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A simple guide to determine elastic properties of films on substrate from nanoindentation experiments

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Abstract

Several models have been developed to extract the intrinsic elastic modulus of thin films from the composite film/substrate modulus value obtained from indentation tests on coated systems. Either analytical, semi-analytical or empirical, they generally propose an expression of the composite modulus as a function of the film and substrate elastic moduli and of the film thickness. When the substrate properties and the film thickness are known, the expression without adjustable parameter contains only the film elastic modulus as unknown parameter, which can thus be deduced.

In this paper, some models from the literature are briefly described and compared with the easy-to-use model we have developed. Based on experimental results obtained with various film/substrate systems, this paper also aims to demonstrate that the error induced by the uncertainty in the knowledge of the substrate Young modulus value and in the film thickness value is often as large as the difference resulting from the choice of the model.

Keywords: Nanoindentation; thin films; Young modulus; elastic model; coatings

1. Introduction

Nanoindentation is extensively used to measure near surface mechanical properties relevant for contact studies. It is also widely used to determine the elastoplastic properties of materials deposited as thin films onto substrates. In this case, although it is still used [1], the simple approach which consists in limiting the indentation depth to a few percent of the film thickness, then to consider the results as if they were obtained on bulk material, is not relevant for the elastic properties. As the matter of fact, the volume of material involved in elastic measurement is large

(half sphere with a radius equal to about ten times the equivalent contact radius [2]) and the elastic deformation is not confined to the film itself. An appropriate model is thus required to extract the intrinsic thin film elastic modulus E_f^* from the composite film/substrate elastic modulus E_{app}^* obtained from indentation tests. E_{app}^* is the calculated reduced Young modulus from indentation curves obtained on the coated system, after correction of the influence of the indenter.

In the literature, the elastic solution of an axisymmetric mixed boundary value model is considered. To be applied to experimental results, numerical treatments are required [3, 4]. Other models were published [5-8] and are described and compared in a paper by Mencik [9] but the list was not exhaustive. Some models are analytical or semi-analytical, other are empirical. They generally propose an expression of the composite modulus as a function of the film and substrate elastic moduli, of the film thickness and, for some of them, of an adjustable parameter noted α . When the substrate properties and the film thickness are known, the expression without adjustable parameter only contains the film elastic modulus as unknown parameter, which can thus be extracted. When an adjustable parameter α is used, the optimum combination of the two unknown parameters - elastic film modulus E_f^* and α - has to be found. However, the precise knowledge of the substrate properties and of the film thickness themselves are not so simple. The aim of the paper is to provide a simple guideline to determine elastic properties of films on substrate from nanoindentation experiments, on samples with approximatively known characteristics, or even totally unknown, such as industrial samples, friction films, For this reason, exact elastic solutions and models which use adjustable parameters (that means additional unknown parameters) will not be considered in the following. Nevertheless, as they are frequently used, a short description of these models with relevant references is given in the first part of the paper, which briefly describes models that can be found in the literature. In the second part, the question of how the uncertainty on the Young modulus value of the substrate may affect the calculated value for the film/substrate system, and thus the determination of the film modulus, is discussed. The third part is devoted to the influence of the film thickness value, as it is not either a parameter which can be easily determined. The need of considering or not an effective thickness diminishing with increasing indentation depth will also be discussed.

2. Existing models

2.1. Short description of some models

The first analytical expression for the compliance of a film/substrate system was established by Doerner and Nix [5], who proposed an empirical expression including exponential terms depending on the relative indentation depth, h_{eff}/t , where h_{eff} is the effective depth and t is the film thickness, multiplied by an empirically determined constant weight factor α . This expression was valid only for the particular case that they studied in their paper, which was sputtered tungsten films on silicon substrates. It was modified by King [6], who used numerical methods to make it applicable to all film/substrate systems and for different indenter geometries. For a given indenter geometry, abacuses were numerically defined to give the weight factor α as a function of normalized equivalent punch size a/t , where a is the square root of the projected contact area divided by π and t the film thickness. As the weight factor is not constant along penetration depth, the use of these abacuses is required to determine the film modulus. Using a finite element method, Battacharya and Nix [7] have obtained results in good agreement with King results for aluminium film on silicon substrate and vice versa. Recently, Saha and Nix [10] have fitted the King numerical α values using a polynomial function and have extrapolated the curve to larger a/t ratios. They also proposed a modified expression of the King's model to extend the analysis to the use of a pyramidal Berkovich indenter. Moreover, the modified model assumes that the flat punch is located at the indenter tip instead of the film surface. This leads to replace, in King equation, the film thickness t by an effective thickness $(t-h)$ equal to the film thickness t minus the total indenter displacement h . This leads to overestimate the contribution of the substrate stiffness and thus to underestimate the film modulus. They applied this model to aluminium films of various thicknesses on different substrates (sapphire, silicon and aluminium). The calculated film modulus value they obtained was in the order of magnitude of the expected value for indentation depths less than 50% of the film thickness, however with rather high scatter.

Other models, leading to rather complex analytical formulations, are based on the work of Gao et al. [8], who derived an analytical expression without adjustable parameters for the film/substrate modulus, from the analysis of the contact between a cylindrical punch and a coated material. This approach was originally developed for the shear modulus. The effective contact

compliance is obtained as a function of the film and substrate shear moduli and Poisson's ratios, through weighting functions I_0 and I_1 , which depend on the ratio t/a , where t is the film thickness and a is the contact radius. The function I_0 also depends on a Poisson's ratio value ν which can be taken either as the film or the substrate value. The ambiguity resulting from the ν dependence of I_0 can be neglected when the ratio of the film and substrate shear moduli is between 0.5 and 2 and if the film and substrate Poisson's ratios are between 0.2 and 0.4, which gives the limit of use for this model. Song and Pharr [11, 12] have recently proposed an extension of the Gao expression, modified to become applicable to a larger range of film and substrate moduli mismatch. As in Gao expression, the effective compliance of the composite system is expressed as a function of film and substrate Poisson's ratios and shear moduli. The same I_0 and I_1 weighting functions are used. Experimental results obtained with a Berkovich indenter were presented with data treated in two different ways. In the first treatment, they kept constant the real film thickness t in the equations, and in second one, they assumed that the appropriate thickness in the equations was the reduced thickness $(t-h_c)$ obtained by subtracting the contact depth h_c to the film thickness t . Comparison between experimental compliance and calculated compliance using the Song/Pharr's model, the Gao's model and numerical results obtained with a commercial software for elastic analysis of indentation problems, were presented on fluorinated silicate glass films on silicon substrates. A good agreement was obtained for the Song/Pharr's model with the reduced thickness assumption and the experimental and numerical data. Even if they give analytical expressions without adjustable parameters, one difficulty in using Gao or Song/Pharr's models comes from the fact that they use film and substrate Poisson's ratios values, which have to be known, contrary to other models which consider the reduced Young modulus $E^*=E/(1-\nu^2)$ for both film and substrate.

Another analytical model was proposed by Bec et al. [13-15], based on the indentation, by a rigid cylindrical punch (radius a), of an homogeneous film (reduced Young modulus E^*_f , thickness t) deposited onto a substrate (semi-infinite half space, reduced Young modulus E^*_s). This system was simply modelled by two springs connected in series (figure 1). The global stiffness K_z of the film/substrate system was then obtained from the reciprocal sum of the film stiffness ($K_f=\pi a^2 E^*_f/t$) and the substrate stiffness ($K_s=2E^*_s a$), each multiplied by correcting polynomial functions $f_1(a)$ and $f_2(a)$, chosen in the form $f_i(a)=1+ka^n$, to ensure correct boundary conditions.

Calculations lead to $f_1(a)=f_2(a)=1+2t/\pi a$. The global stiffness of the film/substrate system is then :

$$\frac{1}{K_z} = \frac{1}{1 + 2t/\pi a} \left(\frac{t}{\pi a^2 E_f^*} + \frac{1}{2a E_s^*} \right) \quad (1)$$

As the global stiffness K_z is related to the apparent reduced modulus E_{app}^* of the film/substrate system through the relation $K_z=2E_{app}^*a$, the film reduced modulus E_f^* can be easily calculated from the apparent modulus E_{app}^* if the reduced Young's modulus of the substrate E_s^* , and the film thickness t are known. For experimental nanoindentation devices with dynamic measurements, the measured global stiffness K_z can be directly used to obtain E_f^* . It is also worth noting that, thanks to its structure, this very simple model can be easily extended to bilayer systems, or even more.

More recently, Perriot and Barthel [4] have calculated the equivalent modulus from numerical treatment of elastic equations, then have proposed an expression to fit the obtained curves, including two parameters, x_0 and n , whose numerical values depend on the elastic mismatch E_s^*/E_f^* .

2.2. Comparison between models

Figure 2 compares the apparent modulus E_{app}^* versus non dimensional equivalent contact radius a/t for 4 models without adjustable parameters in the case of a compliant layer onto a rigid substrate (2a and 2c) and vice versa (2b and 2d), with an elastic mismatch of two (2a and 2b) and with an elastic mismatch of ten (2c and 2d). In the case of the high elastic mismatch (0.1 and 10), Gao expression is out of its application field and the corresponding curves are not plotted. Points resulting from finite element simulation with adaptative remeshing procedure [16] are plotted for comparison in the case of the smaller mismatch (0.5 and 2).

First, this figure confirms that even at small depth compared to the film thickness, the influence of the substrate stiffness leads to a significant difference between the apparent modulus and the film modulus whatever the chosen model is.

Second, in the case of a compliant layer onto a rigid substrate (2a and 2c), the model from Bec et al., Rar et al. (also called Song/Pharr's model) and Perriot et al. give similar curves.

Moreover, there is a good agreement between these curves and the points resulting from finite element simulations. In the case of a rigid layer onto a compliant substrate, the curves plotted using the models of Bec et al., Rar er al. and Perriot et al. are rather similar for small mismatch but the agreement with the points from finite element simulation is not as good as in the preceding case. The best agreement with FEM results is obtained with the model of Perriot et al., which is the most difficult to use in a practical point of view, when the elastic mismatch is unknown. This model can be used to refine the results, after having first estimated the film modulus using models which can be simply inverted. For high mismatch (0.1 and 10), the difference between the models is more pronounced. This may be partially explained by the fact that, in the case of a rigid layer onto a compliant substrate, specific deformation mechanism occurs and a 'membrane effect' should certainly be considered, which is not done in any of the presented models.

In the following, the analysis is focused on the cases where neither the elastic modulus of the substrate, nor the film thickness are precisely known.

3. Effect of uncertainty on the substrate modulus value

When models are used to determine the elastic modulus of a thin layer deposited onto a substrate from nanoindentation tests, it is assumed that the elastic properties of the substrate, considered to be an homogeneous semi-infinite half-space, are known. Actually, some uncertainty often exists on the value that has to be taken for the elastic modulus of the substrate. For example, when this value is measured by nanoindentation, an error of 5 to 10% can be reasonably expected. In the case of non isotropic substrate such as silicon for example, the elastic modulus may depend on the crystal orientation [17]. For polycrystalline materials, such as steel which is frequently used in industrial applications, only a mean value for the elastic modulus can be taken, which can be more than 10% different than the local value, measured for instance by nanoindentation and which depends on the indented grain for instance. Consequently, the determination of the Young modulus of the layer will be affected by this uncertainty.

The curves plotted in figure 3 illustrate how a small deviation of about 5% of the modulus value of the substrate may affect the calculation of the apparent modulus E^*_{app} for a film/substrate system. The experimental results were obtained by indenting a thin gold layer deposited onto a silicon wafer by magnetron sputtering. All the nanoindentation experiments of

this study were performed with the three axial surface force apparatus of the Ecole Centrale de Lyon in continuous stiffness measurement mode [15, 18]. The indenter was a Berkovich diamond tip. On this figure, the measured apparent reduced Young modulus is compared with the apparent modulus calculated from the model of Bec et al. (4a) and from the model of Rar et al. (4b), using the following values for the fixed parameters:

- Young modulus for the gold layer: 74 GPa,
- Poisson's ratio for the gold layer: 0.42,
- Thickness of the gold layer: 65 nm. This value was determined from hardness measurement using a cube corner indenter and confirmed by AFM measurements on a step created in the layer by masking partially the substrate during the deposition process).
- Poisson's ratio for the silicon substrate: 0.25.

Three different values were taken for the reduced Young modulus of the silicon substrate: 180 GPa, 190 GPa (measured by nanoindentation on uncoated silicon wafer) and 200 GPa. The contact radius is calculated from the measured indentation depth, taking into account the tip defect and the pile-up [15].

For the two models, a good agreement is obtained between the calculated apparent modulus and the measured one but for slightly different values for the Young modulus of the substrate, with less than 10% between these two values. This shows that the error that can be induced by the possible uncertainty on the Young modulus value of the substrate, assumed to be known, can be as large as the difference coming from the choice of the model.

The same conclusion can be drawn in the case of the indentation of a rigid thin chromium layer deposited by magnetron sputtering on a less rigid silicon substrate, with the following parameters (figure 4):

- Young modulus for the chromium layer: 310 GPa,
- Poisson's ratio for the chromium layer: 0.3,
- Thickness of the chromium layer: 10 nm,
- Poisson's ratio for the silicon substrate: 0.25.

In this case, the two models give very similar results and the difference between the two models is much smaller than the spreading induced by the uncertainty on the Young's modulus value of the substrate.

4. The film thickness: a complex parameter

4.1. Effect of the possible error on the film thickness value

The film thickness is the second parameter whose value has to be known for the determination of the Young's modulus of a thin film from indentation tests performed on a coated system. But, because of the possible roughness of the substrate in some applications, because of possible local heterogeneity in film thickness or simply because it is difficult to measure precisely the thickness of a thin film, its value is often estimated with an uncertainty which is rarely lower than 10%. Therefore, the value of the apparent modulus of a coated system, calculated using models, can be significantly affected by this uncertainty on the film thickness measurement.

As an illustration, the apparent modulus measured on the same gold layer as figure 3 is calculated using the model of Bec et al. with three different values for the film thickness: 55 nm, 65 nm and 75 nm and plotted in figure 5 (the Young's modulus of the silicon substrate was taken at 190 GPa). It is compared with the apparent modulus calculated with the model of Rar et al. This shows that the difference resulting from the variation of the film thickness value is comparable to the difference due to the choice of the model.

4.2. Effective thickness

The notion of effective thickness was introduced by Saha and Nix [10] who proposed that the equivalent flat punch considered for the elastic analysis should be located at the end of the tip of the Berkovich indenter instead of being located at the film surface like in King's analysis [6]. In this case, the effective thickness ($t-h$) of the thin film diminishes when the penetration depth increases and reaches zero when the penetration depth h is equal to the film thickness t .

In the case of a Berkovich indenter indenting a soft thin film, comparison between calculated apparent modulus and experimentally measured modulus shows a good agreement without introducing such an effective thickness (see for example figure 3). Indeed, the large amount of pile-up around the indenter permits to compensate the reducing quantity of matter under the tip itself. Furthermore, Berkovich indenters are rather 'flat' as far as their geometry is

concerned. On the other hand, the situation is slightly different for cube corner indenters which are much sharper. Nevertheless, if we considered the volume of thin film under the tip (cylinder of radius a and of height t) and if we subtract the volume occupied by the cube corner indenter at a given plastic depth h_p , the remaining matter occupies a cylinder of radius a , whose height is equal to $(t-h_p/3)$. This leads us to propose that the effective thickness to consider, in the case of the indentation of a thin film using a cube corner indenter, should be equal to $(t-h_p/3)$, where t is the film thickness and h_p the plastic depth.

Comparison between experimental results and calculated values of apparent modulus in the case of the indentation of the same gold layer as previously, but using a cube corner indenter, is presented in figure 6 for both cases: the film thickness t is unchanged and an effective film thickness $(t-h_p/3)$ is considered. A good agreement between experimental points and calculated values is obtained when $(t-h_p/3)$ is taken for the effective thickness of the layer.

5. Conclusions

A simple easy-to-use expression giving the elastic response of a coated system during an indentation test is compared to other existing models. It permits to extract the reduced modulus of the thin film from the global elastic measurement and can be easily extended to bilayer systems. This model gives comparable results as more complex ones and finite element simulations. For rigid films on compliant substrate, a 'membrane effect' should be considered to obtain a better agreement between calculation and experiment.

Using experimental results obtained on various thin films, it was shown that the uncertainty on the substrate modulus value or on the film thickness leads to spreading similar to the difference introduced by the choice of a model. Consequently, on samples with approximately known characteristics, such as industrial samples for instance, the simple approach presented in this paper gives good results.

The necessity of considering or not an effective thickness which diminishes when the indentation depth increases was investigated. When a cube corner indenter is used, it is proposed to consider $(t-h_p/3)$ for the effective thickness, where t is the film thickness and h_p is the plastic depth.

To go further, the same writing can be used to model the dissipative response of a coated system (calculation of the loss modulus), or to model the shear modulus.

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FIGURE CAPTION

Figure 1:

Schematic description of the model of Bec et al. based on the indentation, by a rigid cylindrical punch (radius a), of an homogeneous film (reduced Young's modulus E_f^* , thickness t) deposited onto a substrate (semi-infinite half space, reduced Young's modulus E_s^*). This system is modelled by two springs connected in series.

Figure 2:

Evolution of the apparent modulus versus non-dimensional contact radius calculated from different models without adjustable parameters, in the case of different modulus mismatches :

- (a) $E_f = 100$ and $E_s = 200$ ($\nu=0.3$)
- (b) $E_f = 200$ and $E_s = 100$ ($\nu=0.3$)
- (c) $E_f = 20$ and $E_s = 200$ ($\nu=0.3$)
- (d) $E_f = 200$ and $E_s = 20$ ($\nu=0.3$)

Points obtained from Gao's model are not plotted for modulus mismatches of 0.1 and 10 as it is not valid in this case. Points resulting from finite element simulation with adaptative remeshing procedure are plotted for comparison for modulus mismatches of 0.5 and 2.

Figure 3:

Evolution of the apparent modulus E_{app}^* versus plastic depth, calculated with two different models, for three different values for the reduced Young's modulus of the substrate, in the case of the nanoindentation of a gold layer (65 nm thick) deposited by magnetron sputtering onto a silicon wafer:

- (a) from Bec et al. model
- (b) from Rar et al. model

Experimental points for the apparent modulus obtained by nanoindentation using a three-axial surface force apparatus equipped with a Berkovich tip are plotted for comparison.

Figure 4:

Evolution of the apparent modulus E^*_{app} versus plastic depth, calculated with two different models, for three different values for the reduced Young's modulus of the substrate, in the case of the nanoindentation of a chromium layer (10 nm thick) deposited by magnetron sputtering onto a silicon wafer:

(a) from Bec et al. model

(b) from Rar et al. model

Experimental points for the apparent modulus obtained by nanoindentation using a three-axial surface force apparatus equipped with a Berkovich tip are plotted for comparison.

Figure 5:

Evolution of the apparent modulus E^*_{app} versus plastic depth, calculated with the model of Bec et al., for three different values for the film thickness, in the case of the nanoindentation of a gold layer (65 nm thick) deposited by magnetron sputtering onto a silicon wafer ($E^*_s = 190$ GPa) using a Berkovich indenter.

The curve obtained using the model of Rar et al. with 65 nm for the film thickness is plotted for comparison.

Figure 6:

Indentation of a gold layer (65 nm thick) deposited onto a silicon substrate ($E^*_s=190$ GPa) with a cube corner indenter.

Comparison between the calculated apparent modulus E^*_{app} from the model of Bec et al considering the film thickness t and the calculated modulus using $(t-h_p)$ as apparent film thickness, where h_p is the plastic depth.

The evolution of the apparent modulus measured versus plastic depth is plotted for comparison.