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**EUROPEAN WORKING GROUP "HOT LABORATORIES
AND REMOTE HANDLING"**

21-23 September 1998

Windscale UK

THE NUCLEAR MICROPROBE AT THE PIERRE SÛE LABORATORY

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Commissariat à l'Énergie Atomique
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ABSTRACT

The nuclear microprobe of the Pierre Süe Laboratory, located on the Nuclear Center of Saclay is operational since 1993. It can perform highly sensitive analyses based on the use of atomic and nuclear reactions induced by light charged particles (proton, deuteron, helium-3 or helium-4) in the energy range of 0.3 to 3.8 MeV. It provides a efficient means to determine the isotopes of the light elements (H to Si) by using nuclear reactions. Simultaneously, heavier elements (Na to U), are measured by X-ray emission with an accuracy of few wt ppm.

With a beam size resolution of ($1 \times 3 \mu\text{m}^2$), it is also used for non-destructive recording of concentration profiles. It can also map the microscopic distribution of the elements within the regions of interest. Volume analysis can be performed by nuclear resonant reactions.

The beam is analyzed by a magnetic dipole giving two beam lines. The line at 90°, is used for multi purpose studies on non radioactive samples for Material Science (oxidation processes under constraints, impurities in grain boundaries...) and Earth Science (analysis of melt and fluid inclusions in minerals or meteorites). The second beam line, at 45°, is devoted to radioactive sample studies.

The scattering chambers are equipped with solid state detectors, for RBS NRA and ERDA measurements, and an X-ray detector (WDS or EDS). An automatic scanning with minimum step of 0.3 μm is in operation for profiling and mapping.

Radioactive samples are embedded, cut and polished in the EPMA preparation line of Saclay hot laboratories, and sent to Pierre Süe building. The beam line for radioactive samples of this laboratory includes a reception cell, a transfer cell and the shielded hot cell for examination. The d its first analyses are on a highly active, 5-cycle PWR sample by December 1997. The primary goal is the measurement of lithium concentration in the zirconia layer on Zy cladding. The results will be compared to those obtained on a shielded SIMS. In 1998, the analyses will include other oxidized Zircaloy claddings and also irradiated fuel.

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INTRODUCTION

The Pierre Süe Laboratory (LPS) founded in 1970 by *the National Scientific Research Center (CNRS)* and by *the Commission of Atomic Energy (CEA)* had as first vocation the analysis of trace elements within the framework of material studies and of geological research.

By the end of 1980, a growing interest for studies at the scale of the micrometer guided the LPS towards the acquisition of a nuclear microprobe [1]. Operational since 1993, this facility is designed not only for the microscopic study of geological samples, metals or insulators, but also for radioactive samples analysis.

A specific beam line for these samples is operating since December 1997.

MICRO BEAM ANALYSIS

Ion beams of energy ranging between 100 keV and 10 MeV have been extensively used in the last thirty years to study multi-layers materials, profiles of concentration or to seek trace elements in various samples [2-3]. Its great interest comes from the weak disturbance introduced by the beam at the impact point, allowing other analyzes at the same point by complementary techniques. In addition this method examines the matter at depths of several micrometers, making possible the study of small buried material in different states : like solids, liquids and gases.

This method is based on the processes of three types of interactions:

The most probable interaction is the X-ray. When a beam of charged particles interacts with the electronic cloud it can pull out an inner shell electron leaving the atom in an excited state. This atom decays to its ground state by emitting X-ray, characteristic of the atomic number of the element. All the elements from sodium to uranium can be identified by this so called PIXE method (Particles Induced Xray Emission), using proton or helium beams of energy ranging between 2 and 4 MeV.

The elastic scattering of the incident ion on a nucleus of the sample is a second source of information. The energy of the scattered particle is characterized by the ratio of the masses of projectile and the target. In this case, the components of the target are identified by the energy measurement of the scattered particle. This method is applied in the studies of thin layers (10-1000 nm) and of profile along the beam direction. It is also one of the methods to analyze hydrogen.

For incident energies above the Coulomb barrier, nuclear reactions can occur. The detection of the emitted particles or γ -ray allows an isotopic analysis of the target for elements ranging between deuterium and silicon. Among these nuclear interactions, the resonant reactions allow the measurement of concentration profiles with an accuracy of 10 to 100 nm depending on the width of the resonance.

Simultaneous measurements of these kinds of reactions induced by ion beams are very useful for a full analysis of the sample at the impact point.

The development of micro beam is a progress in this technique allowing not only the detection of trace elements with a very great sensitivity (down to 1 ppm) but also a spatial

distribution at the μm level. This last parameter depends entirely on the size of the beam at the impact point.

The field of interest is very wide. We can include astrophysics, biology, mineralogy, medical research, metallurgy and micro-electronics.

NUCLEAR MICROPROBE DESCRIPTION

The beam is provided by a single stage VAN DE GRAAFF accelerator (figure 1), equipped with an RF source to produce sufficiently intense beam. Energy is obtained by adjustment of the high voltage of the terminal electrode between 0.300 and 3.8 MV. This energy range covers almost all the nuclear reactions of common interest.

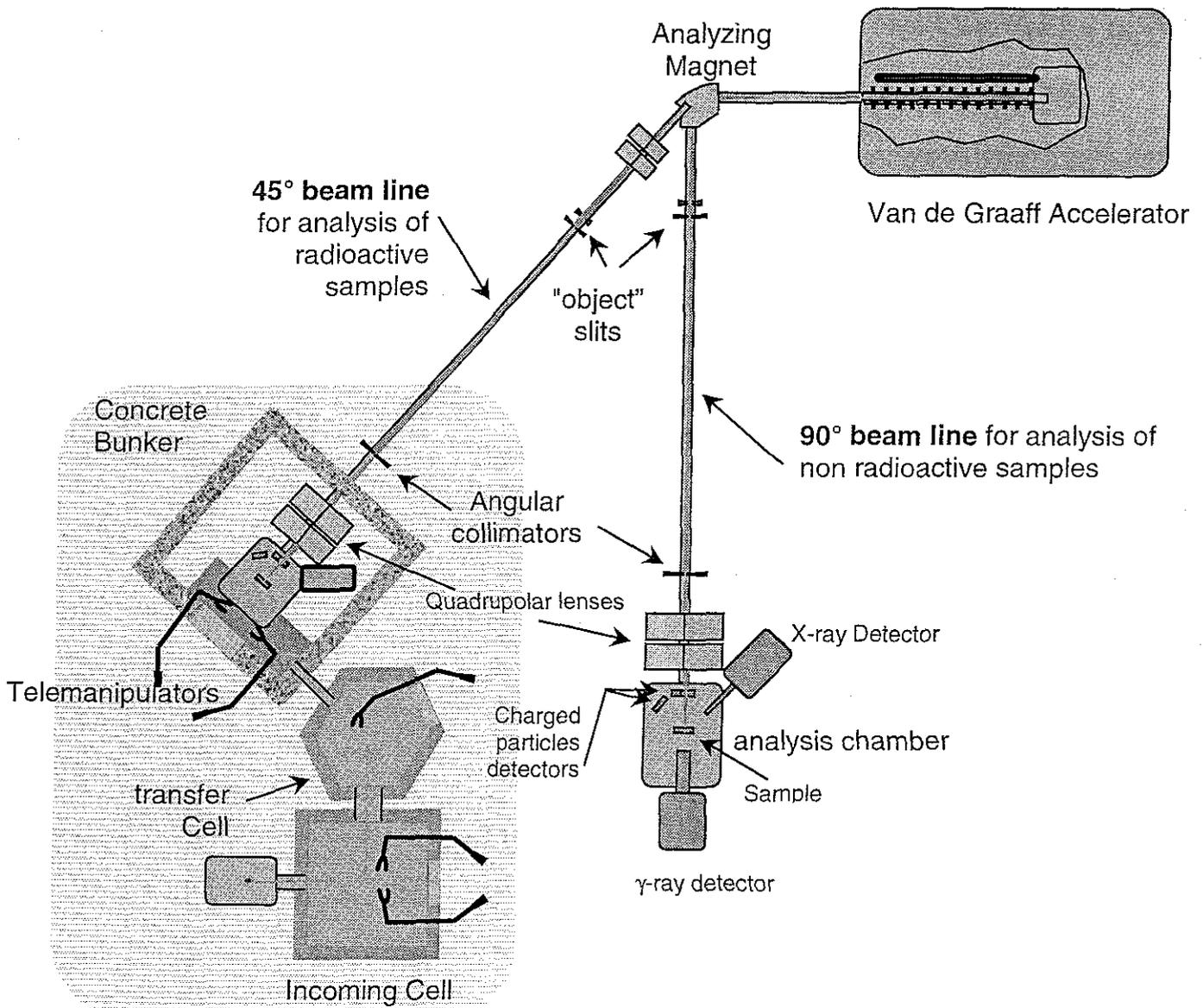


Figure 1 Schematic view of the nuclear microprobe.

Four kinds of ion are accelerated according to the type of gas injected into the bulb of source : protons, deuterons, ^3He and ^4He with intensities at the output of accelerator of several tens of microamperes.

The beam is analyzed by a magnetic dipole to define the energy of the particles and to eliminate the parasitic molecular beams. The energy resolution is typically 5×10^{-3} .

At the output of the dipole magnet, the beam size is of the order of 10 mm^2 and the intensity about 10 microamperes. The image point of the dipole is the starting point of the micro beam lines.

Micro beam lines

They are composed of two four-fold-slit systems. The first one, at the image point of the analyzing magnet, defines the size of the beam at the input of the line. Its aperture is adjustable in the range $\pm 100 \text{ }\mu\text{m}$ to $\pm 0.1 \text{ }\mu\text{m}$. The second one, situated 6 m downstream, defines the convergence of the beam. Its aperture is adjustable between ± 5 and $\pm 0.1 \text{ mm}$. These slits systems are built with stainless steel cylinders. Each X or Y set is mounted in different plans, then each of cylinders is used partially as anti-scattering to the precedent. These slits are cooled in order to minimize variation of the aperture during the experiments. The beam is then focused on the sample by a quadrupole doublet mounted on a adjustable table to optimize the position of the optical axis with respect to the geometrical alignment. The adjustment, opening of the slits and field in the focussing magnetic lens allows to obtain a beam size of a few μm^2 on the sample.

Two electrostatic deflectors are used to adjust the beam position on the sample. These are also used to sweep the beam on the target for mapping or profiling. The maximum swept area is $100 \times 100 \text{ mm}^2$ with a minimum step of $0.3 \text{ }\mu\text{m}$.

Scattering Chambers

Two micro beam lines are installed at the nuclear microprobe of the LPS.

The 90° line is equipped with a multipurpose scattering chamber for studies of no radioactive samples. *CASIMIR*, the chamber of the second line, in operation since end of 1997, is only dedicated to active samples analysis.

Samples are mounted on a target holder which is fixed on a X -Y- Z - θ goniometer. The amplitude and the accuracy of the displacements are summarized in the table below;

AXIS	amplitude (mm)	accuracy (μm)
X	20	0.5
Y	150	0.5
Z	20	0.5
θ Degrees	180	0.01

The goniometer and the electrostatic deflectors allow to adjust the beam position on the target with an accuracy better than $0.5 \text{ }\mu\text{m}$.

The detectors which equip both chambers are somewhat different.

In the 90° line chamber, the X-ray and γ -ray are measured by Energy Dispersive Spectrometers (EDS), Si-Li and high purity Germanium respectively.

In *CASIMIR*, the active samples are high gamma and beta emitters, therefore no nuclear reactions inducing gamma emission are used. For X-ray measurements the EDS detector is replaced by a Wavelength Dispersive Spectrometer (WDS) which has the advantage of not having its active part in direct sight of the sample. The main features of the WDS are the high energy resolution (10 eV) and the easy protection against the parasitic radiation. The disadvantage is its weak efficiency which must be compensated by long time measurement.

In both chambers, nuclear reactions leading to particles emission are measured with solid state silicon detectors. In the case of active samples the detector is embedded in a lead protection.

RADIOACTIVE SAMPLES HANDLING

The samples are prepared in the EPMA line of the Saclay Hot Laboratories. They are inserted in a cup and metallurgically impregnated with a tin-bismuth alloy. In order to reduce the activity, the cup is cut and the remaining part contains the sample with height less than 0.5 mm. The active surface is polished with abrasive papers and diamond or alumina powders. The final granulometry is less than one micrometer. After polishing, the sample is washed in an ultrasonic acetone bath and decontaminated by immersion in an ultrasonic alcohol bath. The sample is then transported to the LPS in a shipping cask.

The left part of figure 1 schematizes the hot line of the LPS facility. The sample is introduced in a hot lead cell. This first step is devoted to the control of the absence of contamination by immersion in a ultrasonic alcohol bath. The non contaminating sample is then transferred into a second cell to be introduced into a shuttle conveyor.

The transfer to the concrete bunker containing the scattering chamber is done by a pneumatic. The last step is the mounting of the sample on the scattering chamber goniometer.

After each analysis, the activity of the whole of the installation is then controlled in order to allow the routine maintenance of the particle detectors, beam diagnostics, reference samples...

FIRST RADIOACTIVE SAMPLE MEASUREMENT

The first analysis in *CASIMIR* was performed on December 17th of 1997. It was devoted to the measurement of lithium content in the zirconia layer of a Zircaloy cladding. Lithium is contained in water of primary cooling system and is a catalyst of the oxidation process. The piece of cladding came from a fuel rod irradiated during five cycles in a PWR nuclear power plant. Its activity was 1 GBq.

Lithium was identified by the ${}^7\text{Li}(p,\alpha){}^4\text{He}$ nuclear reaction. Four points of measurement were done in an undamaged area (figure 2), three points in zirconia layer to deduce an average profile, and one point in Zircaloy to control the sensitivity of the method.

The emitted particles were detected by a telescope consisting of two solid state silicon detectors in order

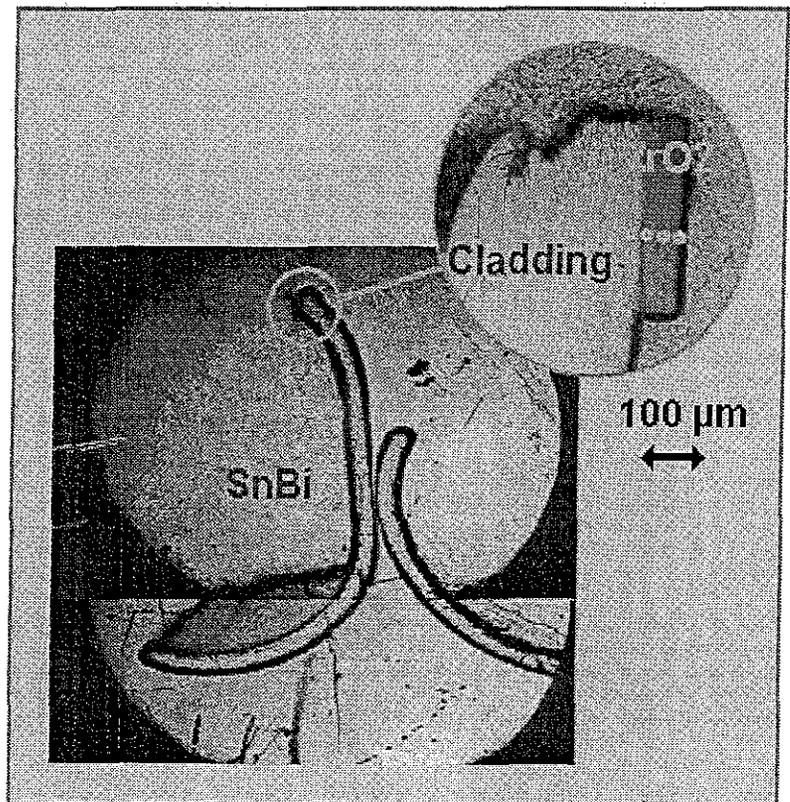
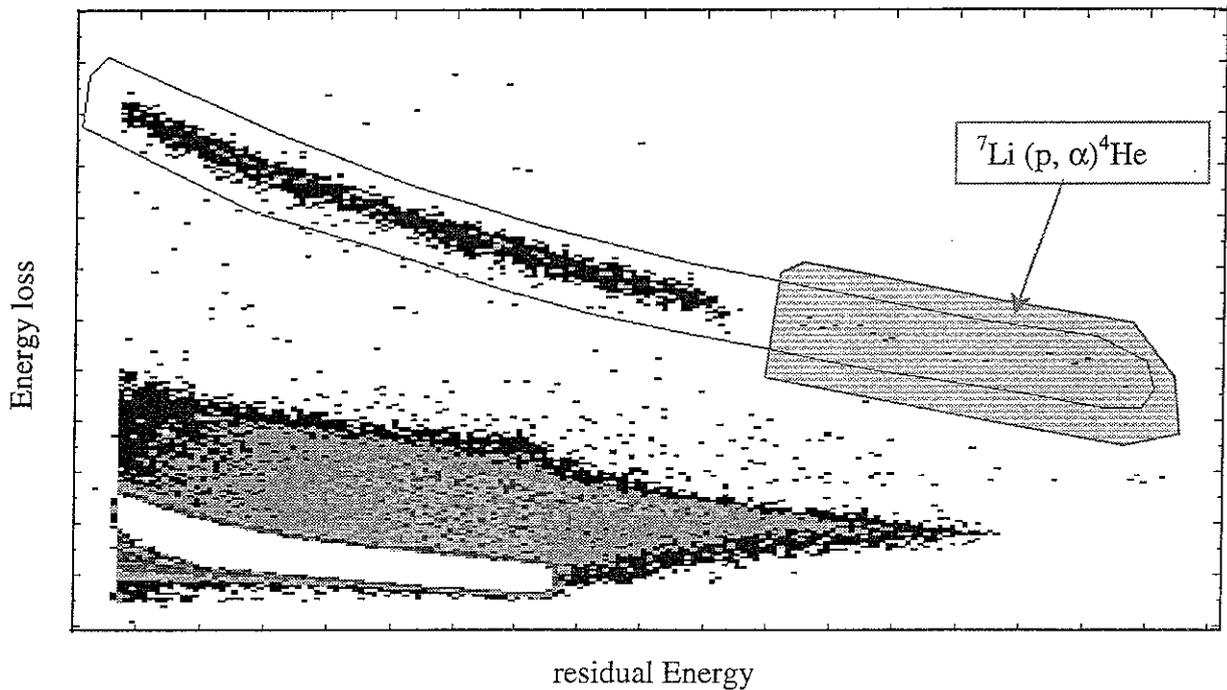


Figure 2 radioactive cladding. The 4 green dots show the measurements.

to identify the nuclear reaction products of interest from the elastic scattering on zirconium and oxygen contained in the sample as well as the α particles emitted by the activation products of a residue of fuel fixed on the analyzed piece of cladding. Figure 3 shows a two dimensional representation of the identified products. The region of interest is delimited by a contour.



Identification of the reaction products measured by a solid state silicon telescope detector (ΔE : $15 \mu\text{m}$, E : $1000 \mu\text{m}$)

The measured profile shown on figure 4 provides an average concentration of 25 wt ppm. This result is comparable with measurements performed with the SIMS method on a similar sample.

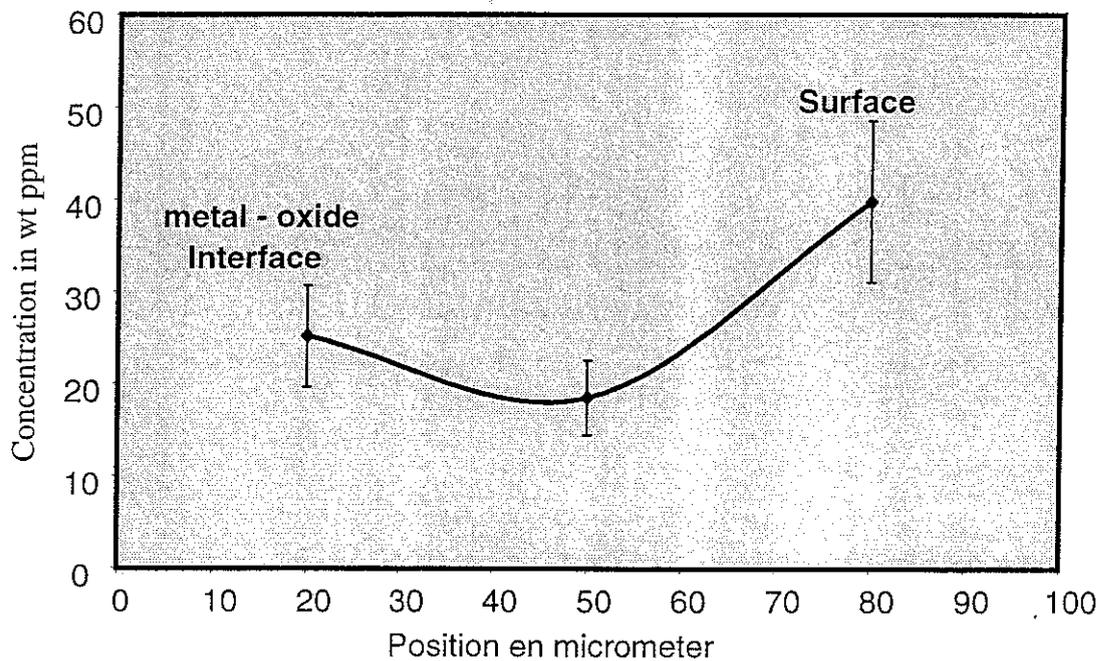


Figure 4 Profile of Lithium in zirconia

SUMMARY

The *CASIMIR* line of the LPS nuclear microprobe facility is operational. A new method now made available analysis of radioactive samples. The first results show the ability to detect very low concentration, down to 25 wt ppm, with the nuclear reactions method for very active samples.

Analysis of other radioactive material is now possible concerning fuel, cladding, control rods, reactor vessel or storage materials.

Our immediate objective, before the end of this year, is to be able to analyze fission products contained in a piece of fuel by the PIXE method, using the WDS X-ray detector.

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