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A new small-footprint external-beam PIXE facility for cultural heritage applications using pulsed proton beams

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ABSTRACT

In the framework of the COBRA project, elemental analyses of cultural heritage objects based on the particle induced X-ray emission (PIXE) are planned in a collaboration between the APAM laboratory of ENEA-Frascati and the LABEC laboratory of INFN in Florence. With this aim a 3–7 MeV pulsed proton beam, driven by the injector of the protontherapy accelerator under construction for the TOP-IMPLART project, will be used to demonstrate the feasibility of the technique with a small-footprint pulsed accelerator to Italian small and medium enterprises interested in the composition analysis of ancient artifacts. The experimental set-up for PIXE analysis on the TOP-IMPLART machine consists of a modified assembly of the vertical beam line usually dedicated to radiobiology experiments: the beam produced by the injector (RFQ + DTL, a PL7 ACCSYSHITACHI model) is bent to 90° by a magnet, is collimated by a 300 μm aperture inserted in the end nozzle and extracted into ambient pressure by an exit window consisting of a Upilex foil 7.5 μm thick. The beam is pulsed with a variable pulse duration of 20–100 μs and a repetition rate variable from 10 to 100 Hz. The X-ray detection system is based on a Ketek Silicon Drift Detector (SDD) with 7 mm^2 active area and 450 μm thickness, with a thin Beryllium entrance window (8 μm). The results of the calibration of this new PIXE set-up using thick target standards and of the analysis of the preliminary measurements on pigments are presented.

1. Introduction

The ENEA Frascati research center develops advanced diagnostic tools and innovative research methods to characterize materials and identify new treatments for the conservation of cultural heritage.

The COBRA project (“Sviluppo e diffusione di metodi, tecnologie e strumenti avanzati per la COnservazione dei Beni culturali, basati sull’applicazione di Radiazioni e di tecnologie Abilitanti”) is aimed at the improvement of ionizing and non-ionizing radiation research activities to support innovation in the Cultural Heritage field. Indeed, the works of art themselves contain the information, for example, on their production techniques, their conservation state and on their usage, thus needing an in-depth study of their composition.

In the field of Cultural Heritage diagnostics, the non-invasive and non-destructive methodologies are the most widely used: diagnostic tests, examinations and measurements have to be performed using methods that do not damage the materials and do not need any sampling.

Non-destructive elemental studies can benefit from the use of ion beam based analysis methods like particle induced X-ray emission (PIXE), backscattering spectrometry and other nuclear reaction analysis where gamma-rays or charged particles are detected. Several studies have been carried out exploiting these complementary methods simultaneously, obtaining a deep insight on the material composition, as widely reported in the literature [1].

In the framework of the COBRA project, a system for ambient pressure PIXE analysis has been realized using proton beams of energy between 3 and 7 MeV. The variable energy allows performing PIXE measurements with protons of different energies (“differential” PIXE technique [2]), in order to obtain information about

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the depth sequence of different elements within the analyzed sample.

This paper reports the characterization and the first measurements with the external beam PIXE system based on a small-footprint proton pulsed accelerator carried out in collaboration between the research laboratories of ENEA-APAM (Development of Particle Accelerators and Medical Applications) and INFN-LABEC (Laboratory of Nuclear Techniques for the Environment and the Cultural Heritage), in the framework of the COBRA program.

2. Experimental set-up

2.1. The particle accelerator

The ENEA-APAM laboratory develops particle accelerators designed to medical applications [3].

The TOP-IMPLART (Terapia Oncologica con Protoni – Intensity Modulated Proton Linear Accelerator for Radio Therapy) facility [4] is a pulsed linear proton accelerator under construction at ENEA Frascati research center and consisting on a 425 MHz injector followed by a 3 GHz booster [5,6].

The injector is a 7 MeV commercial machine (Hitachi-AccSys PL-7 model) composed by a 30 kV Duoplasmatron source followed by two accelerating structures, a radiofrequency quadrupole (RFQ) up to 3 MeV and a drift tube linac (DTL) up to 7 MeV. The pulse width and repetition frequency are variable up to 60 μs and 60 Hz, respectively. The current can be varied by changing the voltage on an einzel lens placed before the RFQ.

A proton beam with continuously variable energy from 3 to 7 MeV can be delivered for low energy experiments in air at the end of a dedicated line: a large 90° bending magnet is inserted on the transport line after the injector and a couple of quadrupoles for a vertical output typically devoted to low doses radiobiology experiments [7,8]. The proton beam energy resolution is around 100 keV, allowing PIXE and eventually PIGE measurements, but obviously not suitable for RBS analyses. The overall length of this accelerator system (ion source, RFQ, DTL and transfer line) is only 6 m.

2.2. The external beam PIXE set-up

The TOP-IMPLART low energy vertical beam line has been arranged in order to reduce the exiting beam spot from 133 mm^2 to 0.07 mm^2. The final part of the beamline is 20 cm long and ends with an aluminium beam exit nozzle. The proton beam passes through a 300 μm graphite collimator (tenths of mm length, inserted directly in the exit nozzle and shielded by a Ta diaphragm), which defines the transverse dimension of the beam in vacuum, and is extracted into air through a 7.5 μm thick Upilex-S ($C_{22}H_{10}N_2O_4$, density 1.47 g/cm^3) foil glued on the exit nozzle [9]. A support connected to the beamline, with an aperture of 1 cm diameter to easily let the beam pass, is used as sample holder to allow horizontal positioning of the samples at 1 cm from the Upilex extraction window (Fig. 1).

The beam size at the sample position after the 1 cm air-gap is about 1.5 mm diameter as measured by the darkening of a HD-V2 radiachromic film [10], conventionally used for high dose dosimetry, realized with the uncovered active polymerization layer and then suitable for the imaging of low energy and not very penetrating protons beams.

The exit nozzle is insulated from the rest of the beamline and the intensity of the proton beam is monitored by the pick-up of the current signal on the graphite collimator.

Work parameters used during the PIXE measurements are summarized in Table 1. The pulse length and average current impinging the samples are 50 μs and about 10 pA, respectively.

The characteristic X-rays emitted by the sample are detected by a Silicon Drift Detector (SDD) supplied by KETEK GmbH, with an active thickness of 450 μm and 10 mm^2 size, collimated to 7 mm^2 by means of a multilayer collimator, equipped with an 8 μm thick Be entrance window (nominal thickness). The measured energy resolution is 160 eV FWHM at the 5.9 keV Mn Kα line with 1 μs shaping time. The detector is placed at 135° with respect to the beam direction, at a distance of about 1.5 cm from the target. It has to be noted that an absorbing layer of 15 mm of air at ambient pressure will attenuate completely Na X-rays and transmit 3%, 11% and 25% of Mg, Al and Si X-rays, respectively.

3. PIXE analysis and discussion

The in-air PIXE system was calibrated for quantitative analysis by irradiating with 2.75 MeV protons (energy on target) thick pure elemental samples (SiO_2, Ti, Cu and Mo, supplied by Goodfellow), followed by analysis of the resulting spectra with the GUPIXWin software [11] to determine the H-factor free parameter, using the peak area of the K X-rays of the Ar component of the air as normalization factor [12]. A constant H-value ($1.5 \cdot 10^{-7}$ in arbitrary units) not dependent on the X-ray energy was determined and it was used in the analysis of a multi-component glass standard (NIST SRM 1412), obtaining a composition of the detectable element oxides (from MgO to PbO) in agreement with the certified values within 10% (Fig. 2). Typical proton beam current values ranged from pA to few tens of pA, in runs lasting typically 300 s. Since the beam is pulsed, instantaneous beam intensities reached up to few nA, causing the appearance of pile-up peaks in the X-ray spectra of the thick pure elemental samples, while the effect resulted negligible for the other samples (multi-component glass standard and pigments). Indeed the use of SDD, having good energy resolution yet at short shaping times, allowed to sustain high count rates [13,14] and to mitigate pile-up effects.

The feasibility of PIXE measurement with the present in-air set-up for cultural heritage studies was demonstrated by analysing three different samples of pigment layers (e.g. red, brown and blue) applied on a gypsum preparation layer. The PIXE spectra obtained at 2.75 MeV protons were compared with those acquired with the XRF portable spectrometer designed at INFN LABEC [15] using a Mo anode (20 kV voltage and 100 μA anode current) and a 17 mm^2 Amptek SDD, showing the good complementary of the two systems in terms of sensibility to low-Z and high-Z elements respectively (Fig. 3).

The red pigment was cinnabar (HgS), whereas the brown pigment was made of ochre (natural earth pigment containing hydrated iron oxide, $FeO(OH)\cdot nH_2O$). The blue pigment was azurite (copper carbonate, $Cu_3(CO_3)_2(OH)_2$) and ultramarine blue [16] as confirmed by the PIXE analysis for the presence of light elements (Al, Si, S, K), which were not detected by XRF. In this way, it is not possible to evaluate if ultramarine blue and azurite are mixed together or if they are separated in two different layers. Calcium and sulphur from the gypsum (calcium sulfate dihydrate, $CaSO_4\cdot 2H_2O$) preparation layer are barely detected due to the strong absorption of their characteristic X-rays in the pigment layer (due to its thickness or due to the presence of high-Z elements), except for Ca in the case of the brown pigment; nevertheless Ca could also be present in the natural earth pigment which is the most likely cause for its presence in the spectrum.

The sequence of paint layers can be inferred, although at a semi-quantitative level, from comparison of PIXE spectra collected on the same spot at various beam energies [17], since in a non-

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