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A MONOCHROMATIC X-RAY SOURCE FOR A PIXE-INDUCED XRF SPECTROMETER

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ABSTRACT

A compact and intense source of proton-induced monochromatic x-rays, tunable in the 5-30 keV energy range, has been obtained using a 3 MeV proton beam in an irradiation chamber equipped with a multiple target system.

The source has been characterised in terms of intensity, monochromaticity and peak to background ratio. Possible applications of the source are described: PIXE-induced XRF analysis (*PI-XRF*) as a complementary tool to PIXE analysis, x-ray photoemission spectroscopy and generation of intense soft x-rays.

Comparison to standard x-ray sources produced by an x-ray tube coupled to secondary anodes is presented.

1. - INTRODUCTION

The production of nearly monochromatic and very intense x-rays in the 1-100 keV energy range by proton irradiation of pure metal targets has been deeply investigated in the past. A set-up for irradiation was realised and installed in the beam-line of the CCR Ispra cyclotron[1,2,3]. The Kα x-ray yields were calculated[4,5,6] for 66 elements from Na to Pb using the ECPSSR theoretical model for proton ionisation cross-section. A great advantage was foreseen by using protons in the energy range of 20-40 MeV, because of the steep increase in the ionisation cross-section, as shown in Fig. 1 (from ref.(4,5)) .

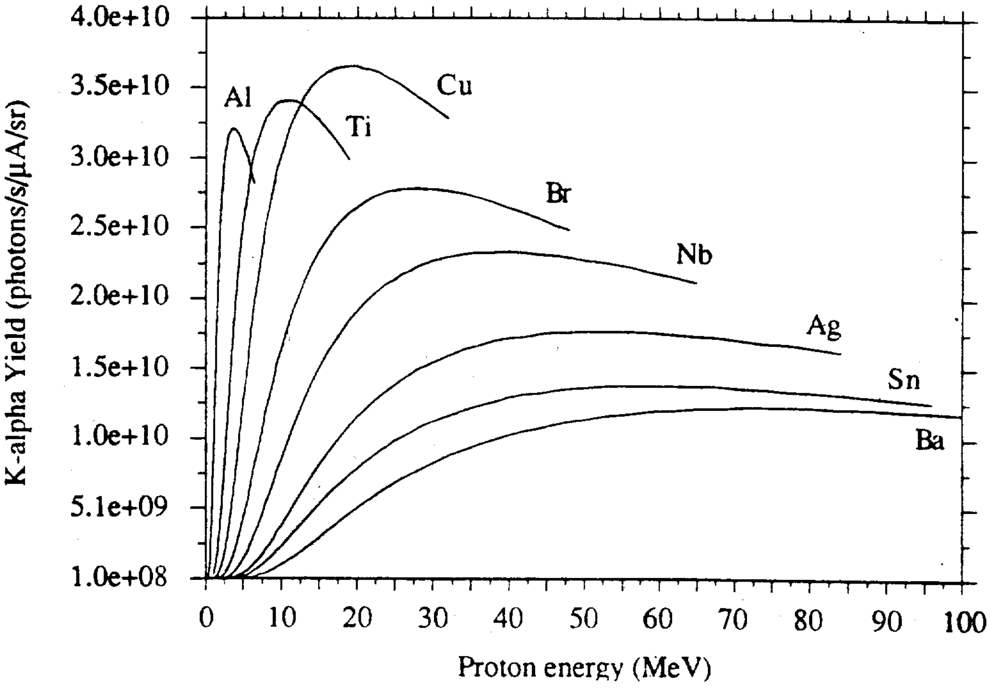


FIG. 1- K_α X-ray yield as a function of proton energy and target element.

The background due to atomic phenomena was calculated too for radiative ionisation, secondary electron bremmstrahlung and proton bremmstrahlung. The expected spectra, obtained by convoluting the continuum background and the Kα and Kβ lines with gaussian shaped peaks, were compared with the experimental spectra. As we can see from Fig. 2 (from ref.(2)), for a Ge target irradiated with 25 MeV protons, the calculated peak to background ratio of about 10^3-10^4 is reduced to about 10^2 because of the contribution to the background due to nuclear reactions that, as expected, is non-negligible at these proton energies. The effort to reduce this nuclear background shielding the detector and choosing the appropriate target thickness didn't success, the peak to background ratio being exceedingly high for practical application.

In order to suppress the nuclear background we decided to use protons with energies below the Coulomb barrier. Of course, the Kα x-ray yields are drastically reduced; nevertheless the system still presents some interesting features :

1. it can be an useful, low cost integration of standard external beam PIXE system
2. the availability at reasonable cost of low energy (few MeV), high current (1-10 mA) proton accelerators as RFQ's should make possible the realisation of high-brilliance x-ray sources[7,8,9]

The aim of the experiment has been to verify the advantage offered by XRF to produce a selective monochromatic excitation of the sample which results in a great simplification in the quantitative analysis. The possibility of focusing the protons over small spot on the primary targets coupled to the use of glass microcapillary for collimating x-rays will allow the achievement of a μ-XRF spectrometer with high lateral resolution and high sensibility.

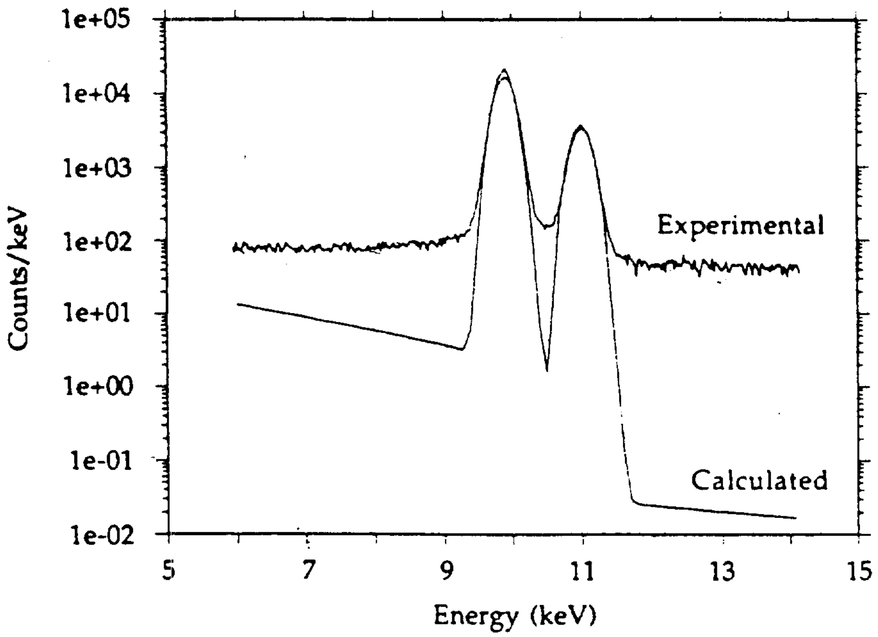


FIG. 2 - Experimental and calculated energy distribution for a Ge target bombarded by 25 MeV protons.

2. - EXPERIMENTAL SET-UP

A semi-automated, remote-controlled, dual-chamber irradiation system was designed and built for PIXE-induced XRF measurements at Ispra cyclotron(1). With some modification the chamber was adapted to the beam-line of KN3000 VDG accelerator in Florence. At the end of the line a PIXE system is already installed.

The experimental set-up consists of two distinct chambers. The first, shown in Fig. 3, is the proton irradiation vacuum chamber: Here the protons impinge on one of six pure metal targets generating the characteristic x-ray fluorescence of the element.

A four sectors diaphragm upstream the target holder with a 2 mm hole allows the proton beam to be centred on the target.

A value of 30° has been chosen for the angle of tilt of the targets with respect to the proton beam direction obtaining a satisfactory compromise between good geometry for the x-ray source and maximum photon intensity (3,6).

The multitarget holder, which is remotely controlled, and the four sectors diaphragm are easily removable from the chamber so allowing the proton beam to go on to the PIXE system without the necessity of entirely removing the XRF installation from the line. This is very useful because in this way the PIXE and the XRF spectrometers can be virtually used together. In fact in the procedure for removing the diaphragm and the target carousel the only time consuming stage is the vacuum repumping.

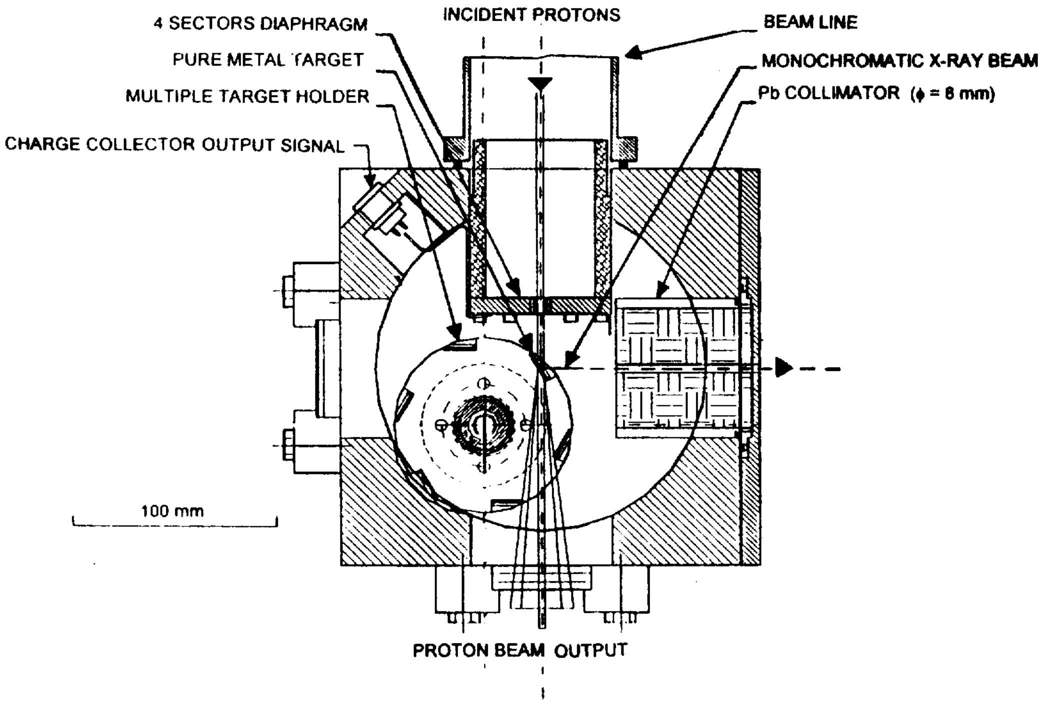


FIG. 3 - Proton irradiation vacuum chamber installed at KN 3000 VDG.

The almost monochromatic x-ray beam (Kα+Kβ lines) produced in the target is collimated through a 8 mm inner diameter lead collimator in the XRF chamber direction. The x-rays emerging from the collimator enter the XRF chamber, shown in Fig. 4, passing through a thin aluminium window (10 μ) and impinges on the sample with 45° glancing angle. Immediately afterwards the window it is possible to put a β-filter made of a pure metal of desired thickness to improve the monochromaticity of the x-ray beam.

The samples are housed on a multiple holder (6 positions) remotely controlled and easily removable from the chamber. The detector (Si(Li) or HPGe) measures the fluorescence x-rays emerging from the sample with a glancing angle of 45° respect to the sample surface. In this way the calculations for quantitative XRF analysis can be simplified also when surface irregularities in the sample are present(10).

The whole system composed of the two chambers is shown in Fig. 5.

The path of the x-rays from the source, in the proton irradiation chamber, to the sample is about 17 cm and it is, obiouvsly, uselessly long. This very long collimating system was necessary when using high energy protons in order to reduce the intensity of γ-rays reaching the detector. Using low energy protons without γ-ray emission, it will be possible to reduce this path of at least 6 or 7 cm improving the whole system by increasing the x-ray flux at the sample surface.

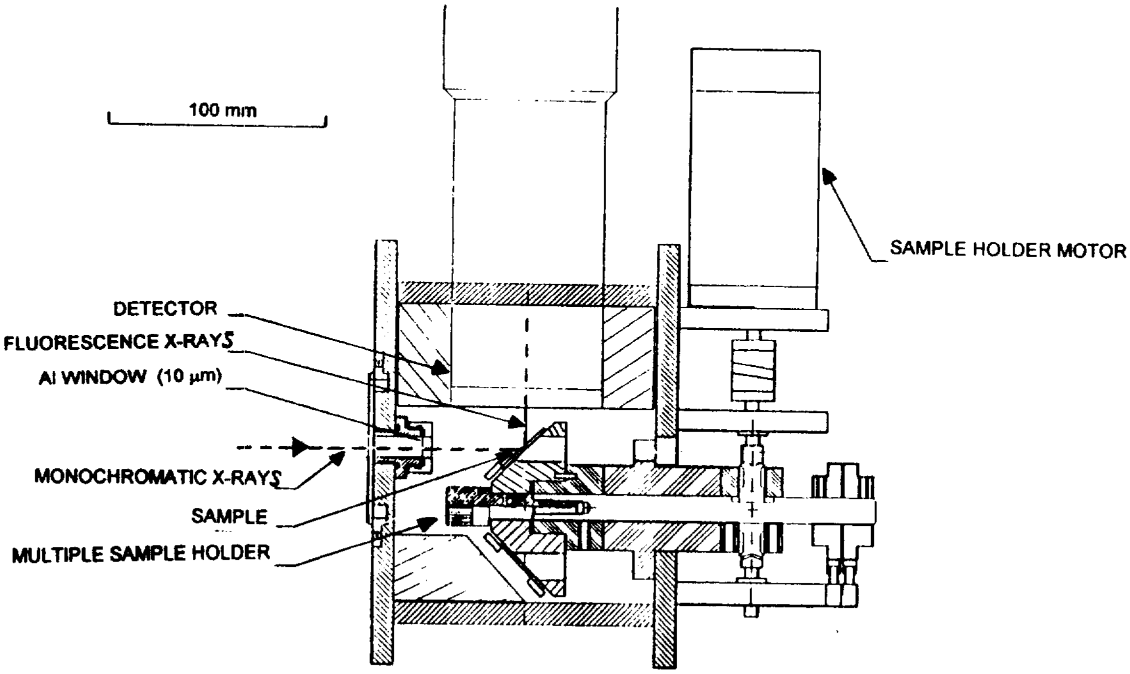


FIG. 4 - XRF chamber installed at KN 3000 VDG.

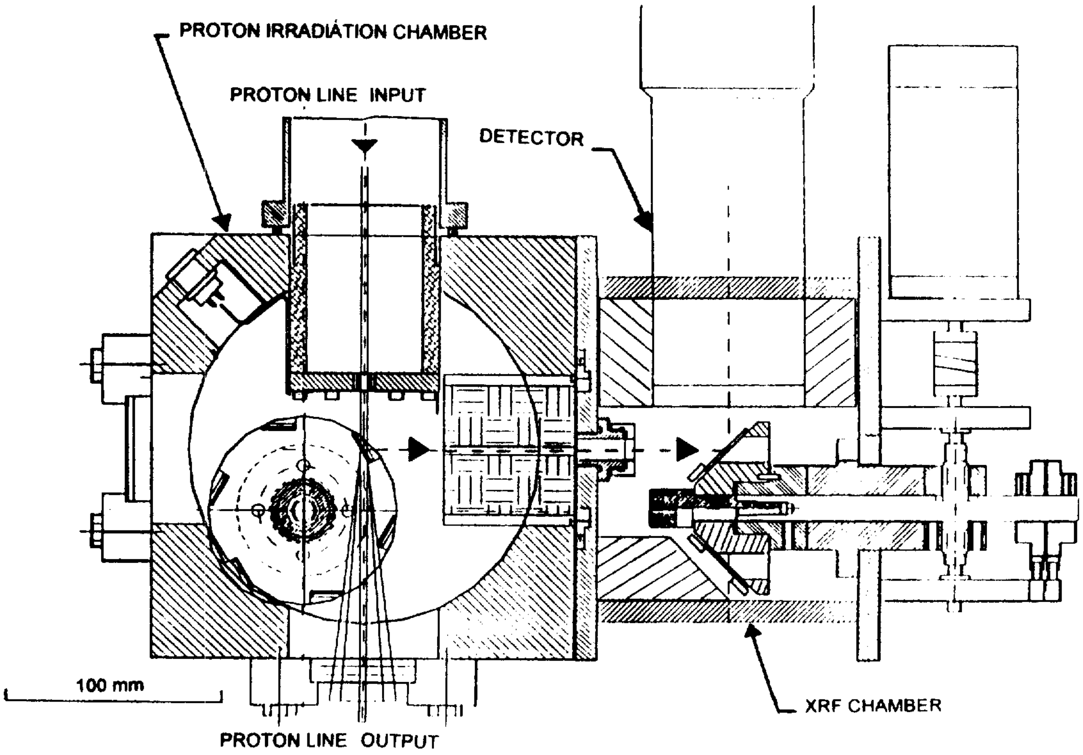


FIG. 5 - Experimental set up for PI-XRF measurements.

3. - MEASUREMENTS AND RESULTS

X-ray yield: The evaluation of the source intensity was the first object for the characterisation of the x-ray beam and was accomplished putting the detector directly in front of the exit window at a distance of about 60 cm from the target to avoid saturation. The yields obtained from the measures on 10 different targets are shown in Fig. 6 and confirm the calculated values(4). In the same figure the PI-XRF and XRF yields are compared referring to the standard operating values for proton current (VDG 2.8 MeV - 10 μA) and electron currents (x-ray tube 25-40 kV - 10 mA). XRF data were obtained with the secondary anode (SA) XRF spectrometer installed in the IFGA X-Ray Laboratory which is sketched in Fig. 7. We can see that, despite of use of low energy protons, the PI-XRF yields are still sufficiently high. Especially when we employ targets with low atomic number we obtain a great advantage on the performances of a standard SA-XRF system (from one order of magnitude for Sr to three order of magnitude for Fe).

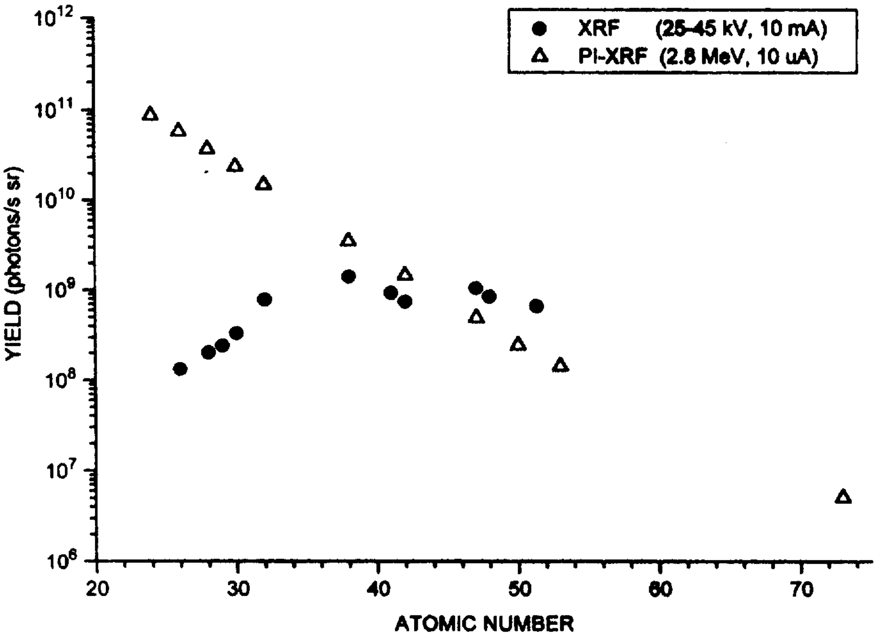


FIG. 6 - Experimental yield values obtained with PI-XRF and AS-XRF systems.

Monochromaticity: The monochromaticity of the source can be improved by the use of the appropriate β-filter for each target. Exploiting the higher intensities from low Z materials it is possible to increase the Kα/Kβ ratio up to values of about 100 still keeping the yield values higher than the corresponding ones obtainable from standard XRF system.

Background: A further improvement in the monochromaticity and in the overall quality of the x-ray source is due to the virtual absence of scattered radiation in the spectra. In fact with SA-XRF system the x-ray tube radiation, besides exciting the characteristic fluorescence of the secondary anode, is scattered coherently and incoherently especially from low Z targets.

This causes a relatively high background mostly when using high Z anode tube (W, e.g.) which generate prevalently continuum bremsstrahlung x-rays. On the contrary employing protons to excite XRF radiation from the pure target there is no scattered radiation but only a very small contribution at low energies (< 5 keV) from secondary-electron bremsstrahlung (SEB) and radiative ionisation (RI-QFEB)$^{(6,8)}$. This effect is manifest in the spectra shown in Fig. 8 from three different targets: Fe-Mo-Ag. The PI-XRF peak to background ratio keeps quite constant at 10^3 value while the SA-XRF ratio is about one order of magnitude lower and decreases using high Z targets.

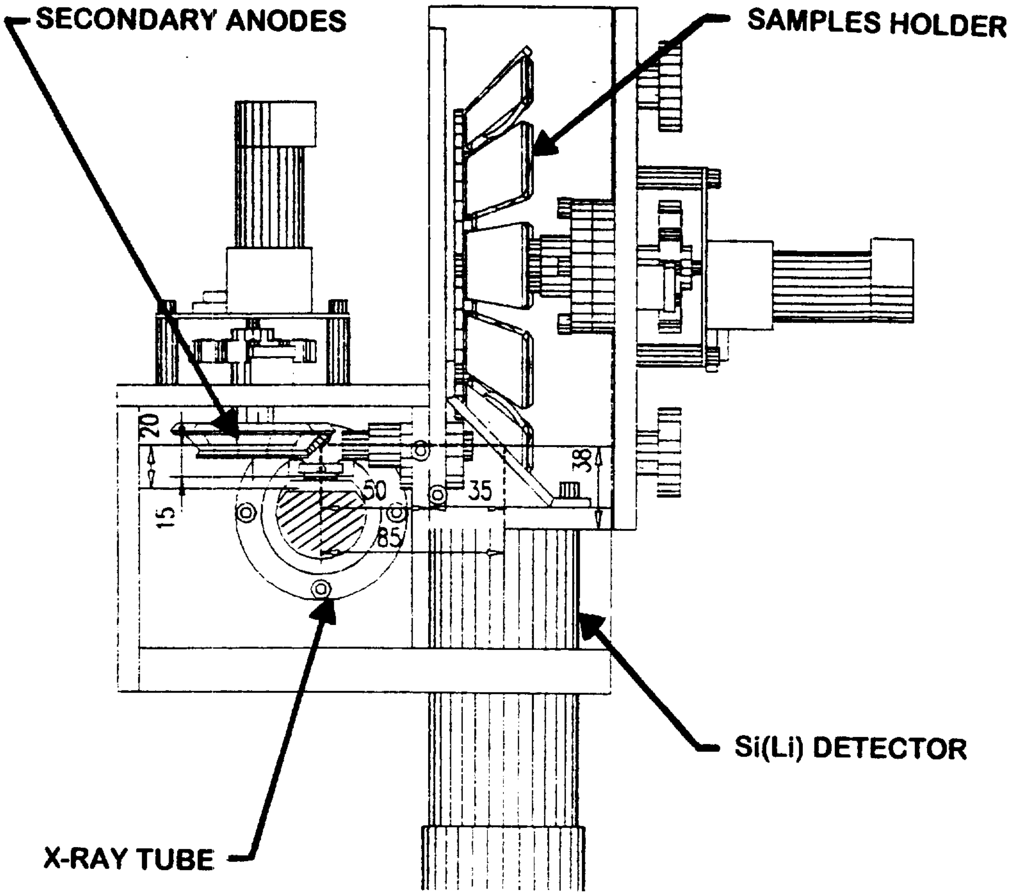
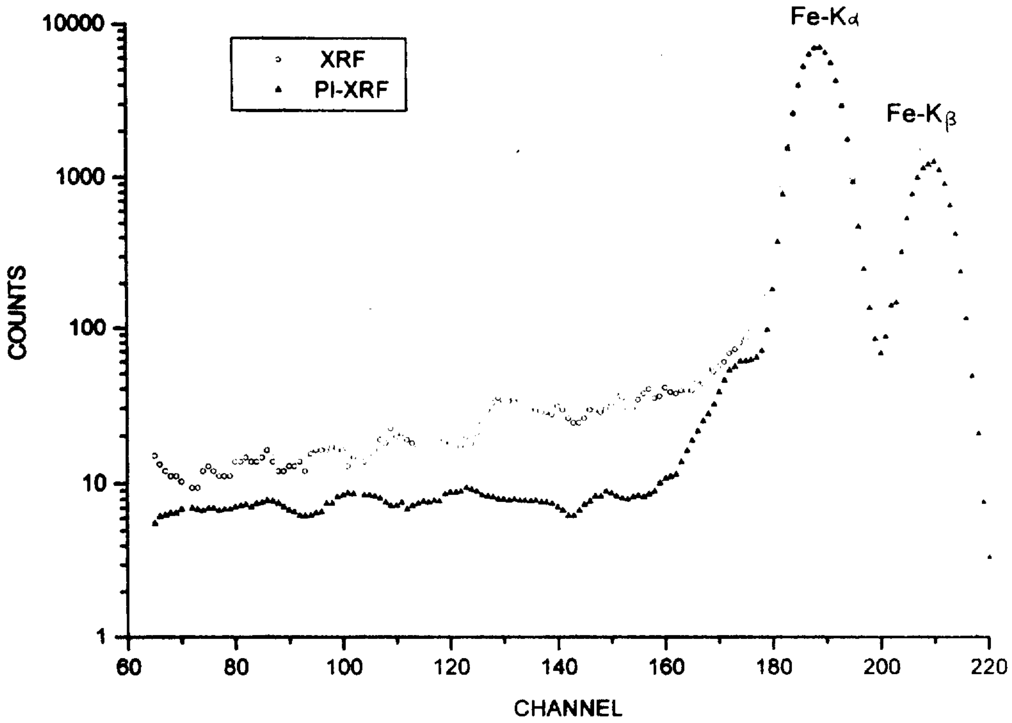


FIG. 7 - AS-XRF spectrometer installed at IFGA X-ray Laboratory.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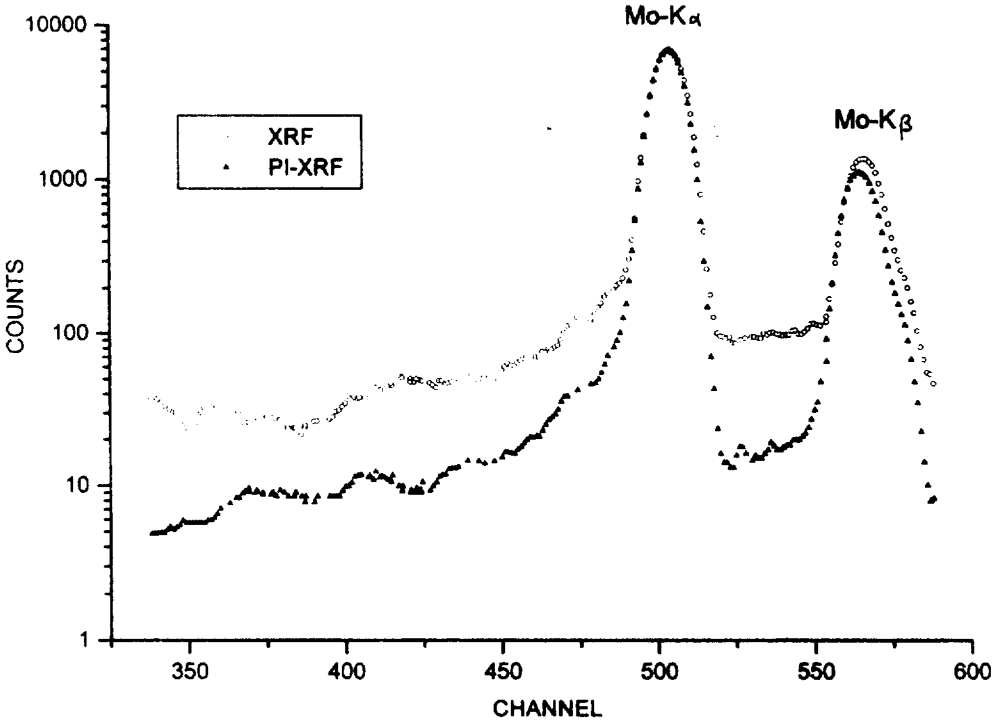
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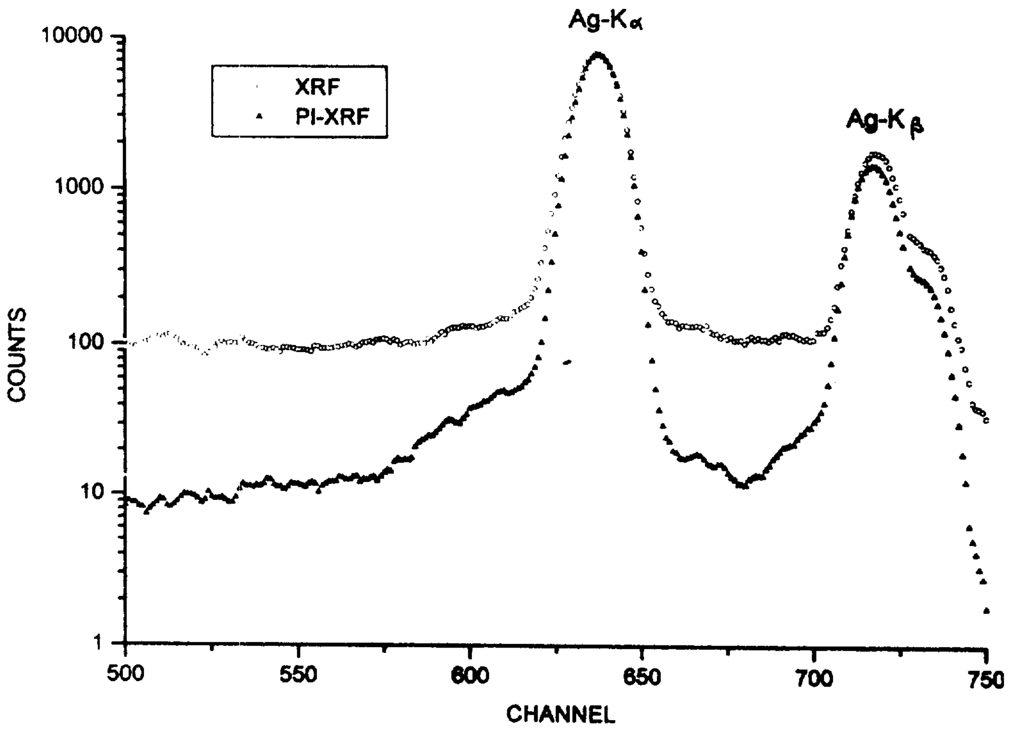

FIG. 8 - Comparison of experimental X-ray spectra obtained with PI-XRF and AS-XRF systems from three different targets: a) Fe; b) Mo; c) Ag.

4. - CONCLUSIONS

We have shown with the measurements at the Ispra cyclotron that the use of protons above a few MeV, in principle attractive because of the steep increase in the ionisation cross-sections with proton energy, is not suitable to produce intense monochromatic x-ray source because of nuclear background radiation interference.

On the contrary the experimental results at Florence VDG accelerator with low energy protons (3 MeV) and low beam currents (1-10 μA) have shown some advantages of the PIXE-induced XRF technique:

- The combined high intensity and monochromaticity of the source with low Z targets could have good applications in the x-ray photoemission spectroscopy (XPS) in the field of surface science. The most commonly used laboratory XPS source are the Kα lines of Al and Mg excited by conventional x-ray tubes. The most appealing feature of the source here discussed should be the low background and the possibility to use targets with higher Z to excite the deeper core levels of the heavier elements(7).
- Also with a low current accelerator we have realised a high brilliance source of monochromatic soft x-rays useful for biomedical applications(7).
- PI-XRF vs. PIXE: Integrating a PIXE facility with a low cost XRF system has some advantages. It is possible to use the high sensitivity of the XRF method, its tunable

monochromaticity and its no-energy deposition in the sample when secondary excitation effects or low sensitivity or possible damage of the sample make the PIXE technique not convenient.

- PI-XRF vs. XRF: The availability of a PIXE facility should suggest to realise a complementary XRF line developing a PI-XRF set-up instead of carrying out a standard XRF spectrometer based on x-ray tubes. This is due to the low cost of the irradiation chambers and the efficiency of the PI-XRF spectrometer higher than the one of SA-XRF especially for low Z primary targets.

For the future, exploiting the high intensity at low energies of the technique, we plan to focalize the proton beam over smaller spots on the primary targets (Φ<1 mm). Using glass microcapillary collimators with inner diameter of 400 μ or less (down to 100 μ), we plan to develop an x-ray source with optical properties well suited for applications to high lateral resolution μ-XRF analysis.

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