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Experimental determination of layers films thicknesses

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The determination of particle induced x-ray emission (PIXE) cross sections and the concentration of elements in a material require the knowledge of the target sample thickness. In this aim, measurements of the thickness by three different methods have been performed. These are absorption of X-rays by a ^{55}Fe source, transmission of alpha particles by a ^{241}Am source and Rutherford backscattering of alpha particles produced by Van de Graff Accelerator with the use of the RUMP simulation code. The results give a thickness with uncertainties ranging from 1 to 8% according to the experimental technique used. The comparison between these methods gives an advantage for the X-rays absorption for its simplicity and accuracy, when backscattering spectrometry is preferred for thin target on backing or as a complementary technique for PIXE analysis.

Key words: Thickness, particle induced x-ray emission (PIXE), Rutherford backscattered (RBS), cross section, rump.

INTRODUCTION

The technique of samples analysis by charged particles induced x-ray emission (PIXE), requires the knowledge of the targets thicknesses in order to determine the concentrations of the elements present in the sample and for the matrix effect correction. The same applies for the calculation of the ionization cross section:

$$d\sigma/d\Omega = dN/AdxI$$

where dN, Adx and I represent respectively the number of emitted X-rays, of target atoms and the intensity of the beam of incident particles.

In the aim of selecting a technique allowing the thickness determination of a target with the best possible precision, several methods of measurement have been undertaken and these are:

(a) Transmission of alpha particles given by a radioactive source or produced by an accelerator.
(b) Rutherford backscattered (RBS) of alpha particles produced by an accelerator.
(c) The attenuation of X-rays resulting from an iron source (^{55}Fe).

Some of these methods are usually used in PIXE measurements (Johansson and Campbell, 1970; Tran et al., 2002; Ekinici and Valles, 2001). The various measurements were carried out at the Nuclear Research Centre of Algiers (CRNA) of the Commission of Atomic Energy (COMENA), in the division of the nuclear techniques. Self-supported targets whose thicknesses were measured by piezoelectric quartz during the evaporation process, commercial targets with thickness is given by the manufacturer and finally targets deposited on a substrate of silicon and of unknown thickness.

Preparation of the targets

The preparation (Ourabah and Amokrane, 2006) of the

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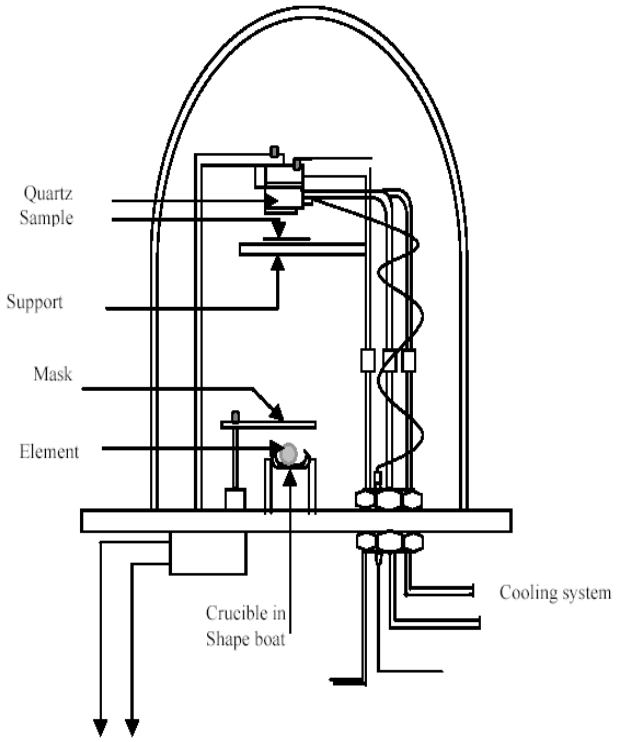

Figure 1. Evaporator with piezoelectric quartz.

targets was carried out in an evaporator composed of a bell provided with a system of pumping and piezoelectric quartz for the measurement of the thicknesses (Figure 1). Two types of targets were elaborated out with and without backing (self-supported target). The self-supported targets were produced by the use of a taking off agent which dissolves easily in distilled water. This agent depends on the material deposited. It should be pointed out that the crystalline shapes of both agent and material to deposit have to be similar.

Determination of the thickness by piezoelectric quartz

Thickness can be measured during the evaporating procedure by a piezoelectric quartz crystal (silicon dioxide crystal) put on the sample in the enclosure of the evaporator. The quartz is subjected to a mechanical pressure during evaporation, giving appearance of an electric potential on its face. The measurement of the resonance frequency of this quartz which varies as function of the thickness allows the determination of the thickness of the target.

RUTHERFORD BACKSCATTERING

Particles backscattering principle is shown in Figure 2.

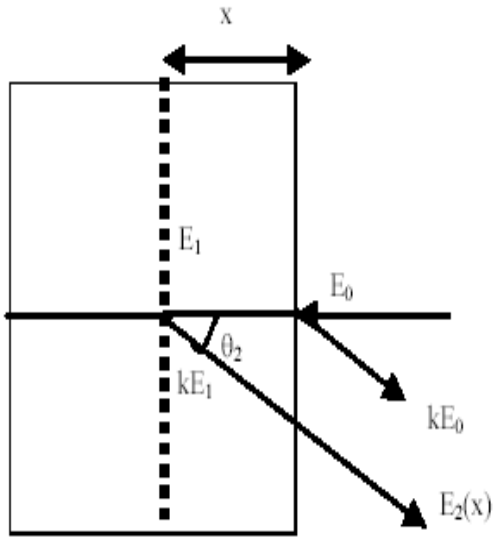

Figure 2. Principle of backscattering.

When a target of thickness x is bombarded with incident particles of energy E_o, their energy after diffusion, at an angle θ, by the nuclei located at the surface of the target is kE_o, where k is the kinematic factor given by:

$$k = \left(\frac{M_1 \cos\theta + \sqrt{M_2^2 - M_1^2 \sin^2\theta}}{M_1 + M_2} \right)^2 \quad (1)$$

M_1 ; M_2 are the masses of incident particle and nucleus of the target, θ being the diffusion angle.

After crossing the sample, the energy of the particle at a depth x is:

$$E_1 = E_0 - \int_0^x \frac{dE}{dx} dx \qquad \text{for the ingoing path} \quad (2)$$

After backscattering on a nucleus of the target at the depth x, its energy will be:

$$E_2(x) = k E_1 - \int_0^{\frac{x}{|\cos\theta_2|}} \frac{dE}{dx} dx \qquad \text{for the outgoing path} \quad (3)$$

θ_2 is the angle of the backscattered ion with the target's normal. The lost energy is then $\Delta E_i = k E_0 - E_2$.
Using Equations 2 and 3, we found:

$$\Delta E = k \int_0^x \frac{dE}{dx} dx + \int_0^{\frac{x}{|\cos\theta_2|}} \frac{dE}{dx} dx \quad (4)$$

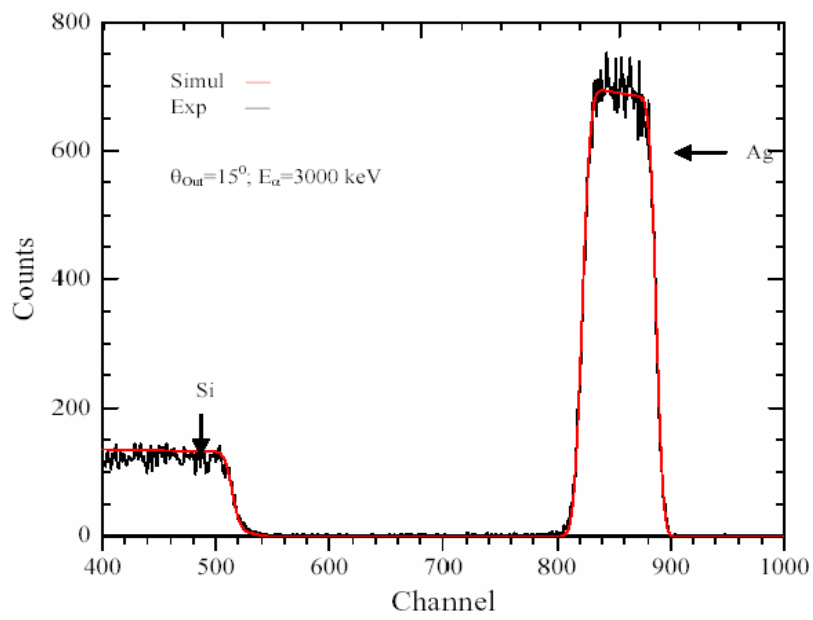

Figure 3. Backscattered spectrum of alpha particles on silver target of 1990°A thickness with silicon backing.

or $\Delta E = k\,\Delta E_{in} + \Delta E_{out}$

Introducing the stopping power [S]= dE/dx and assuming that the energy lost dE/dx is constant and calculated at $\bar{E}_{in}$ and $\bar{E}_{out}$, the integrals give:

$$\Delta E_{in} = \left[S\left(\bar{E}_{in}\right)\right]\Delta x \quad \text{and} \quad \Delta E_{out} = \frac{1}{\cos\theta_2}\left[S\left(\bar{E}_{out}\right)\right]\Delta x \tag{5}$$

Many approximations (Chu et al., 1978) allow calculating [S] and finally Δx.

- Approximating the surface energy for thin target as:

$$\bar{E}_{in} = E_0 - \frac{\Delta E_{in}}{2} \quad \text{and} \quad \bar{E}_{out} = kE_0 - \frac{\Delta E_{out}}{2} \tag{6}$$

- On the other hand, approximating the average energy for appreciable target thickness as:

$$\bar{E}_{in} = \frac{1}{2}\left(E_0 + E_1\right) \quad \text{and} \quad \bar{E}_{out} = \frac{1}{2}\left(kE_1 + E_2\right) \tag{7}$$

E_1 being unknown, one can suppose that the energy loss can be split symmetrically between the ingoing and the outgoing paths, so that $\Delta E_{in} \approx \Delta E_{out}$ and thus the average energies will be:

$$\bar{E}_{in} = \left(E_0 + \frac{\Delta E}{4}\right) \quad \text{and} \quad \bar{E}_{out} = \left(E_2 + \frac{\Delta E}{4}\right) \tag{8}$$

Our measurements were done with alpha particles, produced by the Van de Graff accelerator. The backscattered particles where detected with a surface barrier detector. A typical backscattered spectrum is represented in Figure 3, showing the signal of the silicon backing and that of silver. The width at half maximum (FWHM) of the backscattered peak represents the total energy loss ΔE of he ingoing and outgoing paths. The target thickness can be obtained from:

1. The ratio of the surface of the RBS spectrum of the element over the height of signal of the backing.
2. The analysis of the RBS spectrum with the RUMP code (Doolittle, 1985).
3. The determination of the energy E_1 at depth x by a calculus code using different methods.

However, in this work the determination is limited to cases 1 and 2. Several samples were used:

1. Two samples Ag/Si of different thicknesses, a sample of Au/Ti/Si, and all three with two systems of detection to see if the detection angle influences the thickness determination of the target.
2. Three samples of Ag/Si of different thicknesses, two self supported targets of nickel and aluminium with only one detection system.

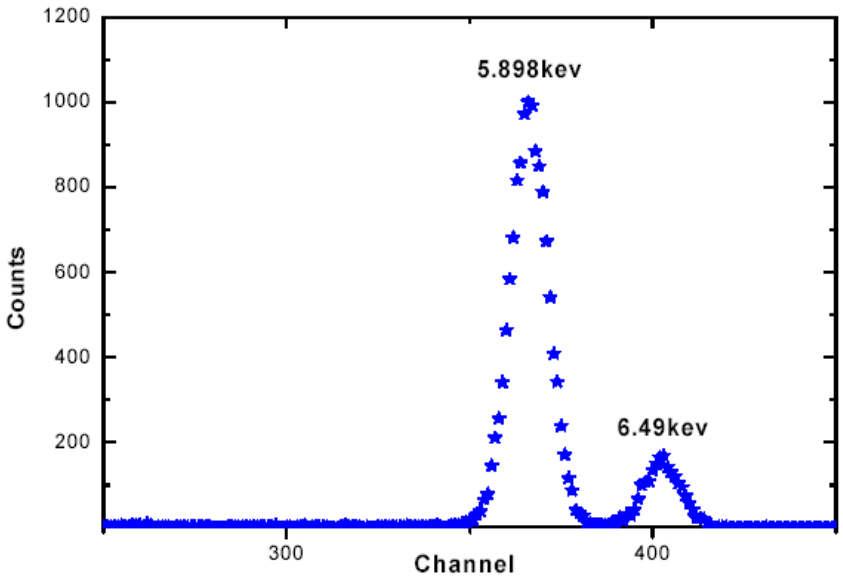

Figure 4. ^{55}Fe spectrum without absorber.

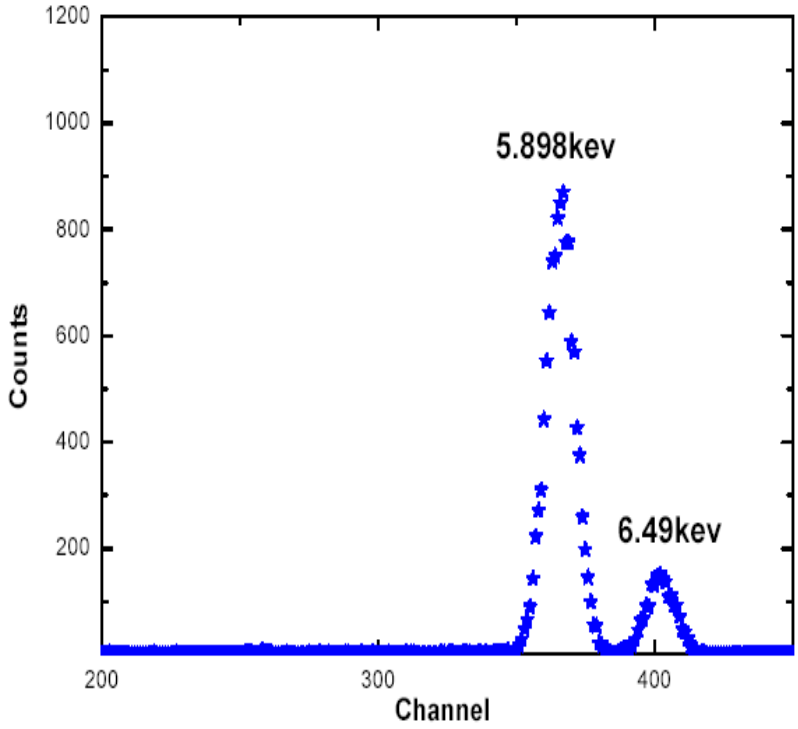

Figure 5. ^{55}Fe spectrum with aluminium absorber.

METHOD OF ATTENUATION OF X-RAYS IN MATTER

Measurements of target thickness were also made from the attenuation of the photons. It is deduced from the Lambert's law (Davisson and Evans, 1952) according to which intensity I of the transmitted photons is given by the relation $I=I_0 \exp(-\mu x)$ where I_o is the initial intensity, μ the linear attenuation coefficient and x the thickness of the absorber. The thickness is then $x = (1/\mu) \ln(I_o/I)$. We performed the experiment on two films of nickel and aluminium.

The X-rays are provided by a 25mCi sealed source of Iron (^{55}Fe), emitting the 5,898 kev and 6,49 kev lines of manganese. The transmitted photons are collected in Si(Li) detector of 220 eV of resolution at 5,898 keV energy. The measurements were conducted three times and gave similar results. Figures 4 and 5

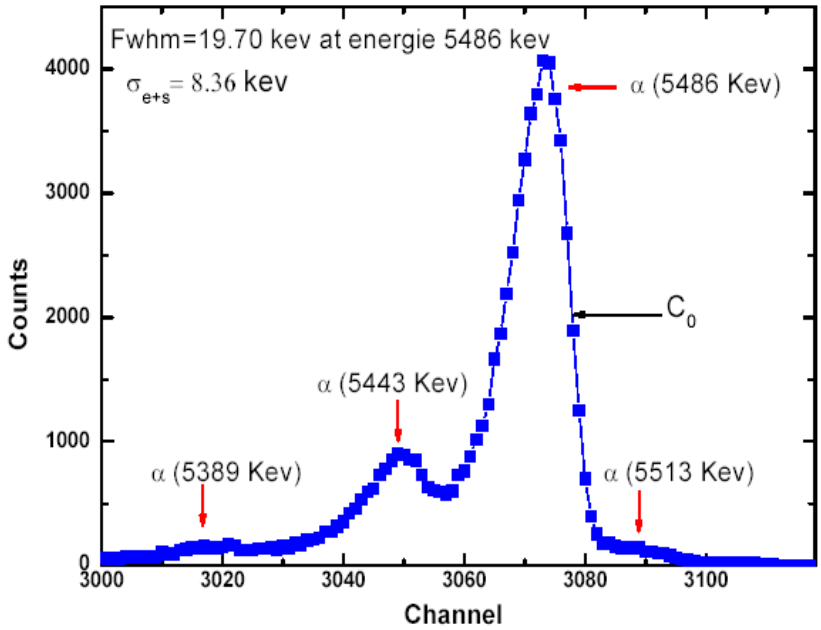

Figure 6. Spectrum of alpha particles issued from the ^{241}Am source without absorber.

show typical X-ray spectra.

Technique by transmission of the He^{++} particles from a ^{241}Am source

The technique consists in measuring the energy loss ΔE of the alpha particles in self supported targets. The stopping power in the approximation of average energy E_M is obtained from code SRIM 2003 (Ziegler et al., 1985), used for the determination of targets thickness. The experimental energy loss is given by:

$$\Delta E[kev] = E_0 - E_1 = a*(C_0 - C_1) \quad (9)$$

where E_0, E_1 and C_0, C_1 are energies and the corresponding channels measured without and with the target, a[keV/channel] is the slope of the calibration straight line. The average energy is

$$E_M = E_0 - \frac{\Delta E}{2} \quad (10)$$

By using the stopping power [ε], one can write

$$\varepsilon(E_M)[\text{keV/micron}] = (\Delta E/x) = a*(C_0 - C_1)/x \quad (11)$$

[ε] is the stopping power at average energy E_M .We thus have

$$x[\mu m] = (\Delta E / \varepsilon(E_M)) \quad (12)$$

Measurements

The experimental set up is composed by an enclosure, the source, a pumping system and a chain of detection constituted by a 50 mm^2 surface barrier detector with 12 keV of resolution at the 5486 keV energy, a preamplifier and an amplifier. Alpha particles are provided by 1μCi ^{241}Am source of 5486 keV energy. Measurements were carried out on targets of nickel and Aluminium manufactured and other aluminium and silver targets that we have realized in the Laboratory for Targets of CRNA. Figures 6 and 7 show spectra of alpha particles resulting from the ^{241}Am source without absorber and after crossing a nickel target of 1.27 μm thickness.

Transmission of the alpha particles provided by an accelerator

The same principle used for transmission for alpha particles resulting from the radioactive source is applied. The surface barrier detector is placed at a detection angle of 30° in order to avoid its deterioration. The experiment is carried out in combination with the RBS. The spectrum obtained is represented in Figure 8.

RESULTS

The resulting thicknesses determined by the two methods; the ratio of the surface of the RBS spectrum of the element over the height of signal of the backing and the analysis of the RBS spectrum with the RUMP code (Doolittle, 1985), are reported in Table 1. We should note that the measurements, carried out with the method of the ratio of the height of the spectrum of the backing over the surface of the target, are not in agreement with the results obtained using the Rump code for samples 1 and 2; the signal of the silicon backing being not well defined because of its bad quality. However, results obtained for the sample 3 are of the same order of magnitude as those obtained with the Rump code. During the simulation by the Rump code for sample 1 and in the range of quoted energies (Table 1), we noticed a light

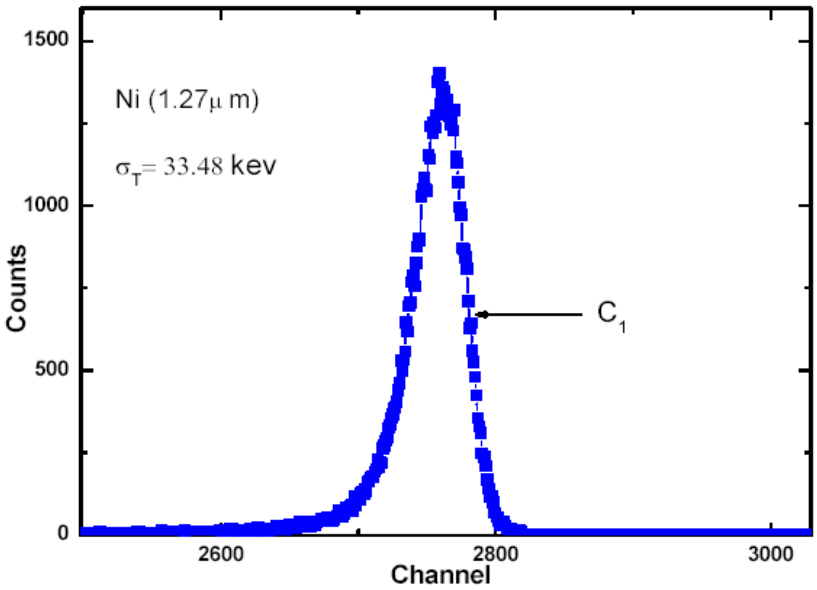

Figure 7. Spectrum of alpha particles after transmission through the absorber.

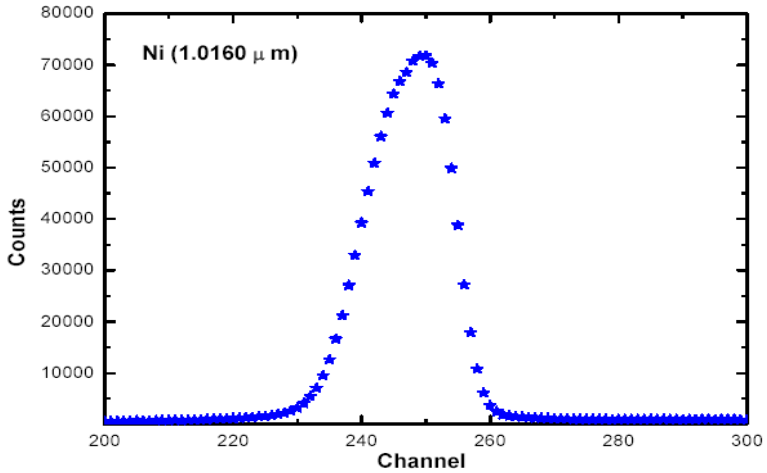

Figure 8. Transmitted alpha spectrum through Nickel target.

Table 1. Thicknesses determined by the method of the ratio of the height of the signal of the backing over that of the element and by simulation with Rump code.

Sample	Target	Energy range of alpha particles	Detection angle θ	(surface of element target) /(Height of backing signal) (Å)	Rump(Å)
1	Ag/Si	[700kev-1100kev]	150°	691.5	1030
		[1200kev-1600kev]		663.9	988
		[700kev-1600kev]	165°	722.5	1055
2	Ag/Si	[1600kev-3000kev]	150°	1587	1930
		[1600kev-3000kev]	165°	1591	1990
3	Au/Ti/Si	[1800kev-3400kev]	150°	1265	1230
			165°	1202	1210

Table 2. Thicknesses obtained with the piezo electric quartz and by the simulation with Rump code. (Q) measured by the piezo electric quartz, (C) given by the manufacturer.

Sample	Target	Energy (keV)	Detection angle θ	Rump (Å)	Given thickness Quartz (Å)	Uncertainties (%)
4	Ag/Si	2000	160°	207	214 (Q)	4.02
5	Ag/Si	2000	160°	433	450 (Q)	3.73
6	Ag/Si	2000	160°	822	859 (Q)	4.31
7	Al	2200	165°	6836	7500 (C)	3.45
8	Ni	2200	165°	6150	6350 (C)	3.00

Table 3. Values for the attenuation coefficients μ.

Elements	Z	μ(m^{-1})
Aluminium	13	32721.22
Nickel	28	101586.61
silver	47	504693.87

Table 4. Measured thicknesses by the attenuation technique compared with those given by manufacturer (C).

Element	Z	Given thickness (μm)	Measured thickness (μm)	Relative uncertainties (%)
Nickel	28	0.635 (C)	0.627±0.019	3
		0.762 (C)	0.757±0.024	3
		1.905 (C)	1.879±0.025	1.3
		3.750 (C)	3.678±0.055	1.5
Aluminium	13	2.000 (C)	1.921±0.038	2
		4.000 (C)	3.780±0.109	3
Silver	47	5.000 (C)	4.818±0.094	2

variation of the thickness, which can be explained by the inclination and the non uniformity of the target. We can also see that the detection angle (θ=150° or θ =165°) does not influence the thickness.

In Table 2, we report the results obtained with the Rump code for self supported targets of nickel, aluminum and for Ag on Si backing. The uncertainties given for the simulation of the spectrum are estimated from the uncertainty on the channel; the resolution of the detector and on the stopping power. The comparison between the values measured with piezoelectric quartz, those obtained with RUMP code and those given by the manufacturer for Ag/Si targets and Nickel indicates a good agreement, except for the aluminium foil for which the light difference can be attributed to the value given by the manufacturer.

Measurements of target thickness were made from the attenuation of the photons using the values for the attenuation coefficients (Berger and Hubbell, 1987) given in the Table 3 for the energy 5,898 keV, we find the results in Table 4. The uncertainties were calculated using the Lambert's law, taking into account the precision on the intensity of the source before and after attenuation (<3%), and the error on the attenuation coefficient (1% for aluminium). They were also made from the technique by transmission of the HE^{++} particles from a ^{241}AM source; the results are reported in the Table 5. The uncertainties were calculated using the equation:

$$\frac{\Delta x}{x} = \frac{\Delta \varepsilon}{\varepsilon} + \frac{\Delta(\Delta E)}{\Delta E}$$

taking into account the precisions of the stopping power (2%) and the energy lost (<0.5%).

Comparison between the thickness measured by transmission of alpha particles given by radioactive source and produced by accelerated particles

The results of the measurements are reported in Table 6. We can notice that the values obtained by transmission of the particles alpha produced by the ^{241}Am radioactive source (Table 5) or coming from the accelerator (Table 6)

Table 5. Measured thicknesses by transmission of alpha particles: (•) prepared in this work (C) manufactured (P) measured by piezoelectric quartz.

Element	Z	ΔE (keV)	Given thickness (μm)	Thickness measured by transmission (μm)	Uncertainties (%)
		264.799	0.635 (c)	0.68±0.03	4.4
		321.290	0.762 (c)	0.82±0.04	4.8
Ni	28	434.271	1.016 (c)	1.10±0.05	4.5
		524.303	1.270 (c)	1.32±0.06	4.5
		835.001	1.905 (c)	2.07±0.09	4.3
Ag	47	54.725	•0.155 (P)	0.16±0.008	5.0
		81.205	•0.500 (P)	0.52±0.03	5.7
Al	13	112.981	0.750 (C)	0.73±0.04	5.4
		631.988	4.000 (C)	3.95±0.2	5.0

Table 6. Measured thicknesses by transmission of 2.2 MeV alpha particles coming from an accelerator (C) thickness given by the manufacturer.

ΔE (kev)	ΔE/E0 (%)	Given thickness (μm)	Measured thickness (μm)	Relative uncertainties (%)
434.696	19.759	0.635 (c)	0.65±0.03	4.6
547.865	24.903	0.762 (c)	0.81±0.05	6.1
744.422	33.837	1.016 (c)	1.08±0.08	7.4
833.766	37.899	1.270 (c)	1.21±0.06	5.0
1298.354	59.016	1.905 (c)	1.81±0.17	9.3

are in agreement with the values given by the manufacturer or those measured by the piezoelectric quartz.

We can see on the Table 5 that for the thicknesses lower than 1.2 μm, the results by transmission of the alpha particles given by the radioactive source and those produced by the accelerator (Table 6) are similar. For the thicknesses above 1.2 μm, the difference between the two measurements can be explained by the use of the approximation of average energy in the calculation of the stopping power for the 2200 keV energy of the alpha particles given by the accelerator, the total energy loss is of 833 keV, the approximation of average energy for the calculation of the stopping power is not good whereas for the alpha particles provided by the source, the energy loss being small, the approximation is more suitable.

Conclusion

In the aim of selecting the technique allowing the thickness determination of a target with the best possible precision, several methods of measurement have been investigated. According to the relative uncertainties made in the determination of the targets thicknesses, the followings can be concluded:

1. The method by attenuation of X-rays is preferable to the other methods for its precision and its simplicity, mainly for large thicknesses, since we found that the uncertainties on the thickness are lower than 3%. This technique is currently used in industry for the measurement of the thicknesses of different materials.
2. In the case of a target deposited on a backing, where the method by attenuation cannot be employed, RBS technique remains the method suitable compared to the two techniques by the attenuation of X-ray and the transmission of the alpha. Only one must take into account that the uncertainties made on the thickness of the target varies between 4 to 6%. This technique is used simultaneously with the PIXE analysis technique (which requires the knowledge of the thicknesses to obtain the absolute concentrations).

In conclusion, the attenuation of X-rays remains the best technique for the determination of large thicknesses targets with a better precision for the self supported ones whereas for very thin targets deposited on a backing, RBS technique remains a good method, since the

difficulty of the self supported thin targets lies in their brittleness to handle them manually and in the fact that certain metal elements of the periodic table cannot be put always in the form of self supported targets.

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