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EUROPEAN WORKING GROUP

"HOT LABORATORIES AND REMOTE HANDLING"

PLENARY MEETING

A shielded SIMS ATOMIKA 4000

AT AEA TECHNOLOGY - WINDSCALE

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1 Introduction

For 15 years the hot-laboratory of PSI has operated a shielded ATOMIKA a-DIDA Secondary Ion Mass Spectrometer (SIMS). During this time it was used for studies of irradiated fuel and structural core components. In 1995 an ATOMIKA SIMS 4000 was acquired. This new machine improves the measurement capabilities and also allows new applications of the SIMS technique in the nuclear materials field.

2 Principle of a SIMS

As shown in figure 1, the SIMS is composed of six main components, which are :

- ☞ the primary ion gun,
- ☞ the target,
- ☞ the energy analyser,
- ☞ the mass analyser,
- ☞ the ion detector,
- ☞ the computer.

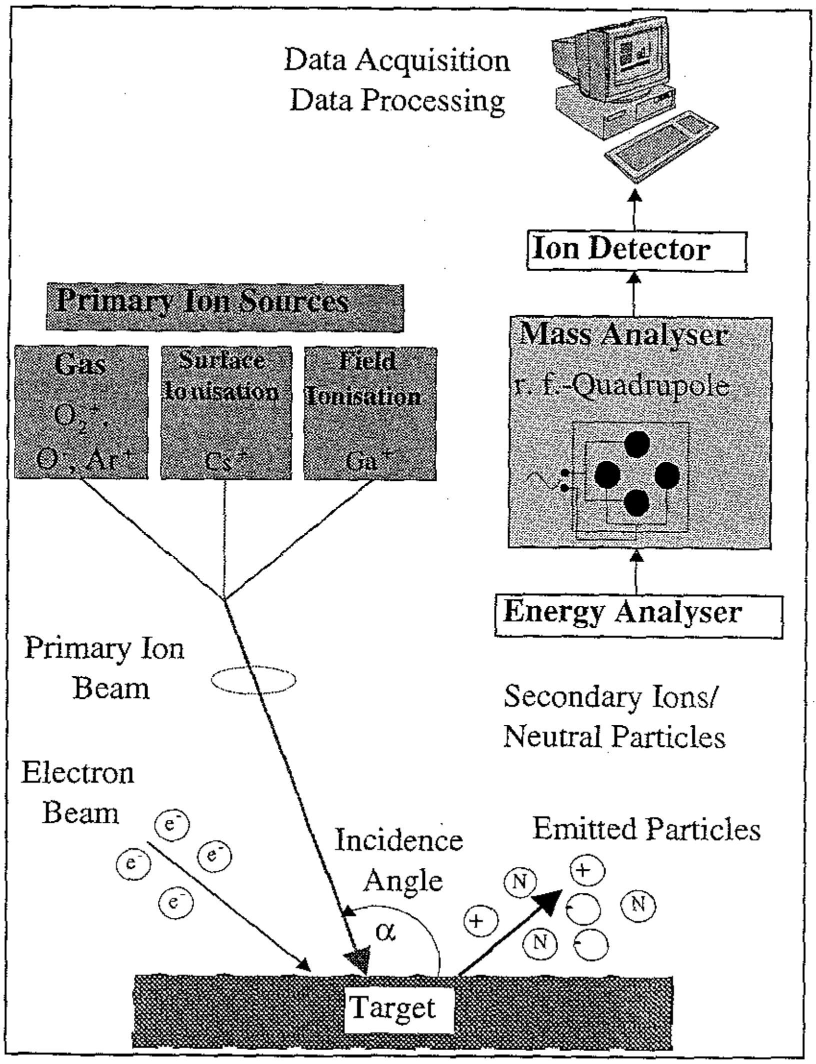

Fig. 1

secondary ions
neutral particles

The ion gun produces a primary ion beam. This beam bombards the surface of the specimen. The consequent effect is the sputtering of secondary ions from the surface specimen. The secondary ions are collected by the secondary optic which consists of the energy analyser which filters the ions of a given energy. The mass analyser make a separation of the secondary ions in function of their mass time charge ratio.

3 The shielded ATOMIKA a-DIDA SIMS

In figure 2 the ATOMIKA a-DIDA SIMS is shown.

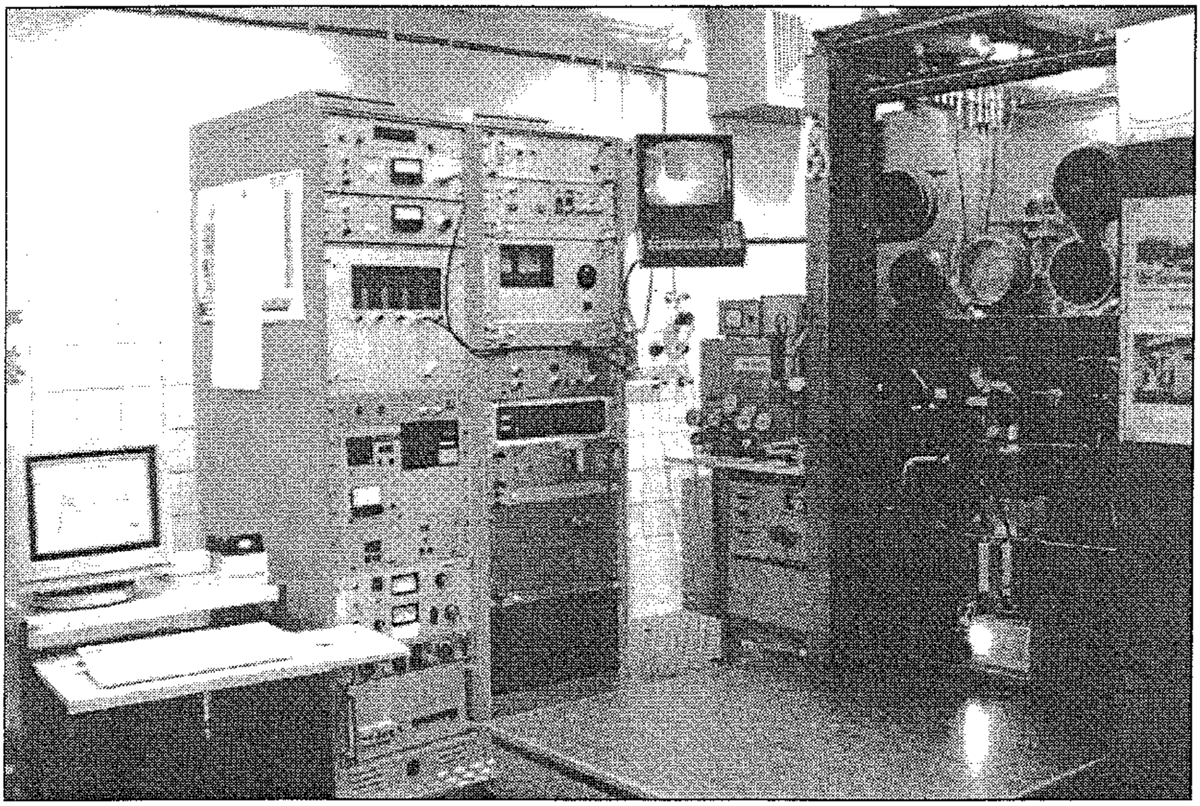

Fig. 2

3.1 The SIMS Ion Guns

The equipment was provided with a cesium (Cs) and a gas-ion source (Ar, O). Only one source can be installed at a given time. For changing the primary ion beam, about eight working hours were needed. The investigations of irradiated fuel and structural core specimens has been primarily made with the oxygen source $(O_{16})^2$.

3.2 The SIMS sample transfer

The sample is transferred in the SIMS alpha box through a double cover lock. The alpha-box is fixed above of the main- and transfer-chambers. The specimen is transferred on the SIMS pneumatic lift system with a simple bar manipulator. The lift system moves the sample down into the chamber. When a good vacuum is obtained in the transfer chamber, the specimen is moved in the main chamber with a simple manipulator operated by hand from outside the shielding. Up to three specimens can be inserted in the main chamber.

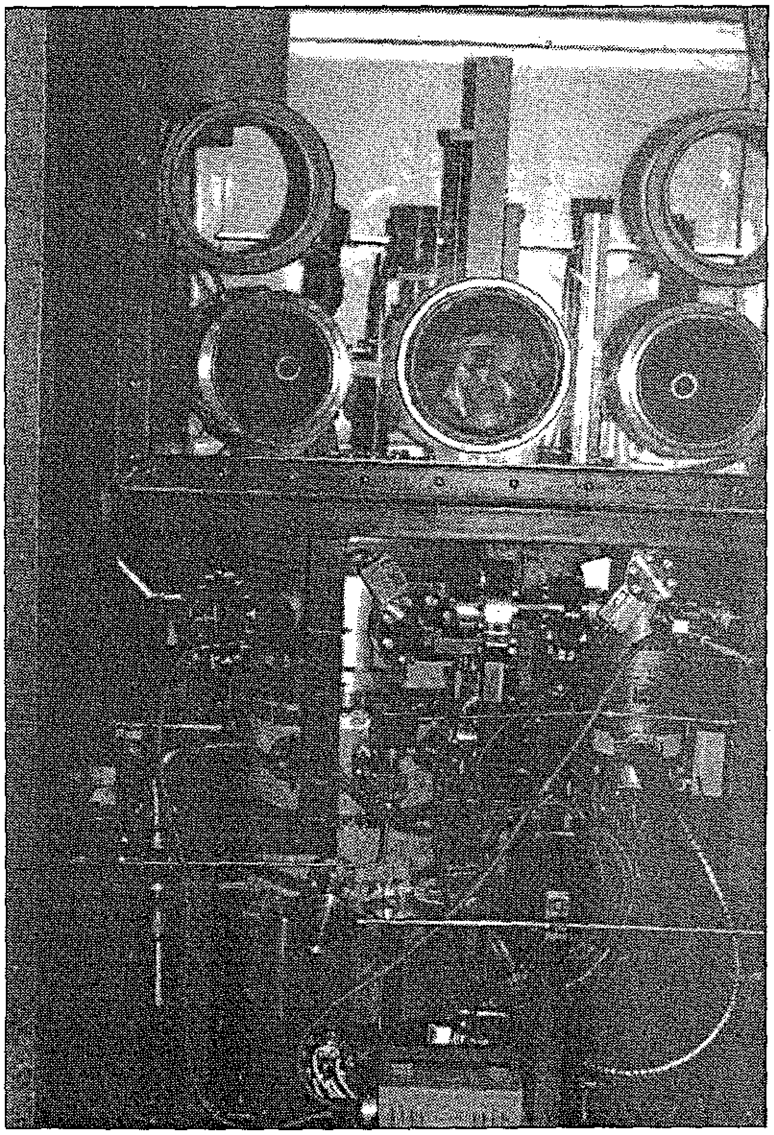

Fig. 3

ATOMIKA SIMS 4000

3.3 The kind of Scan Methods

Standard analyses were made, using the depth profiling method where information is acquired a fixed surface position in function of the depth or the mechanical linescan method. The specimen is physically moved under the primary beam to gain information along a line. The typical ion beam diameter of the oxygen source used in most of the experiments was about 50 µm, with a scan size between 100 x 100 μm^2 to 500 x 500 μm^2.

3.4 The a-DIDA SIMS upgrade

In 1993 the SIMS was upgraded. A new data acquisition software developed by ATOMIKA, running on a Macintosh computer was installed. The stepping motor control was changed to a new system (SELEKTRON), too.

With the new software the beam switch, scan width, Quadrupole Control and the sample position were controlled and supervised. The stepping motor control had an automatic- and manual-mode.

4 The ATOMIKA SIMS 4000

The shielded ATOMIKA SIMS 4000 is shown on figure 4.

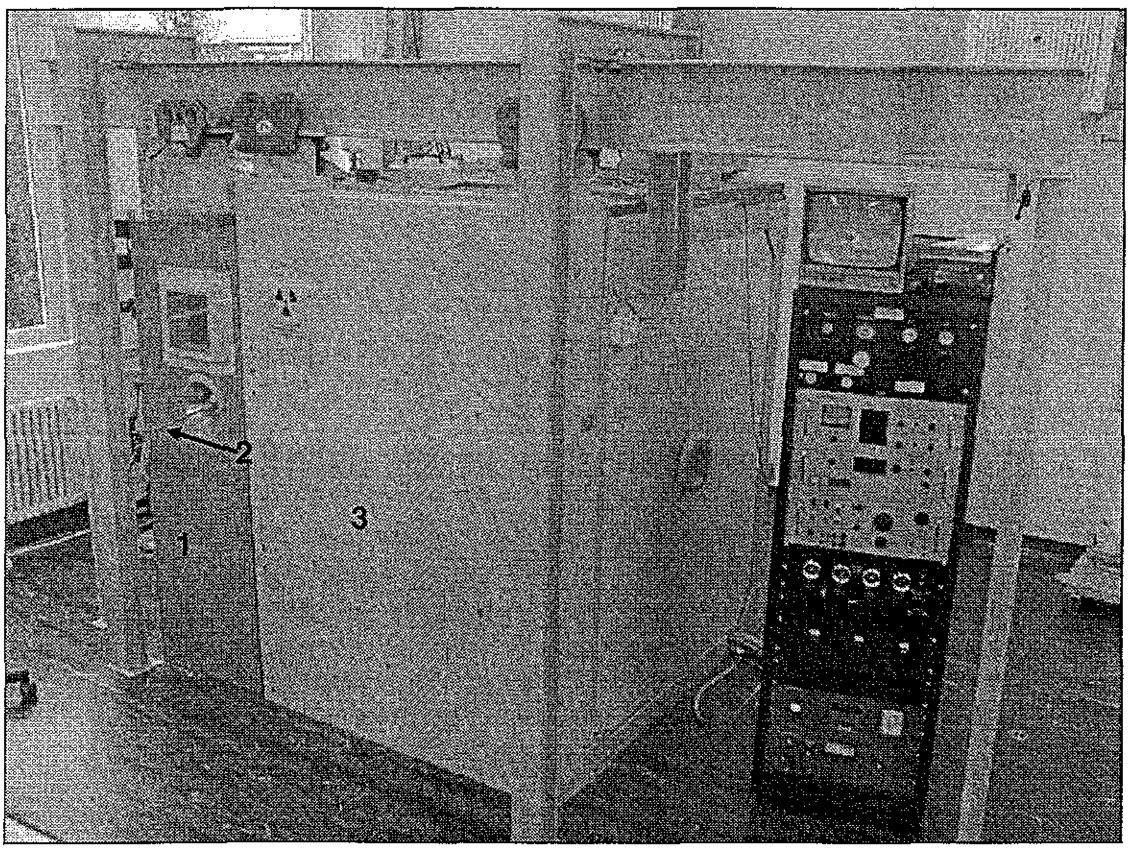

Fig. 4

22 Tons biological shielding

The ATOMIKA SIMS 4000 was delivered at the end of 1995. The instrument has been unshielded tested for about two years. The biological shielding was completed in the beginning of 1998. Since June 1998, highly radioactive specimens, including irradiated fuel pellets can be investigated with the new SIMS.

4.1 The Biological Shielding

The biological shielding is built with lead bricks. The weight of the shielding is about 22 t. When there are n #o irradiate specimens in the sample- or main-chamber the two lead doors can be opened. The main door shielding of the SIMS (figure 4, #3) is moved with electrical motors. Before moving the door (figure 4, #1) in front of the alpha box the simple bar manipulator (figure 4, #2) must be taken out.

4.2 The Ion Guns

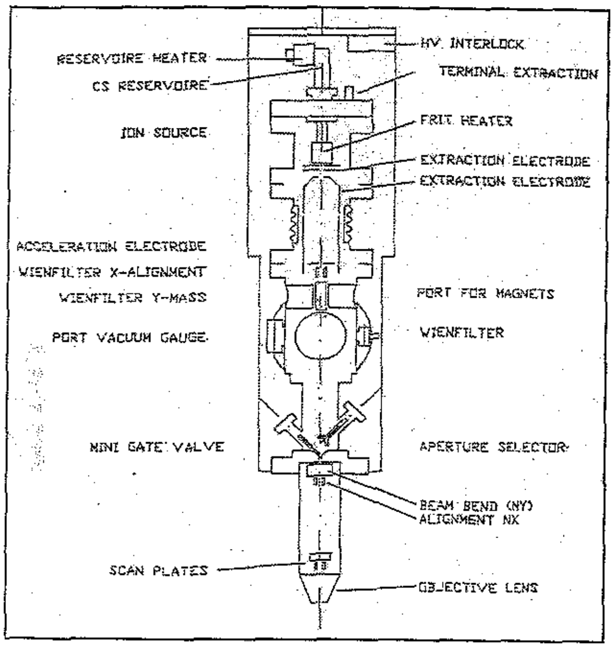

Fig. 5

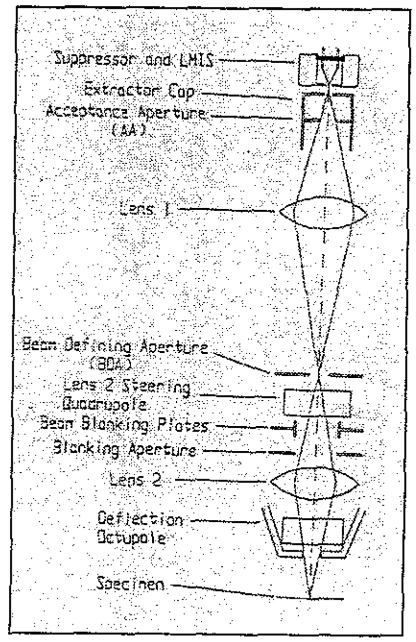

Fig. 6

A sketch of the Cs-ion-gun is shown in figure 5. This gun has no "Wienfilter". These kind of sources is belongs to the class of the surface ionisation sources. The technique is based on the evaporation of alkali-metals like caesium. Cs^+ ions are preferably used for analysing electro negative elements. The alkali-metals are evaporated from porous surfaces like for example the Frit in figure 5.

The gas source has a similar construction but contains a "Wienfilter". The beam is produced by the ionisation of a gas at low pressure. The O_2^+ ions are advantageous for analysis of electro positive elements because of the high secondary ion yield.

The diagram of the liquid metal source is shown in figure 6. The advantage of this source is the extremely high brightness and the very good focusing possibility. The ions of liquid metals (Ga^+, In^+) are extracted from a sharp needle. After then the ions are accelerated to the specimen. These kind of sources are mainly used for imaging analysis or high spatial resolution measurement.

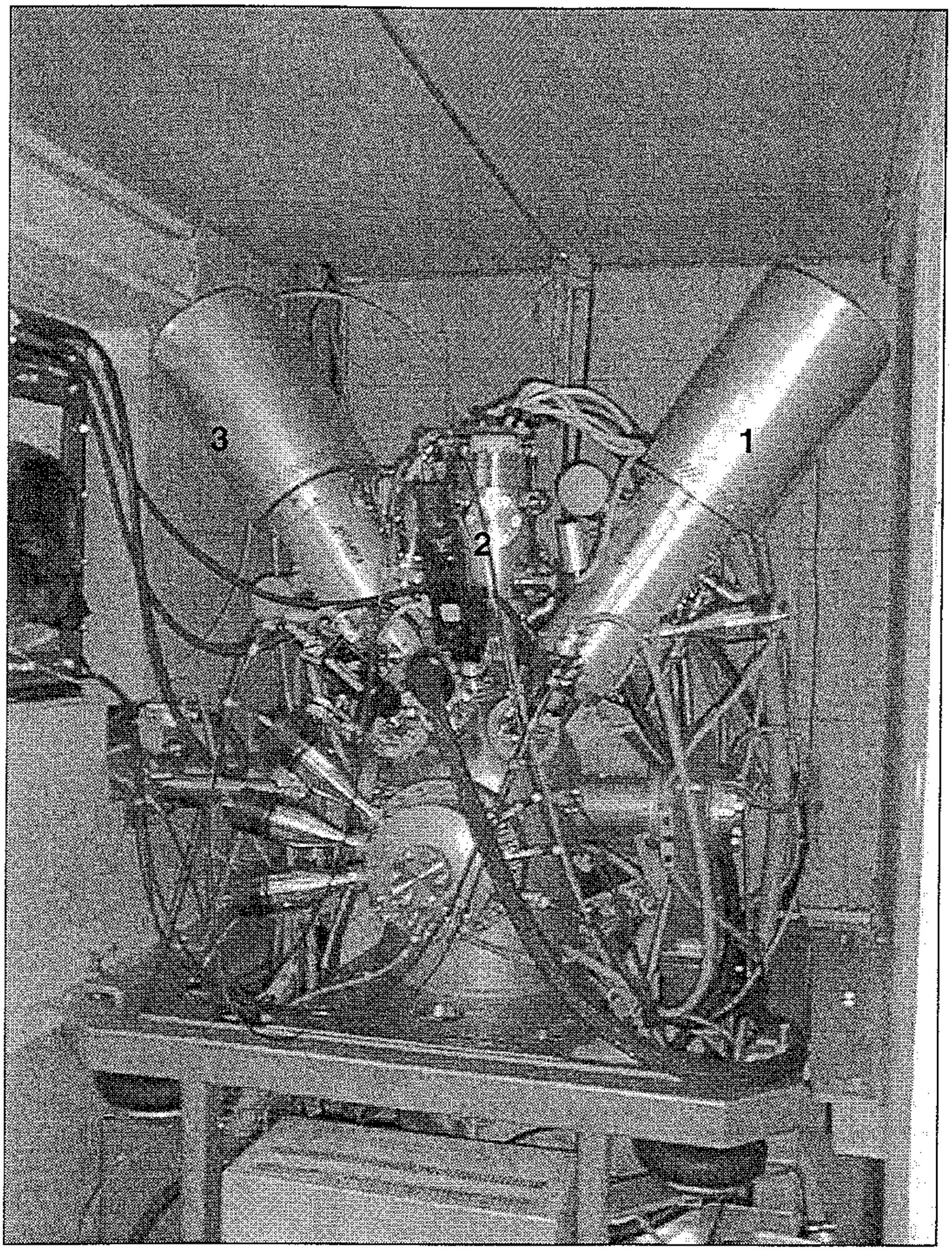

Fig. 7

4.2.1 The SIMS main chamber and sources can be seen in figure 7. The gas ion gun (right, #1), the liquid metal gun (middle, #2) and the Cs ion gun (left, #3) are shown in figure 7. For full service or repairs the main body of the apparatus can be disconnected from the loading system, the alpha-box and the control system. After, it can be moved out of the shielding.

4.3 The Sample Transfer

The sample transfer system is half automatic. Very radioactive or/and alpha contaminated specimen can be transferred from a transport shielding to the alpha-box of the SIMS using a docking station equipped with a double cover lock as shown in Fig. 8, #1.

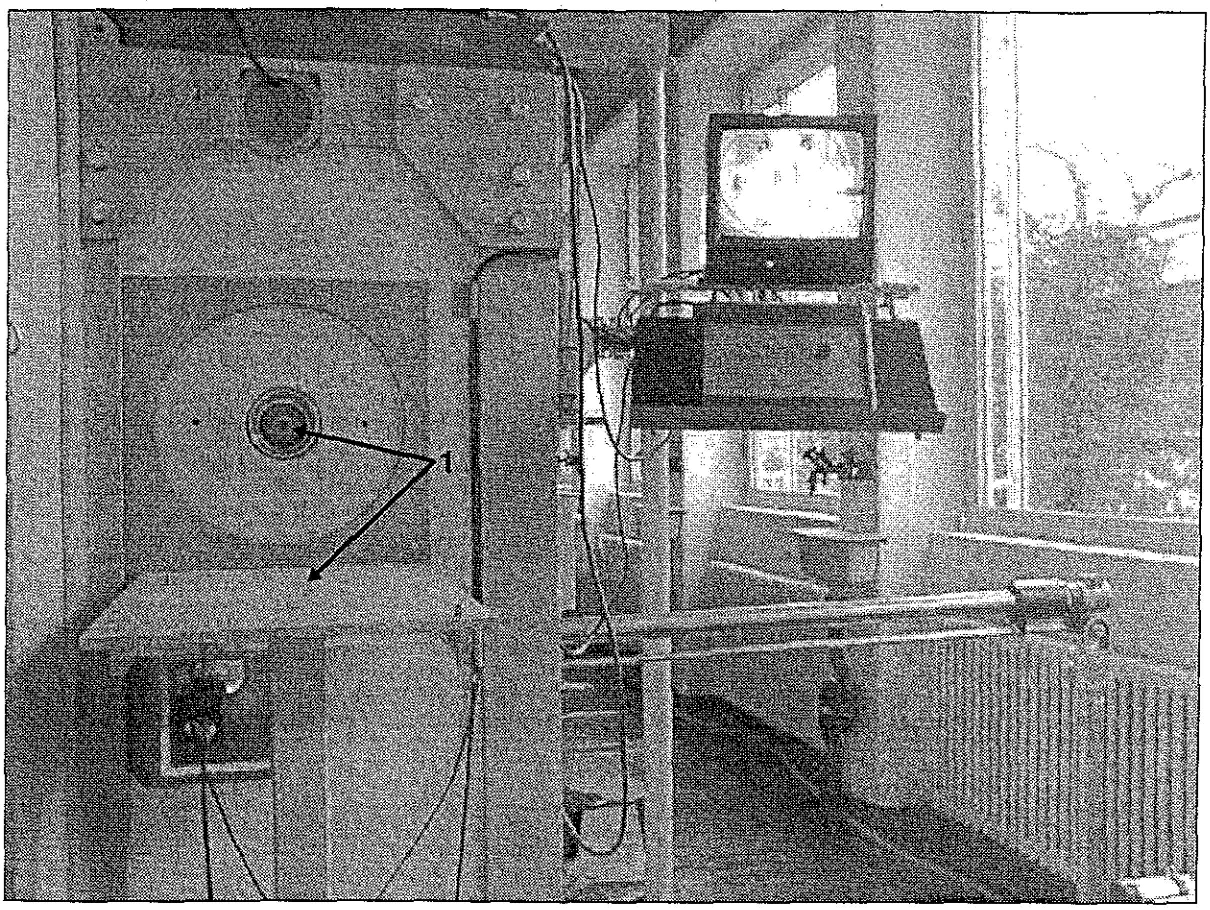

Fig. 8

The specimen is transferred inside the alpha box with a bar manipulator as shown in figure 9, #2. Then the sample is positioned on the gateway lift system (Fig. 9, #3) with the bar manipulator (Fig. 9, #4). The sample chamber gate is opened, and the pneumatically operated lift system moves the specimen into the sample chamber where it can be locked on the transfer bar fork. The lift system is then removed and the chamber closed and pumped down to a vacuum of about 8 x 10^{-8} Torr. When the nominal vacuum (8 x 10^{-8} Torr) is reached, the gateway of the main chamber is opened and the sample is transferred by hand onto the SIMS stage.

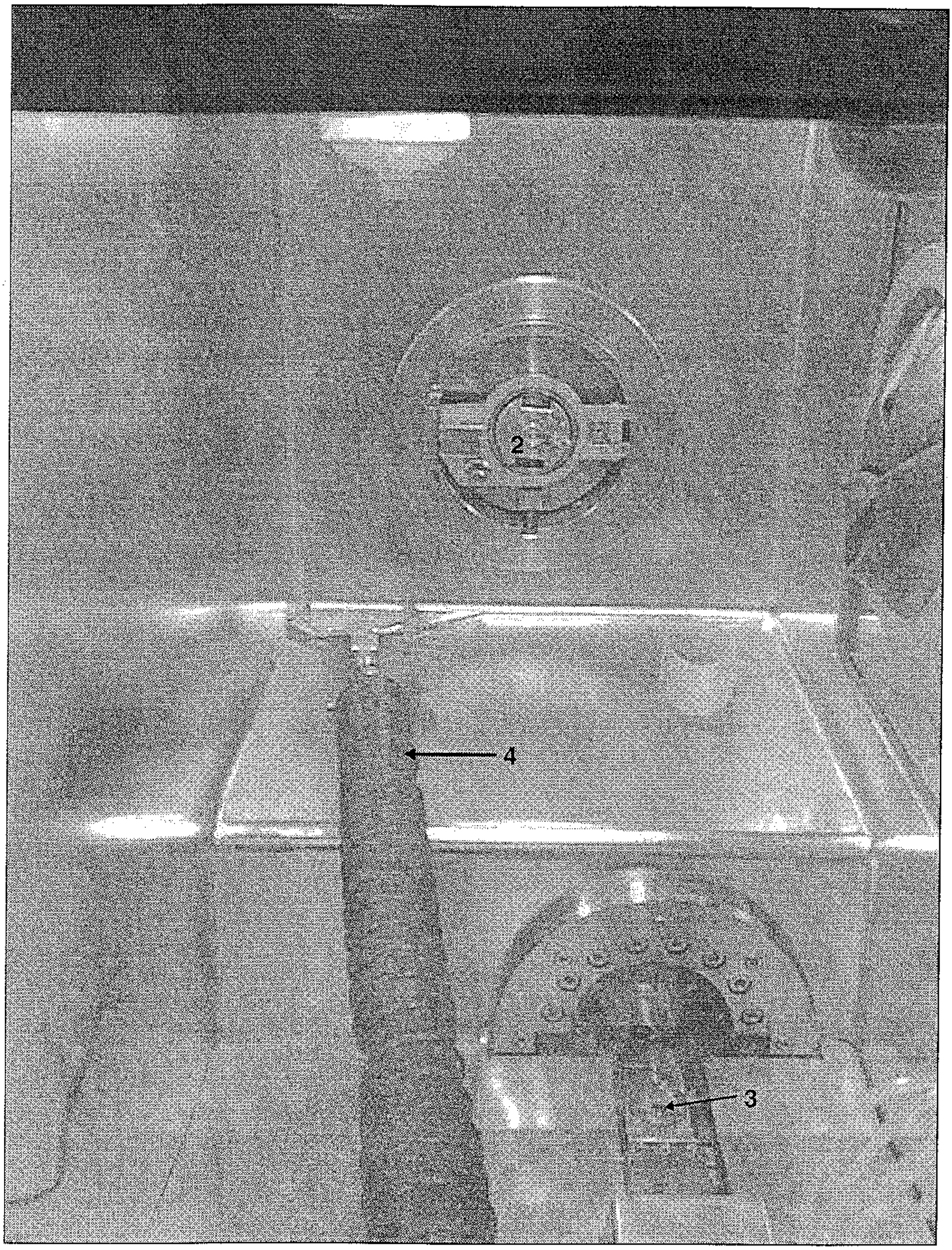

Fig. 9

4.4 The Energy Analyser

The energy analyser has two functions:

- ☞ it selects ions with a defined energy ΔE
- ☞ and separates atoms from molecules

The secondary ions have an energy range between 0 eV and 150 eV , with about 5 eV as the most probable energy. These ions are extracted from the specimen in an electrostatic field and collected in the energy analyser. The ions are filtered in a given energy band that is optimised for each measured mass.

4.5 The Quadrupole Analyser

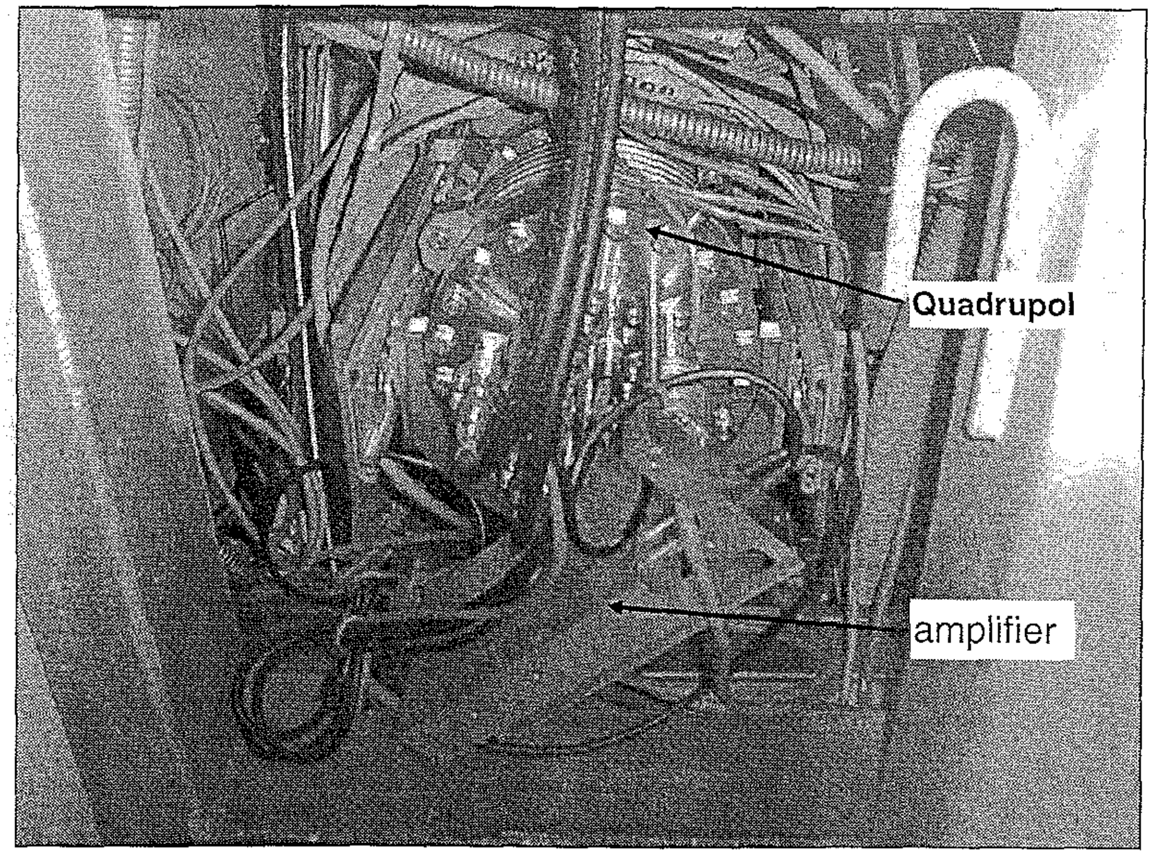

Fig. 10

The mass analyser of this instrument is a quadrupole system. The mass resolution of this system is in the order of 0.5 amu. The mass range of the quadrupole analyser is from 1 to 400 amu. The quadrupole analyser and the amplifier on the back of the SIMS is shown in figure 10.

4.6 The motorised stage

The ATOMIKA SIMS 4000 motorised stage has three movement axis. These axis are:

- ☞ translational axis
- ☞ rotational axis
- ☞ tilt axis

The tilt axis is used for the adjustment of the angle between the specimen surface plane and the primary ion beam. The stage displacement is defined in a polar system centred on the centre of the specimen holder (rotation and translation). The three motors for the movement are shown in figure 11.

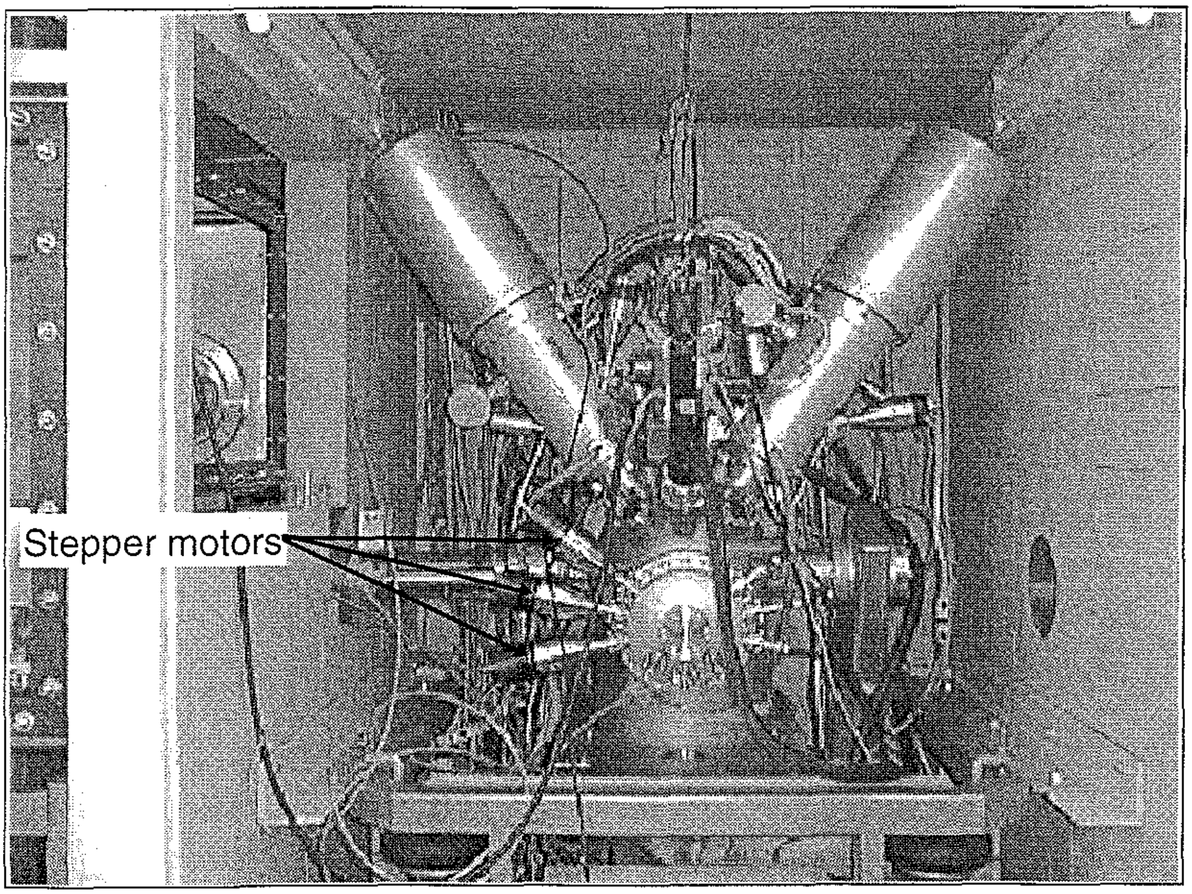

Fig. 11

4.7 The Geometry of Specimens

Specimen surface up to a diameter of 10 mm can be investigated with the instrument. Up to 5 specimens can be mounted simultaneously in a specimen holder. Special holders can be created for non standard specimen shapes. Typical holders are presented in figure 12 and figure 12.1.

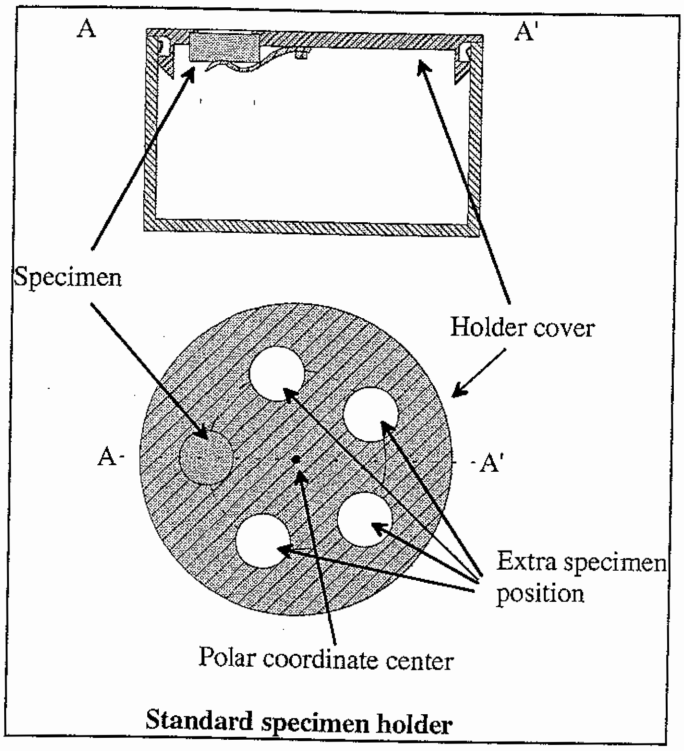

Fig. 12

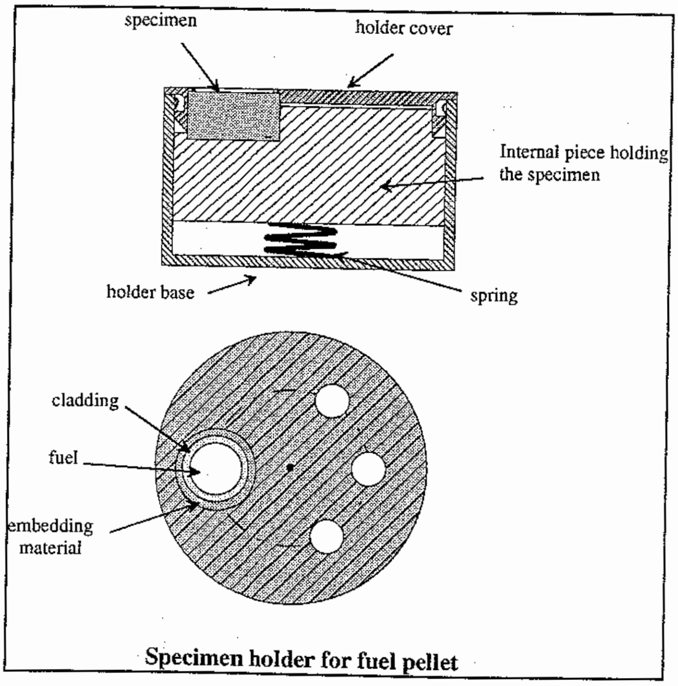

Fig. 12.1

4.8 The Secondary Electron Detector

The secondary electron detector detects secondary electrons which are produced by the interaction of the primary ion beam with the specimen surface. The detector is mainly used for the adjustment of the liquid metal source.

4.9 The Vacuum of the Equipment

For high resolution analysis, a high vacuum quality in the main chamber is needed ($\leq 10^{-9}$ Torr).

The SIMS system is dived in four vacuum zones that can be isolated one from the other. There are the:

- main chamber
- sample chamber
- cesium source
- gas source

✳ The vacuum is produced with seven pumps. The gas source is attached to a turbomolecular pump and a rotary pump. The sample chamber is attached to a turbomolecular pump and the same rotary pump as the gas source. The cesium source is attached to a turbomolecular and a rotary pump. The liquid metal source has no pump of its own but is directly connected to the main chamber. The ultra high vacuum in the main chamber is obtained with an ion pump titan and a sublimation pump. The sample chamber, used for specimen transfer, is pumped with a turbomolecular pump and a rotary pump.

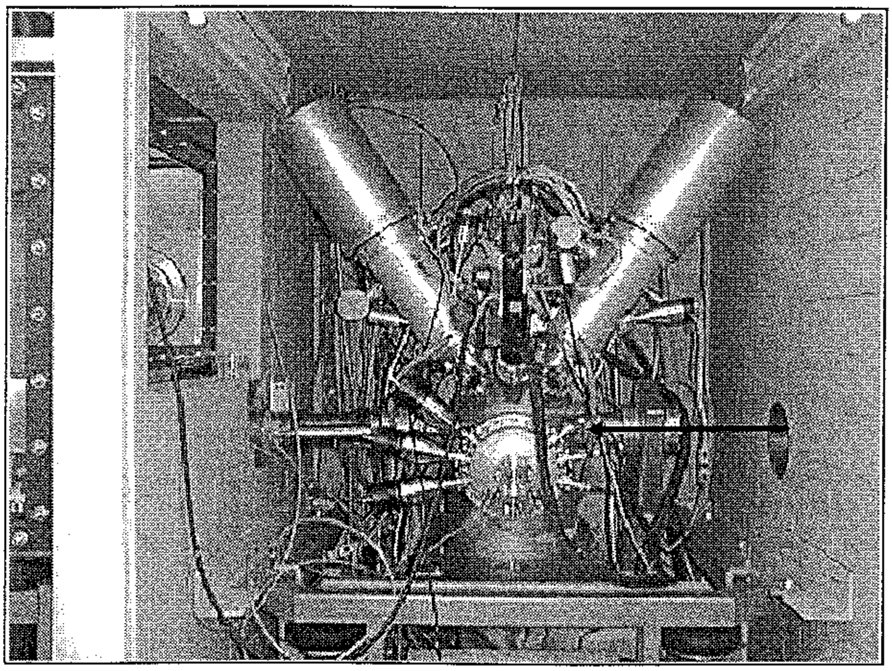

Fig. 13

4.10 Help when an irradiated sample is lost

In the case of a highly radioactive sample falling in the analyse chamber due to of a wrong manipulation or a miss-function, a tool has been developed for the remote handling of the specimen.

The system can be introduced in the main chamber after the vacuum has been broken and a flange of the biological shielding removed. The introduction path into the main chamber is sketched in figure 13.

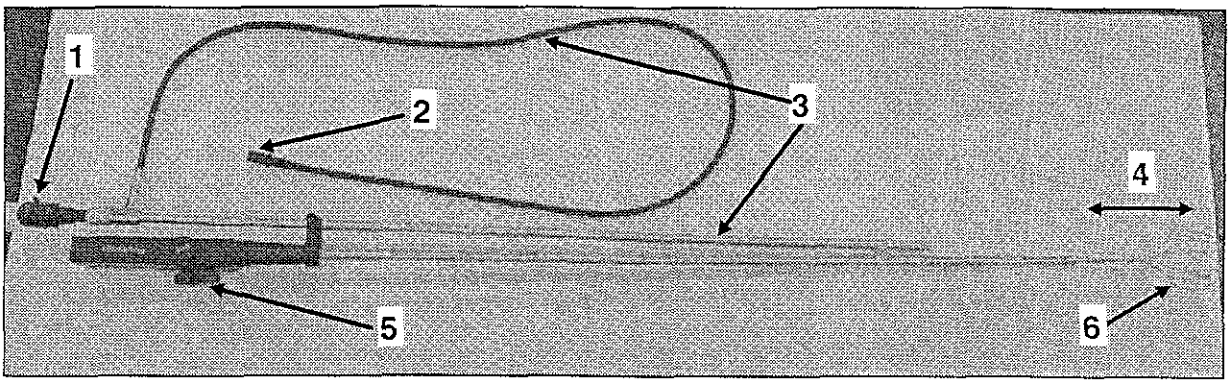

Fig.14

The help tool is presented on figure 14. It is based on a borescope containing a video camera (Fig. 14, 1) and a light conductor (Fig. 14, 3). A light source can be attached to the plug (Fig. 14, 2) and the interior of the SIMS chamber can be inspected. When the specimen is found, it can be caught with a mobile claw (Fig. 14, 4) mounted on the borescope itself and controlled manually (Fig. 14, 5) from outside the biological shielding using a knob. The claw can be moved in all directions up to an angle of 90° from the main axis of the borescope.

4.11 Some examples of results

With SIMS, it is possible to obtain local isotopic information's, in contrast with x-ray spectrometry (EPMA), where only element concentration can be obtained. Important for the analysis is to know on which position the rod was placed in the reactor. In figure 15 the relative plutonium isotope distributions across a high burnup UO_2 pellet (A) and MOX (B) are plotted. The strong Pu enrichment in the rim demonstrates the burnup increase in this region. The influence of the neutron spectra is clearly demonstrated by the different distribution of the Pu isotopes in UO_2 and MOX

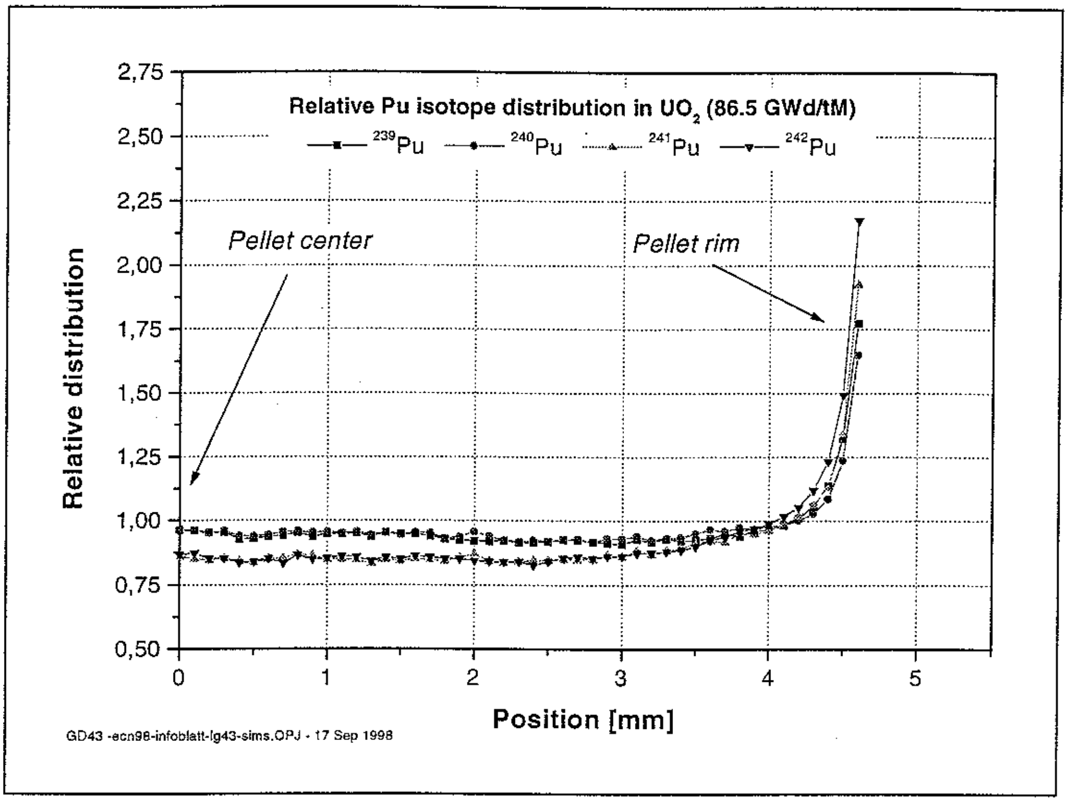

Fig. 15 (A)

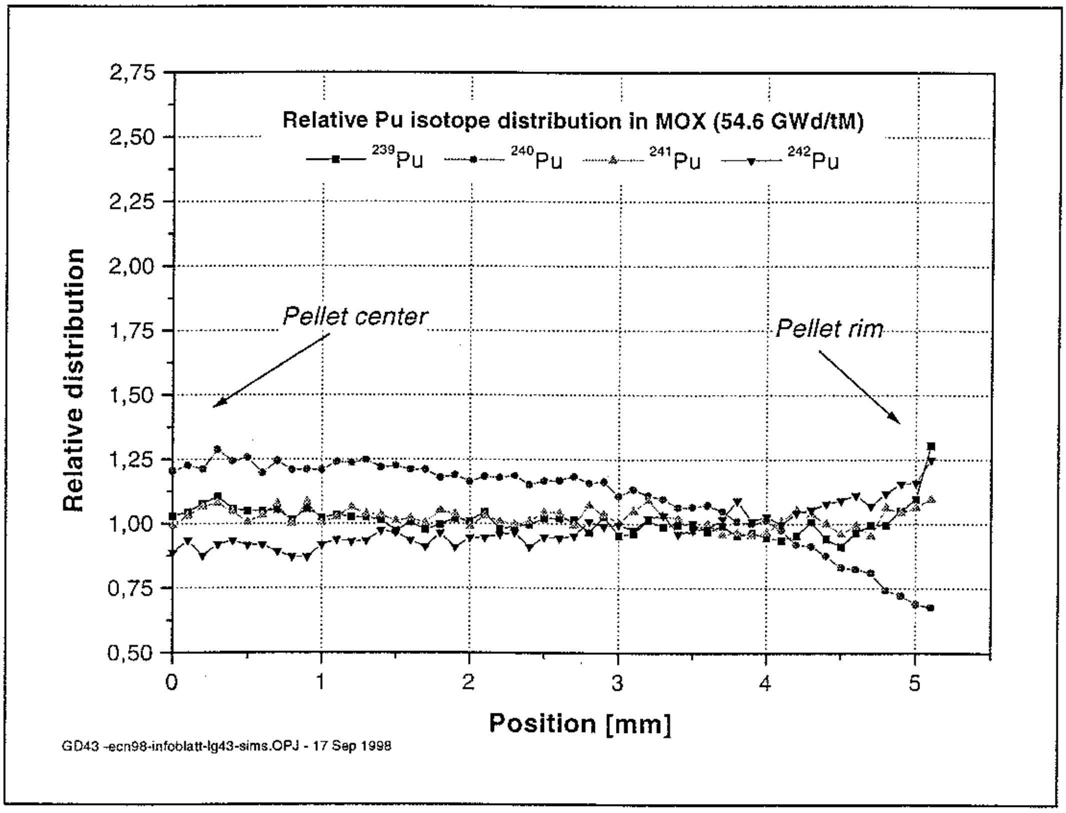

Fig. 15 (B)

The new SIMS capabilities are clearly shown on the two next examples. The high spatial resolution of the instrument has been used to determine the ^{7}Li and ^{11}B in the oxide layer of an in pile irradiated zircaloy cladding. Due to the large thickness at the oxide layer, the standard depth profiling technique is no more useable. The line scan has been performed with the Ga-source on across section at the cladding as seen on figure 16 (A). A resolution better than 1 µm has been obtained as demonstrated on figure 16 (B).

Primary ion : Ga, Ion energy: 25 KeV, Beam size : 0.1 µm Øs, step size : 0.1 µm

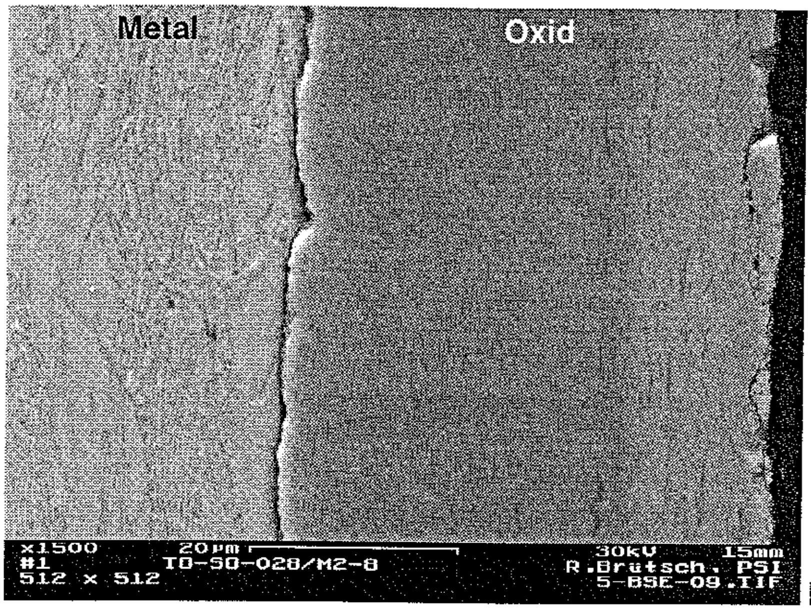

Fig. 16 (A)

inward clad. corrosion

Scanning Electron Image (SE) of the analysed specimen

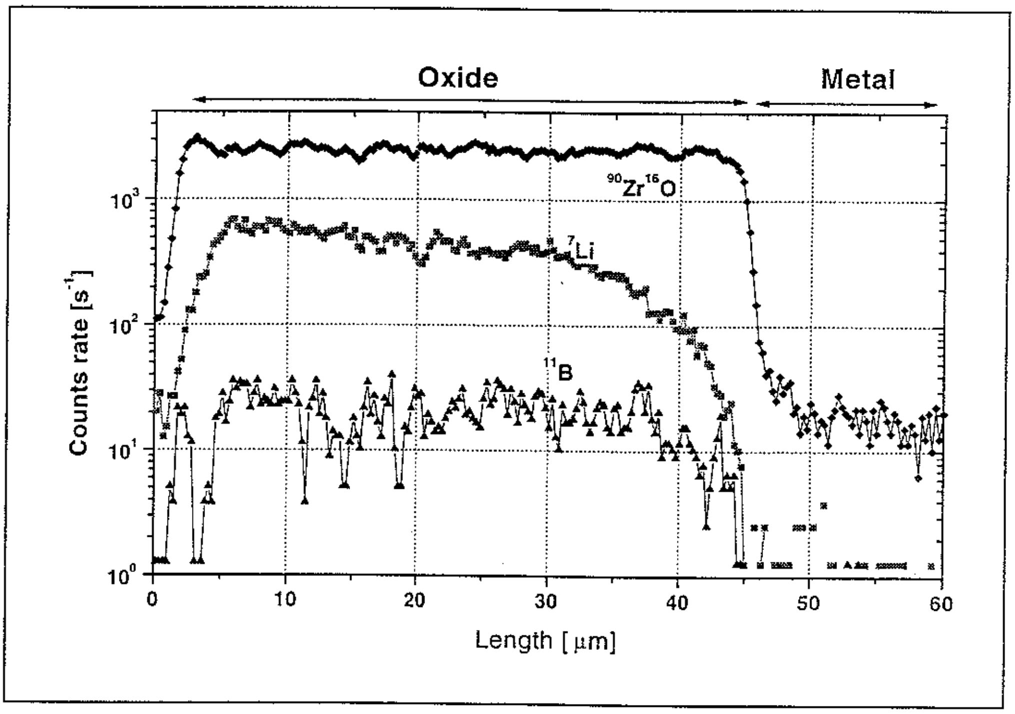

Fig. 16 (B)

The imaging capabilities has been used to determine the ^{10}B distribution on an irradiated $B_{8.5}C$ specimen with an approximate burn-up of 52%. A cross section of the $B_{8.5}C$ (Fig. 17 (A)) specimen has been analysed by scanning the specimen surface with a Cs beam. The results, presented in figure 17 (B), show the burnup of ^{10}B in the periphery of the specimen.

Primary ion: Cs, Ion energy: 12 KeV, Beam size: 30 µm ∅, Scan size: 50 µm x 50 µm, Step size : 50 µm

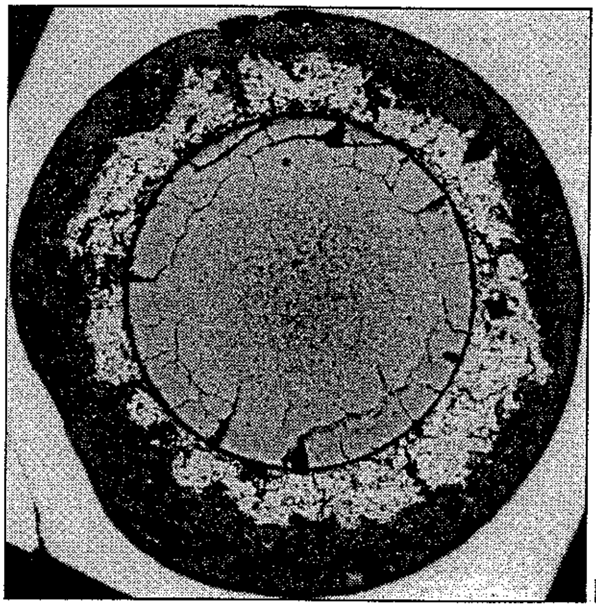

Fig. 17 (A)

Macro-image of the specimen

Bo 10 distribution

* Cs ion beam scanning

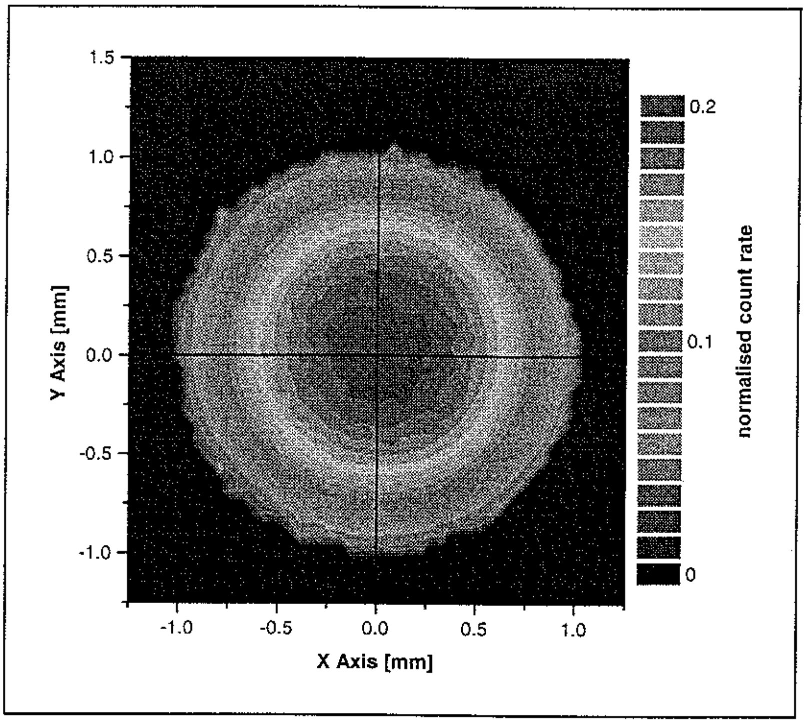

Fig. 17 (B)