A Method to Extract the Intrinsic Mechanical Properties of Soft Metallic Thin Films Based on Nanoindentation Continuous Stiffness Measurement Technique

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Abstract. In order to determine accurately the intrinsic hardness of the soft metallic thin film on a hard substrate using nanoindentation, a proper methodology irrespective of several important effects the Oliver-Pharr method concerns is described. First, the original analysis data such as the load, \( P \), and contact stiffness, \( S \), as a function of the indentation depth, \( h \), are acquired by means of the continuous stiffness measurement (CSM) technique. By CSM, the complicating effects including indentation creep behaviour of metal materials as well as thermal drift on the measured results are avoided effectively. Then, the hardness of film-only is calculated via a material characteristic parameter, \( P/S^2 \), which is independent of the contact area, \( A \), based on the constant modulus assumption method. In this way, the influences of the substrate contribution and material pile-up behaviour needn’t be accounted for. Guided by above ideas, moreover, a 504 nm Au film on the glass substrate system was chosen to study. The results show that the hardness of Au thin film is 1.6±1 GPa, which agree well with the literature. While the composite hardness measured by Oliver-Pharr method is between 2~3GPa, obviously, which is overestimated. This implies the present methodology is a more accurate and simple way for extracting the true hardness of the soft metallic thin films.

1. Introduction

Nanoindentation is now widely used in measuring the hardness and Young’s modulus of thin films [1-10]. In indentation data analysis, the method proposed by Oliver and Pharr [1] is the most popular and has been taken as a standard one [1-5]. However, due to the Oliver-Pharr method was developed for monolithic materials and based on the assumption that the unloading-displacement response is purely elastic, the misleading evaluated results will inevitably occur when it applies to the thin films on the substrates or the metal materials. Especially, for the soft metallic films, measurement of the true mechanical properties becomes more difficult. In this case, one should first take into account the substrate interactions on the indentation response of thin films [2-3]. In addition, much indentation plastic behaviors for the metal materials, such as creep [4], pile-up [5], etc. must be process well in the data interpretation. Otherwise, wrong evaluated results will confuse us.

How to determine the intrinsic mechanical properties of the films has been a focus in nanoindentation studies. However, the proposed methods [2,6,7] by many investigators all have some various degrees of deficiencies so that their applications are limited. For example, the rule of thumb [6] to avoid the substrate effects that controlling indentation depths less than of 10% the total film
thickness is not available to the very thin films, where substrate contribution as well as so-called indentation size effects (ISE) [8] are very remarkable. Also, due to too much information required, the method to extract the intrinsic mechanical properties of thin films from the composite hardness or modulus through various mathematic models is very complex, which cannot meet the needs of daily measurements. Furthermore, the reliability of results obtained by these models needs discussed further, since the effects of the indentation plastic behaviors of metal materials don’t be accounted for. Therefore, the aim of this paper is to attempt to find a more effective and accurate methodology to determine the hardness of film-only applicable to a soft metallic film on hard substrate system. By means of the continuous stiffness measurement (CSM) technique [9] and the constant modulus assumption calculation method [2,10] in couples, the influences of the substrate and some predominated indentation plastic behaviors can be avoided. To achieve this purpose, an Au thin film on glass substrate system is chosen as an investigating object.

2. Methodology

2.1. To acquire the original analysis data by CSM
The CSM technique is utilized to acquire the original analysis data including load, \( P \), and contact stiffness, \( S \), as a function of the indentation depth, \( h \). These data will be used for calculating the hardness of thin film. Since CSM offers the direct measurement of dynamic contact stiffness at any point along the loading curve instead of at the unloading one as the Oliver-Pharr method [9], the influences of creep on the contact stiffness and indentation depth can be avoided. In addition, due to CSM is insensitive to thermal drift [5] so that the errors come from this behavior can also be ignored. To observe the rule the composite hardness and modulus varying with the indentation depths, the maximum indentation depth is designed to two times the film thickness in this study.

2.2. To extract the intrinsic hardness of thin metallic films
An important materials characteristic parameter, \( P/S^2 \), which was first proposed by Josilin and Oliver [10], is applied to extract the true hardness of film-only. By this technique, the influences of substrate and material pile-up behavior on the hardness of thin film determined by the Oliver-Pharr method can be removed. The principle of this method can be generalized as [2,10]:

From the Oliver-Pharr method, we know the definition of hardness is given as:

\[
H = \frac{P}{A} \tag{1}
\]

where \( P \) is the load and \( A \) is the contact area.

Young’s modulus is determined from:

\[
S = \frac{dP}{dh} = \frac{2\beta}{\sqrt{\pi}} E_r \sqrt{A} \tag{2}
\]

where contact stiffness \( S \) is the initial slope of the unloading curve, \( \beta \) is a constant that depends on the geometry of the indenter (\( \beta = 1.034 \) for a Berkovich indenter), and \( E_r \) is the reduced modulus given by:

\[
E_r = \left(\frac{1 - \nu^2}{E} + \frac{1 - \nu_i^2}{E_i}\right)^{-1} \tag{3}
\]

where \( \nu \) and \( \nu_i \) are the Poisson’s ratios of the samples and the indenter respectively, and \( E \) and \( E_i \) are the corresponding elastic moduli. For diamond indenter, \( E_i = 1141 \) GPa and \( \nu_i = 0.07 \) [9].

Combining the equation (1) and (2), we get:

\[
\frac{P}{S^2} = \frac{\pi}{4\beta^2 E_r^2} H \tag{4}
\]

We can see that \( P/S^2 \) is independent of the penetration depth \( h \) or the contact area, \( A \), so it does not depend on the pile-up behavior. Since \( H \) and \( E_i \) are constant for the elastically homogeneous materials,
would also keep invariable with \( h \). Thus, given the modulus of the material is known, one can determine accurately the true hardness, \( H(E) \), by the modified type of equation (4):

\[
H(E) = \frac{4 \beta^2}{\pi E_r} \left( \frac{P}{S^2} \right)
\]

(5)

It should be noted that when this technique applies to a soft film on hard substrate system, the similar modulus of the film and substrate is required. The reason why the glass was chosen as the substrate in our study is just from here. From equation (5), we know that since \( P/S^2 \) is a directly measurable experimental parameter and \( E_r \) is a known one, the determining hardness by this method is irrespective of the substrate effect. In calculation, the similar modulus of the film and substrate is usually utilized as the known input. Saha and Nix [2] has shown this constant modulus assumption method is available for a soft film on hard substrate system.

3. Experimental details

Sputtering technique was used for depositing Au film onto a glass substrate. Before deposition, the substrate’s surface was cleaned ultrasonically with alcohol and acetone. The glass substrate is a rectangular sheet of thickness 860 μm with area of 20 mm×10 mm in size.

A Cold field emission scanning electron microscopy (FE-SEM, JEOL JSM-6700F, Japan) was used to study the microstructure of the film and characterize its thickness. A 10 mm×3 mm SEM specimen was cut from the as-deposited sample and its cross section was applied for observation.

Nanoindentation experiments were carried out using a Nanoinderter XP (MTS Corporation, USA) equipped with a Berkovich diamond indenter. The system has load and displacement resolutions of less than 50nN and 0.1nm, respectively. The CSM mode was used for all experiments of both the film and bare substrate at room temperature of 23˚C. During the penetration, the indenter vibrates at a frequency of 45 Hz for an amplitude of 2 nm. A constant strain rate of 0.05s\(^{-1}\) was applied during loading with a 10s hold period at peak load. Five indentations were made in each sample and the showed results are the mean of these measures. The specimens were indented at a constant loading rate to a depth of 1μm.

The Oliver-Pharr method was used to calculate the hardness and modulus of the bare substrate and first determine both composite values of the film-substrate system. In calculations, the Poisson’s ratios of all materials were uniformly assumed as 0.3 for which has only a minor effect on the evaluated results [2].

4. Results and discussions

4.1. Thin film thickness and substrate characteristics

The section morphology of the Au thin film on the glass substrate can be observed in a cross-section SEM image, as shown in Figure 1. From the SEM image, we can see that the interface between the

![Figure 1. Cross-sectional SEM image of the Au film on glass substrate.](image)

![Figure 2. Hardness and Young’s modulus of glass substrate.](image)
film and substrate is distinct and smooth. Also, the image shows that the thickness of the Au thin film is about 504nm.

The intrinsic mechanical properties of the bare glass substrate were characterized from nanointentation data using the Oliver-Pharr method. From five indentations up to 1000 nm indentation depth, the average hardness and Young’s modulus are 7.6GPa and 90.5GPa, respectively. Figure 2 shows a plot of both mechanical properties as a function of the indentation depth, \( h \). Especially, the data during the small depths are abandoned for there are obvious indentation size effect.

4.2. Composite hardness and modulus

The composite hardness and modulus of the Au film on the glass substrate was calculated using the Oliver-Pharr method from the indentation data. Figure 3 shows an average result as a function of the indentation depth, \( h \), normalized by the film thickness, \( t \), which is from five indentations for the 504 nm film thickness at maximum depth of 1000 nm. For the sake of a better understanding, the results of the bare glass substrate were plotted together. The expected effects or phenomena mentioned above are easily found in Figure 3 (a): (1) at very small depths of \( h/t <0.03 \), the indentation size effect (ISE) is evident where the hardness increases with indentation depth increasing; (2) the constant hardness of 1.2GPa without substrate effects only occurs at a extremely small region occurs region, \( 0.03<h/t<0.05 \); (3) for \( 0.05<h/t<1 \), the hardness increases gradually as indentation depth increases, which is associated with the mixed response of the film and the substrate. When \( h/t >0.5 \), the increasing speed become rapid; (4) when \( h/t >1 \), a more remarkable increase occurs for the hard substrate is penetrated, which tends to near gradually the hardness of the glass substrate.

![Figure 3. (a) Hardness and (b) Young’s modulus of a 504 nm Au film on a glass substrate as a function of the ratio of indentation depth to the film thickness. Both mechanical properties of bare glass substrate have also been plotted.](image)

From Figure 3 (b), one can see the composite modulus of this film-substrate system is around 78GPa at an extremely small indentation region, \( h/t <0.03 \). When \( 0.03<h/t<0.5 \), an appearance of modulus increasing with the indentation depths is observed, again this is due to the substrate effect. When \( h/t >0.5 \), the curve of modulus tends to a level line. This implies that the studied Au film and glass substrate have a similar Young’s modulus. It is noted that the average value of film’s composite moduli is around 122GPa, which is higher than those usually given in literature for Au film and exceeds greatly the measured modulus of glass substrate (\( E_{\text{glass}}=90.5\)GPa). This overestimated result is just from the contribution of pile-up behavior [4-5].

4.3. Intrinsic hardness of Au thin film

As mentioned in section 2.2, for a soft film on a hard substrate system, the rule of the area-independence parameter, \( P/S^2 \), applicable to extract accurately the film intrinsic hardness is based on the constant modulus assumption. Since the Young’s modulus of Au film is very similar to the one glass has, the Au/glass system can be taken as elastically homogeneous and then the intrinsic hardness of Au film can be determined by utilizing the constant parameter, \( P/S^2 \).
From equation (2) we know that the contact stiffness, $S$, is directly proportional to the reduced modulus and the square root of the contact area. So the relation between $S$ and $h$ should be linear for an elastically homogeneous film-substrate system. Figure 4 shows a plot of the dynamic contact stiffness, $S$, versus the indentation depth, $h$, for the 504 nm Au film on the glass substrate. One can see that the contact stiffness is expected increasing linearly with the indentation depth, $h$.

Figure 5 shows the parameter, $P/S^2$, as a function of the ratio of indentation depth divided by film thickness, $h/t$, for the 504 nm Au film on the glass substrate. We observe that although the $P/S^2$ data have some degrees of scatter, it can be approximately regarded as a level line for the region of indentation depths less than the film thickness. This is also expected for the elastically homogeneous film-substrate system we studied. Thus we can use this parameter to extract the intrinsic hardness of Au thin film. Assuming the known modulus of this Au/glass system is approximately equal to 90.5GPa, which is the characterized result of the bare glass substrate, then the intrinsic hardness of Au thin film can be accurately determined utilizing equation (5).

Figure 6 shows the hardness, $H(E)$, as a function of $h/t$ for the 504 nm Au film on the glass substrate. We see that $H(E)$ is between 1.5–1.7GPa until the indentation depth nears 0.6 times the film thickness. Then $H(E)$ starts to increase with increasing indentation depth. Saha and Nix owe this increase to the hardening caused by strong gradients of plastic strain in the film between the indenter and the substrate. Eventually, the plateau values between 1.5–1.7GPa was taken as the intrinsic hardness of Au thin film.
5. Conclusion
We have chosen a 504 nm Au thin film sputter deposited on the glass substrate as a studying object to find a proper methodology determining the intrinsic hardness of the soft metallic thin film on a hard substrate using nanoindentation. We found that the way that the CSM technique and the constant modulus assumption calculation method are applied in couples is excellent. Compared with the general one that the intrinsic hardness of thin film needs to be separated from the composite hardness through various mathematic models, the present methodology is more accurate and easy. Especially, one of the most outstanding advantages this methodology has is that several significant effects that the Oliver-Pharr method concerns can be avoided, such as the substrate contribution, material creep and pile-up behaviors, etc. Therefore, the present methodology is extremely applicable for the soft metallic thin film on the hard substrate systems.

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