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High energy PIXE: improvement of the accuracy of K X-ray production cross sections measurements

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Abstract — High-energy PIXE is a powerful non-destructive method of analysis for the study of ancient paintings. In order to apply this method effectively, it is important to know the X-ray production cross section of a given element when irradiated by a charged particle beam. Due to a lack of published experimental data, theoretical models developed at low energies have not been validated for energies above few MeV. A campaign of experimental measurements of X-ray production cross section was carried out for different elements. Precise knowledge of the various experimental parameters is necessary to obtain a high accuracy in the measurements.

1 Introduction

The investigations on art objects must answer several questions: authenticity, provenance, deterioration and conservation [1]. For example, the study of the pigments in a painting will provide information on its age and origin [2]. Indeed, the composition of the pigments changed over the centuries. Knowing the characteristics of the pigments used, it is possible either to create a database of an artist's palette during its evolution, or to have information on the authentication of an art work by referring the artist's existing database [3]. An in-depth analysis will give information on the different layers of paint to study the painting techniques, but also on the possible deterioration and on the authenticity of the artist's usual techniques [4, 5, 6].

In order to carry out these different analyses, it is possible to use the Particle Induced X-ray Emission (PIXE) [1] technique with high energy beam (HE-PIXE). This technique allows the identification and quantification of inorganic pigments with the exception of the ones containing only light elements [6]. The PIXE method consists in the detection of X-rays emitted by ionized or excited atoms following irradiation with an ion beam. The emitted X-rays are characteristic of the elements constituting the analyzed object. Thus, a quantitative multi-elemental analysis could be performed. A High-energy beam of several tens of MeV can penetrate very deeply inside an art object [7]. With high energy beam, the cross section (probability) of production of energetic X-rays (like K X-rays and L X-rays for medium and heavy elements) is important. These more energetic X-rays will be less attenuated in matter. Therefore, the lower attenuation of the X-rays and the longer path of the ions through matter makes it possible to perform an in-depth analysis of painting layers.

The use of the PIXE method requires on one hand a very good characterization of the experimental parameters, the detector efficiency, the beam energy and its intensity and on the other hand to have access to the fundamental parameter related to the ion-atom inter-

action such as the cross section of K-shell ionization. The latter can be determined by a theoretical model such as RECPSSR initially developed and validated in the case of a low energy beam in the MeV range [8]. However, this model has not yet been validated with high-energy ion beams due to the lack of experimental data.

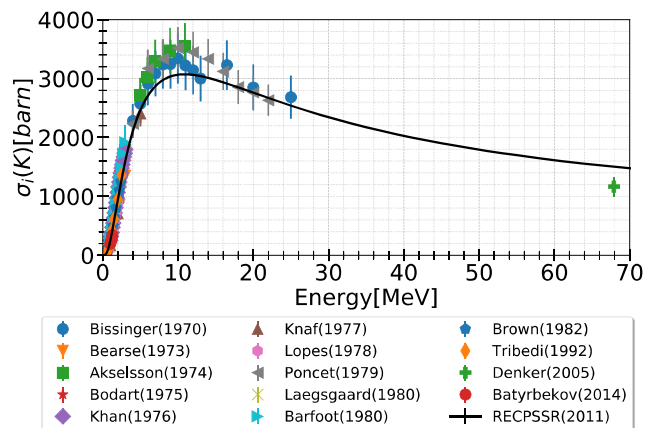


Figure 1: K-shell ionization cross section of titanium. Experimental points [10, 11, 12, 13, 14, 15] ($\omega_K = 0.2148$ [9]) and theoretical model RECPSSR [8]

We have started to measure the K X-ray production cross-sections of Ti, Cu, Ag, and Au with high energy proton beams between 30 MeV and 68 MeV have been performed. To compare with the theoretical model, the X-ray production cross section should be converted to K-shell ionization cross section by using the fluorescence yield ω_K [9]. This latter takes also into account the auger electron yields. Figure 1 shows the K-shell ionization cross section in function of proton energy. The points correspond to the existing experimental data, and the line to RECPSSR model. This shows that high-energy measurements are necessary to complete the database, and to validate the theoretical model. The present document aims to explain the method implemented to measure the K X-ray produc-

tion cross section, the analysis of the X-ray spectra and the detector efficiency.

2 Experimental set-up

The measurements of X-ray production cross sections were carried out at the ARRONAX cyclotron [16], using a high energy proton beam between 30 MeV and 68 MeV. The experimental set-up was described in more detail in Ref. [17]. The beam is focused in order to have an irradiation surface of the order of 1 cm^2 . The intensity was chosen around 100 pA for an irradiation time of a few minutes. During the experiments, the detector dead time is kept below 10% to ensure a linear response. The beam passes through a $50 \mu\text{m}$ thick Kapton exit window, then a 20.5 cm layer of air before reaching the target. The targets are thin foils (approx. $10 \mu\text{m}$) oriented at 45 degrees with respect to the axis of the beam. A Faraday-cup beam stop is positioned at 15.0 cm from the target. This device measures the beam current with a dedicated electronic. At the energies used, the loss of energy in the air and in the target is negligible and the lateral straggling low enough that the entire beam reaches the Faraday cup. Finally, an X-ray Silicon Drift Detector (SDD) is installed normally to the target surface at a distance of 10.3 cm (see Figure 2).

Regarding the experimental conditions (thin target), the K X-ray production cross section can be determined using the following equation:

$$\sigma_P(K_i) = \frac{N_X(K_i)}{N_P \times \varepsilon(K_i)} \times A \quad (1)$$

$$\text{With: } A = \frac{M \times \frac{\mu}{\rho}(K_i) \times \cos(45)}{N_A \times (1 - \exp(-\frac{\mu}{\rho}(K_i) \times \rho \times d))}$$

Where N_X is the number of detected X-rays, N_P the number of incident particles, ε is the X-ray detection efficiency, and $\frac{\mu}{\rho}$ is the attenuation coefficients.

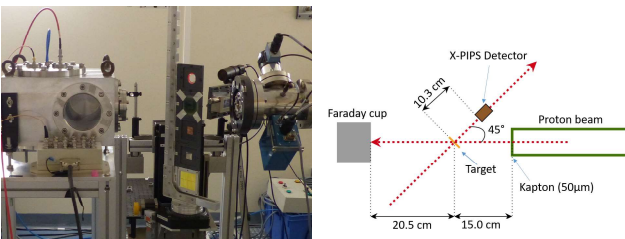


Figure 2: Experimental set-up for measurements of K X-rays production cross sections

3 X-ray spectrum analysis

Several targets were used with the experimental set-up presented above in order to measure the ionisation cross section for different elements. Figure 3 shows an example of an X-ray spectrum obtained with the experimental set-up for a titanium foil of (9.95 ± 0.09)

μm thickness bombarded with a 35 MeV proton beam.

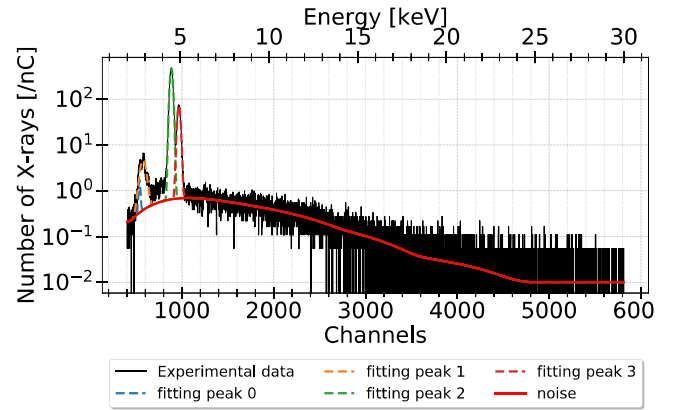


Figure 3: X-ray spectrum of a thin foil of Titanium, bombarded by a 35 MeV proton beam

Spectrum analysis aims to identify characteristic peaks and determine their areas. It is then necessary to estimate the background, mainly composed of Compton scattering from gamma rays emitted in the whole materials (beam lines, beam stop, exit window, collimator ...) and Bremsstrahlung radiation from secondary electron decelerations in matter. The estimation of the background is based on the Statistics-sensitive Nonlinear Iterative Peak-clipping algorithm (SNIP) [18, 19]. In the first step, the algorithm reduces the fluctuation of channels with low statistics by smoothing the spectrum. Then the second step consists in removing the different peaks. The result obtained is shown in Figure 3 with the thick red line.

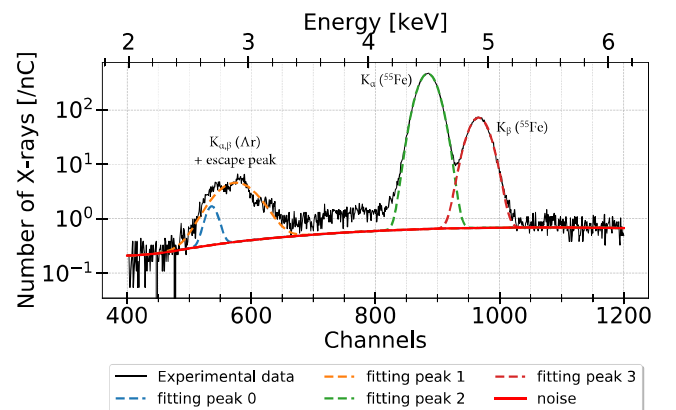


Figure 4: X-ray spectrum of a thin foil of Titanium, bombarded by a 35 MeV proton beam, Zoom on peak adjustment

Once the bottom has been estimated, it is now possible to determine the position of the peaks and to use an adjustment function on each of them to determine the area below the peak. In figure 4, the result of the fitting of the different peaks is represented by colored dotted lines.

Finally, the last step consists in correcting the number of events obtained by the dead time of the detector.

4 Detection efficiency

A Silicon Drift Detector (SDD) from the Canberra/Mirion company was used in this set-up. The manufacturer's characteristics are presented in the Table 1. In order to verify this information, the detector was scanned using an X-ray tomography (Figure 5).



Figure 5: Scanner image of the detector by an X-ray tomography

Nelson Blachman's theoretical model [20] was used to determine the efficiency of X-ray detection. This model takes into account the experimental set-up (detector and source geometry) but also the linear attenuation coefficients extracted from XCOM [21] database.

To optimize the theoretical model, acquisitions were made with a ^{55}Fe radioactive source, produced by LEA CERCA, emitting X-rays of 5.9 keV (K_α) and 6.5 keV (K_β). The absorption of the X-rays in the thin plastic film encapsulating the sealed source was taken into account and the intensity factor provided in reference [22] was applied to weight the two peaks.

Be layer thickness	12.7 μm
Be - Si distance	2.15 mm
Si crystal thickness	0.50 ± 0.02 mm
Si crystal diameter	6.20 ± 0.01 μm

Table 1: Manufacturer's data of X-PIPSTM detector SDD (SXD30M-150-500)

Finally, by taking into account all the previous information, we were able to carry out an optimization of the model parameters. This optimization concerned the detection surface and the dead layer thickness of the detector. The result of the optimization is shown in Figure 6. The black curve corresponds to the theoretical model and the two points to the experimental measurements.

5 Discussion

The estimation of background by the model indicated above can be perturbed by large fluctuations on low-statistical bins. That is why the first step of SNIP algorithm is to perform spectrum smoothing. This step is critical because if the smoothing factor is not correctly

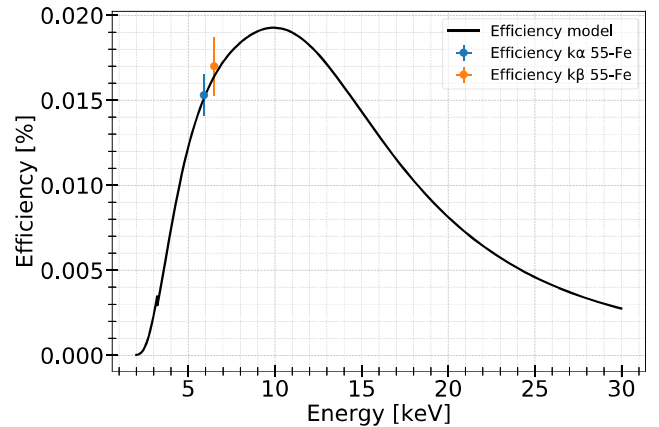


Figure 6: Detection efficiency curve of the X-PIPSTM detector SDD, at a distance of 10.3 cm from the radioactive source

chosen, the tail of the peaks will be modified and therefore leads to a loss of information. The measurement of K X-ray production cross section must be as accurate as possible, an alternative method of determining the background is being investigated. For this purpose, the background of the peak region of a given target would be determined by looking at the background in the same region but on another target whose peaks are not in the same place.

As we can see in Figure 4, using an Gaussian function for peak adjustment is not optimal. With this fitting, there is an underestimation of the peak area. The error is relatively small due to the relatively low statistic, however in order to minimize the uncertainty for the measurement of cross section, another fit function must be found. An Exponentially Modified Gaussian function seems the most suitable to catch up with the tail of the peak.

Acquisition using the X-ray tomography made it possible to verify the characteristics provided by the manufacturer. Nevertheless, the measurement could not be sufficiently accurate to reduce the uncertainties given by the manufacturer. A new acquisition must be carried out in order to determine the characteristics of the detector with a high degree of accuracy.

The use of the ^{55}Fe source allowed constraining the model in its ascending part. New acquisitions must be made to reduce the uncertainties of the efficiency measurement points and to better constrain the model. The acquisition with a ^{109}Cd source emitting X-rays of 22 keV and 25 keV will make it possible to constrain the model in its lower part.

6 Conclusion

The objective is to measure X-ray production cross sections with a high accuracy necessary to perform quantitative HE-PIXE analysis. The methodology applied for the measurement of the X-ray production cross section was presented in this paper. Spectral analysis is

used to identify and quantify the number of X-rays of each characteristic peak of a given element. For this, a method is under development to improve the estimation of background and peak fitting.

The modeling of the efficiency of the detector was optimized in its ascending part thanks to using an X-ray tomography and a radioactive source (^{55}Fe). By using another radioactive source (^{109}Cd), it will be possible to optimize the modeling over the entire energy range.

The knowledge of this information allows us to improve the uncertainties on our measurements of K X-ray production cross section.

References

- [1] Johansson S.A.E., al., Particle-Induced X-ray Emission Spectrometry (PIXE), in Winefordner J.D. (ed) *Chemical analysis: a series of monographs on analytical chemistry and its applications*, vol 133, (1995) Wiley, New York
- [2] Neelmeijer C., al., Depth resolved ion beam analysis of objects of art, *Nucl. Instr. Meth. B118* (1996) 338-345
- [3] Zucchiatti A., al., Building a fingerprint database for modern art materials: PIXE analysis of commercial painting and drawing media, *Nucl. Instr. Meth. B363* (2015) 150-155
- [4] Denker A., al., High-energy PIXE using 68 MeV protons, *Nucl. Instr. Meth. B150* (1999) 118-123
- [5] Denker A., al., Investigation of objects d'art by PIXE with 68 MeV protons, *Nucl. Instr. Meth. B161-163* (2000) 704-708
- [6] Denker A., al., Paintings - high-energy protons detect pigments and paint-layers, *Nucl. Instr. Meth. B213* (2004) 677-682
- [7] Denker A., al., Influence of corrosion layers on quantitative analysis, *Nucl. Instr. Meth. B239* (2005) 65-70
- [8] Lapicki G., al., Universal cross sections for K-shell ionization by low-velocity protons: Importance of relativistic and energy-loss effects, *Phy. Rev. A23* (1981) 2072
- [9] Hazim M., *Mesures des sections efficaces d'ionisations de la couche K induites par des protons de haute énergie pour une large plage de numéros atomiques*, Thesis (2017) Université de Nantes
- [10] Paul H., al., Fitted empirical reference cross sections for K-shell ionization by protons, *Atomic Data and Nucl. data Tables 42* (1989) 105-156
- [11] Kahoul A., al., New procedure calculation fo K-shell ionization cross sections by proton impact, *Rad. Phys. and Chem. 80* (2011) 1300-1311
- [12] Akselsson R., al., X-ray production by 1.5-11 MeV protons, *Zeitschrift für Physik 266* (1974) 245-255
- [13] Poncet M., al., Contribution à l'étude de l'ionisation des éléments par des protons et des particules alpha de 4 à 30 MeV, *Nucl. Instr. Meth. B159* (1979) 455-464
- [14] Bissinger G.A., al., Study of the Production of K X Rays in Ca, Ti, and Ni by 2-28 MeV Protons, *Phys. Rev. A3* (1970) 841-847
- [15] Denker A., al., High-energy PIXE using very energetic protons: quantitative analysis and cross-sections, *X-ray spectrom. 34* (2005) 376-380
- [16] Haddad F., al., Arronax a high-energy and high-intensity cyclotron for nuclear medicine, *Eur. J. Nucl. Med. Mol. Img. 35* (2008) 1377-1387
- [17] Ragheb D., al., Development of a PIXE method at high energy with the ARRONAX cyclotron, *J. Radioanal. Nucl. Chem. 301* (2014) 895-901
- [18] Ryan C.G., al., SNIP, a statistics sensitive background treatment for the quantitative analysis of PIXE spectra in geoscience applications, *Nucl. Instr. Meth. B34* (1988) 396-402
- [19] Morhac M., al., An algorithm for determination of peaks regions and baseline elimination in spectroscopic data, *Nucl. Instr. Meth. A600* (2009) 478-487
- [20] Mohanty B.P., al., Comparison of experimental and theoretical efficiency of HPGe X-ray detector, *Nucl. Instr. Meth. A584* (2008) 186-190
- [21] Berger M.J., al. (2010), XCOM: Photon Cross Section Database (version 1.5), National Institute of Standards and Technology, Gaithersburg, MD.
- [22] Daoudi S., al., Review of experimental photon-induced K_{β}/K_{α} intensity ratios, *Atomic Data and Nucl. data Tables 132* (2020) 101308