EFFECTS OF ADHESION ON THE MEASUREMENT OF THIN FILM MECHANICAL PROPERTIES BY NANOINDENTATION

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ABSTRACT

Experiments have been performed on soft aluminum films deposited on hard ceramic substrates to explore the influences of interfacial adhesion on mechanical property measurement by nanoindentation. The substrate materials included soda-lime silicate glass, aluminum oxynitride (ALON), and (100) sapphire. Thin films of high purity aluminum were sputtered onto each substrate to a thickness of 500 nm. Because the films were deposited simultaneously, the only major difference in the specimens was the nature of the substrate, which exerts an important influence on film adhesion through interfacial chemistry. Of the substrates examined, aluminum adheres strongly to glass and sapphire, but poorly to ALON. In addition, two different types of aluminum films on sapphire were examined—one with and the other without a 10 nm interlayer of amorphous carbon which significantly reduces film adhesion. Testing revealed important differences in the hardness of the specimens when measured by standard nanoindentation methods. Characterization of the residual hardness impressions by high resolution scanning electron microscopy showed that the hardness differences arise from an influence of interfacial debonding and film delamination on pile-up in the film. Furthermore, when the pile-up is accounted for in contact area determinations, the film hardness is actually independent of the substrate, thus indicating that the hardness differences observed in nanoindentation testing are an artifact of the testing analysis procedure. Results of the experiments are documented and discussed.

INTRODUCTION

Nanoindentation is a common technique for measuring the mechanical properties of thin films. Recent experiments have shown that nanoindentation property measurement can be influenced by the adhesion of a film to its substrate [1, 2]. A particularly important observation is that the hardness of a soft film on a hard substrate, when measured by standard nanoindentation methods, increases with increasing film adhesion [3,4]. Models describing this phenomenon are based on the premise that plasticity in a strongly-adhered film is constrained by the hard substrate, thus producing a greater hardness than would be observed if the film material were tested in bulk form or if the film were poorly adhered [1, 2]. It has also been shown, both in finite element studies [5] and in experiments [6,7], that the pile-up behavior of a soft film may be significantly influenced by a hard substrate. Recent experiments conducted by Tsui et al. [7] showed that the relative amount of pile-up of aluminum films deposited on glass substrates varies with indentation depth, reaching a maximum at an indentation depth, h_{max}, of approximately twice the film thickness, t_f. Since methods for measuring mechanical properties by nanoindentation methods do not account for the extra contact area produced by pile-up, this observation has important implications for the accuracy with which the hardness, H, and elastic modulus, E, of the film can be measured by nanoindentation procedures. Tsui et al. showed that nanoindentation methods can overestimate the hardness of a strongly adhered film by as much as 100% [6,7].

In this study, the effects of film adhesion on the measurement of hardness by nanoindentation methods are examined by comparing the behavior of aluminum films sputter-deposited on several different hard substrates. Special attention is paid to pile-up behavior and...
how it is influenced by adhesion. Implications for hardness and elastic modulus measurement by nanoindentation methods are discussed.

EXPERIMENTAL PROCEDURE

Thin films of aluminum were sputter-deposited to a thickness of 500 nm on four different hard ceramic substrates using AC magnetron sputtering. The substrates were soda-lime glass, aluminum oxynitride (ALON), (100) sapphire, and (100) sapphire with 10 nm of carbon sputtered on its surface. Because the films were deposited simultaneously, the only major difference in the specimens was the nature of the substrate, which exerts an important influence on film adhesion through interfacial chemistry. Aluminum adheres strongly to the glass and sapphire but poorly to ALON and carbon, as evidenced by the ease with which the films could be removed by scratching. The hardness and elastic modulus of the bare substrates materials measured by standard nanoindentation methods [3] and are listed in Table 1.

Nanoindentation measurements of each of the soft-film/hard-substrate systems were made using a sharp Berkovich diamond indenter. The load and displacement data obtained in the nanoindentation tests were analyzed using the method of Oliver and Pharr [3] to determine both the nanoindentation contact area ($A_{\text{nano}}$) and the nanoindentation hardness ($H_{\text{nano}}$). Results obtained from this analysis were compared with actual indentation contact areas ($A_{\text{actual}}$) measured from scanning electron micrographs (SEM), and actual hardnesses ($H_{\text{actual}}$) determined by dividing the peak indentation load, $P_{\text{max}}$, by $A_{\text{actual}}$. Care was taken in the measurement of $A_{\text{actual}}$ to include the contact area contained in the pile-up at indentation faces, since this area can contribute significantly to the load-bearing capability of the contact. The amount of indentation pile-up was characterized by the ratio $A_{\text{actual}}/A_{\text{cc}}$, where $A_{\text{cc}}$ is the corner-to-corner area of the indentation; that is, the area of the triangle defined by the positions of the indentation corners as observed in the SEM images. As discussed elsewhere [6], $A_{\text{actual}}/A_{\text{cc}}$ provides a useful measure of the amount of pile-up when there is little or no pile-up at indentation corners, as was the case for most of the indentations in this work. The value $A_{\text{actual}}/A_{\text{cc}} = 1$ implies that there is no pile-up (or sink-in), while values greater than 1 correspond to proportionally larger amounts of pile-up.

RESULTS AND DISCUSSION

The nanoindentation hardness ($H_{\text{nano}}$) for each of the soft-film/hard-substrate

<table>
<thead>
<tr>
<th>Substrate</th>
<th>$E$ (GPa)</th>
<th>$H_s$ (GPa)</th>
<th>$\alpha$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Glass</td>
<td>68</td>
<td>6.4</td>
<td>0.78</td>
</tr>
<tr>
<td>ALON</td>
<td>350</td>
<td>21</td>
<td>0.80</td>
</tr>
<tr>
<td>(100) Sapphire</td>
<td>450</td>
<td>25</td>
<td>0.80</td>
</tr>
</tbody>
</table>

Figure 1. Indentation depth dependence of the nanoindentation hardness of all specimens tested.
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Figure 2. Cross sections of indentations made at the same load in the 500nm Al/ALON specimen.

specimens is plotted as a function the ratio of the maximum indentation depth to the film thickness ($h_{\text{max}} / t_f$) in Figure 1. The hardnesses shown are averages of at least five indentations with error bars corresponding to one standard deviation. For indentation depths less than the film thickness, it is seen that the hardness of each specimen approaches ~0.8 GPa asymptotically with decreasing depth. This value is presumably the intrinsic hardness of the aluminum film. At depths larger than the film thickness, the hardness increases rapidly due to contributions from the much harder substrate. An important feature of the data is that the hardness of the Al/sapphire specimen increases more rapidly than the Al/C/sapphire, with the hardness difference becoming more pronounced as the penetration depth increases. At $h_{\text{max}} / t_f \approx 3$, hardness value of the well adhered Al/Sapphire specimen is approximately 10% higher than the delaminated Al/C/Sapphire specimen. This effect, which has been reported for similar materials in previous studies [1,2], is apparently due to the difference in interfacial adhesion. The behavior of the film deposited on sapphire is similar to the sapphire films, but the hardnesses are slightly smaller due to the intrinsically lower hardness of the ALON substrate (see Table 1). The hardness of the Al/glass specimen rises much more slowly than the other materials because glass is approximately a factor of 4 softer than any of the other substrate materials (see Table 1).

Inspection of the residual hardness impressions in the SEM revealed that there was considerable surface uplift and delamination of the film near the indentation due to interfacial debonding in the specimens with poor adhesion, i.e., Al/ALON and Al/C/sapphire. To illustrate the nature and extent of debonding, Figures 2a and 2b show cross sections prepared by focused ion beam milling of indentations in the Al/ALON specimen. The Al/ALON system proved particularly useful in this study since in limited range of indentation load some of the indentations were found to delaminate while others did not; the figure shows one indentation of each type. The micrographs show how the topographic features of the indentation can be divided into two distinct types: delamination and pile-up. For the delaminated indentation shown in Figure 2a, subsurface voids where the film has blistered upward are observed outside the region of indentation contact. The micrograph also shows that the amount of pile-up around the indentation is small when this blistering occurs. In contrast, the indentation without any delamination shown in Figure 2b exhibits a large amount of pile-up and a correspondingly larger indentation contact area. These observations are important since the studies of Lauren and Simo [5] and Tsui et al [7, 8] have shown that the contact area associated with pile-up can contribute significantly to supporting the indentation load and should therefore be included in the calculation of hardness.

To quantify the pile-up behavior, the actual area of the indentations including pile-up as measured from the SEM images are plotted in Figure 3 as $A_{\text{actual}} / A_{\text{cc}}$ vs. $h_{\text{max}} / t_f$. As discussed earlier, $A_{\text{actual}} / A_{\text{cc}}$ can be used as a measure of the amount of pile-up at an indentation. The values for $A_{\text{actual}} / A_{\text{cc}}$ for the Al/glass, Al/sapphire, and Al/C/sapphire specimens are averages over all the indentations at a particular depth. For the Al/ALON specimen, each data point corresponds to an individual indentation. The reason for plotting the Al/ALON data in this way will be discussed shortly.
Figure 3. Indentation depth dependence of $A_{\text{actual}}/A_{\text{cc}}$ of all specimens tested

Figure 3 shows that the amount of pile-up for specimens with stronger film/substrate interfacial strength, like Al/sapphire and Al/glass, changes with the indentation depth and may exhibit a maximum when the indentation depth, $h_{\text{max}}$, is approximately twice the film thickness, $t_f$. At this depth, $A_{\text{actual}}/A_{\text{cc}}$ is in the range 1.4-1.5, indicating that the indentation contact area is approximately 40-50% greater than it would be if no pile-up occurred. The behavior of the Al/C/sapphire specimen, on the other hand, is quite different. For indentation depths greater than 50% of the film thickness, the values of $A_{\text{actual}}/A_{\text{cc}}$ do not vary with penetration depth but are rather relatively independent of depth at a value of approximately 1.2. SEM examination of the hardness impressions showed that all of the Al/C/sapphire indentations blistered and delaminated while those for Al/sapphire and Al/glass did not. Collectively, these observations suggest that film adhesion plays a significant role in the amount of pile-up which can form during indentation; specifically, when a soft film debonds from the substrate, the amount pile-up is significantly reduced.

To further examine the relationship between film delamination and pile-up, a careful SEM examination of indentations in the Al/ALON specimen was undertaken. This specimen is particularly interesting because at indentation loads near 6 mN, some indentations delaminate while others do not, even when made at exactly the same indentation load. Careful examination of a large number of indentations showed that the fraction of delaminated indentations increases with indentation load, with loads greater than 250 mN producing delamination at virtually all indentations.

As shown in Figure 3, the values of $A_{\text{actual}}/A_{\text{cc}}$ for the Al/ALON specimen exhibit considerable scatter. Inspection of the individual hardness impressions showed that the small values of $A_{\text{actual}}/A_{\text{cc}}$ correspond to complete film delamination, the large values to no delamination at all, and the intermediate values to partial delamination. Furthermore, for those indentations exhibiting no delamination, the $A_{\text{actual}}/A_{\text{cc}}$ values were found to be similar to those for the Al/Glass and Al/Sapphire specimens, increasing from ~1 at small depths to a maximum of 1.4-1.5 at depths close to the film thickness. The very high data points at the upper end of the scatter in Figure 3 correspond to indentations for which the film did not debond. The indentations which fully or partially delaminate produce the lower values of $A_{\text{actual}}/A_{\text{cc}}$, scattered between 1.0 and 1.3.

SEM images of representative indentations in the Al/ALON specimen are presented in Figure 4a-c. The dashed lines indicate the corner-to-corner area of each indentation. Figure 4a illustrates an indentation with no delamination. It has a high $A_{\text{actual}}/A_{\text{cc}}$ value of 1.42, like Al/glass and Al/sapphire. An indentation which partially delaminated and for which $A_{\text{actual}}/A_{\text{cc}}$=1.28 is shown in Figure 4b. Two of the indentation edges show no signs of delamination, but the third is significantly blistered. Close inspection of the contact area outside the dashed line reveals that the two edges with no delamination have more contact area from pile-up than the delaminated edge. A completely delaminated indentation with an
area ratio of 1.08 is shown in Figure 4c. Note that the contact geometry of this indentation is almost a perfect triangle and that $A_{actual}/A_{ec}$ is the smallest of the three indentations. Collectively, these observations suggest that when delamination occurs, pile-up is significantly reduced.

The preceding experimental results show that the indentation contact area and the portion of it associated with pile-up are directly affected by film delamination. Since current methods for measuring hardness by nanoindentation methods do not account for the influences of pile-up on the contact area, it is useful to reconsider some of the film hardness data using the actual contact areas measured from the SEM micrographs for hardness computations. Results for the Al/sapphire and Al/C/sapphire specimens are shown in Figure 5. Recalling that film adhesion is good in Al/sapphire and that no film delamination or blistering were observed at any depth, whereas Al/C/sapphire showed extensive delamination at depths greater than half the film thickness, it is interesting to note that the hardnesses computed from the actual contact areas ($H_{actual}$) are in fact very similar for the two systems at all depths. This is in sharp contrast to the nanoindentation hardnesses (Figure 1), for which a measurable difference in hardness was observed at large depths. The hardness difference in the nanoindentation data is artificial and comes from the fact that the nanoindentation contact areas of the strongly adhered films are underestimated by a larger amount than the poorly adhered films due to the larger amount of pile-up. This then suggests that previous reports based on nanoindentation measurements of a dependence of the hardness on interfacial strength [1,2] may not be real but rather result from measurement errors caused by not accounting for pile-up in the nanoindentation analysis procedures. A similar error would occur in the measurement of elastic modulus, since in nanoindentation methods the modulus is computed in a manner which directly involves the contact area.

Lastly, it is instructive to use some of the data obtained in the current study to illustrate how the hardness of soft thin films on hard substrates can be estimated with a simple composite hardness model. As described in detail elsewhere [6], the model suggests that the composite hardness, $H_c$, for an indentation which resides entirely in the film or penetrates into the substrate, can be determined from a simple area fraction approximation as:

$$H_e = \left( \frac{A_f}{A_{actual}} \right) H_f + \left( \frac{A_s}{A_{actual}} \right) H_s$$  \hspace{1cm} (1)$$

where $A_f$ and $A_s$ are the portions of the projected indentation contact areas in the film and substrate, respectively, and $A_{actual}$ is the total indentation contact area such that $A_{actual} = A_f + A_s$. To implement the model to nanoindentation results, $A_{actual}$ is determined from SEM images of the contact impression by tracing the contact edges in a manner which includes the extra area generated by pile-up. $A_f$ and $A_s$ are evaluated by assuming that at a given

![Figure 4](image-url) Figure 4. SEM images of 500nm Al/ALON indentations made at the same load exhibiting different delamination behavior.
indentation load, the interface between the film and substrate sinks-in to produce the same deflection geometry that would occur if there were no film on the substrate. Such an assumption should hold reasonably well when $H_f < H_s$ and/or $h_{max} > t_f$. With this assumption, the depth along which contact is made between the indenter and the substrate, $h_s$, can be estimated from $h_s = \alpha (h_{max} - t_f)$ where the parameter $\alpha$ is the ratio of the contact depth, $h_c$, to the maximum depth, $h_{max}$, for indentation of the bare substrate. This parameter can be readily determined by standard nanoindentation measurements of the substrate. Once $h_s$ is established, $A_f$ follows by evaluating the area function of the indenter at $h_s$, and $A_f$ can be computed from $A_f = A_{actual} - A_s$.

The data in Figure 5 and 6 suggest that the asymptotic hardness of the aluminum films examined in this study is $H_f = 1.0$ GPa. Using this and the values of the $H_s$ and $\alpha$ shown in Table 1 obtained from nanoindentation measurements of the bare substrate materials, Figure 6 shows the computed composite hardnesses for both the Al/glass and Al/ALON systems and compares them to the actual hardnesses determined experimentally. There is good agreement between the data and model at all indentation depths.

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