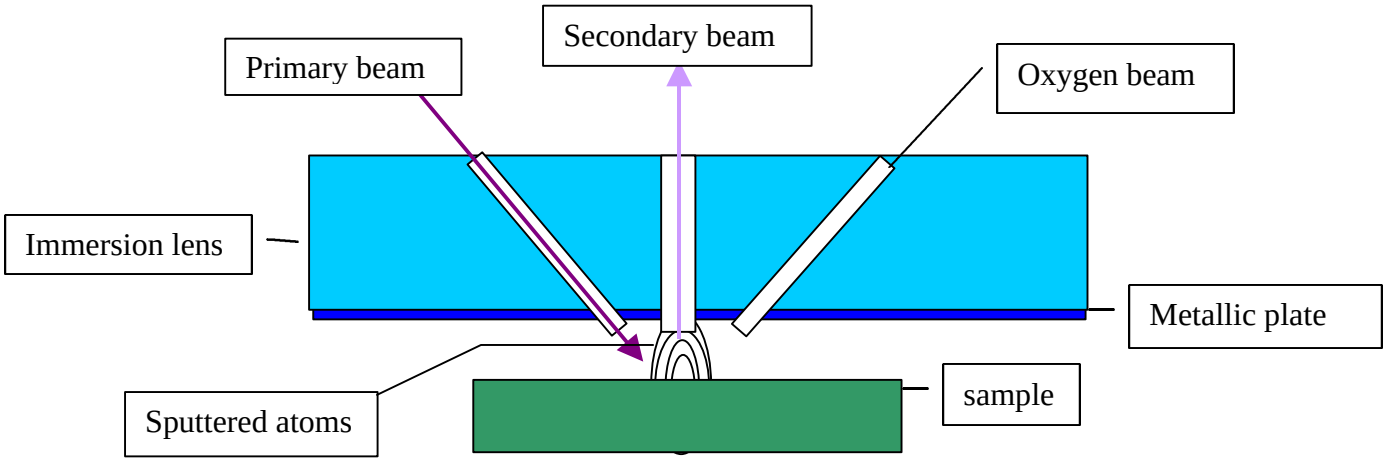

Figure 2 : Schematic view of the immersion lens and the sample in the vacuum chamber.

After 9 months of operation, the vacuum chamber was opened for maintenance. The metallic plate was removed from the immersion lens and a new one was installed. The contaminated metallic plate was checked outside the glove box : its activity was measured equal to 10.000 Bq corresponding to a dose rate of around 150 µGy/h. Three month later the whole immersion lens was changed. The contaminated immersion lens was changed. The contaminated immersion lens is stored in a vinyl bag. The activity measured outside the vinyl bag was about 4000 Bq. Anyhow most of the contamination is fixed : the activity measured on a wipe, dampened with alcohol, after the cleaning of the immersion lens was only a few hundred Bq.
Another wipe test was performed on the bottom of the glove box, the activity of the wipe was 80 Bq, which confirms that the sample itself is not an important source of contamination. This is consistent with the fact the samples are polished slice of fuel rod cleaned with alcohol.

These results evidenced that the level of contamination is not a hurdle for the experiments with SIMS . It level can be kept at low level by changing the immersion or its metallic part at each maintenance cycle. Moreover the irradiation induced by this contamination is low enough to allow any operation maintenance to be performed by manual handling in the glove box. It is also important to notice that it is mainly fixed contamination, which induces that contamination is rather unlikely to move towards inner parts of the apparatus.

3°) Instrumentation

The instrumentation associated to the IMS 6fR is very complex and two main causes of degradation due to the use of radioactive samples were expected : the existence of a radioactive background in the measurements and the degradation of components facing the sample.

In the IMS 6fR, the detectors measuring the secondary ions is located at 40 cm from the radioactive sample itself during the measurements. Although the detectors are protected by a 10 cm thick lead wall and the 3 cm thick stainless steel wall of the vacuum chamber, some gamma radiation could have bring some noise on the measurement. This point was checked by measuring the signal at mass 5 with a radioactive sample in the vacuum chamber. No ion can indeed be detected at mass 5, the signal measured thus only comes from the instrumental background. At mass 5, the signal was around 1 counts/s, and no specific contribution coming from the sample was evidenced.

In the IMS 6fR, most of the instrumentation is made of electrical wires which have a good resistance to irradiation. Two specific pieces need however a special care : the alumina piece which insulate the sample from the vacuum chamber and the glass piece which permit to see the sample surface with a microscope. Up to now, the alumina piece seems to be insensitive to the radiation because the sample voltage has been keeping constant and no breakdown occurred. On the contrary the glass piece darkened and it was necessary to remove it (after one year operation).

II Main scientific issues

In the IMS 6fR SIMS, the ions are mass filtered thanks to magnet which allows to get mass resolution as high as M/ M = 40 000 and also to make ionic images with a lateral resolution better than 1 µm. This two possibilities will be presented and also the first results concerning the measurement of xenon in irradiated fuel.

1°) High resolution mass analysis

The secondary ions produced by the sputtering of the sample are measured as a function of their mass. Anyhow several ions can appear at approximately the same mass. Two types of interferences are to be considered : isobaric and molecular. The isobaric interferences refer to chemically different atoms with the same atomic number, for example ^{241}Pu and ^{241}Am. The resolution needed to resolve the interference is high, around $\Delta M/M=100.000$, and is not achievable with our apparatus. The molecular interferences refer to interference between an atom and a molecule, for example ^{150}Nd and $^{134}Ba^{16}O$, most of them can be resolved with our SIMS. It is very important in SIMS to measure the signal coming from one isotope only, or at least to have negligible interferences.
In the case of irradiated nuclear fuel, the isobaric interference can be obtained from neutronic calculation. The molecular interferences are on the contrary unknown, that is the reason why the mass spectrum was explored with a high resolution tuning in order to evidence them. This exploration evidenced several case of interference, illustrated on figure 3 :

- The ideal situation where no interference exists was observed for a few tens of isotopes,
- A great number of mass evidences interference with molecular ions, the resolution needed to resolve the interference varies from 2000 to the maximum achievable

- ^{237}Np has a non negligible interference but the difference between the mass of the two ions is quite large corresponding to a mass resolution of ΔM/M=400.
- It was possible to resolve the isobaric interference ^{150}Nd = ^{150}Sm because it needs a relatively low mass resolution ΔM/M = 41000

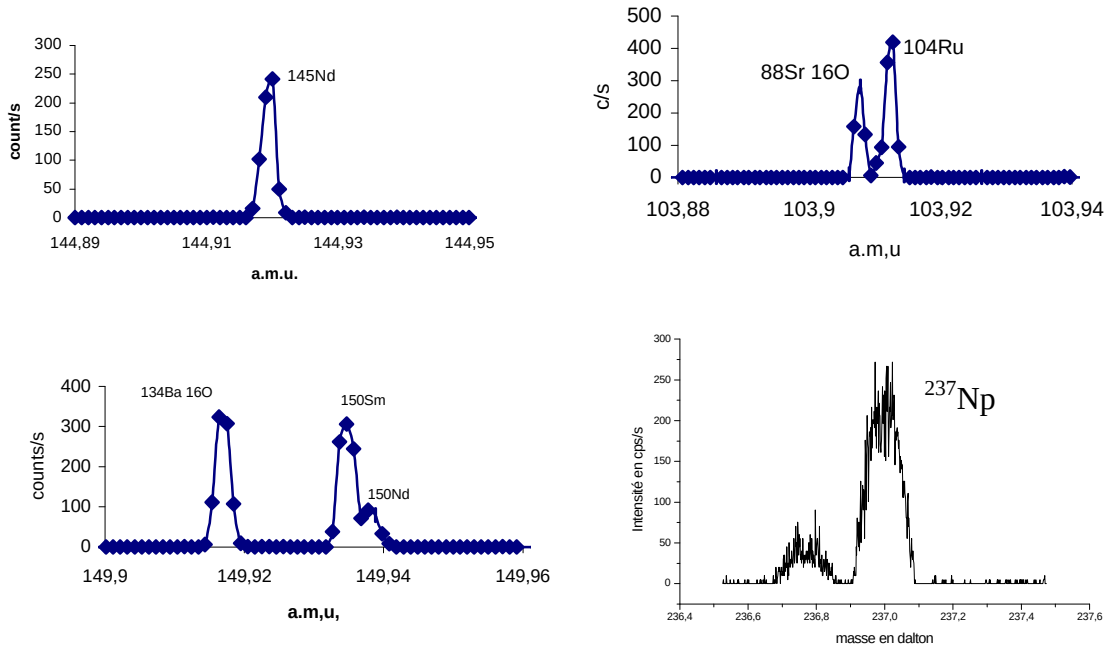

Figure3 : Somme typical high resolution mass spectra.

As a conclusion only a fraction of the isotopes coming from irradiated nuclear fuel can be used for routine analysis, i.e. with low mass resolution and higher signal. For the other isotopes with an interference, only the molecular interferences can be resolved which induces a lower signal and an analysis of poorer quality.

2°) Images

If an isotope has no interference, it is then possible to make images with a good quality because the signal is high enough.

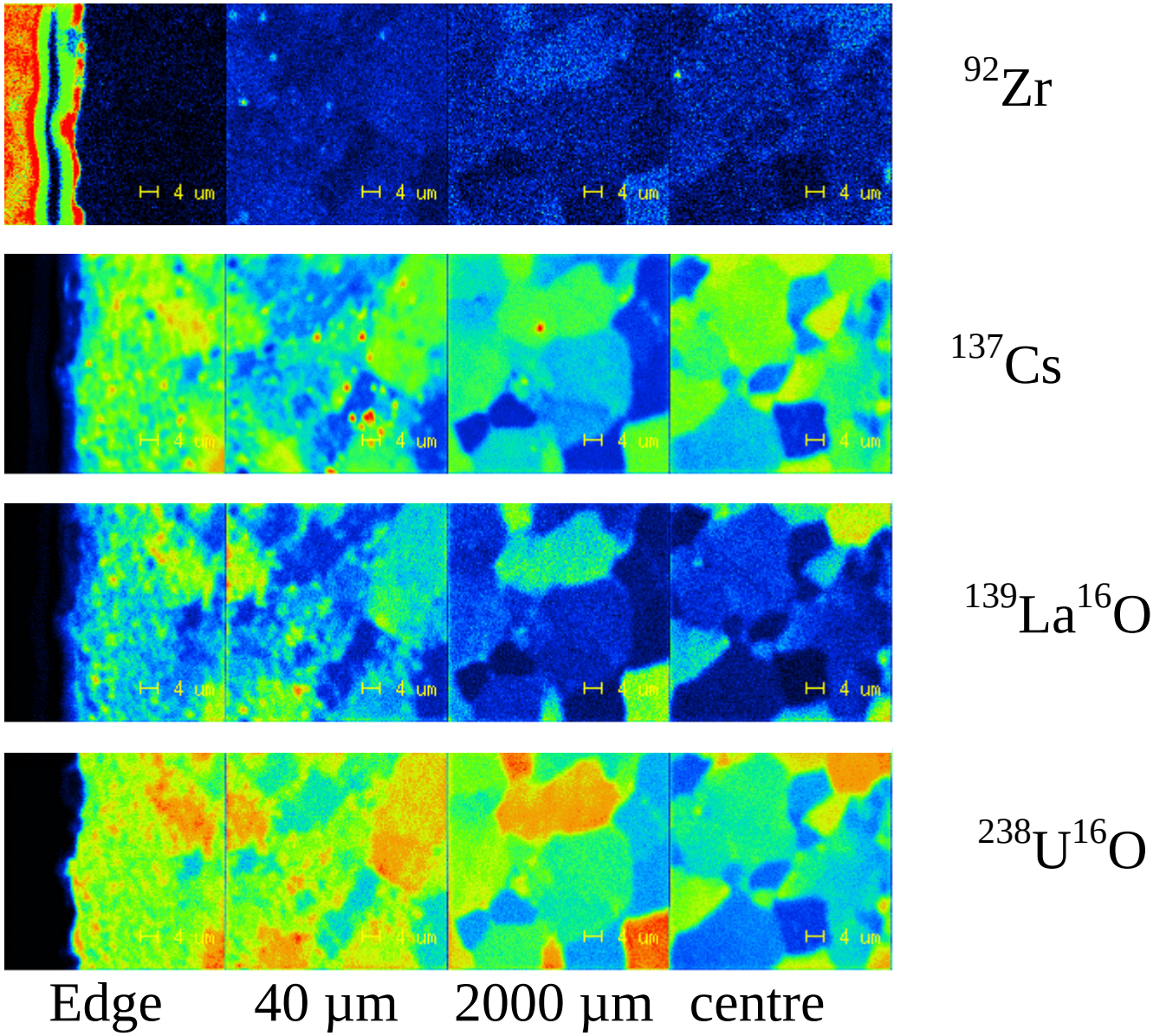

Figure 4 :Ionic images of zirconium, caesium, lanthanum and uranium (see text for details)

An example of such images is given on figure 4. On this figure a line gives the images (50x50µm²) obtained for one ion at different radius on a cross section of a UO_2 5 cycles fuel rod. The location are the same for the four ions : at the pellet edge, 40 and 2000 µm from the pellet edge and in the centre of the pellet. At the centre the grains are clearly evidenced because the secondary ionic yield depends on the crystallographic orientation, so each grain has its own yield. The structure of the grains becomes less and less visible when approaching the pellet edge because of the so called “rim effect”. Due to the higher local burn-up the grain size is lowered from 10 to 0.5 µm. At 40µm from the pellet edge, the image of 137 caesium evidences some spots which can be either precipitates or dust contamination. Further examination are in progress.

3°) Xenon detection

Xenon is a very important fission product for the understanding of the behaviour of nuclear fuel. Up to now it can not be measured with EPMA when it is located in bubbles with a diameter bigger than a fraction of µm. It was proved that xenon can be measured in UO_2 with SIMS [2] and it was shown on an 5 cycles UO_2 fuel that the xenon filling bubbles can be detected with SIMS [3].
The sample was first characterised with EMPA and some bubbles were evidenced. The sample was then sputtered with SIMS and some peaks appeared on the xenon signal. After sputtering the sample was analysed with EPMA once again, and it is clear from the xenon mapping performed before and after, that some bubbles were emptied of their gas during sputtering as shown on figure 5. This gas release is associated to the xenon peak measured with SIMS.

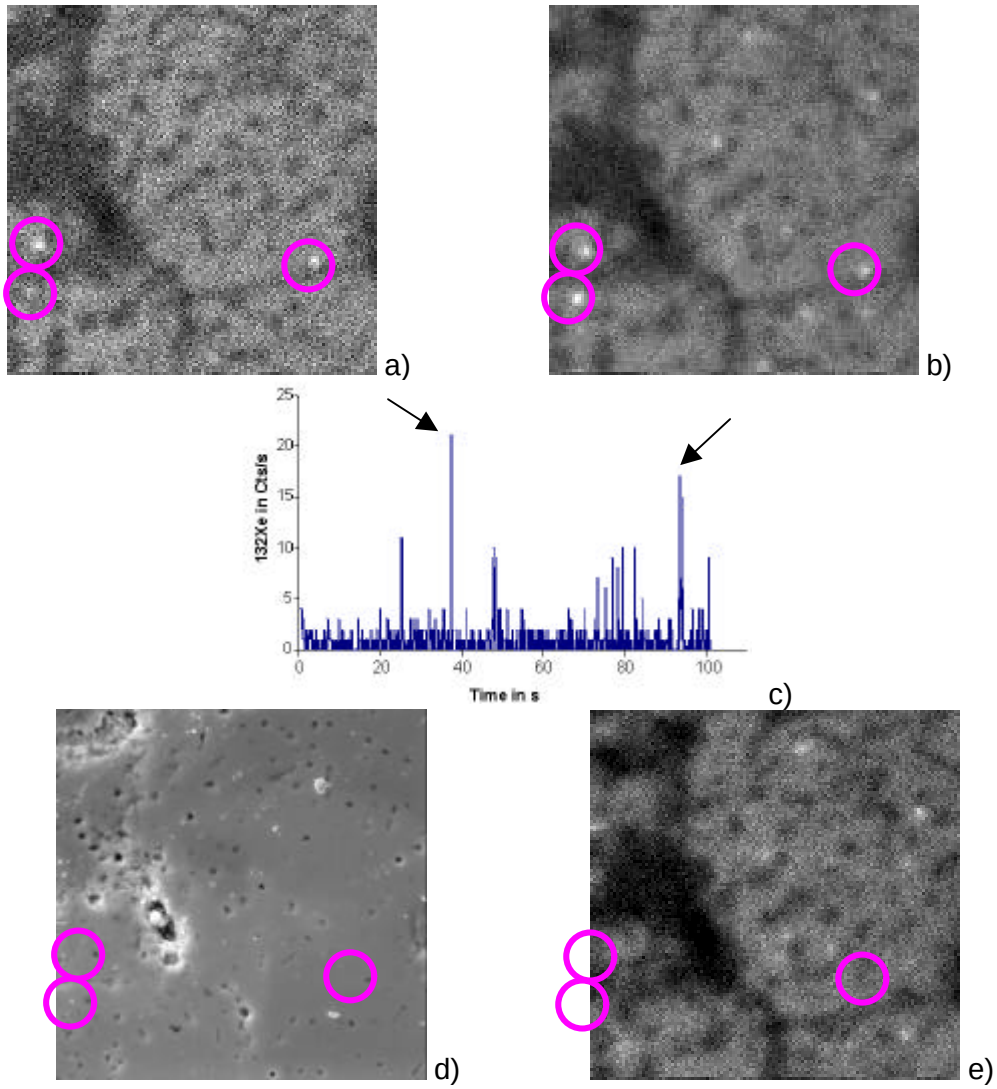

Figure 5 : EPMA mapping of xenon before (a at 15 kV and b at 30 kV) and after (e at 30 kV) and the secondary electron SEM image (d) of the SIMS ion sputtering area. The violet circles point three bubbles which lost their xenon during sputtering. The depth profile of xenon measured SIMS is shown on image c).

Conclusion

The shielded SIMS installed in CEA Cadarache is now fully operational. Its nuclear safety has been demonstrated during one year of operation without any incident. Its analytical performances were demonstrated on irradiated sample for mass resolution, image acquisition and xenon detection. Some more research is in progress to explore further its capabilities and to establish procedure for definite analysis.

[1] B. Pasquet, L. Desgranges « technical aspects of shielded SIMS installation in CEA Cadarache » Proceedings of the 39th European working group on hot labs and remote handling, Madrid, Spain, 22-24 October 2001.

[2] L. Desgranges , B. Pasquet, “Measurement of xenon in uranium dioxide (UO_2) with SIMS” accepted in Nuclear Instrument and Methods B

[3] J. LAMONTAGNE, J. NOIROT, L. DESGRANGES, Th. BLAY, B. PASQUET, I. ROURE “DETECTION OF GAS BUBBLE BY SIMS IN IRRADIATED NUCLEAR FUEL” accepted in mikrochimica acta