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THE FRENCH AEC NUCLEAR MICROPROBE : DESCRIPTION AND FIRST APPLICATION EXAMPLES

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The major components of the microprobe are briefly described.

The performances and the varying possibilities authorized by the instrument are given.

Some application examples concerning especially the determination of concentration profiles in a aqueous leached glass and the distribution of deuterium in a graphite sample exposed to plasma in a Tokamak device are presented.

1. INTRODUCTION

The french AEC nuclear microprobe is in operation since the beginning of 1983.

Our interest for such a instrument is at first because it appears particularly powerful for light elements (Ex : H, D, T, Li, B, C, N, O, F,...) and simultaneously heavy elements ($Z > 12$) local concentration or distribution measurements in solids (geological samples, glasses, metals,...) with a high sensitivity.

It is undoubtedly complementary to other microprobe techniques (Auger, electrons, ions, laser,...).

However, the nuclear microprobe presented some specifif advantages over the letter methods.

Indeed, his calibration for quantitative determinations, especially concerning light elements, is much easier as in the case of ion and laser microprobes; no special prepared standards are required. In addition, the sample is not eroded, this is very important when radioactive materials (Ex : waste glasses containing actinides like neptunium, plutonium,...) are examined.

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On the other hand, the nuclear microprobe is more sensitive (one or two orders of magnitude) as Auger and electrons microprobes.

At last, due to the nature and the energy of the incident particles, the nuclear microprobe can be used for light elements (or their isotopes) concentration measurements in closed vessels provided with a thin (1 to 5 μm thick) wall (Ex : deuterium and tritium determinations in microballons for laser fusion devices).

2. DESCRIPTION

The instrument, localized in the Research Center of BRUYERES LE CHATEL, is constitute by a 4 MV VAN DE GRAAFF, a beam line with usual control and steering devices (bending magnets, collimators, Faraday cups, obturators,...), a adjustable object aperture and magnetic quadrupole lenses in the form of a Russian quadruplet (Harwell system) /1/ /2/.

A general layout of the installation is shown in fig. 1.

The target chamber, represented by the schematic diagram of fig. 2, is equipped with a micromanipulator (XYZ translator and rotary drive system) for sample changing and positioning against beam axis, a lock for the introduction of specimens without vacuum interupption, a monocular microscope (x 40) for rough adjustment of the beam, a binocular microscope (x 200 to 400) for final focussing of the latter and to observe the sample under investigation.

Surface barrier and Si(Li) detectors are used for charged particles (nuclear reactions or Rutherford back-scattering) and X-ray (determination of elements heavier as sodium by PIXE) detection.

The position of the Si(Li) detector, in the front of the target, can be adjust with the aid of a special screw system to optimize his efficiency.

A ion pump (400 l/s) associated with a titanium sublimation device enabled to obtain, if necessary (for example to avoid superficial contamination of high purity metals by hydrogen, carbon, nitrogen or oxygen), ultra-high vacuum ($< 10^{-9}$ Torr) in this target chamber.

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Furthermore, a TN 4000 computer-based (LSI-11/23) multichannel analyser in combination with a color imaging and processing system are used for elemental profiling or mapping; so that the distribution of several elements in the scanned direction or area (1 x 1 mm^2) can be displayed simultaneously.

The sweeping of the beam, by electric deflection (Harwell system) /1/ /2/ , is controlled digitally; the number of pixels can be chosen up to 512 in each direction.

A general view of the target chamber and the above shortly described elemental profiling and mapping equipment is shown in fig. 3.

3. CHARACTERISTICS AND PERFORMANCES

The charged particles used for analytical purposes are :

- protons (1 to 3 MeV) for PIXE measurements or light elements (Li, B, F,...) determination by (p, α) reactions;
- deuterons (1 to 2 MeV) for light elements detection (C,N,O,...) by (d,p) or (d, α) reactions and simultaneously some heavier elements (Ex : Cr, Fe, Ni,...) by PIXE;
- helium-3 (1 to 3 MeV), mainly for the determination of deuterium by the $^{2}H(^{3}He,p)^{4}He$ reaction.

The minimum achievable diameter of the focussed beam spot is about 2 to 3 um; therefore the topographical resolution allowed by the mapping system is of the same order of magnitude.

To avoid secondary effects (local evaporation or destruction, migration, etc...) during bombardment, the beam intensity is limited between 0.1 and 1 nA according to the nature of the examined sample (biological specimen, glass, metal,...).

The detection limits realizable are in the range of 10^{-16} to 10^{-15}g (1 to 10 ppm) for most elements.

4. APPLICATIONS

The first example concern the use of the nuclear microprobe to measure, by PIXE, concentration profiles induced by aqueous leaching in the surface layer of a simulated waste oxides borosilicate glass.

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Samples have been leached during 50 days in granitic water at 90°C and suitably sliced in view of deep profiles determinations.

Line scans (100 pixels, 10 seconds each) across the gel layer and the subsurface region of glass were realized with a 2 MeV proton beam (I = 0.5 nA).

Some typical results are represented by figures 4, 5 and 6.

As seen, the superficial region of the leached glass appears as a complex structure; it is clear that elements such as Zr, Th and U are strongly enriched in the outer layer, whereas Si is depleted in the near surface.

The second example, according to the work done by SOFIELD et al. /3/, is relative to the determination of deuterium distribution in graphite diaphragms from a TOKAMAK device reaction chamber.

For this purpose, the following nuclear reaction has been used :

$$^{2}H\ (^{3}He,\ p)\ ^{4}He$$

The ^{3}He ions energy was 1 MeV (I = 0.5 nA). The emitted protons (13 to 14 MeV) were detected by the annular surface barrier detector (area : 100 mm^2; depletion depht : 100 μm) covered with a 200 um thick tantalum screen slowing the protons energy enough down (< 7 MeV) so that a well defined partial absorption peak characteristic of deuterium can be identified in the spectrum.

The results obtained are shown in figures 7 and 8.

These diagrams represented the deuterium distribution in samples from the ionic and the electronic sides of the graphite diaphragm.

Obviously, as foreseen, the superficial concentration of deuterium is higher in the first type of specimen (fig.7).

Moreover, concerning this latter, as shown by fig.8, some localized concentration peaks are plainly apparents.

5. CONCLUSIONS

Nuclear microprobe is undoubtedly a very powerful tool for investigation about mecanisms of hydrothermal leaching of nuclear waste glasses.

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Concerning this latter matter, one of his main advantage over other methods, like the electron microprobe, lies in the possibility of determining simultaneously concentration profiles of light elements (Li, B, Na,...) by nuclear reactions and heavier elements (Si, Ca, Fe, Ni, Zn, Zr, La, Ce, Nd, Th, U, Np, Pu,...) by PIXE.

It appears also a very interesting technique for some special applications, as light element isotopes detection (Ex : D, T,...) in materials or microvessels used in thermonuclear fusion devices (laser, Tokamak,...).

Among our other research programmes in progress concerning the use of the nuclear microprobe are the following :

- carbon, nitrogen, oxygen, chromium, iron and nickel concentration profiles measurements in steels corroded by liquid sodium or carbon oxides ($CO + CO_2$) atmospheres at high temperatures (500 to 1000°C);
- carbon and oxygen distribution in HLi pellets covered by a thin protective metal layer (Al, Au,...; thickness : 100 to 500 Å);
- light elements (Li, B, C, F,...) detection in localized areas of geological samples (Ex : micro fluid inclusions incide the crystals);
- multielemental analysis of biological microsamples (10^{-6}g);
- traces determination in microsamples take from very precious old paintings.

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FIGURE CAPTIONS

Fig. 1 General layout of the beam line

1 Oil diffusion pump (300 l/s)
2 Line valve
3 Object aperture
4 Magnetic quadrupoles
5 Target chamber
6 Ion pump (400 l/s)
7 Bending magnet
8 Steerer
9 Faraday cup
10 Vacuum gauge
11 Sand bed (20 cm)
12 Concrete blocks (650 and 3000 kg)
13 Beam profile monitor

Fig. 2 Schematic diagram of the target chamber

1 Liquid nitrogen dewar (20 l)
2 Micromanipulator (X Y Z translation)
3 Rotary drive system
4 Si(Li) detector positioning system
5 Si(Li) X-ray detector (A : 12 mm^2; D : 4 mm)
6 Target wheel (5 samples)
7 Removable annular surface barrier (A : 100 mm^2; D : 100μm) charged particles detector
8 Binocular microscope (x 200 to 400)
9 Removable mirror objective
10 Fixed surface barrier (A : 25 mm^2; D : 100 μm) charged particles detector
11 Samples introduction lock
12 Specimens
13 Monocular microscope (x 40)
14 Beam viewing quartz
15 Observation scuttle

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Fig. 3 View of the target chamber and the data acquisition system

Fig. 4 Concentration profiles of Si, Ca, Fe, Ni, Zn and Zr measured by PIXE on the superficial region of a simulated waste glass leached in granitic water during 50 days at 90°C.

Fig. 5 Concentration profiles of La, Nd, Th and U measured by PIXE on the superficial region of a simulated waste glass leached in granitic water during 50 days at 90°C.

Fig. 6 Concentration profiles of Si, Ca, Fe, Zn, Zr, Nd and Th measured by PIXE on the superficial region of a simulated waste glass leached in granitic water during 50 days at 90°C (the above measurements represented by figures 4 and 5, and this one, have been done on the same sample, but at two different places).

Fig. 7 Deuterium distribution on the surface of two samples from a TOKAMAK device graphite diaphragm

.... ionic side

xxxx electronic side

Fig. 8 Deuterium distribution corresponding at three different positions (0.175 and 350 μm) on the surface of the ionic side sample from the TOKAMAK device graphite diaphragm.