Towards a new methodology for measuring the macroscopic Young's modulus of multilayer thin films using Impulse Excitation Technique

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Abstract:

This work presents an enhanced approach to determine the Young's modulus of multilayer composite beam using Impulse Excitation Technique (IET). In this technique, the beam is excited and the corresponding natural frequencies are recorded. The Young's modulus of each individual coating was obtained using different analytical models in the literature and the measured frequencies. Some of the models are limited to a certain film thickness and some others are limited to one-layer coatings. For the purpose to cover all the applicable range of coating's thickness and for multi-coated beam, one-dimensional model was developed based on the flexural theory. A finite element model is developed and used, as a reference, to validate the proposed model. The proposed relation takes into account the shift of the neutral axis after deposition, which makes it applicable for any film thickness. After validation, the model is applied to determine the macroscopic Young's modulus of niobium and titanium films deposited on AISI 316 and glass substrates by conventional magnetron sputtering.

Keywords: Elasticity constants, Multilayers, Coatings, Impulse Excitation Technique, Magnetron sputtering.

1 Introduction

Thin films technology was developed to improve the mechanical and the physicochemical properties of a material. Many surface treatment processes are used to deposit these films among which we can mention PVD, CVD and thermal evaporation. These films can be used individually or in multilayer depending on the required application. The elastic properties of thin films differ from those of bulk materials and depend on the technique of elaboration and its parameters. This paper presents a methodology to determine the macroscopic Young's modulus of thin films in multilayer composited beam. Firstly, a study of the models proposed in the literature will be done to identify the ideal model that can be used to determine the Young's modulus of one layer coating. Then an extension of the models is developed to determine the Young's modulus of each film in a multilayer structure. It is valid for any ratio of coating to substrate thicknesses. Finally, an example of application on niobium and titanium thin films deposited by pulsed-DC magnetron sputtering will be presented.

2 Vibrational Approach

2.1 Impulse Excitation Technique (IET)

The principle is to perform a sequence of frequency measurements, one for the substrate without deposition and one after each film deposited. Knowing the resonance frequencies of the substrate and of the composited beam, the density of each coating and substrate, the Young's modulus of each film can be determined through analytical expressions.

2.2 Analytical models for coated beam

Based on the flexural rigidity of composite beam theory, which assumes that the coating is homogeneous and isotropic and using the solution of Euler-Bernoulli [1]], the resonant frequency of the composite beam can be determined from the following equation:

$$f_N = \frac{X_n^2}{2\pi L^2}\sqrt{\frac{E_t I_t}{\rho_t A_t}} \tag{1}$$

where L is the beam length, X_n is the non-dimensional Eigen frequency and E_tI_t is the flexural rigidity of the entire composite beam expressed as the sum of the flexural rigidity of each layer [2]]. The product ρ_tA_t is expressed as the summation of the corresponding product of each layer.

Lopez's model [3]] was developed for a free composite beam (substrate + coating) with rectangular cross-sectional area to determine the Young's modulus of the single layer coating and assumes that the neutral axis remains fixed after deposition as follows:

$$E_{c1} = \frac{E_s}{4R_{h1}^3 + 6R_{h1}^2 + 3R_{h1}}\left[\left[\left(1 + R_{h1}R_{\rho1}\right)\left(R_{f1}\right)^2\right] - 1\right] \tag{2}$$

where $R_{h1}=h_{c1}/h_s$, $R_{\rho1}=\rho_{c1}/\rho_s$, $R_{f1}=f_1/f_s$. E is the Young's modulus, h is the thickness, ρ is the density, f_s is the substrate's frequency, f_1 is the frequency of the whole beam (substrate + coating) and the indexes s and c_1 correspond respectively to the substrate and the single layer coating.

Pautrot et al. [3] developed a model in order to take into account the shift of the neutral axis after deposition. Using the following equation, the Young's modulus of the coating is obtained:

$$\begin{aligned} E_{c1} &= \frac{E_s}{2R_{h1}^4}\Bigg[\left(R_{h1} + R_{\rho1}R_{h1}^2\right)\left(R_{f1}\right)^2 - 4R_{h1}^3 - 6R_{h1}^2 - 4R_{h1} \\ &+ \sqrt{4R_{h1}^4\left[\left(1 + R_{h1}R_{\rho1}\right)\left(R_{f1}\right)^2 - 1\right] + \left[4R_{h1}^3 + 6R_{h1}^2 + 4R_{h1} - \left(R_{h1} + R_{\rho1}R_{h1}^2\right)\left(R_{f1}\right)^2\right]^2}\,\Bigg] \end{aligned} \tag{3}$$

A simplified approach was provided by Whiting et al. [4] to directly calculate the Young's modulus with further simplification:

$$E_{c1} = \frac{E_s}{3}\left[\frac{2R_{f1} + R_{\rho1}R_{h1} - 2}{R_{h1}}\right] \tag{4}$$

Berry's model was developed by applying the first order Taylor series expansion of either Lopez's or Pautrot's models [5]:

$$E_{c1} = \frac{E_s}{3}\left[\frac{\left(R_{f1}\right)^2 + R_{\rho 1}R_{h1} - 1}{R_{h1}}\right] \tag{5}$$

Based on the classical laminated beam theory (CLBT), which fulfills Kirchhoff's law, another model was proposed and it is applicable to multilayer composite beam [6]:

$$E_{c1} = \frac{-\gamma_a + \sqrt{\gamma_a^2 - 4\gamma_b}}{2} \tag{6}$$

where:

$$\gamma_a = \frac{2\varphi(1 - \nu_s\nu_{c1}) - \phi_R}{H_2} \tag{7}$$

$$\gamma_b = \frac{[\varphi^2(1 - \nu_s^2) - \varphi\phi_R](1 - \nu_{c1}^2)}{H_2^2} \tag{8}$$

with:

$$\varphi = \frac{H_1 E_s}{1 - \nu_s^2} \tag{9}$$

$$\phi_R = \left(\frac{E_s h_s^2}{12}\left(\frac{\rho_s h_s + \rho_{c1}h_{c1}}{\rho_s}\right)\right)\left(R_{f1}\right)^2 \tag{10}$$

$$H_1 = \frac{h_s}{4}\left(\frac{h_s^2}{3} + h_{c1}^2\right) \tag{11}$$

$$H_2 = \frac{h_{c1}}{4}\left(\frac{h_{c1}^2}{3} + h_s^2\right) \tag{12}$$

2.3 Developed models for multilayer composite beam

Lopez's model is extended to a free bi-coated beam (substrate + film 1 + film 2) with a rectangular cross-sectional area. The new expression of the second film Young's modulus is obtained without considering the shift of the neutral axis:

$$E_{c2} = \frac{\left[\left(1 + R_{h1}R_{\rho 1} + R_{h2}R_{\rho 2}\right)\left(R_{f2}\right)^2 - R_{E1}(4R_{h1}^3 + 6R_{h1}^2 + 3R_{h1}) - 1\right]E_s}{4R_{h2}^3 + 6R_{h2}^2 + 3R_{h2} + 12R_{h1}R_{h2}(R_{h1} + R_{h2} + 1)} \tag{13}$$

where $R_{h2}=h_{c2}/h_s$, $R_{\rho 2}=\rho_{c2}/\rho_s$, $R_{f2}=f_2/f_s$.

Another extension is applied to Pautrot's model, where the neutral axis will shift after each layer deposited and can be generalized for *N* isotropic layers. The Young's modulus of the second film is determined:

$$E_{c2} = \frac{-b_1 + \sqrt{b_1^2 - 4a_1c_1}}{2a_1} \tag{14}$$

where:

$$a_1 = R_{h2}^4 \tag{15}$$

$$\begin{aligned} b_1 = 4R_{h2}^3(1 + R_{E1}R_{h1}) + R_{h2}^2\left[6 - R_{\rho 2}\left(R_{f2}\right)^2 + 6R_{h1}(2 + R_{E1}R_{h1})\right] \\ + R_{h2}\left[4 - \left(R_{f2}\right)^2 + 4R_{h1}^2(3 + R_{E1}R_{h1}) \right. \\ \left. + R_{h1}\left[12 - R_{\rho 1}\left(R_{f2}\right)^2\right]\right] \end{aligned} \tag{16}$$

$$\begin{aligned} c_1 = 1 - (1 + R_{E1}R_{h1})\left(1 + R_{\rho 1}R_{h1} + R_{\rho 2}R_{h2}\right)\left(R_{f2}\right)^2 \\ + R_{E1}R_{h1}\left(4 + 6R_{h1} + 4R_{h1}^2 + R_{E1}R_{h1}^3\right) \end{aligned} \tag{17}$$

The CLBT model can also be used for multilayer composite beam and the corresponding expression of the second film Young's modulus is obtained:

$$E_{c2} = \frac{-\lambda_a + \sqrt{\lambda_a^2 - 4\lambda_b}}{2} \tag{18}$$

where:

$$\lambda_a = \frac{2\varphi_1(1 - \nu_s\nu_{c2}) + 2\varphi_2(1 - \nu_{c1}\nu_{c2}) - \phi_R}{Z_3} \tag{19}$$

$$\begin{aligned} \lambda_b = \frac{(1 - \nu_{c2}^2)}{Z_3^2}[\varphi_1^2(1 - \nu_s^2) + \varphi_2^2(1 - \nu_{c1}^2) + 2\varphi_1\varphi_2(1 - \nu_s\nu_{c1}) \\ - \phi_R(\varphi_1 + \varphi_2)] \end{aligned} \tag{20}$$

with:

$$\varphi_1 = \frac{Z_1 E_s}{1 - \nu_s^2} \tag{21}$$

$$\varphi_2 = \frac{Z_2 E_{c1}}{1 - \nu_{c1}^2} \tag{22}$$

$$\phi_R = \left(\frac{E_s h_s^2}{12}\left(\frac{\rho_s h_s + \rho_{c1}h_{c1} + \rho_{c2}h_{c2}}{\rho_s}\right)\right)\left(R_{f2}\right)^2 \tag{23}$$

$$Z_1 = \frac{h_s}{12}[h_s^2 + 3(h_{c1} + h_{c2})^2] \tag{24}$$

$$Z_2 = \frac{h_{c1}}{12}[h_{c1}^2 + 3(h_{c2} - h_s)^2] \tag{25}$$

$$Z_3 = \frac{h_{c2}}{12}[h_{c2}^2 + 3(h_{c1} + h_s)^2] \tag{26}$$

For one coating (N=1), a good agreement between the Pautrot's model and the Finite Element Model (FEM) for any thickness, density and Young's modulus ratios has been noticed by Slim et al. [7-9]. In addition, a divergence between the other models becomes larger for higher ratios. This is due to the assumptions on which is based each model. The extended models for two layers of coatings (N=2) will be analyzed quantitatively by comparing them to FEM which is taken as a reference.

3. Trueness analysis

A parametric comparison of the analytical models with FEM was performed in order to identify the most accurate one. The evolution of the frequency ratio R_{f2} as a function of the thickness R_{h2}, Young's modulus R_{E2} and density $R_{\rho 2}$ ratios is represented in Fig. 1. Four different combinations of materials were chosen where R_{h2} was varied from 0 to 0.55 mm. A good agreement can be seen between the extended Pautrot's model ((14)) and FEM. We can also observe the divergence of the other models due to their supposed assumptions during the derivation of the expressions. According to these results, we can mention that the extended Pautrot's model exhibits results similar to those of FEM and is recommended to determine the Young's modulus of coatings in multilayers.

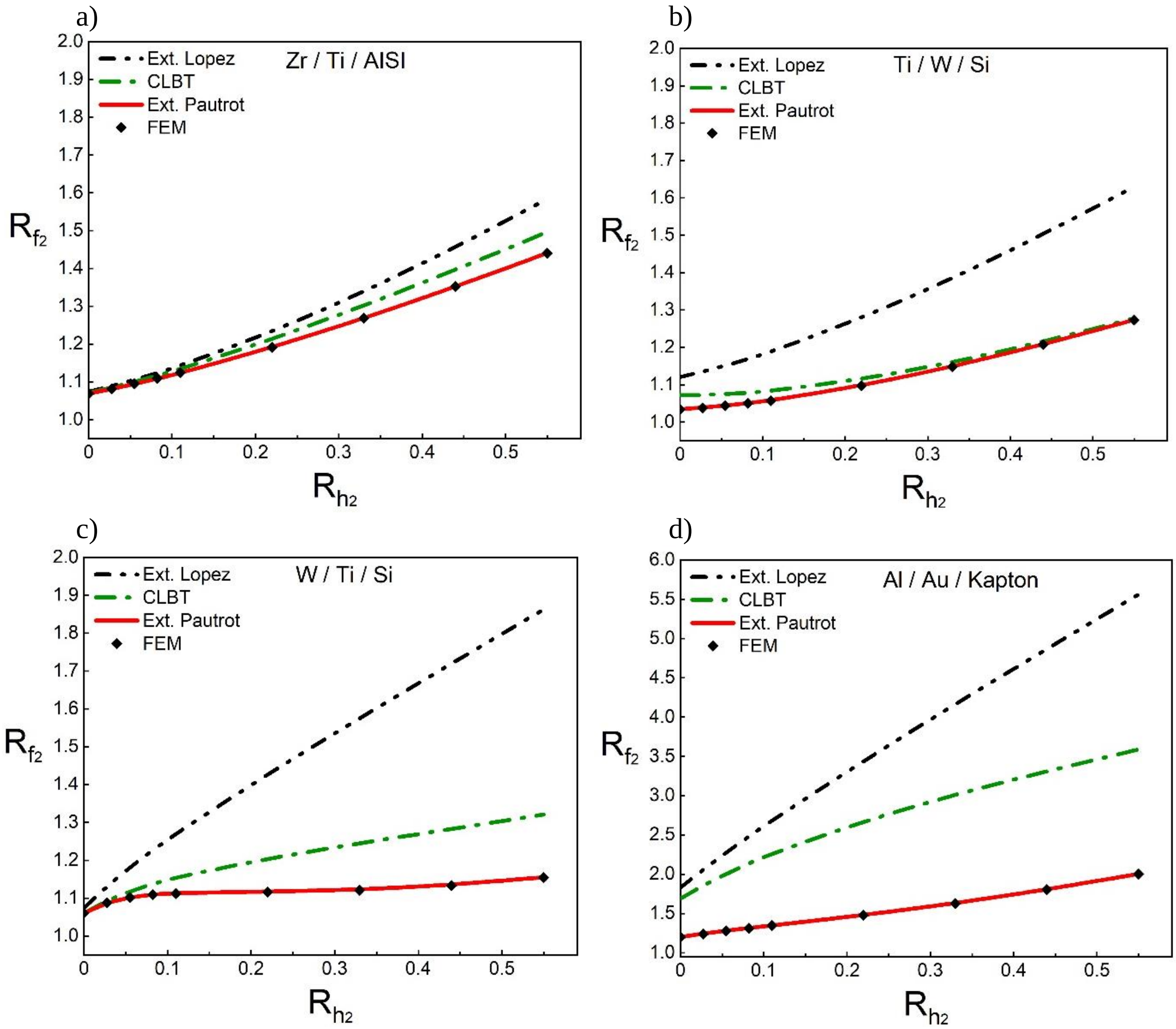

Fig. 1. Evolution of the frequency ratio as a function of thickness, Young's modulus and density ratios: a) R_{E2}=0.5, $R_{\rho2}$=0.81, b) R_{E2}=1.03, $R_{\rho2}$=1.93, c) R_{E2}=3.56, $R_{\rho2}$=8.28 and d) R_{E2}=18.68, $R_{\rho2}$=1.9.

4. Conclusion

The Impulse Excitation Technique (IET) was used to measure the frequencies before and after the deposition of each film. Then, the macroscopic Young's moduli of substrates and coatings were obtained using the best analytical model. Titanium and niobium thin films have been deposited by pulsed-DC magnetron sputtering and their Young's moduli were determined by IET.

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