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The new external beam facility for environmental studies at the Tandetron accelerator of LABEC

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Abstract

At the new 3 MV Tandetron accelerator of the LABEC laboratory of INFN in Florence, an external beam facility fully dedicated to measurements of elemental composition of atmospheric aerosol has been installed. The experimental set-up hitherto used for this kind of applications has been upgraded with the introduction of a silicon drift detector and of a beam current monitor system based on the backscattering of protons on the extraction window. The new facility will be presented, together with some methodological results obtained so far for the analysis of airborne particulate matter samples collected on quartz fiber filters.

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1. Introduction

IBA techniques and mainly PIXE, are a powerful tool to investigate environmental problems: they are indeed particularly suitable for the study of atmospheric aerosol, which nowadays is the pollutant of major concern in many Italian urban areas. Aerosol sampling campaigns produce huge amounts of few μg particulate samples: fast, quantitative, high-sensitivity and multi-elemental analytical methods are thus required. Non-destructiveness is also an important issue to extend the range of detectable elements by complementary techniques applied to the same sample. Scanning possibility may also be very useful, allowing time trend reconstruction by the analysis of time-sequence aerosol deposits collected by continuous samplers.

In the past 10 years our group has been increasingly involved in extensive programs aiming at the characterizations of particulate matter in urban areas [1–3] or industrial districts [4]. This activity was carried out at a laboratory based on a single-ended 3 MV Van de Graaff accelerator.

Recently, a HVEE 3 MV Tandetron was installed in the new LABEC (LAboratorio BEni Culturali) laboratory of INFN in Florence, established with the main purpose of performing applications of nuclear techniques in the field of Cultural Heritage. However, applications of nuclear techniques also to other fields such as studies on environmental problems (mainly, but not only, air quality monitoring) will be performed at the laboratory. Among the IBA beam lines already operating, one is devoted to environmental analysis; the external PIXE–PIGE set-up which we use for aerosol measurements is an upgrade of the one we have used so far. We have introduced the possibility to measure the beam current detecting the protons backscattered by the Upilex extraction window and we have substituted a more traditional Si(Li) detector with a silicon drift

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detector (SDD) for low-energy X-rays. The aim of this paper is to give a description of the current status of the new set-up and to present some tests we have made on quartz fiber filters normally used for aerosol collection, which are known to give some problems in PIXE measurements.

2. Experimental set-up

The basics of our experimental set-up are described in detail in [4]; here its essential features are shortly recalled. The proton beam is extracted in air through a 7.5 μm thick Upilex window and the aerosol samples are positioned at a distance of about 1 cm from the window, perpendicular to the beam. The beam size is typically 1 mm × 2 mm, but can anyway be easily changed by bare collimation in vacuum in the last section of the beam line.

To analyse many samples in relatively short times, a fast automatic sample changer is used. Two different set-ups allow an easy handling, positioning, changing and scanning of samples collected by sequential samplers or streaker samplers [2] as well. Both the scanning of the filters and the change of the sample are automatically controlled by the acquisition system.

Since X-ray production cross sections range over 3 orders of magnitude, to obtain an efficient simultaneous detection of all the elements it is necessary to balance the counting rates produced by the low and medium-high *Z* elements. The adopted solution is the use of two detectors optimised for low and medium-high X-ray energies, respectively. The latter is a Si(Li) detector (80 mm^2 active area, 3 mm thick, 175 eV FWHM energy resolution at the 5.9 keV Mn K_α line and 6 μs shaping time), placed at 135° relative to the beam direction, at a distance of about 2–3 cm from the target. A Mylar foil of ~400 μm in front of the detector is used to attenuate the low energy X-rays (their production cross sections are higher and also low-*Z* elements are generally the most abundant in aerosol samples).

The low-energy X-ray detector is discussed in detail below, in the following section.

Finally, γ-rays for PIGE analysis are detected by a 60 mm × 23 mm Ge detector, with 28% efficiency and 1 keV FWHM energy resolution at 1.33 MeV, respectively. It may be placed at different positions, but usually it is set at about 6 cm from the target at an angle of 135° to the proton beam. The detector is shielded by means of lead bricks from the γ-ray background arising from the interaction of the protons with elements of the beam line (especially collimators).

3. Silicon drift detector

To detect low-*Z* element X-rays a traditional Si(Li) has been substituted by a silicon drift detector, SDD, which offers an excellent energy resolution at moderate cooling (−10 °C, achievable with a Peltier cell) and can sustain much higher X-ray counting rates than Si(Li) detectors, thus allowing, in principle, to work with higher beam currents [5]. The SDD is placed at about 145° relative to the beam direction, at a distance of about 6 cm from the target; it has 10 mm^2 sensitive area, 300 μm thickness, and 145 eV FWHM energy resolution at the 5.9 keV Mn K_α line and 0.5 μs shaping time. Thanks to the ultra-thin entrance window (8 μm beryllium) and to the use of a helium gas flow into the volume in front of the detector, it can detect with good efficiency X-rays of very low energies, down to ~1 keV (Na K_α line).

A common problem in all PIXE set-ups is the backscattering of protons from the target that perturbs the electronic system, increasing the pile-up very significantly and deteriorating the energy resolution, and finally damages the detector itself. This holds significatively true for the SDD – its Be window and the helium flow in front of it allow the backscattered protons to reach the detector itself – as shown in Fig. 1, where the worsening of the detector energy resolution with increasing X-ray count rate, and hence yield of backscattered protons, is unmistakable. To prevent protons from reaching the SDD without employing any shielding material, which would absorb also the low-energy X-rays, we have mounted in front of the detector a magnetic proton deflector placed at less than 1 cm from the active volume of the detector. The length of the deflector and its magnetic field (0.5 T) are chosen to ensure the deflection of backscattered protons with energies up to 3 MeV. Using the proton deflector, the energy resolution maintains its excellent value up to some kHz count rate, whereas a Si(Li) detector, shown for comparison, ceases working already at about 1 kHz due to unsustainable count rate.

Anyway, the use of the proton deflector is mandatory to avoid damaging the SDD. In Table 1 is reported the calculated dose of backscattered protons reaching the SDD in a measurement on different aerosol collecting substrata in

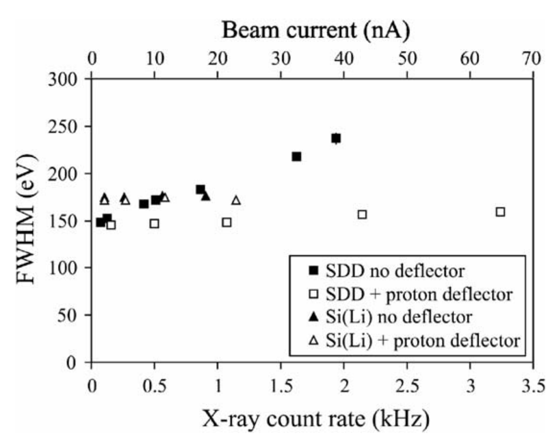

Fig. 1. SDD energy resolution as a function of X-ray count rate, with backscattered proton removal or not. A 50 μg/cm^2 Mn target on a 6.3 μm Mylar foil, whose thickness and composition are similar to those of aerosol collecting filters, was used for the measurement. The results obtained with a Si(Li) are also shown for comparison.

Table 1
Calculated backscattered proton dose in typical measurements on different aerosol collecting substrata

Substratum	Dose (p/cm^2)	Typical measurement conditions
Teflon[a]	0.1×10^6	$E_p = 2.4$ MeV, $I = 5$ nA, $T = 600$ s
Quartz fiber[a]	8×10^6	$E_p = 3.0$ MeV, $I = 5$ nA, $T = 600$ s
Nuclepore[a]	6×10^6	$E_p = 3.0$ MeV, $I = 20$ nA, $T = 600$ s
Kapton/Nuclepore[b]	2.5×10^6	$E_p = 3.0$ MeV, $I = 30$ nA, $T = 180$ s

[a] Daily sample.
[b] Streaker sampler "point"; 168 points in each sample.

our typical experimental conditions. Assuming a conservative limit of 10^9 protons/cm^2 before significative deterioration of the detector performance due to radiation damage [6], and considering the standard load of samples to be analysed in our laboratory in one year (10–15 streaker samples, 1000 daily samples), the SDD would result damaged without proper backscattered proton removal. Note that just the measurement on streaker samples, which can be analysed only with PIXE, would produce a dose beyond damage level.

One of the advantages in using SDD is represented by the possibility of managing high count rates (up to 50 kHz at 0.5 μs shaping time). This implies, in turn, the possibility of using very high beam currents (30–50 nA)

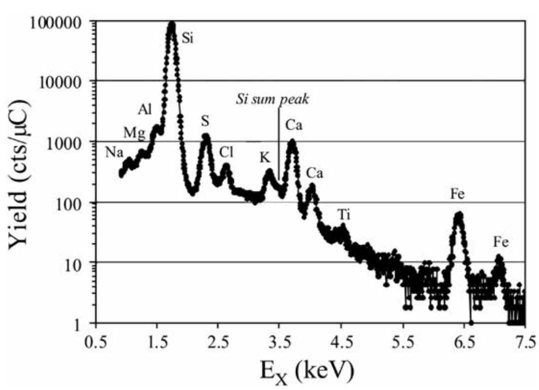

Fig. 2. SDD X-ray spectrum of an aerosol (PM_{10}) daily sample collected on a quartz fiber filter.

thus drastically reducing the measurement time. Nowadays the determination of the contribution of marine aerosols and Saharan dust intrusions to pollution levels in Italy is a still debated important issue. PIXE analysis allows an easy identification and quantification of important tracers of such long-range transport of marine (Na and Cl) and Saharan aerosols (Al, Si, Ca, Ti, Fe, etc.). With a SDD detector PIXE analysis of such elements can be performed in only 2–3 min.

Moreover, airborne particulate matter is often collected on quartz fiber filters since they are suited both for gravimetric measurement of the aerosol mass and for organic carbon and elemental carbon chemical analyses. Unfortunately, the high silicon content of the filters makes it impossible to perform PIXE analysis on low-Z elements with standard Si(Li) detectors. In fact, on these substrata, count rates up to 10 kHz can be easily produced with proton beam currents of a few nA. Thanks to the SDD it is now possible to analyse these samples, as shown in Fig. 2, with a measurement lasting only few minutes at about 5 nA beam current.

4. Beam current monitor

For aerosol samples, the total thickness (aerosol deposit + substrate) is small enough to let the beam pass through; as a consequence the charge can be measured simply integrating the beam current on a Faraday cup placed behind the sample.

The main drawback connected with the use of the Faraday cup in an external set-up (apart from preventing the possibility of placing other detectors behind the sample, i.e. for PESA or STIM analysis [7]) it is the unreliability of its readings at low currents, below 2–3 nA [8]. For this reasons we have implemented a new beam current monitor based on the measurement of the yield of protons backscattered on the exit window.

The device for the beam current measurements was designed in order to reduce as possible its overall dimensions. As a consequence, the detector used (a silicon p–n junction diode having 10×10 mm^2 active area, 300 μm

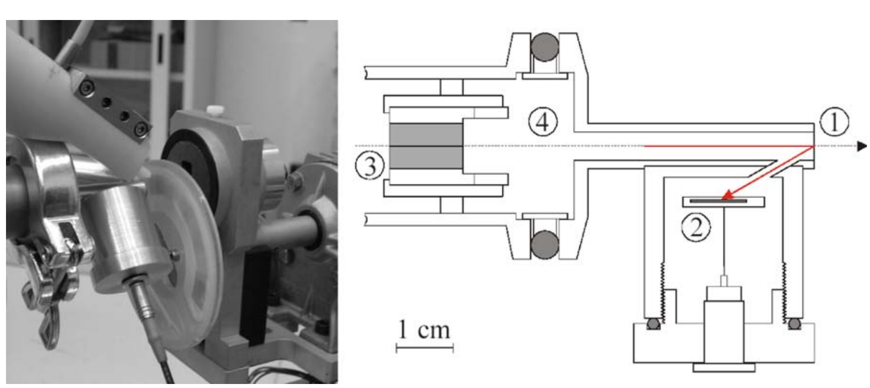

Fig. 3. On the left, picture of the external set-up during a measurement on a streaker sample. On the right, schematic drawing of the device for beam current measurement through particle backscattering: (1) exit window; (2) detector for backscattered particles; (3) collimator; (4) proton beam.

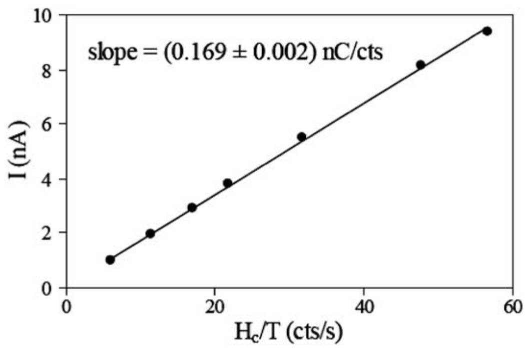

Fig. 4. Beam current versus counts per second and per channel on the plateau left of the leading edge of C in the backscattering spectrum of protons on the Upilex exit window. Also shown are the fit to the data (solid line) and the calibration factor.

thickness, 10 keV FHWM energy resolution) was positioned in vacuum at an angle of 150° with respect to the beam direction, so that its housing structure does not interfere with the positioning of the samples in front of the exit window (Fig. 3).

Protons backscattered by the Upilex extraction window reach the detector through an aperture of 2 mm diameter; the dimensions and position of this hole are such that no protons backscattered by the sample under analysis (placed at 1 cm from the window) can reach the detector. Should backscattering from the sample also be detected, this would in fact produce an incorrect measurement of the beam current since the spectrum would also depend on the local target characteristics and not only on the total charge flown. Anyway, we have checked that such a disturbing effect is absent in our geometry for target–window distances down to 5 mm.

The device has been calibrated by means of a direct and absolute measurement of the beam current, namely by comparison with the X-ray yield from a thin ($\sim$150 $\mu g/cm^2$) Au reference target, which has in turn been normalized to a Faraday cup reading at high beam currents.

The data obtained for 3 MeV protons is reported in Fig. 4, where the beam current is inferred from the height, H_C, of backscattering spectrum averaged over 10 channels situated just on the left of the carbon leading edge; however, one can consider a larger energy region over which integrate the counts, paying attention to not integrate over the channels close to the trailing edges; in fact, even though generally the thickness of the exit window remains constant during a measurement, a possible beam induced thinning of the exit window results in a shift of the trailing edges. This effect can however be controlled during the acquisition and, when starting to show up, the exit window is replaced.

5. Conclusions

The new external PIXE–PIGE beam line dedicated to environmental analysis at the 3 MV Tandetron accelerator of the LABEC laboratory of INFN in Florence was put in operation and some test measurements were performed. The new set-up offers the possibility to analyse in very short measuring time various type of samples, including quartz fiber samples thanks to the introduction of a SDD for low-Z elements detection.

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