Technical Aspect of Shielded SIMS Installation in CEA Cadarache

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Abstract

A shielded IMS 6f has been installed in the LECA facility, CEA Cadarache France. The nuclearisation was performed by CAMECA Company, which sells the standard IMS 6f. Working on nuclear material requires in depth modifications of the apparatus itself. Despite these modifications, the shielded SIMS has the same level of performance as the standard apparatus. The design of the modified apparatus is presented and the safety aspects are emphasised.

The shielded SIMS should be allowed to handle irradiated samples at the end of 2001.

KEYWORDS: SIMS nuclear shielded

1. Introduction

The Department for Fuel Studies of CEA has decided to install a shielded SIMS in Cadarache centre. Sims analysis on highly radioactive samples is not broadly performed in the world[i]/[ii]. This device is designed as a complementary tool for the characterisation of the physical and chemical state of irradiated nuclear fuels together with SEM, EMPA, ceramography and X-ray diffraction. It will provide a local isotopic analysis with a high sensitivity, which becomes more and more necessary to fit experimental needs in R&D of nuclear industry

In this paper the nuclearisation of the shielded SIMS is presented. After a brief description of the apparatus, the constraints attached to irradiated samples are expressed, and the subsequent design is presented.

2. Standard apparatus

The CAMECA IMS 6f is a magnetic sector mass spectrometer based instrument optimized for dynamic SIMS applications, depth profiling and ion microscopy, with ppm to ppb range detection limits. Its mass spectrometer optical system arrangement offers two operating modes: microscope and microprobe.

In the microscope mode, the mass spectrometer provides a mass filtered image of the sample surface with a lateral resolution only determined by the optical properties of the secondary ion optics. The lateral resolution is 10 µm at full mass spectrometer transmission and can be better than 1µm with a transmission reduced. The microscope mode makes possible the use of large primary beam currents in order to run analyses with good detection limits and very short acquisition time.
The microprobe mode is dedicated to applications requiring sub-micron lateral resolution. Lateral resolution down to 0.2 µm can be obtained, but in this case, vibrations must be kept at low level.

The IMS 6f mass spectrometer features high mass resolution (up to 25000), which are of prime importance for resolving mass interferences in the nuclear ceramic. In order to further reduce mass interferences and to avoid sample contamination by residual gas (for example for the analysis of hydrogen in Zircalloy gladding[iii]) UHV conditions are required. The IMS 6f can reach in the sample chamber an ultimate pressure of 7×10^{-10} mbar. For keeping these conditions and for high analysis throughput, samples are introduced through a manual airlock system.

Other key features of the IMS 6f include : electron gun with unique self compensation mode for insulating material analysis, multiple primary ion species (O_2^+, O^-, Cs^+ and Ar^+), variable primary ions incident energy, full automation,...

3. Design of a shielded SIMS

Handling highly radioactive materials requires protections against radiation and against contamination. However these protections does not need to be the same in all situations, that is why two different situations were considered with the goal to analyse 4 $Sv.h^{-1}$ (measured at 5 cm from the source) samples while keeping the dose level outside of the machine at a maximum level of 25 $\mu Sv.h^{-1}$.

The protection during sample introduction and analysis is performed by a 10 cm thick lead wall surrounding the sample chamber and the introduction airlock. This will guaranty the specified dose level of 25 $\mu Sv.h^{-1}$ as soon as the distance from the sample is larger than 50 cm. For the primary and secondary optical axis of the machine, where screening is not possible, additional 10 cm thick iron wall are used. Detailed description is done in the gamma shield description §4.3.

During maintenance operation, because of irradiation, the sample is removed. However, as SIMS analysis is a destructive method, different parts of the machine could be contaminated. Most of the contamination (about 95 %) is deposited in front of the sample, on the immersion lens, but part of it can also reach the primary column diaphragm and the entrance slit of the spectrometer. These parts will thus have to be dismounted in a confined α environment. Maximum dose level for maintenance operation is 2 $mSv.h^{-1}$ at 20 cm. In typical analysis conditions with high activity samples, this corresponds to about 1500 hours of sputtering. In order to permanently control the contamination of the machine, real time monitoring of the sputtered equivalent dose is performed during analysis and instrument tuning. It uses a database with data of sample erosion rates in different analytical conditions and specific sample doses.

Special care has to be taken for protecting the different detectors of the machine. This concern the electron multiplier for quantitative measurement as well as the microchannelplate-fluorescent screen assembly used for direct imaging, the resistive anode encoder and video cameras.

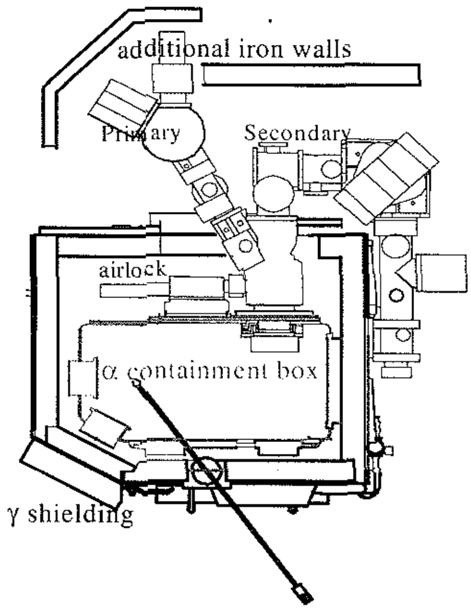

Figure 1 General synoptic of the IMS 6fR.

4. Ion microprobe nuclear adaptation

Nuclear adaptation of the ion microprobe concerns the basic machine itself and the new parts: introduction system, gamma shield and containment box. General synoptic of the instrument is shown on Figure 1 with in black the gamma shielding, in red the containment box and in blue the apparatus and introduction system.

4.1. Adaptation of the basic machine

As already emphasised, the immersion lens is the most contaminated part of the machine when analysing highly radioactive samples. It has been modified for easy and rapid non contact exchange by

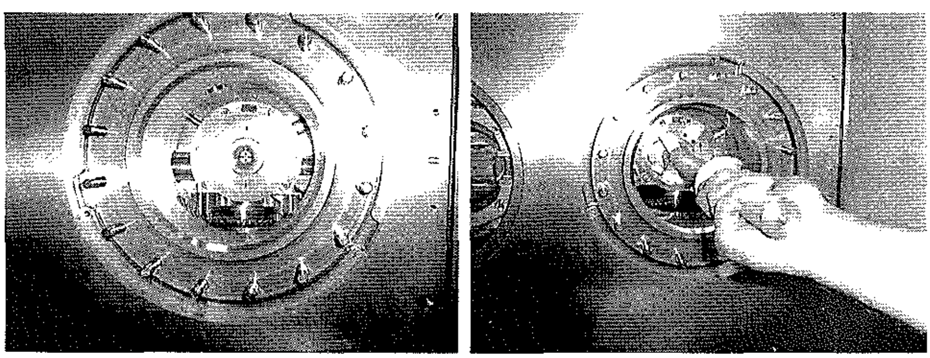

Figure 1 Exchange of the immersion lens.

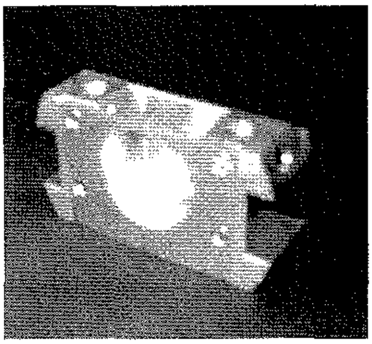

Figure 3 Sample holder

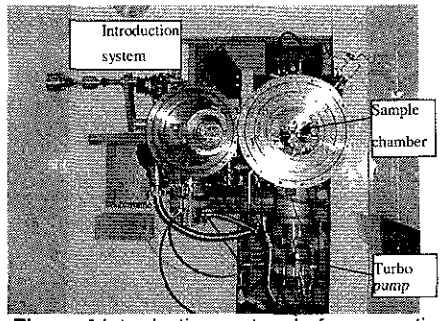

Figure 4 Introduction system before mounting of the α containment box

means of a dedicated tool as seen in the Figure 2.

As no access to the machine is possible during analysis, all the diaphragms and slits have been fully motorised. Slits and adjustments diaphragms can be set, their tuning saved and after recalled by specific alignment files. Position reproducibility is better than 5 µm.
The primary aperture diaphragm and the entrance slit assembly, which can also be contaminated by sputtered particles have been adapted for α tight dismounting. This concerns also the spectrometer isolation valve.

Other modifications have been done for the sample stage, the sample illuminator (by optical fibre), and the oxygen leak (with manual remote command).

4.2. Introduction system

The manual airlock of the IMS 6f cannot be used for highly radioactive samples. A new introduction system has been realised. It includes:

- A new airlock in which the opening/closing introduction door and the loading/unloading sample holder is done by remote manipulations.
- A motorised transfer rod with torque limitation. This ensures an automatic and precise positioning of the sample holder on the sample stage as well as security in case of incident.

The sample holder in the Figure 3 has been modified for a very safe sample mounting. It is gravitationally mounted on the transfer rod, two locating pins maintaining it in the right position. It accepts 25 mm diameter with a maximum of 13 mm thickness.

Transfer into the sample chamber is done by a combination of transfer rod and sample stage movements. Automatic procedures for sample loading/unloading control all the involved functions (transfer, vacuum, sample stage...). Manual security operation (for both the introduction rod and the sample stage) is also possible for sample recovery in case of trouble. Introduction system is shown on Figure 4.

4.3. Gamma shielding

The IMS 6fR Gamma Shielding has been designed to reduce the dose emitted by a high activity sample to an acceptable dose level for the laboratory. Its basically consists of a 10 cm thick lead cask with the components feedthroughs and openings necessary for the operation and service of the machine. The shielding is completed by additional iron walls for the primary and secondary column axis.

It is realised in several independent walls tightly assembled and fixed on a thick stainless steel base plate. When necessary, chicanery ensures an efficient radiation tightness. The lead walls are realised in molten lead. External part of the lead walls is covered by 5 to 10 mm thick steel plates, with decontaminable painting. Total mass of the Gamma Shielding is 12.4 tons.

A general view of the lead castle is shown on Figure 5.

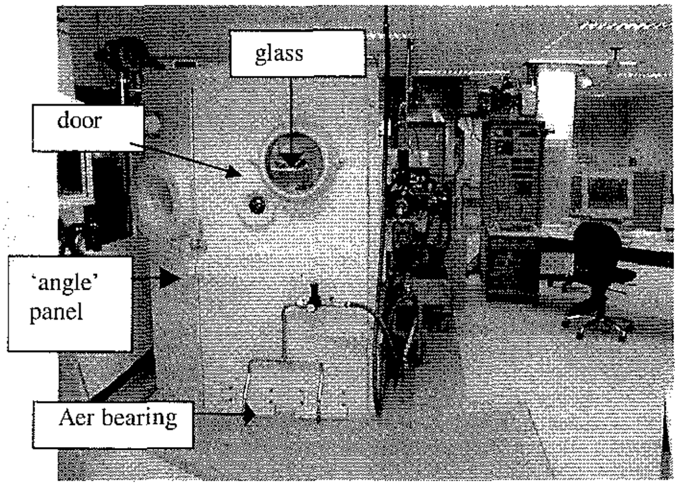

Figure 2 IMS 6fR implantation

The front door is movable on air bearings and includes a manipulator and a lead glass window (20 cm thick). The lateral and rear walls include doors and opening for accessing the different parts of the machine which could require maintenance (sample stage, spectrometer isolation valve, oxygen leak, primary diaphragms, Titanium Sublimation Pump,...). The right side wall protects the detectors (electron multiplier, microchannel plate) from the radiation. Possibility for additional shielding of the detector has been managed on that wall. The entry for sample transfer can be done either on the 'angle' panel or on the left wall. The first configuration has been chose.

A tunnel connected to an external preparation chamber is underway.

All the design is earth quake resistant up to 9.8 $m^2 \cdot s^{-1}$ horizontal acceleration.
The thickness of the Gamma Shield as well as its geometry guaranty a very efficient protection for the radiation. Assuming a 1 MeV gamma radiation with an initial dose of 4 Sv/h at 5 cm, the resultant dose at the operator current positions are:

- 2.2 μSv/h at the front door (sample manipulation operations).
- 1 μSv/h at the command desk (control of the machine and analysis position).

Those are calculated values. Precise measurements have not yet been done with a real high activity source. However, preliminary radiation leak test carried with a Cs 137 source (0.4 Gy/h) and babyline detectors have shown no detectable leaks.

4.4. Alpha containment glove box

The IMS 6fR glove box has a leak rate < 10^{-2} volume of the glove box / hour.

A general view of the containment box is shown on Figure 6. The glove box is a 5 mm thick stainless steel box. It is made of welded panels and a plexiglass transparent panel. All the angles of the glove box are rounded for easy decontamination. The bottom of the glove box has 15° inclined edges for easy recovery of sample holder or other pieces accidentally fallen in the box. The front panel (10 mm thick polycarbonate) has feedthroughs for two gloves and a manipulator. The glove box also has electrical feedthroughs for supplying the sample high voltage and sample stage motorisation. Sample introduction is done through an alpha door. A 400 mm feedthrough in a lateral panel allow the introduction or extraction of big materials like sample holder and its joint. The glove box is equipped with a gamma detector for radiation control.

The top of the glove box includes the ventilation system and a lighting system. The ventilation component is complete with comprising filters, valves and securities. It can support a flow of 30 m^3/h, i.e. a

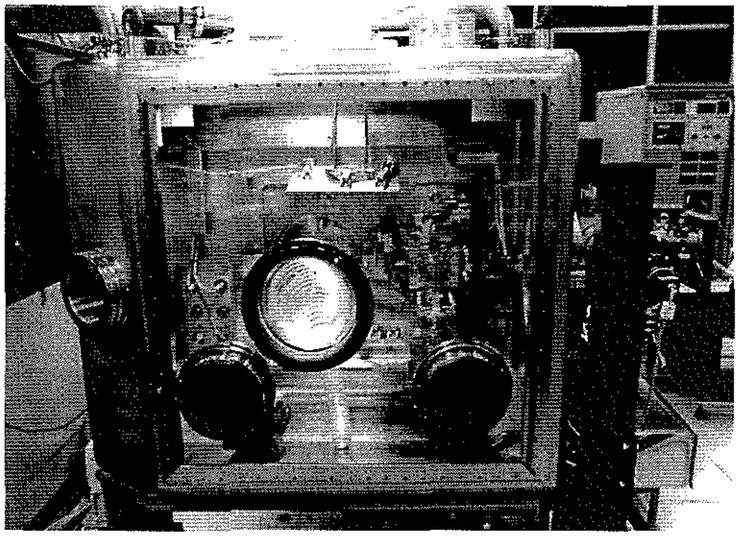

Figure 6 α containment box.

renewing rate of 60 volumes /h. This allows to use the glove box even when baking the machine. Safety valve will maintain gas flow even in case of glove breaking. An other safety valve preserve the box from implosion A flow meter and a manometer display the effective flow through the box and the depression in it. The ventilation can induce vibration on the machine. Also metallic bellows between the containment box and the apparatus (sample chamber and introduction system) has been mounted.

A set of different tools is provided for operation and servicing inside of the glove box. Some of these tools can be adapted to the manipulator when used with a sample inside the glove box or the sample chamber; others are manipulated directly with the gloves, when no sample is present in the machine and after having opened the gamma shield front door.

5. Receipts and specifications

Despite these modifications the performances of the IMS 6f were kept equivalent to those of a standard apparatus, and were evaluated during the testing period. The more significant performances obtained are:

- 7.10^{-7} bar vacuum in the sample chamber
- Mass resolution M/ΔM better than 25 000
- Detection limit of 10^{13} atoms/cm^3 of boron and 10^{14} atoms/cm^3 of phosphor in implanted silicon
- Lateral resolution of 0.2 µm with a caesium beam in scanning mode

All maintenance operations similar to real conditions of contamination were realized (exchange of the immersion lens, diaphragms...).

The accessibility of different organs has been also verified. The extraction of the titanium sublimator is shown on Figure 7.

A main part of the receipts was to verify operations in breakdown conditions. These operations are the extraction of a sample without motors or automations, the extraction of the motors in the α containment gloves box and the extraction of the door of the sample chamber in a α tight bag. Because its weight this

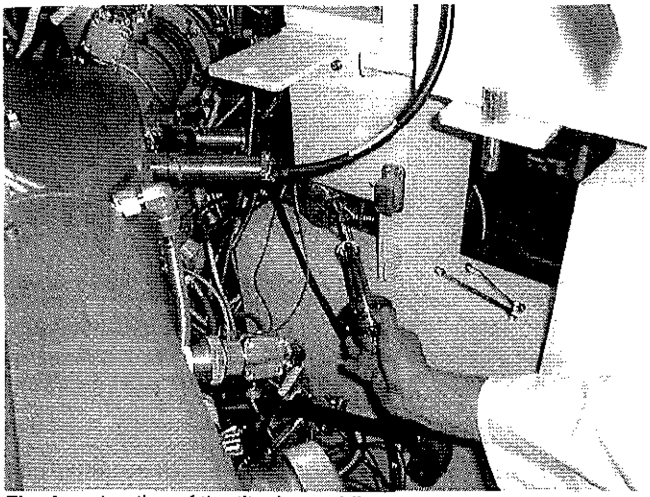

Fig. 1 : extraction of the titanium sublimator

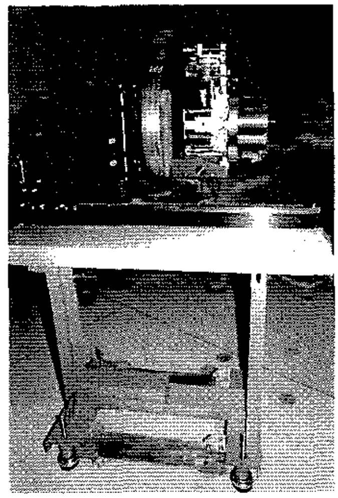

Fig. 2: table with sample chamber

last operation requires a table realise by CAMECA and shown on Figure 8.

The apparatus was installed in 1999 and has been working on unirradiated samples since May 99. The examination of nuclear samples requires to be connected with the ventilation of the laboratory and to realize a channel connected to an external preparation chamber. This has not been done when installing the IMS 6fR because the whole facility is in a renewal process, which implies that no new apparatus can be installed without a renewal of the room where it is located in. These operations are underway and we will be able to start the examination on irradiated samples at the end of the year 2001.

This delay was used as a training period in order to test the capabilities of the SIMS method on nuclear topics. Some obtained results are presented at Dimitrovgrad [iv] and shown the versatility of the apparatus.

6. Conclusions

A shielded SIMS, the IMS 6fR from CAMECA allows to handle and analyse highly radioactive samples. Its design was optimised to preserved all analytical performances. Its capabilities were tested with unirradiated samples. The shielded SIMS should be fully operational on irradiated specimens at the end of the year 2001.

7. References

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