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Film Thickness Determination by SEM-EDX Measurements of the Attenuation of X-Rays Generated in a Specially Designed Target

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ABSTRACT
The controlled deposition of thin films on a variety of substrates is a fundamental step in many technological applications, being of great importance to know their thickness. The thickness of a film with known composition, can be determined by using the intensity attenuation of a monochromatic X-Ray beam which traverses the film. In this work, we propose to use a SEM-EDX system to detect the characteristic X-Rays generated by a specially designed target, and that traverse a metal film deposited on a thin polymeric substrate. The target is a Cu film with 800 nm of thickness, deposited on a Ti plate which upon the incidence of a 30 keV electron beam generates three lines of comparable intensity, well separated in energy. EDX spectrums are acquired from this target and, depending on the film thickness, it was selected one of these signals to measure its attenuation, choosing the one which gives the best measure for the particular film under study. The films under study were three Ag films deposited with different deposition times, by magnetron sputtering on transparent polyethylene sheets. The target and the sheets were mounted on a specially designed sample holder, which allows to determine the X-Ray attenuation by simple 90° rotations, in order to face the detector and acquire the non-attenuated emitted signals, the signals attenuated by the uncoated sheet and the signals attenuated by the Ag coated sheet. The film thickness determined were 0.20, 0.59 and 0.99 μm, which agrees linearly with the corresponding 5, 15 and 25 min sputtering deposition times.

Keywords: Film-thickness, X-Ray-attenuation, X-Ray-target, SEM-EDX.

Determinación del Espesor de Películas mediante Mediciones con SEM-EDX de la Atenuación de Rayos-X Generados en un Blanco Especialmente Diseñado

RESUMEN
La deposición controlada de películas delgadas sobre variedad de sustratos, es un paso fundamental en muchas aplicaciones tecnológicas, siendo de gran importancia conocer el espesor de estas películas. Puede determinarse el espesor de una capa de composición conocida, utilizando la atenuación de la intensidad de un haz de Rayos-X monocromático que atraviese la capa. Proponemos un sistema SEM-EDX para detectar los Rayos-X característicos, generados por un blanco especialmente diseñado, que atraviesan una película metálica depositada sobre una lámina delgada de material polimérico. El blanco consiste en una película de Cu, de 800 nm de espesor, depositada sobre una placa de Ti que, ante la incidencia de un haz de electrones de 30 keV, genera tres líneas de intensidad comparable, bien separadas en energía. A partir de este blanco, se obtienen los espectros de EDX y, dependiendo del espesor de la película, se elige una de estas señales para medir su atenuación, escogiendo la que resulte en la mejor medida para la película particular de estudio. Sobre láminas transparentes de polietileno se depositaron tres películas de Ag mediante pulverización catódica con magnetrón con tres diferentes tiempos de deposición. El blanco y las láminas se montaron en un portamuestras que permite determinar la atenuación de los rayos X mediante simples rotaciones de 90° para enfrentar al detector las señales emitidas no-atenuadas, las señales atenuadas por el substrato polimérico no recubierto y las señales atenuadas por el substrato recubierto de Ag. Los espesores determinados fueron de 0.20, 0.59 y 0.99 μm, lo que concuerda linealmente con los correspondientes tiempos de deposición de 5, 15 y 25 min.

Palabras claves: Espesor-de-película, atenuación-de-Rayos-X, blanco-de-Rayos-X, SEM-EDX.

INTRODUCTION

The controlled deposition of thin films on a variety of substrates is a fundamental step in diverse branches of technology which encompass optical, microelectronic, magnetic recording and decorative applications. Besides knowing the film chemical composition and density, the determination of its thickness is of a major importance, since usually the behaviour of a particular device depends on it.

The measurement of X-Rays intensity attenuation by matter is a well known method for the determination of the thickness and/or density of a material traversed by monochromatic X-Ray beams [1]. When an X-ray beam of energy E and intensity I_0 traverses a film of thickness t, its intensity is exponentially attenuated to a value I according to the expression (1).

$$(I/I_0) = \exp[-(t/\tau)] \qquad (1)$$

Where τ is the attenuation length of the X-rays, a parameter that depends on the photon energy E and on the particular material which the film is made. It corresponds to the depth into the material measured along the film surface normal where the X-rays intensity falls to 1/e of its value at the entrance surface and represents the photon mean free path in that medium. If both I_0 and I are measured, its ratio $R = (I_0/I)$ can be calculated and the thickness t of the film can be determined by using the expression (2).

$$t = \tau \ln(R) \qquad (2)$$

Values of the attenuation length τ for a given material as a function of the X-Ray energy E, can be obtained from public domain data bases [2]. Related parameters, also easily available, are the linear attenuation coefficient $\mu=(1/\tau)$ and the mass attenuation coefficient (μ/ρ) [3, 4].

In the present work, it is proposed to use a Scanning Electron Microscope with an X-ray Spectrometer (SEM-EDX system) as an X-rays generator from a specially designed target. The generated X-rays are used to determine the thickness of films of known composition by means of measurements of the attenuation that these X-rays undergo when passing through them before reaching the EDX detector. In general, X-ray attenuation is dominated by both Compton scattering and photoelectric absorption but for the small X-Ray energies used in the SEM-EDX systems (0.1-15 keV) the attenuation is basically due to the photoelectric effect.

MATERIALS AND METHODIS

An X-Ray target was specially designed in order to generate three characteristic X-Rays well separated in energy and with similar intensities, using a 30 keV electron beam as the excitation source. For that purpose, a target was made consisting of an 800 nm thick Cu film deposited by vacuum thermal evaporation on a 0.25 mm thick Ti plate. Cu generates a low energy (Lα) and a high energy (Kα) characteristic peaks and Ti generates a peak (Kα) at an intermediate energy. The required thickness of the Cu film to be deposited on Ti was previously calculated from Monte Carlo EDX spectra simulations using the public domain software NIST DTSA-II [4]. With this target one can generate three characteristic signals conveniently separated in energy: Cu Lα at 0.930 keV (peak 1), Ti Kα at 4,511 keV (peak 2) and Cu Kα at 8,048 keV (peak 3). These signals could now be used in the attenuation measurements, choosing the one that provides the best sensitivity for the particular film to be studied.

The films which thickness were determined by this X-Ray attenuation method, were Ag films deposited by magnetron sputtering on 1 cm x 1 cm x 80 µm sheets of transparent polyethylene (PE) substrates. Three Ag films of different thickness were prepared using deposition times of 5, 15 and 25 min, respectively, under the same sputtering conditions (Ag disk target, Ar working gas at 0.1 mBar, 600 V, 40 mA, 3 cm target-substrate separation, 19 rpm).

For the X-Ray attenuation analysis, the Cu/Ti X-Ray target and the samples, were mounted on a specially designed sample holder, which is shown in Fig. 1. The sample holder was made from an Al block and has the shape of a truncated pyramid with an horizontal square base, two opposite vertical surfaces and two tilted symmetrical surfaces cut at 55° relative to the horizontal basal plane. The Cu/Ti target is located horizontally on the top surface, next to a Faraday cup perforated in the block for measuring the electron beam current, if required. The uncoated and the coated PE sheets can be placed on the opposite tilted surfaces, sticking out of the top surface without touching each other, leaving between them a gap of a few mm to allow the electron beam to be focused on the target.

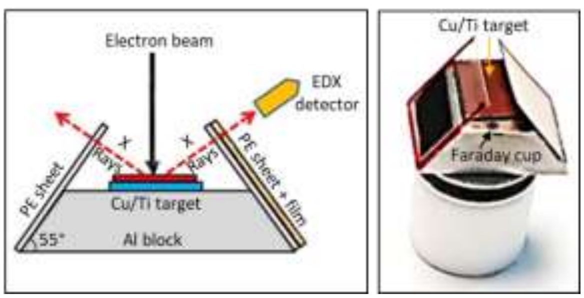

Fig. 1. Diagram and photograph of the sample holder.

In the Si(Li) SEM-EDX system used the X-Ray detector is located on the column at a 35° elevation angle, so when the PE sheets are placed on the tilted surfaces, they are traversed perpendicularly by the X-Rays that reach the detector. The normal sequence for carrying out the attenuation measurements is the following: a) place the sample holder into the microscope with no tilt, b) rotate the sample holder in order to face the unobstructed side of the target to the EDX detector (this position is defined as the reference or 0° rotation position), c) at the chosen working distance for EDX, measure the electron beam current using the Faraday cup and focus the electron beam on the target in a region close to its center, d) acquire the EDX spectrum of the target, e) rotate the sample holder to the +90° position (clockwise) and acquire the EDX spectrum corresponding to the target signals attenuated by the clean substrate sheet, f) rotate the sample holder to the -90° position (counterclockwise) and acquire the EDX spectrum corresponding to the target signals attenuated by both the substrate sheet and the film deposited on it.

These last two spectra must be acquired under the same conditions. The essential steps to determine the X-Ray attenuation are then step e) to obtain the value I_0 and step f) to obtain the value I.

RESULTS AND DISCUSSION

Fig. 2 shows the experimental EDX spectrum obtained from the designed Cu/Ti target using a 30 keV electron beam. As expected, three well separated peaks of comparable intensity were obtained.

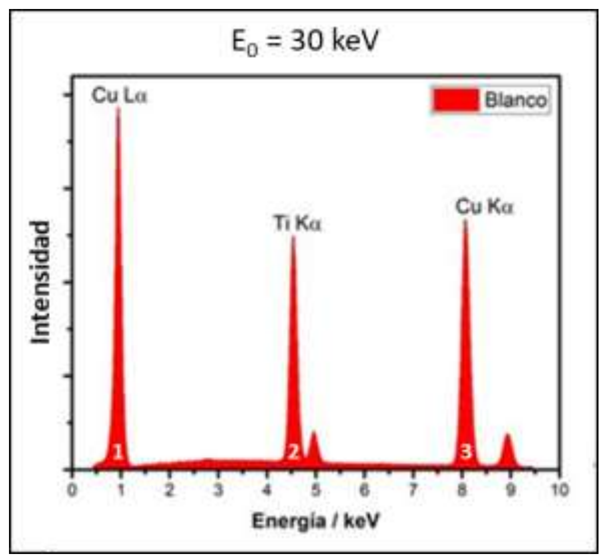

Fig. 2. EDX spectrum generated by the designed target.

The 80 μm thick PE substrate sheet used in this case completely attenuates the low energy peak 1 but allows peaks 2 and 3 to pass through slightly attenuated, been then further attenuated by the Ag films deposited on the substrate.

Fig. 3 shows a graph of the attenuation length in Ag as a function of the photon X-Ray energy, provided by the application of reference [2].

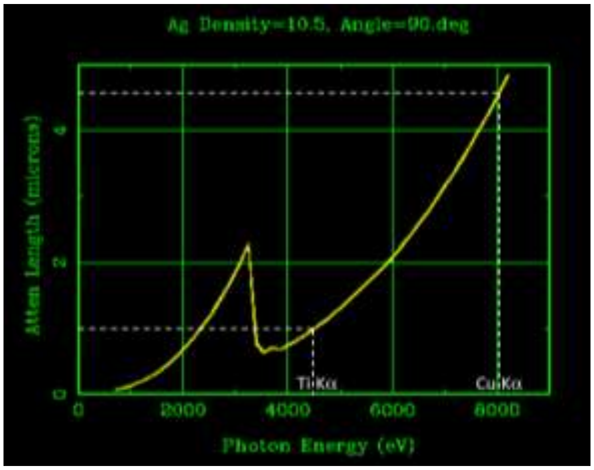

Fig. 3. Attenuation length τ in Ag vs. photon energy.

Except for the Ag L_3 absorption edge at 3351 eV, it is a monotonic growing function. From Table 14.3 in reference [4] one obtains the mass absorption coefficients for Ti Kα and Cu Kα traversing Ag, from which one gets the values τ = 0.956 ± 0.009 µm for the attenuation of Ti Kα and τ=4.361± 0.004 µm for the attenuation of Cu Kα.

Figure 4 shows the expected variation of the ratio $R = (I_0/I)$ of the Ti Kα peak as a function of the thickness of the deposited Ag film, as given by equation (2).

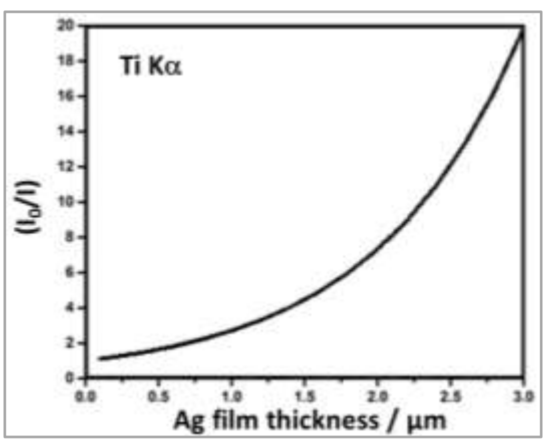

Fig. 4. Thickness dependence of the ratio R = (I0/I).

As can be observed in figure 4, this ratio is quite sensitive to the thickness of the silver film in the range 0 - 3 µm. For thicker films it is more convenient to use the more energetic Cu Kα peak.

Figure 5 shows the EDX signals of the Ti Kα peak obtained for the case of the uncoated (deposition time t = 0 min) PE sheet and for the three sheets coated with Ag films of different thickness (deposition times t = 5, 15 and 25 min). The respective intensities correspond to the area under each one of these curves.In this case the attenuation length is τ = 0.956 µm, using equation (2) one obtains a thickness of 0.20± 0.03 µm for the first film, 0.59±0.03 µm for the second film and 0.99±0.03 µm for the third one, which are proportional values to the corresponding deposition times, as expected. The uncertainty in the film thickness was obtained by error propagation in equation (2) and it is determined by both the uncertainty in the tabulated τ value and the uncertainty in the X-Ray peak intensity ratio R.

The uncertainties in the I_0 and I intensity values can be calculated by repeating several times the acquisition of a given EDX spectrum and comparing the variations in intensity in order to determine the mean value and the standard deviation, what can be performed either experimentally or by Monte Carlo simulations. In the present example we used Monte Carlo simulations with the NIST TDSA-II application [5].

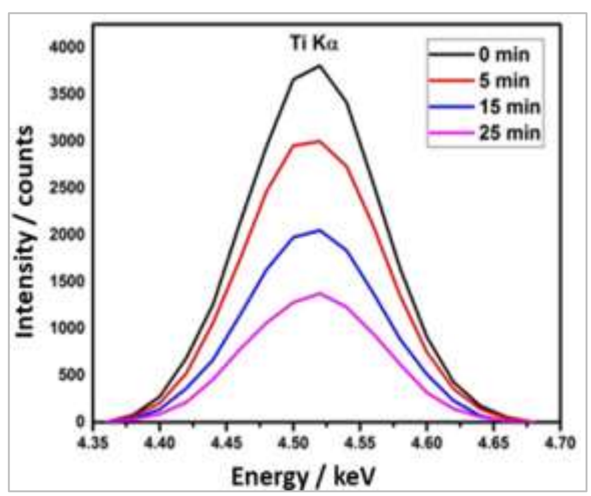

Fig. 5. Attenuation of the Ti Kα peak when traversing PE substrates with Ag films of different thickness.

It is important that the electron beam current be the same when the spectra are acquired for both the substrate sheet without the film and with the deposited film on it.

Films deposited by sputtering at room temperature usually present a columnar structure, like the one shown in Fig. 6. This image corresponds to an Ag film deposited on Si. As can be observed, the film is much less compact than a bulk solid. What is important for the attenuation of the X-Ray beam by photoelectric effect is the number and identity of the atoms that the photons encounter in their passage through the film, not how close or separated these atoms are from each other. Normally the compiled attenuation length values t_{att} assume that the film density corresponds to the density ρ_{bulk} of the bulk material, what is not usually true for most films.

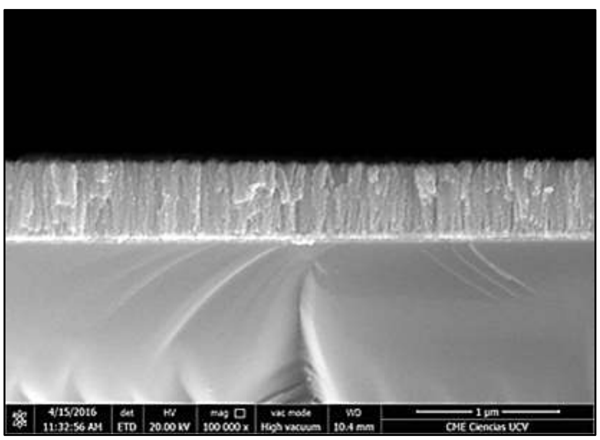

Fig. 6. Secondary electron image of a Ag/Si film observed in cross section.

The thickness values t_{att} to be obtained by this attenuation method would then correspond to the thickness of a film with de density ρ_{bulk}. If the true thickness t_{film} is directly measured, for instance by acquiring a cross section SEM image like the one in Fig. 6, then the true density ρ_{film} can be calculated from the equation (3).

$$\rho_{film}\, t_{film} = \rho_{bulk}\, t_{att} \tag{3}$$

CONCLUSIONS

Using the presented methodology, it has been possible to increase the analytical potential of a SEM-EDX system in order to determine the thickness of films by means of spectroscopic measurements of the attenuation of X-rays generated in a properly designed target.

In addition to the Cu/Ti target presented here, targets of different materials can be designed to choose the characteristic X-ray signals that best suit the particular films of interest. For this purpose the NIST DTSA-II application is a very useful tool.

When films are being deposited by physical methods, it is then recommended to also use, in addition to the substrate of interest, a thin polymeric substrate to collect the film and to take it to the SEM-EDX system in order to be analyzed by the present method. The use of a Si substrate is also recommended in order to fracture it and measure the film thickness by SEM so the film density can be determined using equation (3).

In the case of alloys, the film deposited on the polymeric substrate can be analyzed by EDX to determine its composition and to introduce it, together with an estimated value of its density, in the application of reference [2] in order to obtain the value of the attenuation length τ.

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