Indentation Hardness of Film/Substrate System: Discovery of the Unconventional Overshoot and Undershoot Behaviors*

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Abstract
When a thin film/substrate system is indented, the measured indentation hardness is always regarded as a weighted average of the hardness of film and that of the substrate material. That is, one usually takes it for granted that the measured hardness should be bounded between that of the film and substrate, and with the increase of indentation depth, the measured hardness should vary monotonically from the intrinsic film hardness to the intrinsic substrate hardness. Using finite element simulations of sharp indentation on film/substrate systems, here we show an “abnormal” behavior that if the film and substrate have close hardness but different plastic behaviors, within a certain range of indentation depth, in some cases the measured hardness may “overshoot” and be higher than both the film and substrate hardness; when the film and substrate materials are reversed, then the measured hardness may “undershoot” and be lower than both the film and substrate hardness. In both cases, the indentation hardness varies non-monotonically with indentation depth. This unconventional behavior may provide some physical insights on correctly interpreting the indentation measurements on thin film/substrate systems.

Key words: Indentation, Film/Substrate Composite, Hardness, Finite Element Analysis, Substrate Effect

1. Introduction
Nanoindentation tests are widely used to determine the mechanical properties of small-sized materials from the measured force-depth curves, which is in particular attractive for extracting the properties of thin film deposited on a substrate. During the indentation on the film/substrate system, the substrate inevitably influences the deformation field, with the magnitude depending on the indentation depth relative to the film thickness. Usually, the measured mechanical property/response is a mixture of that of the film and substrate, and as the indentation depth gets larger, the contribution of the substrate becomes more prominent. In many practical applications, such a substrate effect is unfavorable when the users would like to obtain only the intrinsic properties of the film.

Two approaches are currently employed to “mitigate” the substrate effect. The most widely used one is the 10 % rule [1]: In general the intrinsic film properties may be extracted with negligible substrate effect if the indentation depth is less than about 10 % of the film thickness. While such a threshold may be acceptable for soft film on hard substrates, for
hard film on soft substrates, the 10% rule does not hold well and the substrate effect may be significant even at small penetration depth (2). Moreover, the continuous decrease of film thickness used in electronic devices makes the application of this criterion too restrictive, especially for nanometer scale films (2). Another approach is based on the indentation response of the “composite” at large indentation depth, which requires one to first figure out the substrate effect, and then subtracts its contribution from the indentation measurement. For example, an analytical closed-form expression for the elastic stiffness of the composite was obtained (3); for elastoplastic deformation various methods were proposed to evaluate the indentation hardness of the “composite”, by taking into account the different plastic zones of the two materials beneath the indenter (4-7). Extensive experiments and finite element studies have been carried out to determine the thin film’s hardness, based on the subtraction of the substrate effect from the measurement at deep indentation depth (8-11).

Almost all previous works have assumed (or take it for granted) that the measured indentation parameter, such as the indentation hardness, is a weighted average of that of the film and substrate materials. That is, the indentation hardness of the “composite” material can be expressed as an interpolation of the intrinsic hardness of film and substrate, and the weighting factor depends monotonically on the indentation depth relative to the film thickness; consequently, the measured indentation hardness should be bounded by the intrinsic film hardness and substrate hardness, and as the indentation depth increases, the composite hardness varies monotonically from the intrinsic film hardness to the intrinsic substrate hardness. Such a monotonic variation assumption is the basis of previous indentation approaches (4-7).

In what follows, we challenge the monotonic variation of indentation hardness and show that when the hardness of film and substrate are close, in some cases the resulting indentation hardness of the “composite” can vary in a non-monotonic way with respect to indentation depth. Within certain ranges of indentation depth, for some film/substrate systems the indentation hardness may be higher than both the hardness of film and substrate, termed as the “overshoot” behavior, whereas in for some other film/substrate combinations, the indentation hardness may be below both of the film and substrate hardness, denoted as the “undershoot” characteristic. This unconventional behavior, which to our knowledge has not been reported before, may provide some insights on correctly interpreting the indentation measurements on thin film/substrate systems.

2. Model description

2.1. Relevant relations

We employ a sharp rigid conical indenter with a half apex angle 70.3°, which is widely used to simulate Berkovich or Vickers indenters (2, 11). If the specimen is homogeneous and semi-infinite (i.e. bulk material), then a quadratic law exists between the indentation load \( P \) and the indentation depth \( h \), owing to the self-similarity of the entire deformation field (12):

\[
P = Ch^2
\]  

(1)

The coefficient \( C \) is also referred to as the loading curvature and it is a constant for given material elastoplastic properties (it also depends on the apex angle, which is fixed in this paper). The true hardness and the nominal hardness of the material are defined by the following expressions, respectively (13):

\[
H = P / A \quad \text{(true hardness)}
\]  

(2)

\[
H_o = P / A_0 \quad \text{(nominal hardness)}
\]  

(3)

where \( A \) and \( A_0 \) represent the true and nominal projected contact area, respectively (Fig. 1). The former takes account of local deformation of the indented material, which is known as “pile-up” or “sink-in”, and the latter is a simple geometrical relation ignoring all the surface deformation due to indentation (\( A_0 = 24.5h^2 \) in this study). The normalized contact area
$A/A_0 = H_0/H$ gives a measure of a local deformation tendency for pile-up or sink-in; such that $A/A_0 > 1$ for pile-up and $A/A_0 < 1$ for sink-in.

If the indented material is a homogeneous bulk material, then all of the indentation quantities $C, H_0, H$ and $A/A_0$ are kept constant during the indentation process; that is, they are invariant with $h$. When the specimen is a film-substrate system, $C, H_0, H$ and $A/A_0$ are functions of the indentation depth $h$, and this is the focus of the present paper.

Figure 1. Schematic view of indentation on thin film/semi-infinite substrate system with a sharp conical indenter.

Figure 2. Finite element mesh.

2.2. Computational model

The model setup of the film/substrate system is shown in Fig. 1, where the film thickness is $d$, and it is bonded perfectly to a semi-infinite substrate. The dimensionless parameter $h/d$ is now the only length variable which governs the indentation process (13). In the current analysis, we also assume that the uniaxial stress-strain ($\sigma$-$\varepsilon$) curve of both the film and substrate materials obeys the power hardening law:
\[ \sigma = E \varepsilon \text{ for } \varepsilon \leq Y/E \quad \text{and} \quad \sigma = Y(E/Y)\varepsilon^n \text{ for } \varepsilon \geq Y/E \]  

for \( E, Y \) and \( n \) represent the Young’s modulus, the yield stress, and the work-hardening exponent of the material.

The commercial code ANSYS is used for the finite element simulations\(^{(15)}\). Von Mises’s yield surface with isotropic hardening is used, and the option for finite deformation and strain is employed. The friction coefficient between the indenter and the contact surfaces is taken to be 0.15, simulating the contact condition between the diamond indenter and metal surface\(^{(14)}\) (friction has some effect on the detailed characteristic of the overshoot/undershoot behaviors, but it does not change the general trend; the effect of friction will be explored elsewhere). The overall finite element model is shown in Fig. 2. The axisymmetric mesh is comprised of 30,750 four-node elements, where the bottom of the specimen is constrained. In our analysis, the radius \( R \) and the height \( L \) of the model are chosen as \( R=100\mu m \) and \( L=200\mu m \) (which effectively simulates the semi-infinite substrate), and the film thickness is \( d=2\mu m \). For indentation on a given specimen, the selection of maximum indentation depth \( h_{\text{max}} \) is based on that proposed by Xu and Li\(^{(16)}\).

A practical problem of finite element simulation is that when the indentation depth is small, the calculated hardness often fluctuates with depth, which is mainly attributed to the small number of nodes under the contact, and also to the discrete nature of the contact conditions\(^{(17)}\). To overcome the hurdle, we employ a new approach with stratified mesh to compute the hardness, where the computational procedure is divided into several sub steps, see Appendix A for details. In addition, we also use an interpolation procedure to deduce the contact point (and the projected contact area) more precisely, illustrated in Fig. 3 (note that the number of nodes in contact is typically more than 500 in our analysis, and only a small number of nodal data located near the contact point is shown in the figure). To determine the exact contact radius \( x \) where the pressure value just reaches zero, which is located between the nodes \( l \) and \( l+1 \), we choose \( l \) nodes. Extrapolation using a \( m \)-th order polynomial function based on these \( l \) values would give us the contact radius with higher resolution, shown in the figure as \( x_{\text{contact}} \). It is also validated that the exact contact radius determined from pressure distribution and the contact radius determined from gap distribution are very close and therefore we use hereafter only the pressure distribution to determine \( x_{\text{contact}} \). Our practice shows that \( l=10 \) and \( m=3 \) works fairly accurately for deducing the contact radius. Using these improved numerical techniques, the measured indentation hardness vs. indentation depth show negligible scatter, illustrated below.

![Figure 3. Pressure and gap distributions along the contact surface nodes (only those near the contact rim are shown), and the interpolation procedure of using \( l \)-nodes to determine the exact contact point.](image-url)
3. Results and discussion

3.1. First set of film/substrate material combinations

We illustrate the hardness undershoot and overshoot behaviors in two series of material combinations. In the first series, shown in Table 1(a), four different materials for the film (A, B, C and D), and a single material for the substrate (U) are chosen. The uniaxial true stress-strain curves of the five materials are shown in Fig. 4. The four combinations of the materials are denoted as AU, BU, CU and DU, where the former character corresponds to the film material and the latter to the substrate material, respectively. The Young’s modulus \( E \) of the films and the substrate are chosen to imitate the tungsten film on the sapphire substrate \(^{(6, 18)}\). The plastic properties, the yield stress \( Y \) and hardening exponent \( n \), are determined on a trial-and-error basis: Since for most metals and alloys, \( n \) is between 0.1 and 0.5 \(^{(19)}\), we chose \( n=0.1 \) as the two extremes for the hardening exponent (\( n=0.1 \) for the substrate U and \( n=0.5 \) for the material D). The other materials A, B and C are determined such that their properties vary slightly different in the hardness. In the table, the computed hardness ratio of the film and substrate \( H_f/H_s \) is also shown. In what follows, the subscripts \( f \) and \( s \) represent the film and the substrate, respectively. The reversed combinations of the materials from Table 1(a), the single film (U) and the four different substrate materials (A, B, C and D), are shown in Table 1(b); these combinations are denoted as UA, UB, UC and UD, respectively.

<table>
<thead>
<tr>
<th>Combination</th>
<th>Film</th>
<th>Substrate</th>
<th>( H_f/H_s )</th>
</tr>
</thead>
<tbody>
<tr>
<td>AU</td>
<td>A</td>
<td>U</td>
<td>0.833</td>
</tr>
<tr>
<td>BU</td>
<td>B</td>
<td>U</td>
<td>0.919</td>
</tr>
<tr>
<td>CU</td>
<td>C</td>
<td>U</td>
<td>1.007</td>
</tr>
<tr>
<td>DU</td>
<td>D</td>
<td>U</td>
<td>1.115</td>
</tr>
</tbody>
</table>

Table 1. Material properties for the first series: (a) 4 combinations, AU, BU, CU and DU, (b) the reversed combinations from (a), UA, UB, UC and UD.

![Figure 4. Uniaxial true stress-strain curves for materials A, B, C, D and U.](image-url)
The computed results are shown in Fig. 5, where the normalized indentation hardness of the “composite” bi-layer system \( H/H_s \) is plotted against the normalized indentation depth \( h/d \). With increasing indentation depth, the indentation hardness \( H \) almost remains a constant at the film hardness \( H_f \) in a relatively shallow depth range \( (h/d < 0.15) \). When the penetration becomes deeper, say \( h/d < 0.5 \), the indentation hardness tends to converge to the substrate hardness \( H_s \). The behavior between these two regions, however, is very different in the cases AU and BU, and cases CU and DU. In the cases of AU and BU (and similarly for UA and UB), the hardness monotonically changes with increasing depth, and finally converges to \( H_s \) in Fig. 5(a) (and to \( H_f \) in Fig. 5(b)). These are conventional behaviors.

However, the behavior becomes unconventional for cases CU and DU (and likewise for UC and UD). In cases CU and DU, after the initial shallow indentation region where \( H=H_f \), the hardness overshoots both values of \( H_f \) and \( H_s \) with increasing depth (\( H/H_s = 1.15 \) at \( h/d = 0.34 \) in DU, and \( H/H_s = 1.07 \) at \( h/d = 0.47 \) in CU). Instead of overshoot, undershoot is observed in UC and UD (\( H/H_s = 0.96 \) at \( h/d = 0.34 \) in UD, and \( H/H_s = 0.94 \) at \( h/d = 0.52 \) in UC). Although these peak values of overshoot and undershoot are not large, these “abnormal” phenomena are to our knowledge reported for the first time. For reference, in Fig. 5, the relative hardness for the film-bulk material is shown both for the materials A and D, indicated as AA and DD, respectively, which are indeed constants for homogeneous materials.

![Figure 5](image_url)
There is a possibility that the local deformation around the indenter is responsible for overshoot or undershoot. In Fig. 6, the ratio between the true and nominal projected contact area is plotted against the normalized indentation depth $h/d$, in cases of AU, BU, CU and DU (a), and in cases of UA, UB, UC and UD (b). The true projected contact area $A$ is calculated from the actual contact radius by the interpolation procedure described in 2.2. As expected from the conventional behavior of hardness, $A/A_0$ starts at its own value for the film and then gradually converge to that of the substrate. Although the results show indentation depth dependence, the values of $A/A_0$ keep remaining in the “sink-in” zone ($A/A_0 < 1$); in other words, with the increased penetration during the indentation process for these material sets, the local deformation mode around the contact rim does not change from sink-in to pile-up. Therefore, it may be concluded that the overshoot or undershoot behavior of the hardness is not attributed to the local deformation mode around the indenter.

Figure 6. Normalized projected contact area $A/A_0$ as a function of the normalized indentation depth $h/d$: (a) AU, BU, CU and DU; (b) UA, UB, UC and UD, where $A_0$ means the nominal contact area (see Figure 1).
3.2. Second set of film/substrate material combinations

The second series of materials are shown in Table 2, with four different material candidates of film (E, F, G and H) and one single material of substrate (V). The Young’s modulus of the films and the substrate are chosen to mimic the copper film on silicon substrate \(^{(20)}\). The four combinations of the bi-layer system are EV, FV, GV and HV, given in Table 2(a). In addition when the film and substrate materials are reversed, new composite systems VE, VF, VG and VH are formed in Table 2(b). Again, in these combinations, the former character corresponds to the film material and the latter to the substrate material (same as that described in 3.1). The uniaxial stress strain curves of the five materials are shown in Fig. 7.

<table>
<thead>
<tr>
<th>Combination</th>
<th>Film Material</th>
<th>$E$ (GPa)</th>
<th>$v$</th>
<th>$Y$ (GPa)</th>
<th>$n$</th>
<th>Substrate Material</th>
<th>$E$ (GPa)</th>
<th>$v$</th>
<th>$Y$ (GPa)</th>
<th>$n$</th>
</tr>
</thead>
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<tr>
<td>EV</td>
<td>E</td>
<td>3.0</td>
<td>0.50</td>
<td>0.844</td>
<td></td>
<td>V</td>
<td>168</td>
<td>0.278</td>
<td>7.0</td>
<td>0.10</td>
</tr>
<tr>
<td>FV</td>
<td>F</td>
<td>4.0</td>
<td>0.45</td>
<td>0.900</td>
<td></td>
<td>V</td>
<td>168</td>
<td>0.278</td>
<td>7.0</td>
<td>0.10</td>
</tr>
<tr>
<td>GV</td>
<td>G</td>
<td>4.0</td>
<td>0.50</td>
<td>0.948</td>
<td></td>
<td>V</td>
<td>168</td>
<td>0.278</td>
<td>7.0</td>
<td>0.10</td>
</tr>
<tr>
<td>HV</td>
<td>H</td>
<td>5.0</td>
<td>0.50</td>
<td>1.034</td>
<td></td>
<td>V</td>
<td>168</td>
<td>0.278</td>
<td>7.0</td>
<td>0.10</td>
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</table>

<table>
<thead>
<tr>
<th>Combination</th>
<th>Film Material</th>
<th>$E$ (GPa)</th>
<th>$v$</th>
<th>$Y$ (GPa)</th>
<th>$n$</th>
<th>Substrate Material</th>
<th>$E$ (GPa)</th>
<th>$v$</th>
<th>$Y$ (GPa)</th>
<th>$n$</th>
</tr>
</thead>
<tbody>
<tr>
<td>VE</td>
<td>E</td>
<td>3.0</td>
<td>0.50</td>
<td>1.185</td>
<td></td>
<td>E</td>
<td>3.0</td>
<td>0.50</td>
<td>1.185</td>
<td></td>
</tr>
<tr>
<td>VF</td>
<td>F</td>
<td>4.0</td>
<td>0.45</td>
<td>1.111</td>
<td></td>
<td>F</td>
<td>4.0</td>
<td>0.45</td>
<td>1.111</td>
<td></td>
</tr>
<tr>
<td>VG</td>
<td>G</td>
<td>4.0</td>
<td>0.50</td>
<td>1.055</td>
<td></td>
<td>G</td>
<td>4.0</td>
<td>0.50</td>
<td>1.055</td>
<td></td>
</tr>
<tr>
<td>VH</td>
<td>H</td>
<td>5.0</td>
<td>0.50</td>
<td>0.967</td>
<td></td>
<td>H</td>
<td>5.0</td>
<td>0.50</td>
<td>0.967</td>
<td></td>
</tr>
</tbody>
</table>

Figure 7. Uniaxial true stress-strain curves for materials E, F, G, H and V.
The computed results for the depth-dependent hardness are shown in Fig. 8. In the cases of EV and FV, and VE and VF, the indentation hardness equals to that of the film hardness ($H_f$) at small depth range ($h/d < 0.15$). It then monotonically changes with increasing depth, and finally converges to $H_s$, following the conventional behavior. Whereas, in the cases GV and HV, the indentation hardness is observed to overshoot both values of $H_f$ and $H_s$ with increasing depth ($H/H_s = 1.06$ at $h/d = 0.45$ in HV, and $H/H_s = 0.99$ at $h/d = 0.50$ in GV). Instead of overshoot, undershoot phenomenon is observed for VG and VH ($H/H_s = 0.98$ at $h/d = 0.52$ in VH, and $H/H_s = 0.96$ at $h/d = 0.52$ in VG). These are once again unconventional behaviors.

Figure 8. Normalized hardness $H/H_s$ as a function of the normalized indentation depth $h/d$: (a) EV, FV, GV and HV; (b) VE, VF, VG and VH, where $H_s$ is the intrinsic hardness of the substrate material, respectively.
The behaviors in these second series of material combinations are similar to those of the first series, and thus the overshoot and undershoot are common phenomena that could be observed when the hardness of the film and substrate are not too far away from each other. For the second set of material properties, the ratio between true and nominal projected contact area is plotted against indentation depth in Fig. 9, the cases of EV, FV, GV and HV (a), and the cases of VE, VF, VG and VH (b). Again, the evolution of the contact area is “normal”.

Figure 9. Normalized projected contact area $A/A_0$ as a function of the normalized indentation depth $h/d$: (a) EV, FV, GV and HV; (b) VE, VF, VG and VH, where $A_0$ means the nominal contact area (see Figure 1).
3.3 Parametric study

The computed results in the above sections showed that the overshoot behavior and the undershoot behavior appear for some material sets. In this section, the condition which causes those behaviors will be shown more clearly with parametric study.

3.3.1. The yield stress of the film  
For the first parametric analyses, the yield stress $Y$ of the film is changed as a parameter, and the Young's modulus $E$ and the hardening exponent $n$ of the film, and the elastoplastic properties of the substrate are fixed. The material sets are shown in Table 3 and the computed results of the indentation hardness are shown in Fig. 10. The yield stress has a strong relation with the indentation hardness, therefore the discussion is based on the indentation hardness instead of the yield stress. In the case of that the film hardness is close to the substrate hardness, the overshoot behavior seems to occur. In addition, Fig. 10 shows a new behavior. When the film hardness is larger than the substrate hardness, the indentation hardness usually decreases monotonically from the intrinsic film hardness to the intrinsic substrate hardness. But, in Fig. 10, when the film hardness is slightly larger than the substrate hardness, the indentation hardness goes over the film hardness once and then decreases into the substrate hardness.

Table 3. Material properties for the first parametric analyses: The parameter is the yield stress of the film.

<table>
<thead>
<tr>
<th>Combination</th>
<th>Film</th>
<th>Substrate</th>
</tr>
</thead>
<tbody>
<tr>
<td>Material</td>
<td>$E$</td>
<td>$v$</td>
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<tr>
<td></td>
<td>GPa</td>
<td></td>
</tr>
<tr>
<td>IW</td>
<td>I</td>
<td>168</td>
</tr>
<tr>
<td>JW</td>
<td>J</td>
<td>168</td>
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<tr>
<td>KW</td>
<td>K</td>
<td>168</td>
</tr>
<tr>
<td>LW</td>
<td>L</td>
<td>168</td>
</tr>
</tbody>
</table>

Figure 10. Normalized hardness $H/H_s$ as a function of the normalized indentation depth $h/d$: The parameter is the yield stress of the film.
3.3.2. The hardening exponent of the film

Only the hardening exponent $n$ of the film is changed as a parameter, and the other properties for the film/substrate system are fixed. In order to keep the same hardness ratio between the film and the substrate, the film yield stresses $Y$ are chosen. The material sets are shown in Table 4 and the computed results are shown in Fig. 11. Although the hardness relations between the film and the substrate are the same, only the indentation hardness for the case $n=0.5$ has the overshoot behavior.

Table 4. Material properties for the first parametric analyses:
The parameter is the hardening exponent of the film.

<table>
<thead>
<tr>
<th>Combination</th>
<th>Film Material</th>
<th>$E$ (GPa)</th>
<th>$v$</th>
<th>$Y$ (GPa)</th>
<th>$n$</th>
<th>Substrate Material</th>
<th>$E$ (GPa)</th>
<th>$v$</th>
<th>$Y$ (GPa)</th>
<th>$n$</th>
<th>$H_f/H_s$</th>
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<tr>
<td>MW</td>
<td>M</td>
<td>168</td>
<td>0.30</td>
<td>0.432</td>
<td>0.25</td>
<td>NW</td>
<td>0.121</td>
<td>0.50</td>
<td>0.875</td>
<td></td>
<td></td>
</tr>
<tr>
<td>NW</td>
<td>N</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>W</td>
<td>168</td>
<td>0.30</td>
<td>1.00</td>
<td>0.00</td>
<td>0.875</td>
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</table>

Figure 11. Normalized hardness $H_f/H_s$ as a function of the normalized indentation depth $h/d$:
The parameter is the hardening exponent of the film.

3.3.3. The hardening exponent of the substrate

The next parameter is the hardening exponent of the substrate, and the other properties for the film/substrate system are fixed. Like the subsection 3.3.2., to keep the same hardness relation, the yield stresses of the substrates are determined. The material sets are shown in Table 5. According to the computed results shown in Fig. 12, the hardening exponent of the substrate also affects the overshoot behavior.

Same as the above section, by switching the film and the substrate material which shows the overshoot behavior, the undershoot behavior appears. When the Young's modulus and the friction coefficient were changed as parameters, they did not affect the overshoot behavior very much. As results, the conditions of the overshoot behavior are the following two;
1) The film hardness should be close to the substrate hardness.
2) The hardening exponent of the film should be significantly larger than the substrate exponent.
Table 5. Material properties for the first parametric analyses:
The parameter is the hardening exponent of the substrate.

<table>
<thead>
<tr>
<th>Combination</th>
<th>Film Material</th>
<th>Substrate Material</th>
<th>$H_f/H_s$</th>
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<tr>
<td></td>
<td>$E$ (GPa)</td>
<td>$v$</td>
<td>$Y$ (GPa)</td>
</tr>
<tr>
<td>OX</td>
<td>O</td>
<td>168</td>
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<tr>
<td>OY</td>
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<tr>
<td>OZ</td>
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</tr>
</tbody>
</table>

Figure 12. Normalized hardness $H/H_s$ as a function of the normalized indentation depth $h/d$:
The parameter is the hardening exponent of the substrate.

3.4. Discussion

It is envisioned that with the mismatched plastic behaviors (yielding and hardening) of the film and substrate materials, as the plastic flow of the two materials impedes each other during indentation, the hardness of the “composite” may sometimes become higher than both the film and substrate (causing “overshoot”), and at other times become lower than both the film and substrate (causing “undershoot”). This may occur to a large range of film/substrate systems, with a necessary (but not sufficient) condition that they have similar hardness but different elastoplastic properties. Overshoot tends to be favored when the hardening exponent of the film is significantly larger than that of the substrate. If overshoot occurs in one particular film/substrate system, then if one switches the film and substrate materials while keep other conditions/parameters the same, then undershoot is bound to appear, and vice versa.

The mechanism of the overshoot behavior is investigated by comparing the stress distributions. Figures 13 (a), (b), (c) and (d) show the axial stress distributions and the radial stress distributions; (a) for the materials KW ($n_f >> n_s$ and $H_f \approx H_s$) in Table 3 with the overshoot behavior, (b) for the materials LW ($n_f >> n_s$ and $H_f < H_s$) in Table 3 with the non-overshoot behavior, (c) for the materials MW ($n_f > n_s$ and $H_f \approx H_s$) in Table 4 with the non-overshoot behavior and (d) for the materials OZ ($n_f = n_s$ and $H_f \approx H_s$) in Table 5 with the non-overshoot behavior. All hardness ratios, $H_f/H_s$, except the case (b) are almost same and are close to 1.0. All indenters in Fig. 13 are located in the same depth, $h/d=0.40$, where the overshoot behavior occurs for the case KW. The gray lines indicate the borders between the film and the substrate. Regardless of occurrence of overshoot behavior, the axial stress distributions are similar in all material combinations. On the other hand, for all radial stress distributions, compressive stresses are observed right under the indenters, but only the overshoot case (a) shows a complicated stress distribution near the border under the indenter rim.
This difference of the radial stress distribution is also observed in the indentation analyses for an elastoplastic film/rigid substrate system. When all of the bottom nodes of the film are fixed, see Fig. 14 (a), the stress distribution is similar to the non-overshoot case. On the other hand, when the bottom nodes except the center vicinity are fixed, see Fig. 14 (b), the film shows a bending mode and the radial stress distribution is similar to the overshoot case. These may imply that the overshoot behavior is caused by the bending mode of the film. In other words, for the overshoot behavior, the indentation hardness increases temporarily by adding the bending component into the ordinary hardness. In addition, the reasons why the bending mode occurs are the following: the film with a larger hardening exponent deforms as sink-in form, and the substrate with a smaller hardening exponent shows pile-up deformation. This deformation combination evokes a large
curvature in the film under the indenter rim; therefore the overshoot behavior may occur. According to Figs (13), even if the hardening exponent of film is significantly larger than the hardening exponent of substrate, the overshoot behavior is not observed if the hardness ratio, $H_f/H_s$, is not close to 1. This may be because the bending mode effect is not large enough and disappears with the difference of hardness. Note that the overshoot behavior needs both conditions; 1) a small difference between film hardness and substrate hardness and 2) a significantly larger hardening exponent of film than substrate. It is further remarked that the overshoot and undershoot characteristics are not just limited to the true hardness $H$ as focused in this study. The nominal hardness $H_0$ also exhibits similar overshoot and undershoot behaviors, so does the loading curvature $C$. Meanwhile, the elastic stiffness (contact stiffness) of the “composite” still behaves in the conventional way and does not exhibit overshoot nor undershoot. Unfortunately, the undershoot mechanism which relates with overshoot mechanism could not be made clear in this section, but more detailed analyses will be needed to resolve the undershoot mechanism.

4. Concluding remarks

We have shown using finite element simulation that the “unconventional” overshoot or undershoot phenomenon can be observed during the indentation hardness measurement of some film/substrate systems. In particular, if two materials with similar hardness but different plastic properties are chosen for the film and the substrate, sometimes (especially when the work hardening exponent of the film is significantly larger than that of the substrate) one may observe the overshoot phenomenon where the measured indentation hardness of the “composite” within a range of indentation depth is higher than that of both film and substrate; in some other cases, the undershoot behavior occurs where the measured composite hardness within a range of indentation depth is lower than that of both film and substrate. If overshoot occurs in one system, then by switching the film and substrate materials (while keep other conditions the same), undershoot is guaranteed to occur, and vice versa. Therefore, we can conclude that the composite indentation hardness of the film/substrate systems cannot always be expressed by a monotonic function that changes from the intrinsic film hardness to the intrinsic substrate hardness. This is a new type of substrate effect owing to the strong interaction of the different plastic flow behaviors in the film and substrate.

Hardness test is a material test with a long history: If overshoot and undershoot are real phenomena, why have there been no previous reports published on these phenomena? We can think of the following possible explanations. First of all, the amount of overshoot or undershoot is relatively small although they are distinctive in hardness-depth relationship. The percentage of overshoot is at most 15 % in the first series of materials, and 6 % in the second series of materials studied in this paper. Therefore, these overshoot/undershoot phenomena may be overlooked even if they existed during real hardness tests, and for people who were not aware of the importance of these phenomena, they may be regarded as simply fluctuation during experiment. Moreover, for very thin films the indentation size effect may start – this effect is ignored in our analysis, however in practice the size effect may make the evaluation of the hardness data more complicated at small indentation depth, making it hard to observe the overshoot/undershoot behaviors. Secondly, in practical industrial applications, the mechanical properties of the film and substrate materials are often very different. Perhaps, that makes it hard to discover the unconventional overshoot or undershoot behaviors – it is remarked that regardless whether the film/substrate systems with overshoot or undershoot behaviors may receive wide potential applications or not, they still stand for a significant theoretical value and challenge that remains to be explored in future. Future work also involves manufacturing the “special” unconventional film/substrate systems to validate the overshoot or undershoot phenomena experimentally via indentation.
or nanoindentation hardness test.

Appendix

In conventional finite element simulations of the indentation process, when the indentation depth is small, the number of contact nodes in contact with the indenter is few, which, in addition to the discrete nature of nodes, may cause fluctuation of the computed contact area and severely affect the accuracy of hardness. In this paper, we propose a novel numerical technique “stratified mesh” so as to circumvent this difficulty. With this technique, it is possible to calculate the projected contact area and hardness with very high resolution (ensuring at least 500 nodes in contact with the indenter at any time). The indentation process is divided into several substeps described below, and only one mesh is needed without re-meshing, thus not sacrificing computational efficiency. This technique takes full advantage of the fact that the parameter $h/d$, the normalized indentation depth, is the only length variable which governs whole the conical indentation process on the semi-infinite film/substrate system.

The stratified mesh structure is illustrated in Fig. A1. The computational system is comprised of seven material layers (each with the same radius $R$ but different thickness). In what follows, material layer 1 starts from the surface to thickness 0.32 $\mu$m, material layer 2 starts from thickness 0.32 $\mu$m to 0.8 $\mu$m, material layer 3 starts from thickness 0.8 $\mu$m to 2 $\mu$m, material layer 4 starts from thickness 2 $\mu$m to 5 $\mu$m, material layer 5 starts from thickness 5 $\mu$m to 12.5 $\mu$m, material layer 6 starts from thickness 12.5 $\mu$m to 31.25 $\mu$m, and material layer 7 starts from thickness 31.25 $\mu$m to 200 $\mu$m. For each layer, either the material properties of the film (f) or the substrate (s) is assigned according to the chart in (a) and (b), creating film/substrate “specimens” 1-6 (each combination of material layer properties would create a film/substrate specimen to be used in a particular analysis substep; for instance, in substep 5 the specimen has its first 2 material layers having film properties and the bottom 5 layers having substrate properties). Although these specimens have different film thickness, note that with scaling of the length unit, one can easily convert any film/substrate specimen (in any substep) back to the real film/substrate system with real $d=2\ \mu$m. If all layers are assigned with the same properties, then a homogeneous bulk specimen is created.

Six substeps are employed in indentation simulation, in each substep indenting a different “specimen” with the maximum indentation depth $h_{\text{max}}=1\ \mu$m. In the results obtained from each analysis step, the initial 40 % of the computed information ($=1/k$, $k=2.5$) is discarded and only the latter 60 % of data is used, so as to ensure a large number of nodes are in contact with the indenter. We use the 4th substep to illustrate the calculation procedure: In this substep the range of indentation covers from $h=0.4$ to 1 $\mu$m, which corresponds to $h/d=0.2$ to 0.5. With the scaling of length unit, switching the material assignment to that for the different substeps is equivalent to changing the real indentation depth. For example, the indentation of $h=1\ \mu$m (real $h/d=0.5$) in the 4th substep is equivalent to that of real $h/d=0.2$ ($=0.5/k$) in the 3rd substep. Therefore, all the quantities with the length dimension in the 3rd step must be multiplied by 0.4 = 1/k, and that with force dimension must be multiplied by $0.4^2 = (1/k)^2$; the latter quadratic factor comes from the dimensional requirement when the expression for the stress is kept the same in the different steps. Thus, applying the similar relation to other steps, we can eventually cover a relatively wide range of real indentation depth $h/d=0.0128$ to 3.125 with almost the same accuracy kept in every substep. Small gaps in $P$-$h/d$ curves indicate the connection of substeps. Data allocation for obtaining the relation between the indentation force $P$ and the normalized indentation depth $h/d$ is schematically shown in Figure A1, and the stable results of the contact area and hardness are given in Section 3, which validates the present approach.
Figure A1. Stratified material layers, where f and s mean the layers with film and the substrate properties, respectively: (a) Six analysis substeps and material assignment for the layers 4 to 7; (b) Those for layers 1 to 3; (c) Data allocation for a complete run, which superimposes the results from the six analysis substeps shown in $P$ vs. the real (converted) $h/d$ for a given film/substrate system.
References

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