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AN ANALYTICAL METHOD OF DETERMINING THICKNESS OF MULTI-LAYER FILMS WITH ELECTRON MICROPROBE

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Abstract

In the previous work we have developed a series of theoretical corrections for calculating the emitted X-ray intensity in multi-layer films. By the use of these theories, along with careful experimental operation of the electron probe microanalysis (EPMA) and Monte Carlo iteration calculation, the thickness of each layer in multi-layer films can be determined.

To test the reliability of this method, the multi-layer film specimens Au/Cu/Si, Cu/Au/Si and Ag/Cr/Si of known thicknesses were analyzed at 20, 25, 30 and 35 keV. The percentage relative errors between the thicknesses determined using the correction procedures and those measured using nuclear backscattering are less than 10%, the average value of the errors is 4.6%.

The method may be extended to the calculations of determining element concentrations for the multi-layer specimens of known thicknesses.

KEY WORDS: Multi-layer film thickness, Electron scattering, Electron probe microanalysis, Monte Carlo simulation, Iteration calculation.

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Introduction

The thickness of a thin film on a substrate can be determined using the electron microprobe analysis and Monte Carlo simulation.[1,2,7] However, the quantitative analysis of multi-layer films on a substrate has proved to be quite difficult.

This paper describes the preparation of multi-layer specimens, the experimental technique of electron microprobe analysis and theoretical correction procedures for determining thicknesses of multi-layer films. Finally, the percentage relative errors and the limitations of this work are discussed.

Preparation of Multi-layer Specimens

In order to reduce the experimental errors, multi-layer film specimens must be carefully prepared.

Choice of Substrate

The substrate on which the multi-layer films are deposited should be smooth, flat, conductive, and stable under electron beam, and not contain the same elements which are to be analyzed for in the multi-layer films. A silicon wafer meets these requirements.

The multi-layer films specimens were deposited on a suitable substrate. For example, the preparation process of the films Au/Cu/Si is: Si substrate was finely polished, then Cu and Au films were deposited on Si plate by vacuum evaporation.

The single-layer film specimens Au/Si and Cu/Si (on Si substrate) were simultaneously prepared for measuring the thicknesses of each thin layer in the sample Au/Cu/Si (Fig. 1a.). The thicknesses of the single-layer films Au/Si and Cu/Si (Fig. 1b) were measured by using nuclear backscattering and shown in table 1. Since nuclear backscattering method can accurately give the mass thicknesses of the single-layer films, the calculated error resulted from the difference between the film density and theoretical density may be avoided. Therefore, it is suitable to take the single-layer films as the standards for determining thicknesses of the multi-layer specimen Au/Cu/Si, i.e. the thicknesses of the single-layer films are those of the corresponding thin layer in the films Au/Cu/Si.

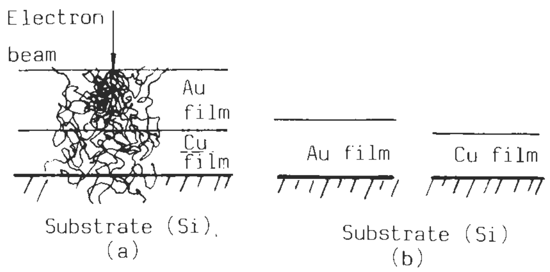

Fig. 1 Schematic diagram of multi-layer films Au/Cu/Si and single film specimens Au/Si, Cu/Si.

Analysis by Electron Microprobe

Choice of Standard

Thick polished specimens have been used as standards for multi-layer film analysis. The standards have the same compositions and physical state as every thin layer of the multi-layer films being analyzed.

Experimental Condition

Usually, in EPMA of film specimens, use of a defocussed beam covering a larger surface of multi-layer films tends to average out some of the effects of variation in film composition and thickness. In this work, the evaporated multi-layer film specimens show such homogeneity that the measured intensity value of X-rays for the specimens maintains constancy while the diameter of electron beam is changed from 1 μm to 50 μm. We take the diameter of electron beam to be 5 μm. Probe current is 2×10^{-8} A.

Experiment of Microprobe

The composition of every layer in the analyzed multi-layer films is known. Beam energy is high enough to penetrate well into the substrate (Fig. 2), and the intensities of X-rays Au-L_α and Cu-K_α from the first layer and the second layer are measured, respectively. Then, the ratios, k_{Au} and k_{Cu}, of X-ray intensities of elements Au and Cu in the specimen to those in the standards are obtained. Finally, the thicknesses of Au film and Cu film in the specimen Au/Cu/Si can be determined by using Monte Carlo iteration method.

Calculation Procedure

X-ray Intensity of Multi-layer Films

For the multi-layer film specimen of known compositions, when the thicknesses of each layer have been given, we can evaluate the X-ray intensities emitted from the elements in each thin layer using our theoretical calculation method[3]. Considering the absorption and fluorescence of

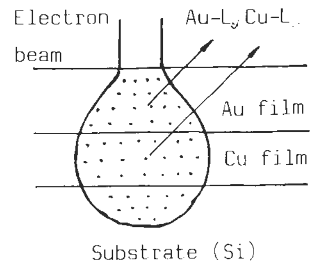

Fig. 2 Schematic diagram of emission of X-rays in multi-layer specimen Au/Cu/Si

characteristic X-rays, the general equation of the emitted X-ray intensity of element i from the nth layer film of multi-layer specimens is:

$$I_{n.i} = \Delta I_{n.i.f} + \int_{(n-1)\text{th layer}}^{n\text{th layer}} \phi_n(\rho_n z)\, d\rho_n z \cdot \exp\left(-\sum_{m=1}^{n} \mu_m \rho_m d_m \csc\psi\right), \quad (1)$$

where $\Delta I_{n.i.f}$ and the integration term are the emitted fluorescence intensity and the generated intensity of characteristic X-rays, respectively, and $\exp(-\sum_{m=1}^{n} \mu_m \rho_m d_m \csc\psi)$ is the absorption term; ϕ_n , $\rho_{n.m}$, μ_m and d_m are the in- stribution in depth of X-rays, the density of the nth (or mth) layer medium, the mass attenuation coefficient of the mth layer medium, and the thickness of the mth layer film respectively, and ψ is the X-ray take-off angle.

A detailed calculation process of Eq.(1) is shown in Refs. (3), (4) and (5): electron scattering in multi-layer media is evaluated by using 'equivalent single medium scattering model'4, and Monte Carlo method, the fluorescence intensity $\Delta I_{n.i.f}$ emitted from the nth layer is strictly calculated by using the formulae developed by us[5].

Iteration Calculation

Since there is a certain proportional relationship between the X-ray intensity ratio, $k_{n.i}$ of element i in the nth layer to that of standard i and the thickness of the nth layer, we may take $k_{n.i}.d_i$ as the first approximate value T_n' of thickness of the nth layer, d_i is the excitation depth of X-ray of element i in solids, it can be calculated by using Monte Carlo method[6]. The electron scattering and X-ray excitation are simulated by Monte Carlo technique for the multi-layer specimen with thickness T_n' to get the ratio value $k'_{n.i}$. Let $\Delta k'_{n.i} = k'_{n.i} - k_{n.i}$, then the secondary approximate value of T_n is

$$T_n^2 = T_n'\left(1 - \frac{\Delta k'_{n.i}}{k_{n.i}}\right). \quad (2)$$

Repeat the above Monte Carlo calculation until the nth iteration, for a given arbitrarily small quantity ε ,
if

$$\left| T_n^n - T_n^{n-1} \right| < \varepsilon , \quad (3)$$

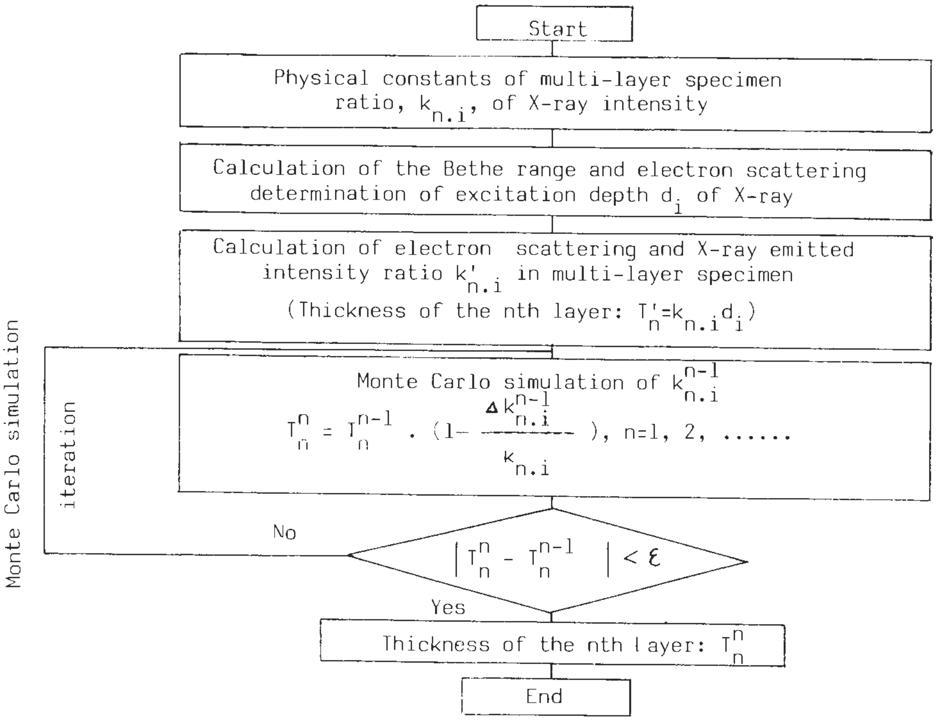

Fig. 3 Schematic flow chart of determining thicknesses of multi-layer films using Monte Carlo simulation.

then T^n_n is the desired thickness of the nth layer film of the multi-layer specimen.

In this work, we let ε be 0.02. This precision is high enough for determination of film thicknesses. Generally, the simulated result will converge after iterating four or five times. Figure 3 shows a diagram of the method employed.

Results and Discussion

In order to examine the reliability of the method, we have carried out the electron microprobe experiments and theoretical calculations for the multi-layer films specimens Au/Cu/Si, Cu/Au/Si and Ag/Cr/Si of known thicknesses. As noted above, the specimen have been carefully prepared with vacuum evaporation. The thicknesses analyzed by using our method are in good agreement with those measured with a nuclear backscattering equipment for the multi-layer specimens (table 1).

We know from table 1 that the differences between the thicknesses determined using our method and those measured using nuclear backscattering for the multi-layer specimens are less than 10%, and the average value of the errors is 4.6%. The accuracy may be higher for film thickness measurement in EPMA. In addition, agreement among the analyzed results for a multi-layer specimen at different accelerated voltages is fairly good. It will be seen from this that the method proposed in this paper is reliable.

However, if thickness of a multi-layer specimen is very thin, it is quite difficult to determine the thickness of each layer. In X-ray quantitative analysis of thin film specimens, the thinner the thickness of any layer, the larger the error will be. The error chiefly results from the experiment of electron microprobe analysis. The X-ray intensity emitted from a very thin layer is quite low, so that there is a large error in measuring X-ray intensity. According to thickness of a layer and incident electron energy, in order to obtain enough scattering step number, one should select a suitable step length to reduce the statistical error in Monte Carlo simulation.

The work may be easily extended to the analysis of EPMA for multi-layer film specimens with polybasic composition.

EPMA experiments were carried out in JCXA-733 Electron Probe. Monte Carlo simulation was calculated on Vax 8350 Computer, the simulated electron number is 2000.

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Table 1. Comparison between the thicknesses analyzed using our method and those determined using nuclear back-scattering analysis.

No.	Multi-layer specimen	Thin layer	Measured ratio $k_{n.i}$ of X-ray intensity			Thickness calculated by Monte Carlo method (mg/cm^2)			Thickness measured with nuclear back-scattering equipment (mg/cm^2)
			20 keV	25 keV	30 keV	20 keV	25 keV	30 keV	
1	Au/Cu/Si	Au film	0.3874	0.2253	0.1487	0.1506	0.1473	0.1488	0.1500
		Cu film	0.1979	0.1428	0.1035	0.1090	0.1028	0.1058	0.0990
2	Ag/Cr/Si	Ag film	0.2681	0.1799	0.1306	0.1410	0.1390	0.1384	0.1370
		Cr film	0.1383	0.0984	0.0676	0.0789	0.0830	0.0843	0.0870
			30 keV	35 keV		30 keV	35 keV		
3	Au/Cu/Si	Au film	0.2331	0.1736		0.2135	0.2256		0.2365
		Cu film	0.2605	0.2128		0.2795	0.3121		0.2982
4	Cu/Au/Si	Cu film	0.3292	0.2407		0.2963	0.3066		0.2982
		Au film	0.2030	0.1688		0.2220	0.2300		0.2365
5	Cu/Au/Si	Cu film	0.2748	0.2072		0.2506	0.2709		0.2609
		Au film	0.2072	0.1568		0.2209	0.2080		0.2256
6	Cu/Au/Si	Cu film	0.0829	0.0635		0.0786	0.0815		0.0746
		Au film	0.4339	0.3416		0.4096	0.4382		0.4513

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Discussion with Reviewers

D.E. Newbury: Please specify the formulation for $\phi_n(\rho_n Z)$ that you are using.

Authers: The distribution function, $\phi_n(\rho_n Z)$, or characteristic X-rays in the nth layer film can be evaluated by using our electron scattering model in multi-layer media based on the Rutherford cross section and Bethe's energy loss equation[4]. The excited probability, $Q_{K,L}$, of X-rays for k or l shell expressed by:

$$Q_{k,l}=q_{k,l}\ \frac{\ln U}{U}\cdot\frac{1}{E_{k,l}^2}, \qquad (4)$$

in which $E_{k,l}$ is the minimum ionization potential, $q_{k,l}$ is a constant, and U is overvoltage.

G. Love: Presumably it is necessary to first know that gold is the outer coating and copper is the inner coating -- this is not arrived at independently from the calculations.

Authors: Yes, we must know the sequence of evaporated multi-layer films on a substrate and the intensity of X-rays emitted from each layer before Monte Carlo calculation. The component of film can be obtained from characteristic X-rays of element, and the sequence can be known with the method of gradually increasing voltage in the electron probe experiment.

G. Love: Is the Monte Carlo method a practical approach for determining coating thickness? How long does it take to determine the thickness using the iteration method described?

Authors: Yes, the Monte Carlo procedure may be a practical and accurate method in EPMA of multi-layer films, since the CPU time we spend is less in calculating thicknesses for a multi-layer specimen. For example, the CPU time to calculate the thicknesses of sample 1 (table 1) at 30 keV is about 12 minutes on Vax 8350 computer.

P. Rez: How is your method affected by the well known diffusion of gold into silicon?

Authors: When an element in a thin layer diffuses into the substrate, it isn't suitable to use the method reported here. For this case, a series of further researches should be done, which include improvement of physical model, study of experimental technique and so on.