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Laboratori Nazionali di Frascati

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THE ^{15}N BEAM FACILITY FOR HYDROGEN PROFILE STUDIES AT THE LABORATORI NAZIONALI DI LEGNARO

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ABSTRACT

Several modifications to the 7 MV CN Van de Graaff of the LNL have been performed to allow 15N beam acceleration. Hydrogen depth profile measurements have been performed. The structural changes, experimental apparatus and sample data are shown.

1. - INTRODUCTION

Several nuclear laboratories have devoted considerable efforts, over the last years, towards the application of nuclear techniques in the solution of solid state and material research problems. Rutherford backscattering and the use of low-energy nuclear reactions for elemental depth profiling are of wide use. In particular, several authors[1, 2] have stressed the importance of hydrogen profiling to investigate the properties of the material surfaces.

A particular interesting example, concerns the study of the surface behavior of glasses. The increasing interest in these techniques is due to the important scientific and technological problems dealing with the long-term durability of materials exposed to acqueous corrosion in natural environments, such as geological glasses or glasses incorporating high-level radioactive wastes.

In this paper we report preliminary results of hydrogen profile measurements performed at the 7 MV Van de Graaff of the LNL using the ^{15}N beam.

2. - EXPERIMENTAL SET-UP

The 7 MeV Van de Graaff accelerator of the LNL is a CN type High Voltage Corporation machine. The CN accelerator has been operating since 1962 and has been extensively used in nuclear physics experiments.

The ion source is a home-made standard radiofrequency model coupled to a magnetic triplet for the separation of singly and doubly charged ions[3].

The old magnetic triplet (Fig. 1) was designed to focus ions of H, D, ^3He, ^4He into a 1.6 mm exit hole before injection in the accelerating tube.

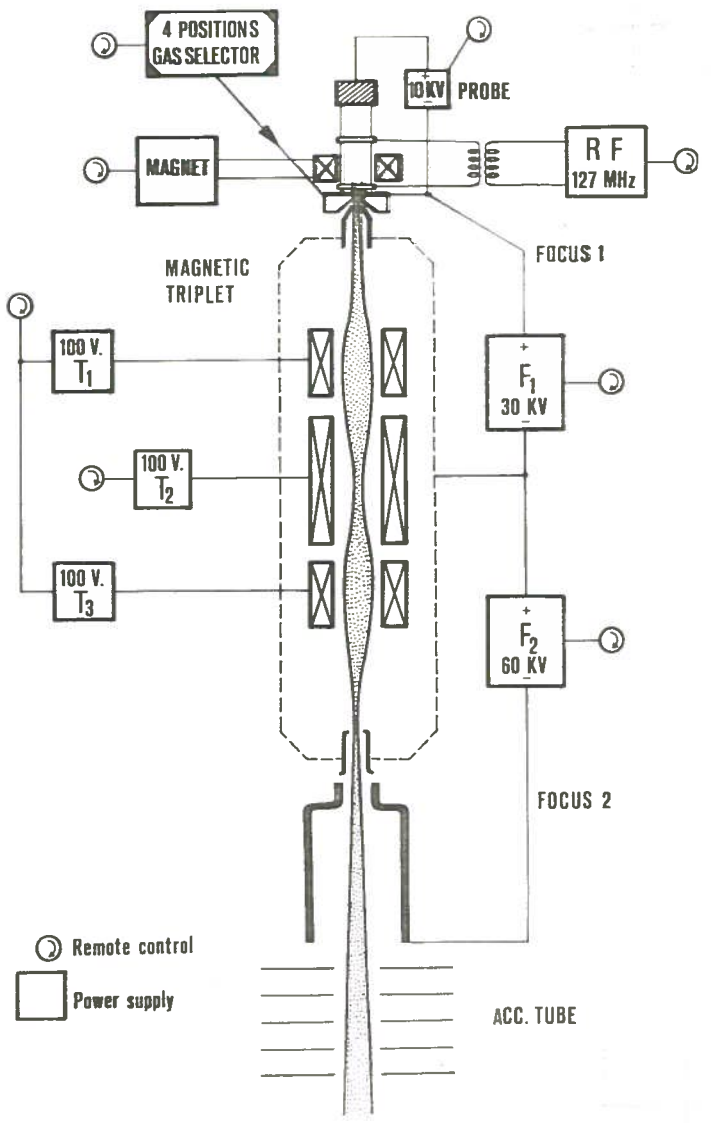

FIG. 1 - Schematic representation of the CN ion source.

While the external dimensions of the triplet (60 mm for T1 and T3, 110 mm for T2) were maintained, the number of coils of the magnet has been increased from 1000 to 1450 and the maximum gradient from 400 to 600 Gauss/cm. The increased focusing capability of the magnetic lens allows now the extraction of a beam of ions with a Q/M ratio of 1/8 and acceptable intensity. A beam of 150 nA of analysed doubly charged ^{15}N has been obtained using a 99.9% enriched ^{15}N gas.

The accelerator is equipped with a 90 degree 70 cm radius analysing magnet of the doubly focusing type, with a EM/Z^2 product of 30, that allows the analysis of a full energy (7.5 MeV) $^4He^+$ beam. Under these conditions the upper limit in energy for the $^{15}N^{++}$ ions is about 8 MeV limiting the depth profile measurements to less than 10^4 Å. In the most common cases, the analysis of the hydrogen concentration concerns the near surface of the sample, and 10^4 Å is deep enough to give useful information.

Nevertheless, in order to extend the energy range and consequently the depth of analysis, a carbon stripper, equipped with 20 foils of 10 μgr/cm^2, has been installed 10 cm after the entrance slits of the magnet. The stripper increases the charge state of the ions to be analysed with a loss in intensity of a factor two, and an increase in energy spread of the same factor. In Fig. 2 is shown a picture of the stripper.

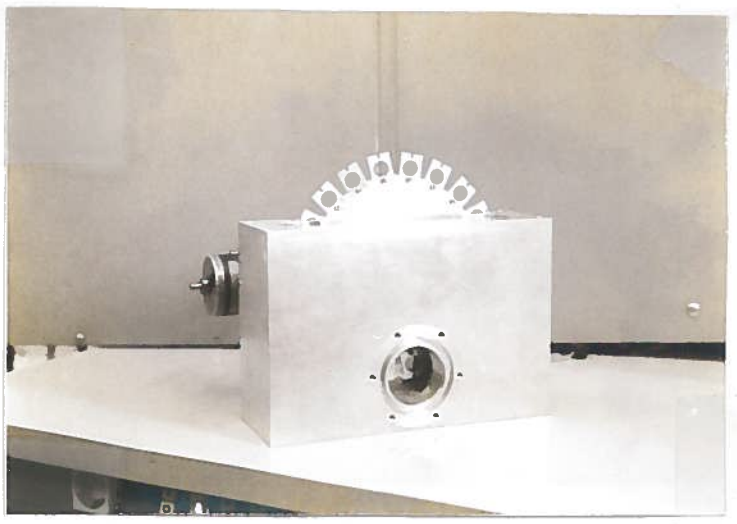

FIG. 2 - The beam stripper.

The schematic experimental set-up for the H profile measurements is shown in Fig. 3. The vacuum in the target chamber is 3×10^{-7} mbar and a liquid nitrogen trap is used in order to condensate organic vapor residues. A tantalum diaphragm with a hole of 2 mm is located 12 cm before the target.

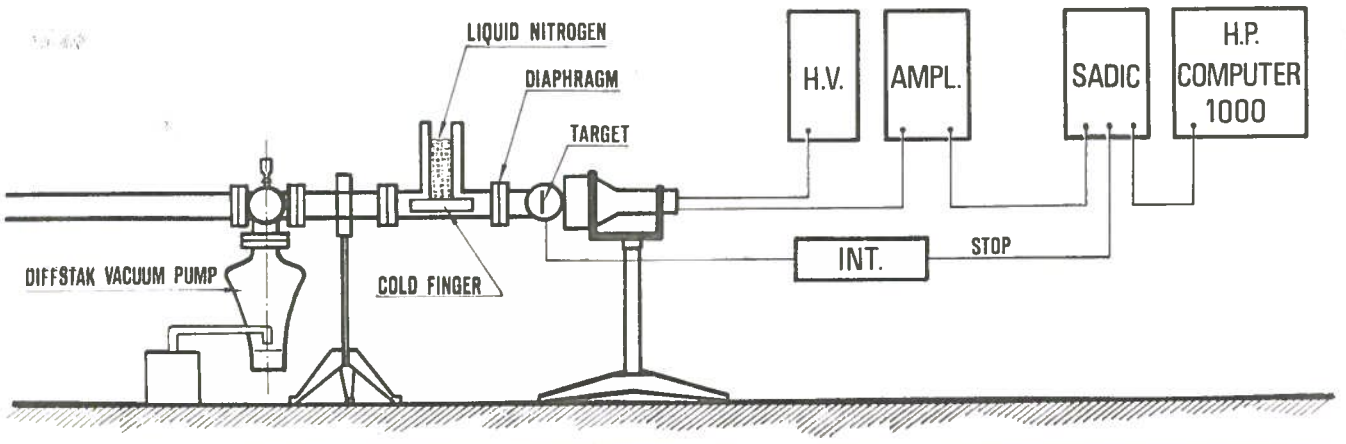

FIG. 3 - Schematic representation of the experimental apparatus and electronics.

The target chamber is 2 cm in diameter and contains 5 different targets. Every sample used in the measurements shown in the following was 10 mm in diameter and the non conductive samples were coated with a 100 Å of evaporated aluminum in order to prevent charge build-up on the surface.

A NaI(Tl) detector of 4" x 4", placed at 2 cm from the target and at 0 degrees with respect to the beam direction, has been used to detect the 4.43 MeV gamma rays produced by the H(^{15}N, α, γ)^{12}C reaction. As shown in Ref. (4), the angular distribution of the gamma line is 30% more intense at 0 degrees than at 90 degrees. The detector has been shielded from the gamma rays produced along the beam line with 5 cm

of iron, not shown in Fig. 3.

In Fig. 4 are shown the gamma spectra measured off-resonance and at the resonance energy.

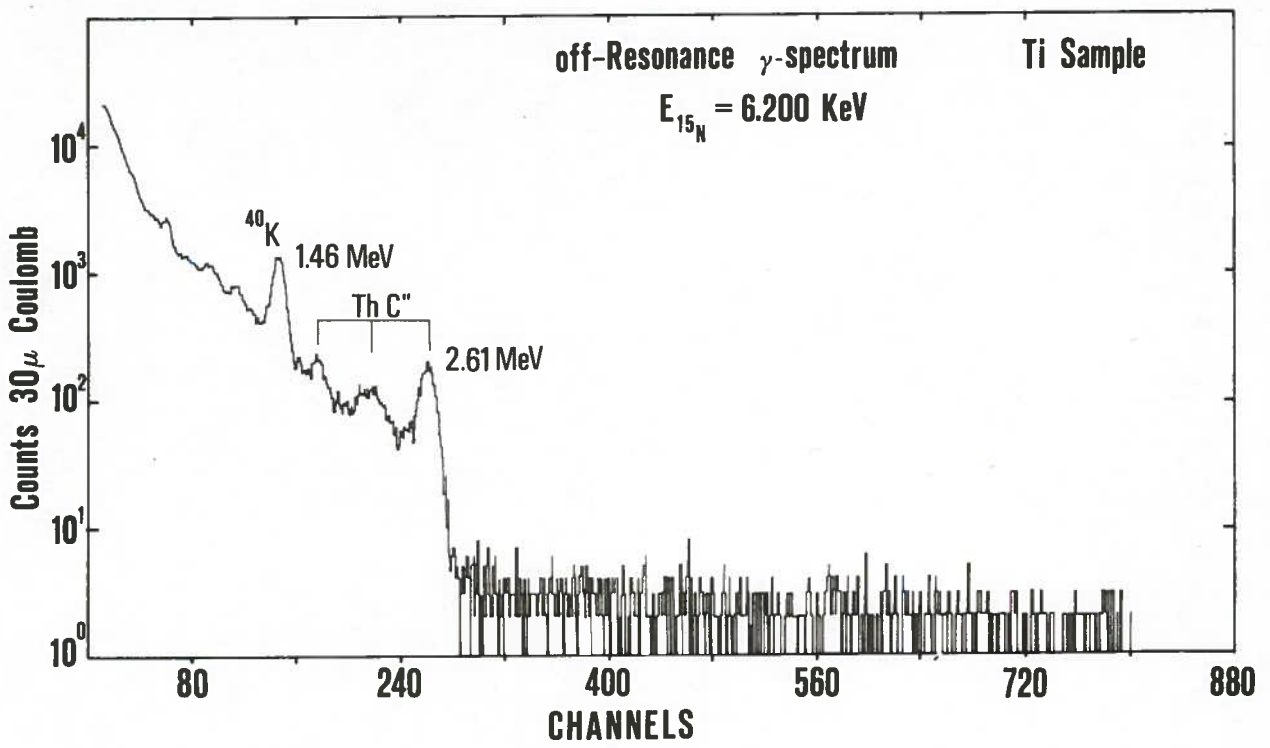

FIG. 4 - Gamma spectra obtained: a) off-resonace; b) on-resonance. The sample was natural Ti with unknown contents of H on the surface.

In Fig. 5 is shown the resonance shape of the E(^{15}N) = 6385 keV resonance obtained with a natural Ti sample.

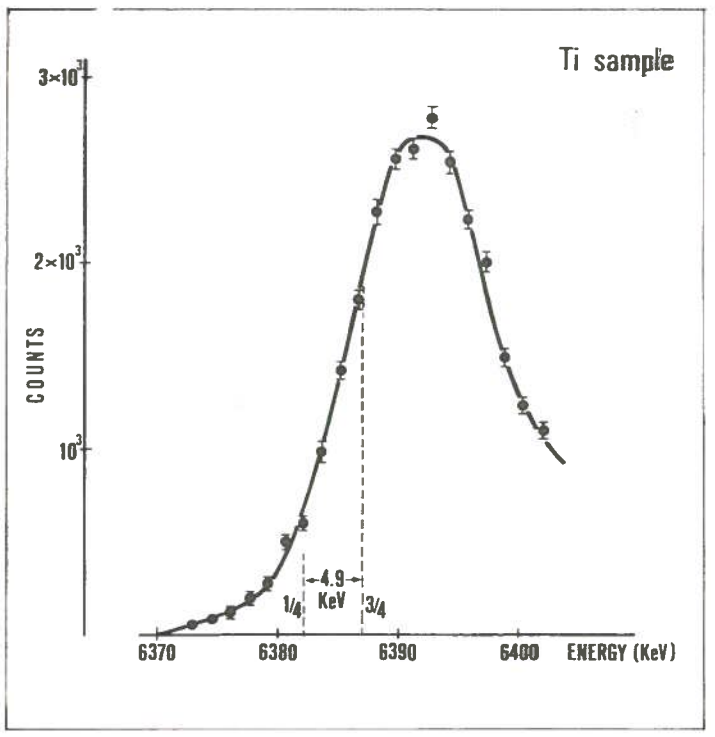

FIG. 5 - Resonant yield of the 3.5-4.5 MeV range gamma rays obtained with the same sample of Fig. 3.

Although the target used does not completely saturate the resonace,from the slope of the low energy side of the resonance it is possible to infer that the overall energy resolution is about 5 keV. Taking into account that 4-5 keV is a reasonable estimate of the Doppler effect, and the resonance width is 1.8 keV[5], the ultimate beam energy spread is less than 3 keV.

The data collection system[6] is equipped with a 5 nsec ADC linked to a HP 1000/E computer. The general purpose acquisition code (SADIC) has been adapted to perform automatic yield measurements with computation of the intensity of the gamma line at the end of each ^{15}N energy point. The resulting yield curve can be plotted at the end of the profile measurement. A current integrator interfaced to the data collection system has been used for charge normalization in the yield measurements.

In the standard experimental conditions no dead time corrections are necessary. In fact, with a H concentration less than 20%, the counting rate, in the gamma energy range 100-6000 keV, is lower than 0.5 KHz.

The profile curves have been obtained increasing the beam energy by steps of 15 keV and measuring at each step the gamma yield of the radiation, with energy in the 3.5-4.5 MeV range, integrating a total beam charge of 2 to 6 micro-Coulombs (20 to 60 seconds of duration). This means that for a standard profile (20 energy points) a total dose of about 10^{15} ^{15}N atoms/cm^2 is implanted on the sample.

As pointed out in Ref. (7), the background yield is the ultimate limit to the concentration sensibility. With the present set-up the background is of 30 counts/MeV/ /min, with the beam out of resonance, and of 40 counts/MeV/min, with a beam intensity of 50 nAmp deviated on the diaphragm at the energy of the resonance.

The profile curve giving the H concentration versus depth can be obtained from the yield curve, taking into account the energy versus depth relation (Stopping power)

and the yield of a standard reference sample of known contents in H. Using the usual stopping power values in MeV/at/cm^2, the unknown concentration can be easily deduced.

One sample of a single crystal of Si (implanted with a total dose of 2×10^{17} H atoms/cm^2 at 30 keV (Fig. 6) and a sample of Hornblende (with a 4% in atoms constant in depth of H concentration) have been used as references. The two references give the same results for unknown samples within 10%.

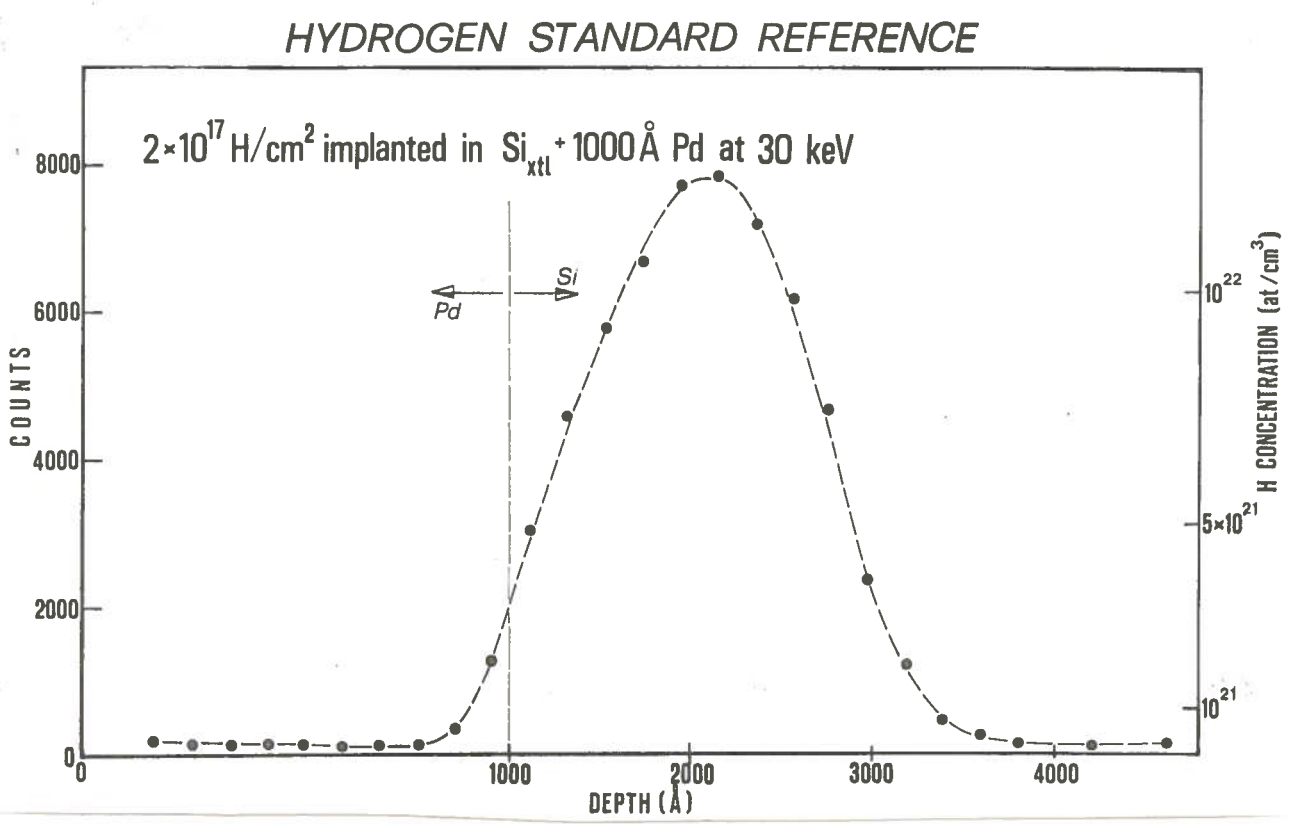

FIG. 6 - H profile of a Silicon sample implanted with 2×10^{17} H atoms cm^2 at 30 keV. A layer of Pd of 1000 Å has been evaporated before implantation.

3. - EXPERIMENTAL RESULTS

Preliminary data have been collected in order to test the ability of the technique to detect the H content of the surface of some materials.

The ion induced modifications of acqueous dissolution has been investigated in several silicates (soda-lime, amorphous silica, etc.) implanted with Pb ions and leached in standard conditions. The details of the target preparation and the interpretation of the results are described in Refs. (8, 9).

The correlation between Na depletion and hydrogen absorption has been indicated as the main result of the process of dissolution of the glass surface exposed to acqueous corrosion.

Special care has been devoted to verify the reproducibility of the measurements. Long runs of duration equivalent to a profile measurement have been performed at fixed energy where the profile concentration was steeper. No variation of the yield has been observed.

Other samples of ionic salts and of $LiNbO_2$ showed a marked dependence of the yield as a function of the beam intensity.

The measurements on SiO_2 implanted with different doses of Pb ions at 1 keV/amu showed that the H penetration, after implantation and leaching, increased when the ion dose exceeded a critical value between 10^{13} and 10^{14}. Fig. 7, and that, for long leaching times, the hydrogen decorated the defect distribution produced by the implanted ions Fig. 8.

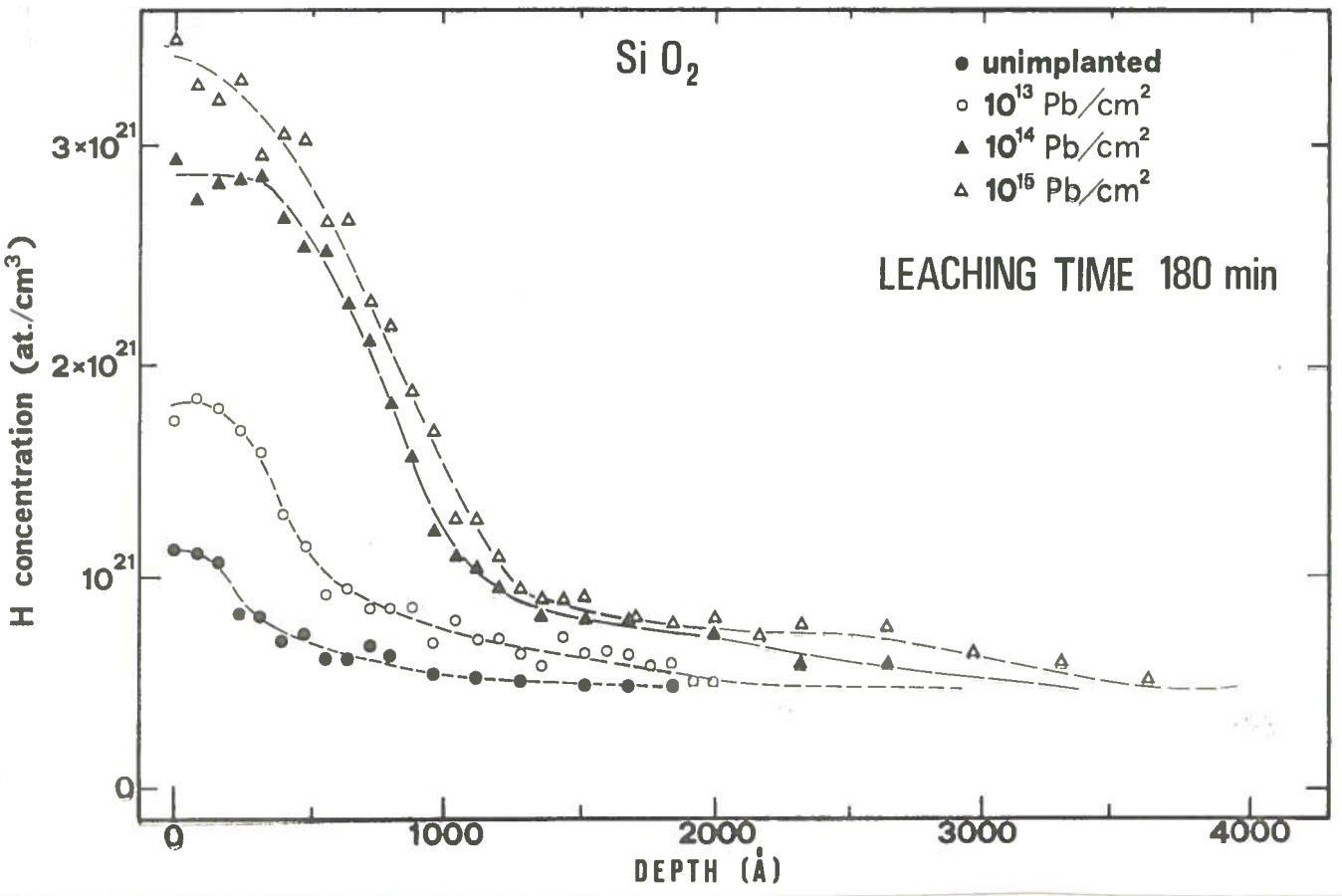

FIG. 7 - Hydrogen depth profiles of amorphous silica implanted with Pb ions at increasing doses and leached 3 hrs in water at 100 °C.

The experimental results showed that the hydrogen profile measurements are possible also on non-conductive samples.

The sensibility of the experimental set-up was able to detect 0.2% of hydrogen content.

Hydrogen buil-up due to organic residues was absent.

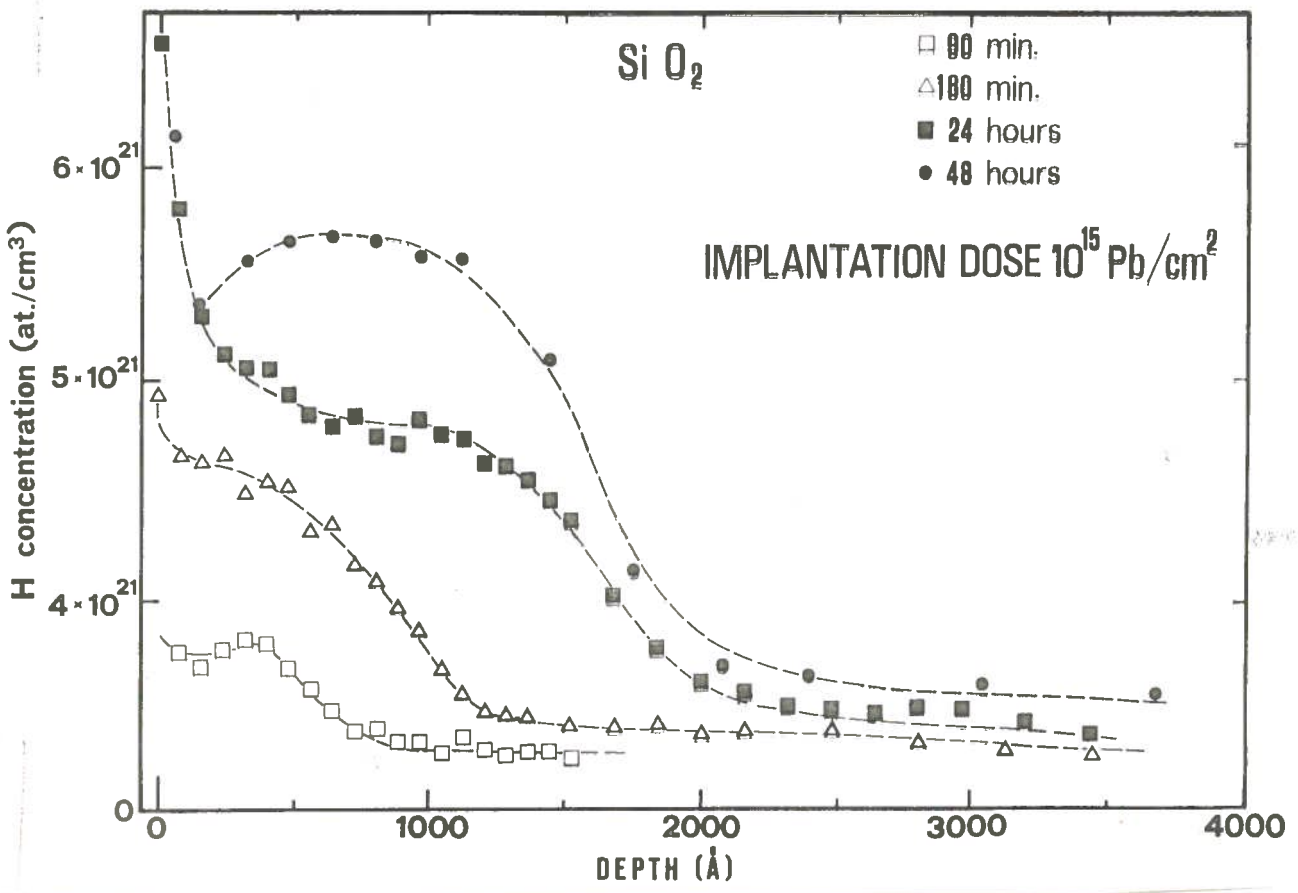

FIG. 8 - Hydrogen depth profiles of amorphous silica implanted with 10^{15} Pb ions/cm^2 and leached in water at 100°C for increasing times.

Further experimental work is in progress, the investigation is related to polycrystaline silicon, $LiNbO_2$ for optical applications, etc.

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