

High energy PIXE: A tool to characterize multi-layer thick samples

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ABSTRACT

High energy PIXE is a useful and non-destructive tool to characterize multi-layer thick samples such as cultural heritage objects. In a previous work, we demonstrated the possibility to perform quantitative analysis of simple multi-layer samples using high energy PIXE, without any assumption on their composition. In this work an in-depth study of the parameters involved in the method previously published is proposed. Its extension to more complex samples with a repeated layer is also presented. Experiments have been performed at the ARRONAX cyclotron using 68 MeV protons. The thicknesses and sequences of a multi-layer sample including two different layers of the same element have been determined. Performances and limits of this method are presented and discussed.

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1. Introduction

Analysis of cultural heritage objects is a very active application field for the PIXE method [1–3], especially thanks to its ability to determine their elemental composition in a non-destructive way. In many cases, those objects consist of a stack of many layers (paintings, coins...) with sometimes several layers made of the same element. Differential PIXE [4], using low energy beam, was developed in order to provide depth concentration profile, composition and ordering of the layers. The principle of this technique relies on the comparison between PIXE spectra recorded at different beam energies [5] (e.g from 1 to 4 MeV) or at different incident angles [6]. Complementary information about the target is obtained for each measurement due to the strong variation of the ionization cross sections, at low energy, within the target. It is, therefore, possible to use a de-convolution algorithm to extract the contribution of each layer. Differential PIXE has been used to study multi-layer samples [7] with a thickness up to tens of microns [8].

High-energy PIXE (HEPIXE) is suitable to perform non destructive analysis of thick samples, in normal air, due to the high penetration range of energetic light ions in matter and their ability to excite the energetic K X-rays. HEPixe has already been used in the field of cultural heritage [9–11] and especially for the characterization of ancient paintings with several superposed layers

[12]. Metallic archeological objects can be also investigated without removing the patina layer present on their surface. The investigated thicknesses in those works were between few dozens and few hundred of micrometers. But in this case, contrary to the usual differential PIXE method, ionization cross sections change smoothly with the energy and incident angle variation. However, HEPixe analysis using $\frac{K_\alpha}{K_\beta}$ (or $\frac{K_\alpha}{L_\alpha}, \frac{L_\beta}{L_\alpha}$) ratio provides qualitative information about the thicknesses and the sequences of several layers but assumptions on the sample composition is required [12]. In a recent work [13], we demonstrate the possibility to perform quantitative analysis of simple multi-layer samples without any assumption on their composition. This method, based on the relative variation of $\frac{K_\alpha}{K_\beta}$ (or $\frac{K_\alpha}{L_\alpha}, \frac{L_\beta}{L_\alpha}$) when the sample is rotated, had given good results for patterns of different pure material foils. But in more realistic applications, there might be some repetition of a layer inside the samples. The present work aims at studying samples with repeated layers based also on the relative variation of $\frac{K_\alpha}{K_\beta}$

(or $\frac{K_\alpha}{L_\alpha}, \frac{L_\beta}{L_\alpha}$) ratio. This study is an extension of the previous method [13] and it is a step further to develop an accurate quantitative method in order to analyze cultural heritage samples later on. Firstly the experimental setup is described. Secondly, the key parameters of the method such as the self attenuation and the impact of the layer placed in front of the emitting one are described. Finally, the extension of the previous method [13] to the analysis of a multi-layer sample with a repeated layer is presented.

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2. Experimental procedure

2.1. Experimental set-up

High energy experiments were carried out at ARRANAX cyclotron [14] using 68 MeV protons. The beam diameter on the sample was in the range of 1 cm. The X-rays are detected by a High Purity Germanium (HPGe) detector. The distance between the sample and the detector is 25 cm. The sample holder can be rotated with respect to the incident beam in order to change the angle between the sample and the detector, with a precision of 0.01°. An ionization chamber is used in order to monitor the number of particles penetrating the samples. The ionization chamber current is measured by a high precision commercial electrometer (MULTIDOS-PTW). Prior to the samples irradiations, the ionization chamber is calibrated using a faraday cup. During the experiments, the beam intensity is kept around 100 pA and the irradiations last about 15 min. The high energy PIXE platform is described in more details in [15].

2.2. Studied samples

The composition and the thickness of the samples prepared for this work are listed in Table 1. The samples are composed by a stack of several pure material foils. Those foils are provided by GoodFellow and their dimensions are 25 × 25 mm². The Al-Ti sample is used to study the linearity of $\Delta\mu \cdot d$ (see Section 3.2) involved in the determination of the layers thicknesses. The Ti-Ag₁-Au-Ag₂ is prepared as a multi-layer sample with a repeated layer in order to perform the associated analysis. Reference samples for each elements are also irradiated using the same experimental configuration as the Ti-Ag₁-Au-Ag₂ irradiation. Those reference samples are used to reduce the experimental errors (such as detection efficiency) on the quantification of the layers thicknesses.

The thicknesses of the foils presented in Table 1 are given by GoodFellow. Accurate values obtained using a scanned and a high precision scale for the Ti of Ti-Ag₁-Au-Ag₂ are presented in Table 4.

2.3. Multi-layer analysis method

The analysis method [13] is based on the relative variation of $\frac{K_x}{K_\beta}$ (or $\frac{K_x}{L_\alpha}, \frac{L_\beta}{K_\alpha}$) ratio (K_x is the number of detected K_x X-rays), named $\frac{\Delta R}{R}$, as a function of the angle between the target and the detector, $\theta = 0$ when the detector axis is normal to the sample, i.e their surfaces are parallel. This relative variation is given by:

$$\frac{\Delta R}{R} = \frac{R(\theta_1) - R(\theta_2)}{R(\theta_1)} = 1 - f_{\text{self}} \cdot \frac{e^{\left(\frac{\Delta\mu \cdot d}{\cos(\theta_2)}\right)}}{e^{\left(\frac{\Delta\mu \cdot d}{\cos(\theta_1)}\right)}}, \quad (1)$$

where $\Delta\mu = \mu_{K_\beta} - \mu_{K_\alpha}$ with μ the attenuation coefficient [16] of the layers placed in front of the emitting one, d is X-rays path in this layer and f_{self} is given by the equation:

$$f_{\text{self}} = \frac{\left(1 - e^{\left(\frac{-\mu'_{K_\beta} \cdot d'}{\cos(\theta_2)}\right)}\right)}{\left(1 - e^{\left(\frac{-\mu'_{K_\beta} \cdot d'}{\cos(\theta_1)}\right)}\right)}, \quad (2)$$

where μ' is the attenuation coefficient of the emitting layer and d' the X-rays path inside it. The function f_{self} represents the ratio of the $\frac{K_x}{K_\beta}$ attenuation in the emitter layer itself, in other words the self attenuation. The impact of this factor is presented in the next section.

When f_{self} is negligible, we can extract from Eq. (1):

$$\Delta\mu \cdot d = \frac{1}{\frac{1}{\cos(\theta_2)} - \frac{1}{\cos(\theta_1)}} \cdot \ln\left(1 - \frac{\Delta R}{R}\right). \quad (3)$$

$\Delta\mu \cdot d$ contains the attenuation of the effective layer located between the emitting one and the detector (the effective layer has the same effect on the X-ray attenuation than real layers). Using the photoelectric mass attenuation coefficient (far from the shell edge), $\frac{\mu}{\rho}$ given in [17] and Eq. (3) we can calculate the attenuation of K_α lines (or $K_\beta, L_\alpha, L_\beta$) M_{K_x}

$$M_{K_x} = \sum_{i=0}^n e^{(-\mu'_{K_x} \cdot d^i)} = e^{-\Delta\mu \cdot d \cdot \left(\frac{1}{\frac{1}{(E_{K_\beta})^{7/2}} - \frac{1}{(E_{K_\alpha})^{7/2}}}\right)}, \quad (4)$$

where n is the number of layers placed in front of the emitting one and E the energy of the considered X-ray. The detected number of K_x X-rays (or $K_\beta, L_\alpha, L_\beta$), $N_{K_x}^{\text{layer}}$, can then be corrected from this attenuation. Therefore we can determine the thickness of a layer, e_{layer}

$$e_{\text{layer}} = -\frac{1}{\mu'_{K_x}} \cdot \ln\left(1 - \frac{N_{K_x}^{\text{layer}}}{N_{K_x}^{\text{Standard}} \cdot M_{K_x}}\right), \quad (5)$$

where $N_{K_x}^{\text{Standard}}$ is the X-ray intensity of a standard sample irradiated in the same condition.

To determine the sequence of a multi-layer sample, $\Delta\mu \cdot d$ can't be used because of its dependency on the energy of the X-rays. Using the definition of $\frac{\mu}{\rho}$ given in [17], we can calculate a new term $(\Delta\mu \cdot d)'$ defined by Eq. (6).

$$(\Delta\mu \cdot d)' = \frac{1}{\frac{1}{E_{K_\beta}^{7/2}} - \frac{1}{E_{K_\alpha}^{7/2}}} \cdot \frac{1}{\frac{1}{\cos(\theta_2)} - \frac{1}{\cos(\theta_1)}} \cdot \ln\left(1 - \frac{\Delta R}{R}\right) \quad (6)$$

$(\Delta\mu \cdot d)'$ is also equal to: $\rho \cdot d \cdot Z^5 \cdot \frac{N_A}{A}$, where E is the energy of the considered X-ray, N_A is the Avogadro constant and Z, A, ρ, d are the effective atomic number, mass number, density and thickness of the layers placed in front of the emitting one. The comparison between $(\Delta\mu \cdot d)'$ values for each detected elements allows to determine the sequences of the layers (demonstrated in [13]).

3. Robustness of the method

In this section, the main parameters (described in the previous section) of the analysis method are studied.

3.1. Self attenuation

The relative variation of $R, \frac{\Delta R}{R}$, is function of the self attenuation and of the attenuation inside the layers placed above the emitting one. In order to use $\frac{\Delta R}{R}$ to determine the thickness and the position of the emitting layer, the self attenuation factor has to be small

Table 1
Composition of the irradiated samples for this study. The value in the parenthesis is the thickness of the given pure foil.

Sample name	Composition
Al-Ti	Al(20 μm)/Ti(10 μm)
Ti-Ag ₁ -Au-Ag ₂	Ti(10 μm)/Ag(10 μm)/Au(10 μm)/Ag(25 μm)
Ti _{ref}	Ti(10 μm)
Ag _{ref}	Ag(10 μm)
Au _{ref}	Au(10 μm)

compared to the attenuation inside the layers. The variation of the self attenuation factor, f_{self} , as a function of the emitting layer thickness ($\text{g}\cdot\text{cm}^{-2}$) and for $\theta_1 = 45^\circ$ and $\theta_2 = 0^\circ$ is shown in Fig. 1. These angles have been chosen because they lead to the maximum variation of the self attenuation factor (i.e the worst case).

For these three elements (Ti, Ag and Au representing a wide range of Z), the self attenuation factor is less than 1.04.

For the titanium, due to its low energy X-ray, even a low amount of light element in front of it will induce a value of $\frac{\Delta R}{R}$ far larger than 1.04. For example, 20 μm of aluminium gives $\frac{\Delta R}{R} = 1.15$ for the titanium. So the self attenuation is negligible and is not an issue for light element.

In the silver case, a low value of $\frac{\Delta R}{R}$ (1–3%) can be due to the self attenuation of a thick Ag layer or to the attenuation by the layers above in case of a thin Ag layer. We can solve this issue using the value of the K_α intensity. In fact a thick layer of Ag will induce a higher K_α intensity compared to a thin one.

In the gold case, the self attenuation factor is lower than 1.01, but in the majority of the cases the $\frac{K_\alpha}{K_\beta}$ ratio will not vary so much due to the high energy K X-rays. Therefore this ratio cannot be used (or in very specific cases like very thick samples), $\frac{L_\alpha}{K_\alpha}$ or $\frac{L_\beta}{K_\alpha}$ ratios can be used instead. The variation of f_{self} in this case is represented in Fig. 2. For the couple of angles $(45^\circ, 0^\circ)$, the self attenuation factor is too high (up to 1.35) but by choosing a smaller angle variation (25°) we can reduce f_{self} . Due to the low energy of the L X-rays, even for a narrowed angular range, the angular variations of those ratio are sensitive to thin layer (few dozens of μm) contrary to the $\frac{K_\alpha}{K_\beta}$ ratio. Therefore for heavy elements we can use the $\frac{L_\alpha}{K_\alpha}$ (or $\frac{L_\beta}{K_\alpha}$) ratio and a narrowed angular range to determine the sequences and the thicknesses with a negligible influence of the self attenuation.

Looking at the self attenuation factor for different elements, we have defined a framework for the method. Analysis of light elements is not affected by the self attenuation. For mid-Z elements and low value of $\frac{\Delta R}{R}$, the value of the K_α intensity has to be considered. For heavy elements, the ratio $\frac{L_\alpha}{K_\alpha}$ has to be used instead of the $\frac{K_\alpha}{K_\beta}$ with a lower angles ($\theta_2 - \theta_1$).

3.2. Attenuation in layers placed in front of the emitting layer: $\Delta\mu \cdot d$

The normalization of $\Delta\mu \cdot d$ by $\frac{1}{\cos(\theta_2)} - \frac{1}{\cos(\theta_1)}$ remove the angular dependency. This point has been already experimentally demonstrated [13].

The Sample Al(20 μm)/Ti(10 μm) is irradiated in order to compare the value of $\Delta\mu \cdot d$ with those previously obtained with a Al

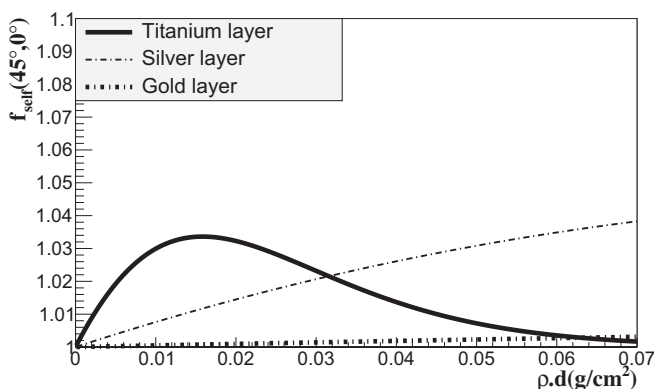


Fig. 1. f_{self} as a function of $\rho \cdot d$ calculated using Eq. (2) for titanium, silver and gold and for $\theta_1 = 45^\circ$ and $\theta_2 = 0^\circ$. The self attenuation factor is calculated for the $\frac{K_\alpha}{K_\beta}$ ratio.

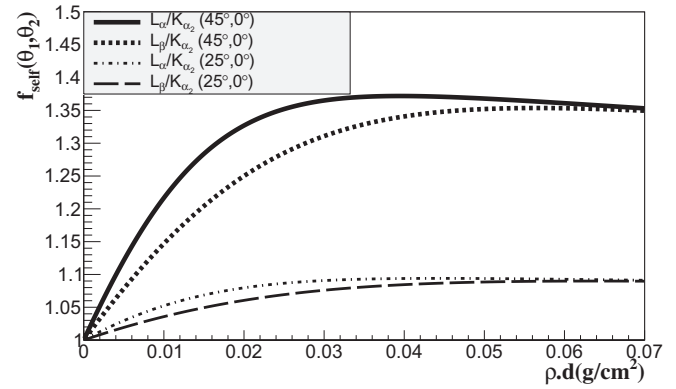


Fig. 2. Self attenuation factor as a function $\rho \cdot d$ calculated using Eq. (2) for the gold and for $\theta_1 = 45$ or 25° and $\theta_2 = 0^\circ$. The self attenuation factor is calculated for the $\frac{L_\alpha}{K_\alpha}$ and $\frac{L_\beta}{K_\alpha}$ ratio.

(50 μm)/Ti(10 μm) sample [13]. The mean value of $\Delta\mu \cdot d$ (calculated with 3 different couples of angles θ_1, θ_2) for 20 μm aluminium is equal to -0.35 ± 0.03 , whereas the mean value was equal to -0.90 ± 0.06 for the thicker sample in [13]. The ratio of this two values, 2.57 ± 0.09 , is equal to the ratio of the aluminium thicknesses which is equal to 2.5 (50/20). Therefore, results show a linear behavior of $\Delta\mu \cdot d$ with the attenuation layer thickness.

If the emitting element is repeated in several separated layers, Eq. (3) is no longer valid, and the calculated $\Delta\mu \cdot d$ will vary with the choice of θ_1 and θ_2 . Therefore it's a way to determine if an element is repeated or not. For this purpose a minimum of 3 irradiations with 3 different detection angles are required to check the variation of $\Delta\mu \cdot d$.

4. Analysis of a multi-layer sample with two layers of the same element

A sample have been irradiated with a 68 MeV proton beam with three different detection angles. Fig. 3 shows one of the obtained spectra. Three elements (titanium, silver and gold) are detected thanks to their characteristic X-rays.

For the titanium, the ratio of the $\frac{K_\alpha}{K_\beta}$ intensity is constant when θ varies (see Table 2). We can then, deduce that there is no layer in front of the titanium, and its thickness is determined (see Table 4) using Eq. (5) without the attenuation correction.

In the gold case, R (calculated using L_α and K_α lines) varies with the detection angle so Eq. (5) is used to determine its thickness (see Table 4).

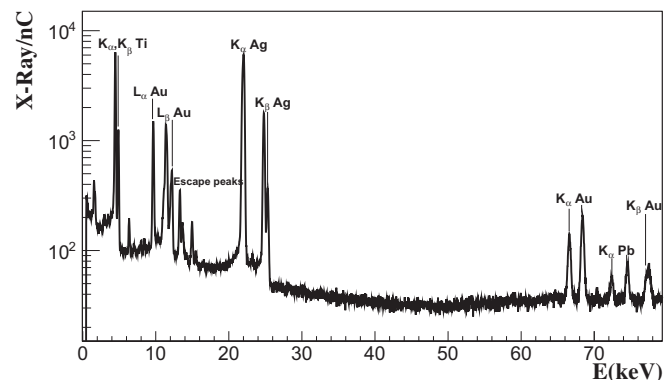


Fig. 3. Spectra of the investigated target obtained using 68 MeV proton beam with the detector normal to the target surface ($\theta = 0^\circ$).

Table 2
Values of R for the titanium as a function of θ .

$\theta(^{\circ})$	R
0	5.43 ± 0.22
22	5.43 ± 0.22
45	5.42 ± 0.22

For the silver, in contrary to the others layers, $\Delta\mu \cdot d$ is not constant with θ (see Table 3).

This variation signs more than one silver layer inside the sample. All those information (three elements detected in the spectrum, the titanium is the first layer and the silver is the only element to be repeated) give us the sequences of the layers (see Fig. 4).

Therefore the following sequence is the only one possible: titanium is the first layer, the second and the last are made of silver and gold is the third layer.

Therefore the attenuation, A^{Au} , of the X-ray from the gold layer is the product of the attenuation by the titanium and the first layer of silver and is given by:

$$A^{Au} = e^{-\mu_{Ti} \cdot d_{Ti}} \cdot e^{-\mu_{Ag1} \cdot d_{Ag1}} \quad (7)$$

Eq. (4) is used to calculate A^{Au} from the value of $\Delta\mu \cdot d$. We can calculate the thickness (see Table 4) of the first layer of silver using:

$$d_{Ag1} = \frac{-1}{\mu_{Ag}} \cdot \ln \left(\frac{A^{Au}}{e^{-\mu_{Ti} \cdot d_{Ti}}} \right). \quad (8)$$

In the spectra, the intensity of the silver X-ray is the sum of the contribution from the two layers. Knowing the thickness of the first layer, we can calculate its X-ray intensity and therefore determine the intensity of the second layer I_{Ag2} . This intensity, I_{Ag2} , is then corrected by the attenuation of the other layers (titanium, silver, gold) to determine the second layer thickness by using the same method as for titanium (see Table 4).

The calculated thicknesses presented in Table 4 are in good agreement with the real thicknesses of each layers of our samples, even for the repeated layer.

Some limitations to be worked out:

We were able to determine the thickness of the second silver layer with a small relative error, because its contribution to the total X-ray intensity is significant. In the case of a non repeated layer, using $\Delta\mu \cdot d$ to determine the thickness of each layer [13], the error doesn't depend of the number of layers.

To calculate the thickness of the repeated layers, the thickness of each layer in front of the considered ones is required. The uncertainty calculated using the propagation uncertainty formula will increase with the number of layers in front of the repeated one. The performance of this method in those cases depends on the studied object (number of layers, sequences, thicknesses of the repeated layers).

In more complex cases, with multiple repetition, this method is no longer applicable. But the use at the same time of complementary IBA techniques, such as PIGE or detection of charged particles,

Table 3
Values of $\Delta\mu \cdot d$ for the silver as a function of θ .

$\theta_1(^{\circ})$	$\theta_2(^{\circ})$	$\Delta\mu \cdot d$
22	0	-0.623 ± 0.009
45	22	-0.104 ± 0.001
45	0	-0.207 ± 0.003

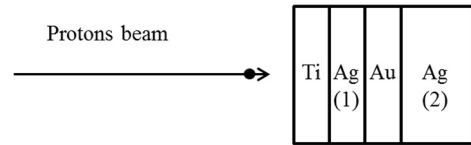


Fig. 4. Scheme of the layers sequences inside the irradiated target.

Table 4
Results of the multi-layers analysis with the repetition of an element. The elements are listed in their real position inside the sample.

Element	Calculated thickness (μm)	Real thickness (μm)
Titanium	10.13 ± 1.23	10.3 ± 0.02
Silver (1)	9.23 ± 0.91	10.4 ± 0.02
Gold	9.8 ± 1.08	10.79 ± 0.02
Silver (2)	23.3 ± 2.4	24.59 ± 0.01

could provides additional useful information to deal with those more complex cases.

5. Conclusion

We demonstrate the possibility to perform quantitative analysis of multi-layer targets using high energy PIXE, without any assumption on their composition. A proper study of the parameters involved in the method previously published [13] is presented and more complex cases are investigated. The framework of the method depending on the atomic number of the analyzed element has been defined by looking at the self attenuation factor. We demonstrate the linearity of $\Delta\mu \cdot d$ involved in the thickness calculation. An extension of this analysis method also based on the relative variation of $\frac{K_{\alpha}}{K_{\beta}}$ (or $\frac{K_{\alpha}}{L_{\alpha}} \cdot \frac{L_{\alpha}}{L_{\beta}}$) when the sample is rotated, has been applied to a multi-layer sample with a repeated layer. The sequences and the thicknesses of the layers without assumption on the sample composition have been determined even for the repeated layer. For samples without any repeated layer, the error on the thickness is low. In the case of a repeated layer, the X-ray intensity has to be significant to apply this method. The thicknesses determined in this work are in the same order of magnitude as in cultural heritage samples analysed by PIXE. The performance of this method will be tested on real cases and especially on art objects already analysed by HEPIXE in the near future.

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