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Elemental mapping of large samples by external ion beam analysis with sub-millimeter resolution and its applications

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ABSTRACT

The elemental mapping of large areas using ion beam techniques is a desired capability for several scientific communities, involved on topics ranging from geoscience to cultural heritage. Usually, the constraints for large-area mapping are not met in setups employing micro- and nano-probes implemented all over the world. A novel setup for mapping large sized samples in an external beam was recently built at the University of São Paulo employing a broad MeV-proton probe with sub-millimeter dimension, coupled to a high-precision large range XYZ robotic stage (60 cm range in all axis and precision of 5 μm ensured by optical sensors). An important issue on large area mapping is how to deal with the irregularities of the sample's surface, that may introduce artifacts in the images due to the variation of the measuring conditions. In our setup, we implemented an automatic system based on machine vision to correct the position of the sample to compensate for its surface irregularities. As an additional benefit, a 3D digital reconstruction of the scanned surface can also be obtained. Using this new and unique setup, we have produced large-area elemental maps of ceramics, stones, fossils, and other sort of samples.

1. Introduction

The use of ion beam analysis (IBA) techniques in external beam setups appeals to many applications, mostly because it is very convenient in practice. Among the strongest benefits is the virtual absence of sample preparation, vastly increasing the variety of specimens that can be analyzed by including delicate materials and also those where dehydration and outgassing in the vacuum environment are of concern. Additionally, in air measurements, when compared to in-vacuum, greatly reduce sample charging and heating, facilitate sample handling and positioning, and meet constraints of non-destructiveness [1].

Since it permits the direct analysis of objects, with no need of sampling, many applications of external IBA address studies on cultural and archaeological artifacts. However, the applications in such areas are extremely challenging in many aspects, mainly because most of the objects of artistic or historical interest have quite heterogeneous structures. When not properly treated, the analysis of heterogeneous samples can easily result in a biased determination of the compositions [2]. This pushes the limits not just for the performance of the equipment, fostering many developments [3–6], but also for the data processing and modeling, promoting a series of innovations [7–9,2].

Considering the case of single spot analysis, the information of different phases of a material may be affected by the probe size since it can be smoothed out when using too large probe sizes, or may not be representative of the real composition of interest if the probe size is too small [10]. However, IBA methods when coupled to mapping capability provide highly meaningful composition maps enabling a better understanding of the space distribution of elements, and an improved characterization of the objects [10]. The use of statistical tools (like principal component analysis, dynamic analysis, multivariate curve resolution, etc.) to process the space distribution data, help clarifying questions related to mixtures and underlying layers. Besides, the use of elemental maps strongly contributes to a common base to share knowledge among researchers of different areas, which is a very attractive aspect for a tool to be used in a multidisciplinary work.

Concerning the eventual damage of the sample caused by the ion beam irradiation, PIXE mapping has been demonstrated a valuable tool, even for sensitive and irreplaceable objects, like paintings [11]. There

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are ongoing studies aiming at a better understanding of possible damages of the pigments and investigations on strategies for its mitigation at doses typically used in IBA techniques [11–13]. Nevertheless, the advantages of using PIXE are unquestionable, as demonstrated by Calligaro et al. [11]. Comparing elemental mappings obtained by X-ray Fluorescence (XRF) and PIXE, it was found a clear superiority of the latter in the detection of some elements (for details see reference [11]). In spite of XRF having the advantage of an easily movable equipment.

Despite mapping capabilities using proton beams with micrometer dimensions having attracted great attention in the last 30-years, Grassi et al. [10] showed several interesting problems analyzed with broad sized proton beams. In the present work, we demonstrate some cases where several characteristics have been fully revealed thanks to the macroscopic scale analysis. As examples we mention the composition of pigments on decorative tiles to determine their origin, or the chemical substitution of the original composition promoted by micro-organisms during fossilization processes.

Mapping large areas can be accomplished with sub-millimeter dimension beams by moving the object in front of the beam [10], however, two drawbacks must be overcome. The first one is the need of compensation for beam instabilities, that may arise during long scanning times. For this, it is necessary to identify some signal proportional to the integrated dose and use it for normalizing the data. The second one is related to the risk of damages induced by the ion beam. In this case, the setup should allow simultaneous measurements with multiple techniques, maximizing the information obtained and minimizing the time of irradiation, hence a possible damage.

Here, we discuss the characteristics and the performance parameters of the external beam setup that was built and installed at the Laboratory for Material Analysis with Ion Beams of the University of São Paulo (LAMFI-USP). The setup combines a sub-millimeter beam with a large range XYZ robotic stage (60.0000 cm) with high-precision/reproducibility (0.0005 cm) XYZ computer controlled stage. Although the mapping of large areas has already been reported [6] elsewhere, despite differences in approaches of the mapping procedure, here we demonstrate that a 3-axis stage is necessary to continuously correct the position of the sample relative to the exit window to compensate for sample irregularities, thus avoiding artifacts in the composition maps.

2. Methodology

The external beam line built at LAMFI-USP, is a versatile setup for multi-technique analysis by in-air IBA. With continuous improvements, it is becoming a robust multi-analytical station for major and trace elements analysis ($Z > 5$). In this section, we describe its specifications and its imaging capabilities.

2.1. Beam transport

The LAMFI-USP is a laboratory fully dedicated to material analysis. It uses a NEC-5DSH tandem accelerator, with a nominal maximum accelerating voltage of 1.7-MV. Two ion sources, an RF alphatross and a SNICS-II, provide H, He, and heavier ions. The external beam setup is installed at the 30°-line fed by a switching magnet. A standard doublet of magnetic-quadrupole-lenses is located approximately at 2/3 of the distance between the switching magnet and the external beam setup.

2.2. The beam exit window

In the initial stage of design and testing, the beam exit window was made of a 3 μm thick Kapton foil. However, the degradation of this material did not allow long irradiation periods required for large-area mapping. Hence, after testing several materials, we adopted 6 μm thick Aluminum foils as beam exit window, taking advantage of the mechanical stability, heat conductance and radiation hardness that provide long life-times, while the γ produced by the $^{27}Al(p,\gamma)^{27}Al$ reaction at 844-keV and 1014-keV can be conveniently used for integrated dose measurement (see Section 2.5 on dose control).

One of the inconveniences of choosing a 6 μm thick Aluminum foil as an exit window is the beam energy spread at the analyzing spot. For example, an internal proton beam with 2.6-MeV energy (a typical value used in our setup), emerges with 2.38 MeV and 15-keV FWHM energy spread at the analyzing spot (see Section 2.3), of which 68% is the energy loss straggling in the Aluminum foil according to SRIM2003 calculations [14]. This affects the resolution of EBS (Elastic Back-scattering Spectrometry) measurements but, it enables very-long irradiation periods with no significant damage of the beam exit foils. For PIXE analysis the beam energy straggling is not a concern due to the smooth and slow dependence of ionization cross sections on the beam energy.

Another inconvenience could be a background in the PIXE spectra, when compared to the case of a Kapton window [15], due to the γ-rays produced in the foil and reaching the X-ray detector either directly or after interacting in the surrounding materials. We did not observe a significant degradation of the PIXE detection limits.

Replacement of the Aluminum foil by a thin silicon-nitrite window is being considered in a next upgrade of the setup, depending on its resilience and on the implementation of a different dose control method. If long irradiation proves to be impractical with silicon-nitrite windows, new materials shall be tested. Good results have been reported using Chromium coated on self-supported silicon-nitrite films as a beam exit window, presenting an increment of 54% in the burst-pressure (highest differential pressure supported by the film) and a better power dissipation when using high-current electron beams. Performance tests for proton beams still have to be done but the results observed using electron beams are promising [16].

An internal collimator with a circular hole is installed before the beam exit window. Available options are 0.1, 0.3, 0.5 and 1.0 mm diameter. All measurements presented in Section 3 were done using the 1.0-mm collimator.

As a safety measure, we installed an automatic system to close the pneumatic valve to prevent serious damage to the accelerator and the ion sources due to sudden ruptures of the beam exit window. The valve is located near the switching magnet, almost 4 m away from the exit window, and closes when the vacuum gets worse than 1×10^{-4} Torr in a region near the exit window.

2.3. Detectors configuration

The exit window and the detectors are mounted in a solid self-aligned end station with 7 slots to hold the detectors or other devices, all facing the analysis spot as shown in Fig. 1. The detectors are mounted in a star-like configuration, optimized to allow the simultaneous acquisition of data with different techniques during the ion beam irradiation, see Fig. 1. A similar setup can be found in [17]. The current available techniques are: PIXE, EBS and IBIL. PIGE can also be employed, and can be performed simultaneously, but the γ-ray detector is mounted further away. All detectors, with exception of the γ-ray one, are geometrically shielded from the signals directly originating from the beam exit window.

Two Si-PIN detectors (both are Amptek XR-100CR - with a 12.5-μm thick Beryllium window, 4.4-mm^2 active area, 500-μm depletion depth and 145-eV resolution at the Mn-Kα line) are dedicated to PIXE measurement (see schematics in Fig. 2(a)). An upgrade to the next-generation technology of silicon-drift detectors (SDD) is planned in a future development phase. One of the detectors (on the right side of Fig. 1) is optimized to measure low energy X-rays (for elements with $13 < Z < 30$). Its solid angle is restricted to approximately 2-msr by a conical collimator, and is kept in rough vacuum by a 3-μm Kapton foil that seals the collimator. This conical collimator was designed not just to reduce the X-rays path in air from the sample to the detector, but also to shield the detector from X-rays not produced by the sample, e.g. mitigating the x-rays counts from Argon in the air. The other detector

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