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A Comparison of Nanoindentation Pile-up in Bulk Materials and Thin Films

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Abstract

During nanoindentation testing there are many issues that need to be considered if high quality data is to be obtained when testing both bulk and thin film materials. For soft materials, one of the main issues in determining mechanical properties based on the Oliver and Pharr method is the accuracy of the determined contact area due to the pile-up around the indenter leading to a significant increase in the contact area. During nanoindentation tests for both thin films and bulk materials, the deformation mechanisms, and therefore the governing dislocation nucleation and propagation events, are complex and hence the volume of the pile-up is not always proportional to the indentation load and its shape can vary. Therefore accurate measurement of the Young's modulus and hardness requires the determination of the contact area using another technique such as atomic force microscopy (AFM) or scanning electron microscopy (SEM) images.

In this study, AFM images obtained using the indenter tip after the main indentation cycle was completed were analysed to measure the pile-up heights and widths obtained in bulk materials (copper, gold and aluminium) and the results were compared to those from their respective thin films under similar indentation conditions. It was observed that the amount of pile-up that appeared in the thin films was considerably higher than in the bulk materials. Thin films with low hardness values deposited on harder substrates show a different plastic response under the indenter. During the indentation tests, the harder substrate does not deform to the same extent as the softer deposited coating and consequently it has an extreme effect on the degree of pile-up formation for the thin film.

1. Introduction

Nanoindentation is a widely adopted method to measure the elastic, plastic, and time-dependent mechanical properties, including the hardness (H) and Young's modulus (E) of thin films and small volumes, of bulk materials. The nanoindentation method gained

popularity with the development of machines that were capable of recording very small loads and displacements to a high level of precision and accuracy [1]. Analytical models were also developed to estimate contact modulus and hardness using load-displacement data [2-8]. The most commonly employed method to determine the modulus and hardness of the indented material is the Oliver and Pharr method which was first proposed in 1992 [2, 4, 8-15]. This has become the standard procedure to extract the elastic modulus and hardness of the specimen material from load-displacement measurements [9, 10, 16, 17].

There are however potential issues that need to be considered when testing both bulk materials and thin films. For example, when the thickness of the film is less than $1 \mu\text{m}$, there are some errors, especially at greater depths, in the obtained mechanical properties due to the influence of the substrate on the results, consequently affecting the load-displacement curve [16, 18]. Furthermore, issues such as pile-up are significant when dealing with soft coatings on hard substrates and the obtained results are usually overestimated [19, 20]. This is because the Oliver and Pharr method cannot account for the effect of pile-up on the measured data at greater depths; the method is most accurate when material deforms elastically and sink-ins rather than piling-up. The appearance of pile-up and its quantity as well as sink-in behaviour in various materials depends on the work-hardening characteristics of the material undergoing the indentation test [9].

Based on the Oliver and Pharr method, the hardness is inversely proportional to the contact area and the Young's modulus is inversely proportional to the square root of the contact area [2]. Therefore, the appearance of pile-up around the edges of the contact area results in an underestimation of the contact area causing an overestimation of the Young's modulus and hardness values [21]. Therefore, when pile up occurs, the values of hardness and reduced modulus determined by the Oliver and Pharr method are too high since this method is based on the contact area in the plane of the original surface, rather than the true contact area.

During nanoindentation tests for both thin films and bulk materials, the deformation mechanisms, and therefore the governing dislocation nucleation and propagation events, are complex and not fully understood [22, 23]. In general, Young's modulus, initial yield stress and work-hardening exponents are known to be major influences in controlling the piling-up or sinking-in behaviour of materials in response to indentation forces [8]. According to Oliver and Pharr [9], the ratio of the effective modulus to the yield stress as well as the work-hardening behaviour of materials can influence the pile-up formation. Materials with a

greater ratio of the effective modulus to the yield stress with less capacity for work-hardening show larger pile-up. Cheng and Cheng [24-26] also used finite element analysis to show that pile-up depends on the work-hardening behaviour of elastic-plastic materials by analysing various types of materials with different work-hardening behaviour. Based on their method, there is also a relationship, independent of the work-hardening behaviour of materials, between the work of indentation and the effective modulus over hardness (E_{eff}/H) [9]. Soft, easily hardened materials sink-in whereas harder, work-hardened materials pile-up [6]. The physical explanation of this is that dislocations are generated below the contact and are propagated by the high shear stresses which occur at about 45° to the loading axis. If the stress is high enough these dislocations will propagate on the slip planes closest to the maximum shear and will move downwards into the materials. If there is no barrier to their motion (i.e. in soft materials) the dislocations continue into the material and sink-in is observed. However, when the material is work-hardened the dislocation mobility is reduced and the dislocations are confined in proximity to the surface. In this case cross slip can occur on slip planes which allows dislocations to move to the surface causing pile-up. The extent of pile-up will thus depend on how far the dislocations move into the material. Therefore, the volume of the pile-ups is not always proportional to the indentation load and true measurement of the Young's modulus and hardness values requires the calculation of the contact area from the nanoindentation load-displacement curves as well as AFM or SEM images. For that reason, AFM images were obtained using the indenter probe after each indentation in this work to accurately measure the true contact area and apply the pile-up correction to the obtained data for comparison. In some cases pre-indentation AFM images were obtained to measure the roughness of the sample surface before any indentation tests took place.

2. Experimental

2.1. Materials

Due to the differences in the mechanical properties of bulk materials and thin films and the different responses that they exhibit during nanoindentation testing, both forms of materials were investigated in this work. Three different face centred cubic metals, gold (Au), copper (Cu) and aluminium (Al), were chosen to investigate the appearance of pile-up for thin films and bulk samples as well as the effect of pile-up on the nanoindentation test results. Prior to indentation testing, AFM images obtained from $10\ \mu\text{m} \times 10\ \mu\text{m}$ areas were used to measure

the surface roughness of the samples. It was confirmed that the Cu, Au and Al thin films have average surface roughnesses of 0.18, 0.22 and 0.16 *nm* respectively. According to Bobji et al. [27] when the penetration depth is more than 3 times the root mean-square (RMS) roughness of the surface, the roughness effect on the hardness and modulus data can be considered to be negligible, which is applicable in all cases here. The AFM images also confirmed that the roughnesses of the bulk samples tested in this work are less than 0.25 *nm*.

2.1.1. Copper

0.5 *mm* thick <100> Si wafers were thermally oxidised to produce a 1 μm thick silicon oxide layer. The oxidised silicon was coated with a 25 *nm* TiW inter-layer diffusion barrier layer, then sputtered to produce a 20 *nm* Cu seed layer followed by electrodeposition of an 800 *nm* thick blanket Cu metallisation with an average grain size of 0.5 μm . The grain size was measured using the electron backscatter diffraction (EBSD) technique. An 8 *nm* sputtered TiW layer was applied to passivate the material with regards to oxidation. A bulk Cu sample with 1 μm grain size was also studied to provide a comparison. This sample, which was 99.9% pure but had a small quantity of oxygen impurity (0.03%), was rolled to an 8% reduction and subsequently vacuum annealed for 1 hour at 300 °C with the aim of reducing the defect density whilst promoting recrystallisation and grain growth.

2.1.2. Gold

Pure gold thin films with a 1 μm thickness having 1 μm average grain size were vapour deposited onto 0.5 *mm* thick <100> Si substrates. The Si substrates were oxidised before film deposition to produce a 2.3 μm oxide layer. Following this, a 25 *nm* thick TiW layer was deposited onto the oxidised silicon substrate to improve the adhesion between the substrate and the Au films as well as to create a diffusion barrier layer. Gold films were then deposited onto the TiW layer. Finally, similar to the Cu thin films, the Au thin films were coated with an 8 *nm* sputtered TiW layer in order to be directly comparable to the Cu samples. The results were compared to that of pure single crystal bulk Au.

2.1.3. Aluminium

In addition to the Cu and Au thin films, nanoindentation tests were also performed on Al thin films. Two Al thin film samples were sputter coated onto 0.8 *mm* thick glass substrates with different Al thicknesses of 375 *nm* and 1400 *nm* with 0.4 μm and 1 μm average grain sizes

respectively. The obtained results were compared to that of a pure bulk Al (100) single crystal sample.

2.2. Nanoindentation testing

In the current study, all depth sensing nanoindentation tests were performed using a Hysitron Triboindenter fitted with a Berkovich indenter having a tip radius of 150 nm. The nanoindentation tests were carried out under both the open loop mode and displacement control mode using a single cycle indentation (load-hold-unload) test method. During each indentation cycle a 4 s hold was applied at the maximum load to minimise the effect of creep on the unloading curve and its resulting effect on the Young's modulus and hardness. Prior to applying each set of indentation tests, samples were kept for 24 h in the nanoindentation chamber to stabilise the temperature of the sample with the surroundings. Furthermore, similar to any high accuracy measurement technique, the nanoindentation instrument was calibrated before applying the indentation tests using the standard aluminium, tungsten and fused silica samples for tip area function and machine compliance calibrations. This was carried out to ensure that the obtained data were not affected by errors due to the indenter tip shape or errors on the machine compliance.

3. Results and Discussion

3.1. Effect of Pile-up on the Mechanical Properties of Cu and Au

Initially, the effect of pile-up on the hardness and modulus values obtained from the nanoindentation tests on bulk Cu was investigated. The first set of indentation tests were performed under high loads (1 to 10 mN) to observe the magnified effects of pile-ups on the hardness and Young's modulus values of bulk Cu obtained from nanoindentation tests. In addition to this, the tests were also carried out using high loads to allow for a comparison of the pile-up shapes of harder bulk Cu samples with softer Al bulk samples. Figure 1 (a) and (b) illustrate two AFM images ($10\ \mu\text{m} \times 10\ \mu\text{m}$ areas) obtained from a polished bulk Cu sample after nanoindentation tests in open loop mode under 10 and 9 mN loads respectively. Additionally, the cross-sectional curves corresponding to the AFM images shown in (a) and (b) obtained from the three different sides of the indentation edges are shown in Figure 1 (c) and (d) respectively.

The cross-sectional curves assist in measuring the amount of pile-up, as well as its height and width around the indentation edges. The combination of AFM images and cross-sectional

curves confirms both that the pile-ups are not symmetrical around the indentation edges and that the height and width of the pile-ups differ from each other at the three different sides of the indentations. They also confirm that during the formation of pile-ups, the material protrudes upwards in proximity to the indentation edges, building narrow but high pile-ups.

The hardness and Young's modulus values obtained before and after pile-up correction for high load indentation tests on bulk Cu under open loop mode are shown in Figure 2. It was observed that as the indentation size is increased, the pile-ups show a greater influence on the obtained hardness and modulus values. When quantifying the effect of pile-up from lower loads to higher loads it was found that the effect on the obtained data increased from 5 to 15% for the Young's modulus and 10 to 35% for the hardness values. The hardness of bulk Cu is quite high compared to other bulk results in this study, probably as a result of polishing damage.

To determine the relationship between the pile-up appearance and contact depth as well as the effect of pile-up on the obtained data for lower loads, the height and width of the pile-ups were measured using the AFM images. The measured height and width of the pile-ups obtained from 100 nanoindentation tests applied on bulk Cu for open loop mode under low load ranges are shown in Figure 3 (a) and (b). As can be seen from Figure 3, both the pile-up heights and widths increase as the contact depth increases. Moreover, the width of the pile-up can represent the plastic radius zone around the indentation contact radius. For the indentations with contact depths of less than 50 nm, the pile-ups were extremely small and not even observable. The effect of pile-up in the calculated hardness and modulus values at very small indentation depths can therefore be negligible. However, at small indentation depths the effect of tip end shape is more pronounced, as these small indentations do not involve the self-similar part of the indenter tip.

When pile-up heights and widths obtained from bulk Cu were compared to the thin films under the same indentation conditions, it was observed that the amount of pile-up that appeared in the thin films was considerably higher than in the bulk Cu. Thin films such as Cu, Au or Al with low hardness values deposited on substrates such as glass ($H=5$ to 8 GPa) and silicon ($H=12$ GPa), which are harder than the deposited materials by nearly one order of magnitude, show a different plastic deformation under the impression of the hard indenter [18]. During the indentation tests, the harder substrate does not deform to the same extent as the softer deposited coating and consequently it has an extreme effect on the degree of pile-

up formation on the thin film. For comparison, the height and width of pile-ups obtained from Cu thin film deposited on an oxidised silicon substrate are shown in Figure 4.

It should be noted that although the effect of pile-up in the hardness and modulus values is important, it is also important to understand when this effect starts and whether the pile-up area carries the applied load or not. Sometimes the effect is minor and the actual area in contact with indenter tip that carries the load is not related to the measured pile-up height directly. For example, the pile-up influence in the obtained data for the Cu thin film at contact depths less than 50 nm did not have any effect on the measured contact area, but for the depths greater than 85 nm, the effect is significant.

Further investigation was carried out on the 1 μm Au thin film deposited on oxidised silicon and bulk Au for comparison using the same indentation conditions as the Cu samples. Au was chosen due to the identical crystal structure with Cu which can facilitate identifying and also confirming the effect of pile-up on the nanoindentation test results using the Oliver and Pharr method under same indentation conditions. Figure 5 shows two different AFM images (3D views and top-down views) obtained from typical indentation tests carried out on a Au thin film. The appearance of pile-up for Au films was clear from the AFM images under the range of loads tested, even at low loads. The obtained results from the indentation tests are therefore affected by pile-up. The experiment was carried out on the bulk Au single crystal under the same test conditions for pile-up correction. Au single crystals were chosen for comparison due to the simple structure of the material and the reduction in the grain boundary effect on the experimental results. The average hardness and modulus values obtained from single crystal Au are 1.05 ± 0.2 and 84.91 ± 1 GPa respectively. The corresponding AFM images confirmed that the appearance of pile-up for Au even at low loads is large, however as the pile-ups are more symmetrical, pile-up correction is more practical. Using the Gwyddion software [28] to calculate the true contact area from the obtained AFM images, the hardness and modulus values were reduced to 0.65 ± 0.1 GPa and 80 ± 1 GPa which are more in agreement with the results previously reported [29-31]. In comparison to the bulk Au, the thin films are more likely to have even larger pile-ups. The average hardness and contact modulus results determined from 10 single cycle nanoindentation tests (with standard deviations shown as the error bars) on Au thin films after pile-up correction are shown in Figure 6.

The average hardness value obtained from single indentation tests is 1.08 ± 0.2 GPa after pile-up correction and the average modulus value is 71.45 ± 5 GPa, which is lower than that of bulk Au but in the range of values given by the elastic anisotropy of gold. As the Au thin films were deposited on a Si/SiO₂ substrate, the effect of the substrate can influence the obtained modulus values. Thin Au films are harder than pure bulk Au but can be affected by the substrate [27], the indentation size effect [32], strain gradient plasticity phenomena [33], work-hardening and different microstructures (grain sizes) [34]. Thin films with smaller grain sizes and consequently lower dislocation movements compared to that of bulk materials are harder. This is primarily due to the small grain size of the coating compared to the bulk single crystal.

In general, the values vary in a similar manner to that of thin Cu films and bulk Cu under the similar indentation conditions. As with the Cu thin films, the hardness graphs shown in Figure 6 can be divided into two regions after pile-up correction; the contact depths of less than 40 nm and the contact depths greater than 40 nm. It can be seen that due to the indentation size effect, the hardness values are higher at the contact depths lower than 40 nm. However, in the second region they remain constant at around 1 GPa.

3.2. Effect of Pile-up on the Mechanical Properties of Aluminium

To further investigate the effect of pile-up in the mechanical properties obtained from nanoindentation tests, work was carried out on an Al single crystal (100) sample. High purity bulk Al was chosen as it has a well-known modulus value of 70 GPa and a low hardness value. These properties make Al one of the ideal materials used for load frame compliance calibration of nanoindentation machines. Moreover, Al is nearly elastically isotropic and its modulus value is independent of indentation depth [35]. Therefore, Al can be used to identify any changes in the modulus value due to the effect of pile-up.

To investigate the difference in pile-up appearance as well as its effect on the hardness and modulus values of thin films with different thicknesses, Al films were deposited on a hard glass substrate. A glass substrate was chosen as the Young's modulus of the glass and Al are relatively similar, thereby ensuring that any unusual behaviour in the obtained modulus data cannot be related to a substrate effect. This consequently means that any unexpected behaviour can be attributed to differences in the plastic flow characteristics only. Also, despite the modulus values of these two components being approximately the same, the great difference in the hardness values (approximately 0.5 to 1 GPa for Al thin films compared

with 7 *GPa* for glass [36]) makes them an ideal example system of a soft coating on a hard substrate.

3.2.1 Bulk Aluminium

A series of indentations were applied on the Al bulk sample under open loop mode for loads ranging from 4 to 0.1 *mN* and also displacement control for contact depths of less than 130 *nm*. The obtained hardness and modulus values for the Al bulk sample are shown in Figure 7. These tests were applied to investigate the hardness and Young's modulus of Al under different loads and contact depths and compare these results to those of Al thin films. The average hardness and Young's modulus values obtained from both tests for bulk Al are 0.45 *GPa* and 70.2 *GPa* respectively. The slight increases in the obtained data at shallow depths can be due to the thin layer of Al oxide near the surface for modulus data and the indentation size effect with regards to the hardness values.

The displacement control data were obtained for the contact depths less than 130 *nm*, and it was confirmed through the obtained AFM images that appreciable pile-up did not occur. Al has a low hardness value and consequently the aforementioned contact depths can be produced at very low loads. Nonetheless, for the data obtained under open loop mode, it was expected that some pile-up would be observed around the indenter imprint edges when the applied load is high. However, the hardness and Young's modulus values obtained from open loop mode are almost constant even at high loads. When the AFM images were reviewed, it was observed that there were some evident, broad pile-ups around the indentation impressions at high loads. However, the shapes of pile-ups were different from those found on the bulk Cu and Au samples and the effect on the hardness and Young's modulus values was extremely small. This confirms that the dislocation movements under the indentation tests are different from each other. The pile-up shapes (heights and widths) are shown in Figure 8 which illustrates several AFM images and the associated cross-sectional curves obtained from the single crystal Al sample indented at high loads.

The pile-up effects for the obtained Al data differ from those associated with Cu and Au. It should be noted that the volume of the pile-ups is related to the indentation volume for all materials, however the height and width characteristics of the pile-ups cause the differences in the effect of pile-up on the obtained hardness and modulus values. Since pile-up formation and its effect on the accuracy of the contact area measurement has been shown to have

considerable influence on the obtained data, further work was carried out on the Al thin films to detect the presence of any substrate-induced enhancement of pile-up.

3.2.2 Aluminium Thin Film

Two different high purity Al thin films with 375 nm and 1400 nm thickness, deposited on a glass substrate, were investigated under open loop mode to detect the substrate and pile-up effects on the hardness and Young's modulus values obtained using nanoindentation tests. These two films were chosen to study the effect of the substrate on the appearance of pile-up in two different situations. The first situation was when the indenter penetration is greater than the film thickness (using the 375 nm thick film) and the second is when the indentation remains in the thin film but is affected by the harder substrate (using the 1400 nm thick film). A series of indentations were conducted under open loop mode at very high loads for both thin films using the same indentation conditions. The hardness and Young's modulus values of the 1400 nm Al film obtained from both the Oliver and Pharr method and the actual contact area determined using the AFM images are shown in Figure 9 as a function of penetration depth.

The obtained hardness values from the Oliver and Pharr method (shown with open circles in Figure 9) at small depths are around 0.7 GPa and increase to 1.8 GPa as the contact depth increases. The Young's modulus calculated using the Oliver and Pharr method also increases from 65 GPa at small contact depths to 112 GPa at higher contact depths. However, the results calculated using the actual contact areas obtained from AFM images (shown in Figure 9 with filled circles) are much lower than those from the Oliver and Pharr method. These results show that the effects of pile-up on the Young's modulus and hardness values can alter the results from 5 to 30% and 10 to 45% respectively, depending on the contact depth. The average values of hardness and Young's modulus measured from the actual contact area were almost constant at 0.81 ± 0.07 GPa and 69 ± 2 GPa respectively. These results are in agreement with nanoindentation measurements of thin Al films on a glass substrate reported by Tsui and Pharr [18].

When the data was compared to that of the 375 nm Al film shown in Figure 10, it was observed that the hardness values are higher than those of the 1400 nm film even at very small indentation depths and that they increase as the contact depth increases. This is in good agreement with the hardness and Young's modulus measurements of Al films deposited on glass substrate stated by Saha and Nix [36]. This increase is due to the influence of the hard

glass substrate beneath the Al film. However, the increase in the hardness values for the contact depths of less than 300 *nm* is almost steady and is mainly due to the pile-up effects on the actual contact area measurement.

At a contact depth of approximately 300 *nm*, which is roughly a maximum indentation depth of 325 *nm*, the hardness increases rapidly while the pile-ups get slightly smaller and the effect on the actual contact area reduces. When approaching the film thickness, the glass substrate begins to have an even bigger effect on the obtained data and suddenly starts to control the dislocation movements.

Similar behaviour is also observable in the Young's modulus results. For the contact depths lower than 300 *nm*, the obtained Young's modulus data increases with increasing contact depth and is similar to that of the 1400 *nm* Al film. However, the obtained values start to plateau for about 40 *nm* after this and eventually decrease when the substrate influence dominates over the pile-up effect. This is due to the residual contact impression as well as a transition in the characteristic profile from indentations in the soft Al film with straight-sides to indentations in the hard bulk glass with a cusp-like shape at the bottom of the unloading curve [18].

Although both the thin film and glass have similar Young's moduli, the glass has a higher hardness value, requiring a higher contact pressure and consequently a greater fraction of the total displacement is elastic. Therefore, during the indentation, the elastic displacement recovery of the Al is much smaller than that of the glass. During the indentation process when the maximum contact depth is less than the film thickness, the loading and unloading of the indentation tip is entirely reliant on the Al film and its displacement recovery. However, as there is a hard glass substrate, the quantity of pile-up appearing around the indenter is higher than when examining bulk Al. The obtained results are therefore highly dependent on the degree of pile-up appearance. When the indenter is closer to the substrate but still remaining in the film, the hardness and modulus values depend on the thin film's elastic recovery with influences from the hard substrate and pile-up. However, in the scenario in which the indentation penetrates deeper than film thickness, both the film and the substrate affect the loading and unloading parts of the load-displacement curve. As the Oliver and Pharr method relies on the unloading part of the load-displacement curve, it is important to understand what physically occurs during the unloading of the indentation. After a small amount of unloading, the indenter comes out of the contact with the Al thin film around the

edges of the indenter as Al has less elastic recovery than glass. Consequently, the subsequent elastic recovery is controlled by the harder glass substrate that quickly dominates the unloading curve. Because the contact stiffness is obtained from the unloading part of the load-displacement curve and the obtained mechanical properties are strongly dependent on the measured stiffness, any changes in the curve can have an effect on the obtained data when using the Oliver and Pharr method.

Comparison of the pile-up heights and widths obtained from Al, Cu and Au in this study has confirmed the results of Cheng and Cheng [8] in that softer materials such as bulk Al with low H/E ratios (0.006) show less pile-up effect than harder materials such as Cu with higher H/E ratios (0.012) where cross slip during indentation is more pronounced (Figure 11). In general, thin films have a smaller grain size than comparable bulk materials and dislocation mobility below the indenter is more limited. This leads to more cross slip close to the indenter and higher pile-up surrounding the indenter. The presence of a harder substrate greatly exacerbates this affect when the coating is thin and dislocations emitted from under the indenter can interact with it. Practically, this means that pile-up will dramatically affect the hardness and modulus of thin metal films when the indenter penetration is a significant fraction of the coating thickness. For submicron thin films the Oliver and Pharr method will overestimate these properties unless a direct measurement of contact area is obtained. Calibration method may be used in some circumstances [37] but if the properties of the material change, the shape and extent of pile-up will change too and the approach will no longer be valid.

4. Conclusion

The initial purpose of this study was to perform a comparison between the pile-up appearance for various materials in both bulk and thin film forms. The effect of pile-up on the accuracy of the hardness and Young's modulus values obtained from the nanoindentation tests was identified for both types. When the appearance of pile-up for bulk Al was compared with Cu and Au, it was found that the dislocation movements during the indentation tests differ for Al, Cu and Au bulk samples due to their different microstructure, and the effect of pile-up on the mechanical properties of bulk Al is much smaller in comparison to that of bulk Cu and Au under similar indentation conditions. The shape of the pile-up played a significant role on its effect, as narrow and high pile-ups generated next to the indenter tip were obtained for bulk

Cu with a greater effect on the data while wide but shorter pile-ups seen for bulk Al had less effect on the nanoindentation test results.

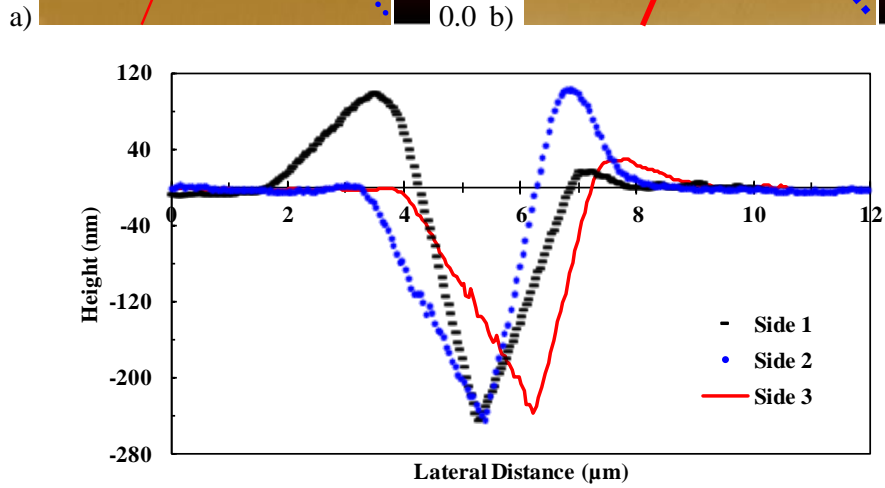
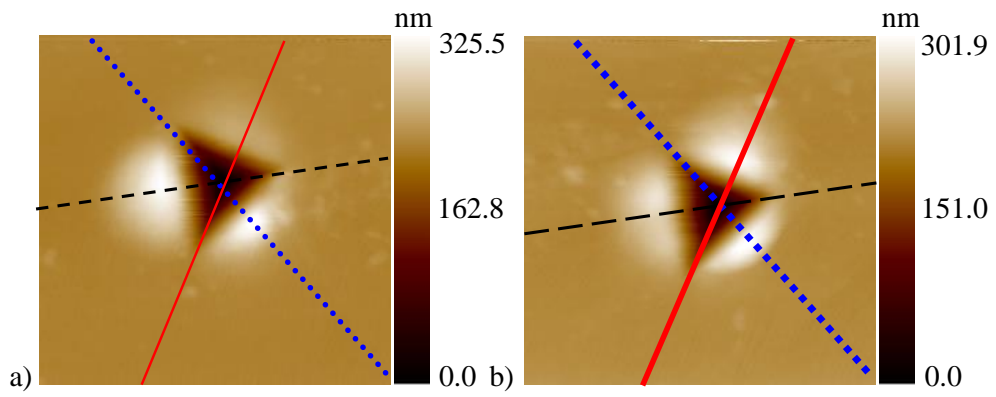
When the pile-up heights and widths of the Cu, Au and Al thin films were compared to that of the respective bulk materials, the bulk materials tend to form less pile-up than thin films. This confirmed that thin films show different plastic deformation under the indentation tests due to the substrate effect as well as the hardening effect of their finer microstructures. When dealing with soft coatings on hard substrates, the effect of substrate on the formed pile-up as well as its effect on the load-displacement curve can be crucial. Therefore it is important to recognise the contact depth at which the substrate effect dominates over the pile-up effect on the data. For this reason nanoindentation test results were illustrated for Al thin films deposited on glass substrates with different film thickness.

It was also found that the pile-up appears asymmetrically in most of the indentation tests due to local microstructural conditions and consequently, the pile-up correction methods using constant factors that are suggested in literature are not practical unless the AFM or SEM images after indentation tests are available and the true contact area can be measured.

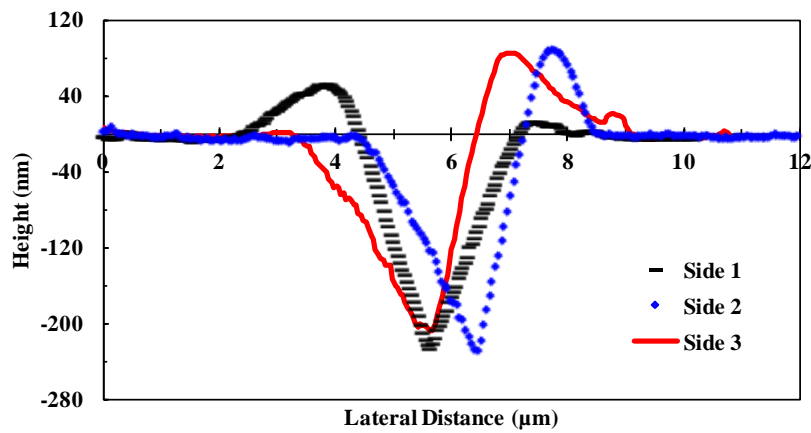
References

- [1] G. Constantinides, E.C.C.M. Silva, G.S. Blackman, K.J. Van Vliet, *Nanotechnology* 18/30 (2007).
- [2] W.C. Oliver, G.M. Pharr, *Journal of Materials Research* 7/6 (1992) 1564.
- [3] N. Axén, G.A. Botton, H.Q. Lou, R.E. Somekh, I.M. Hutchings, *Surface and Coatings Technology* 81/23 (1996) 262.
- [4] M.F. Doerner, W.D. Nix, *Journal of Materials Research* 1 (1986) 601.
- [5] S.V. Hainsworth, H.W. Chandler, T.F. Page, *Journal of Materials Research* 11/8 (1996) 1987.
- [6] Y.-T. Cheng, C.-M. Cheng, *Surface and Coatings Technology* 133-134/0 (2000) 417.
- [7] S.J. Bull, *Zeitschrift Fur Metallkunde* 93/9 (2002) 870.
- [8] Y.T. Cheng, C.M. Cheng, *Materials Science & Engineering R-Reports* 44/4-5 (2004) 91.
- [9] W.C. Oliver, G.M. Pharr, *Journal of Materials Research* 19/1 (2004) 3.
- [10] A.C. Fischer-Cripps, *Nanoindentation*, Springer, 2011.
- [11] M. Dao, N. Chollacoop, K.J. Van Vliet, T.A. Venkatesh, S. Suresh, *Acta Materialia* 49/19 (2001) 3899.
- [12] A.E. Giannakopoulos, P.L. Larsson, R. Vestergaard, *International Journal of Solids and Structures* 31/19 (1994) 2679.
- [13] F.P. Ganneau, G. Constantinides, F.J. Ulm, *International Journal of Solids and Structures* 43/6 (2006) 1727.
- [14] A.E. Giannakopoulos, S. Suresh, *Scripta Materialia* 40/10 (1999) 1191.
- [15] D. Tabor, *The Hardness of Metals*, Oxford University Press, USA, 2000.
- [16] H. Li, J.J. Vlassak, *Materials Research Society*, 506 Keystone Drive, Warrendale, PA 15086, United States, 2009, p. 1114.
- [17] C.A. Schuh, *Materials Today* 9/5 (2006) 32.
- [18] T.Y. Tsui, W.C. Oliver, G.M. Pharr, in: W.W. Gerberich, H.J. Gao, J.E. Sundgren, S.P. Baker (Eds.), *Thin Films: Stresses and Mechanical Properties Vi*, vol. 436, 1997.
- [19] N.X. Randall, *Philosophical Magazine a-Physics of Condensed Matter Structure Defects and Mechanical Properties* 82/10 (2002) 1883.
- [20] T.Y. Tsui, G.M. Pharr, *Journal of Materials Research* 14/1 (1999) 292.

- [21] A. Bolshakov, G.M. Pharr, *Journal of Materials Research* 13/4 (1998) 1049.
- [22] S. Suresh, T.G. Nieh, B.W. Choi, *Scripta Materialia* 41/9 (1999) 951.
- [23] S. Soare, S.J. Bull, A.G. O'Neil, N. Wright, A. Horsfall, J.M.M. dos Santos, *Surface & Coatings Technology* 177 (2004) 497.
- [24] Y.T. Cheng, C.M. Cheng, *J. Appl. Phys.* 84/3 (1998) 1284.
- [25] Y.T. Cheng, C.M. Cheng, *International Journal of Solids and Structures* 36/8 (1999) 1231.
- [26] Y.T. Cheng, C.M. Cheng, *Applied Physics Letters* 73/5 (1998) 614.
- [27] M.S. Bobji, S.K. Biswas, *Journal of Materials Research* 14/6 (1999) 2259.
- [28] D. Necas, P. Klapetek, *Central European Journal of Physics* 10/1 (2012) 181.
- [29] L. Siller, N. Peltekis, S. Krishnamurthy, Y. Chao, S.J. Bull, M.R.C. Hunt, *Applied Physics Letters* 86/22 (2005).
- [30] G.B. Jahromi, *Investigation of the Near Surface Mechanical Properties of Gold-titanium Thin Films*, Oklahoma State University, 2008.
- [31] A.A. Volinsky, N.R. Moody, W.W. Gerberich, *Journal of Materials Research* 19/9 (2004) 2650.
- [32] S.J. Bull, *Zeitschrift Fur Metallkunde* 94/7 (2003) 787.
- [33] Y. Cao, S. Allameh, D. Nankivil, S. Sethiaraj, T. Otit, W. Soboyejo, *Materials Science and Engineering a-Structural Materials Properties Microstructure and Processing* 427/1-2 (2006) 232.
- [34] S.-Y. Chang, T.-K. Chang, *J. Appl. Phys.* 101/3 (2007).
- [35] B. Bhushan, X.D. Li, *International Materials Reviews* 48/3 (2003) 125.
- [36] R. Saha, W.D. Nix, *Acta Materialia* 50/1 (2002) 23.
- [37] A. Oila, S.J. Bull, *Zeitschrift Fur Metallkunde* 94/7 (2003) 793.



c)



d)

Figure 1. (a) and (b) AFM images as well as respective (c) and (d) cross-sectional curves of the drawn lines in the images obtained from bulk Cu after indentation tests under 10 and 9 *mN* loads for open loop mode.

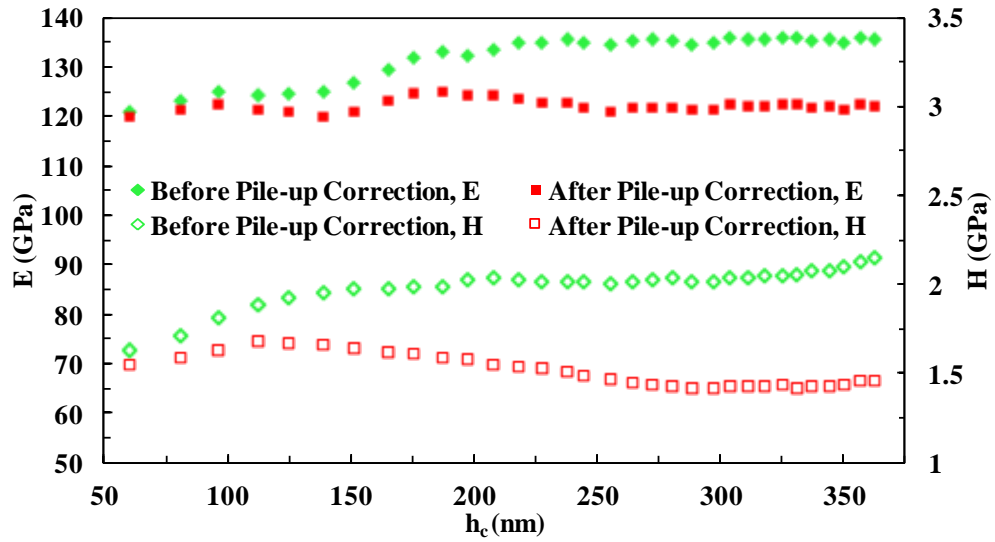


Figure 2. Hardness and Young's modulus values obtained under high load indentation tests for bulk Cu using open loop mode showing data before and after pile-up correction.

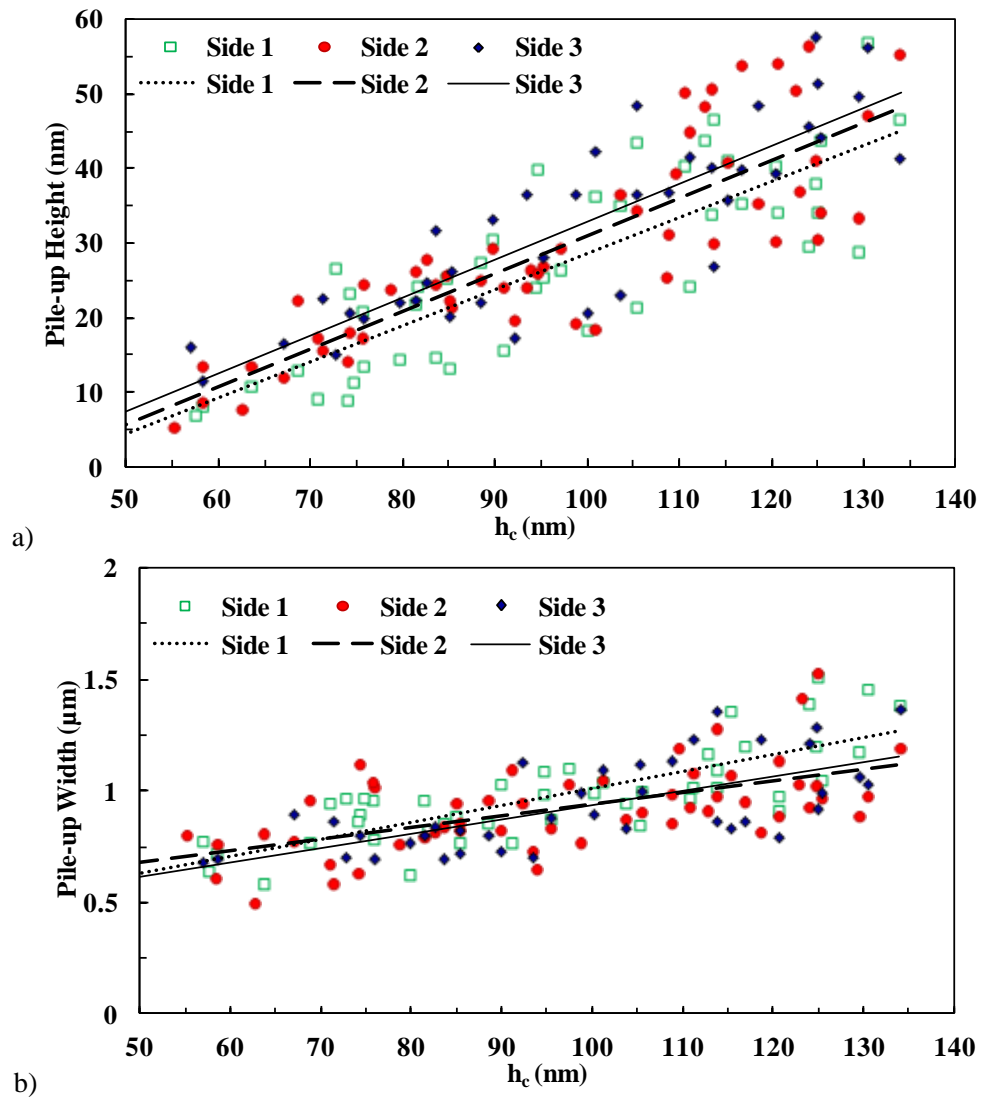


Figure 3. (a) height and (b) width of the pile-ups obtained from a single cycle nanoindentation test under open loop mode from bulk Cu.

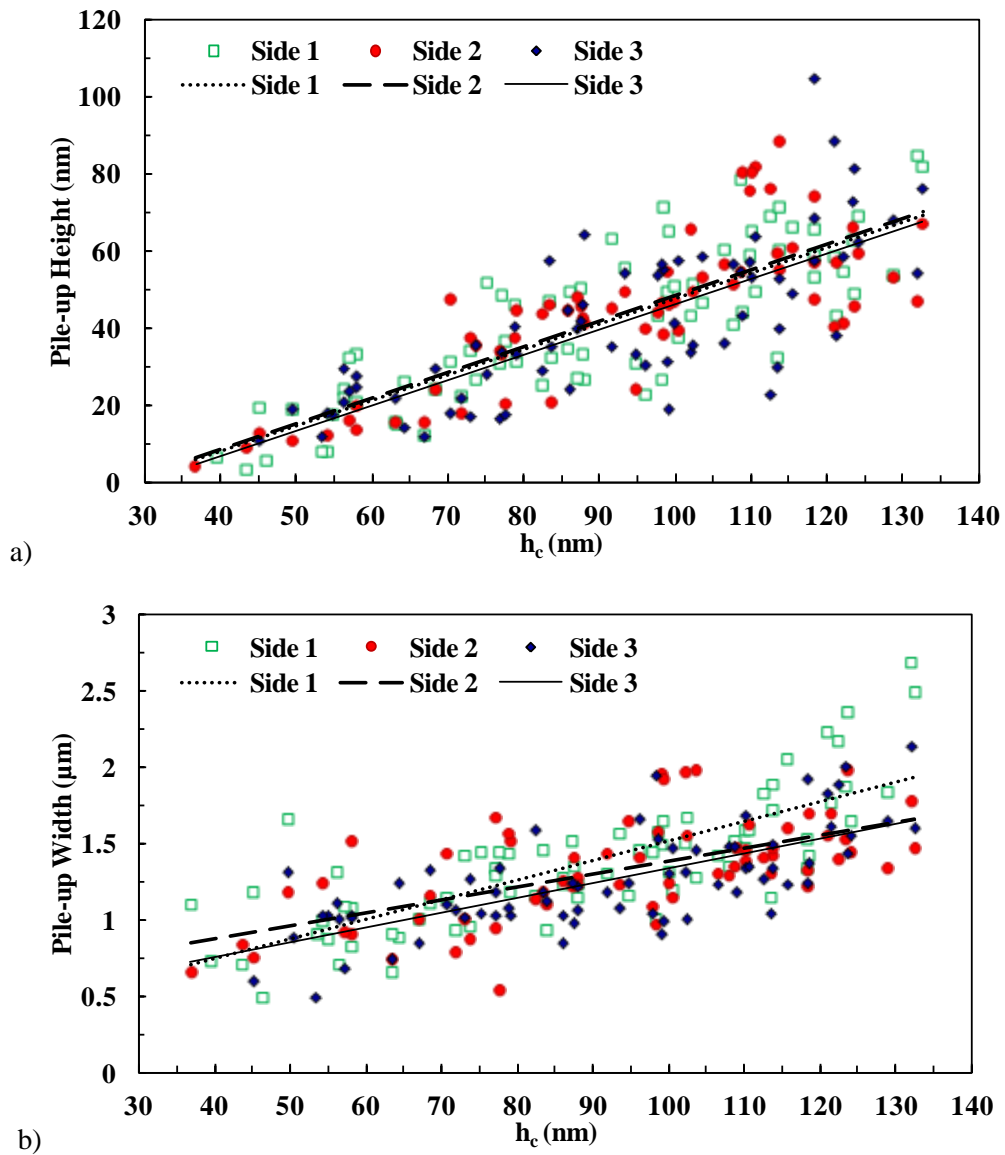


Figure 4. (a) height and (b) width of the pile-ups obtained from single cycle nanoindentation test under open loop mode on Cu thin film.

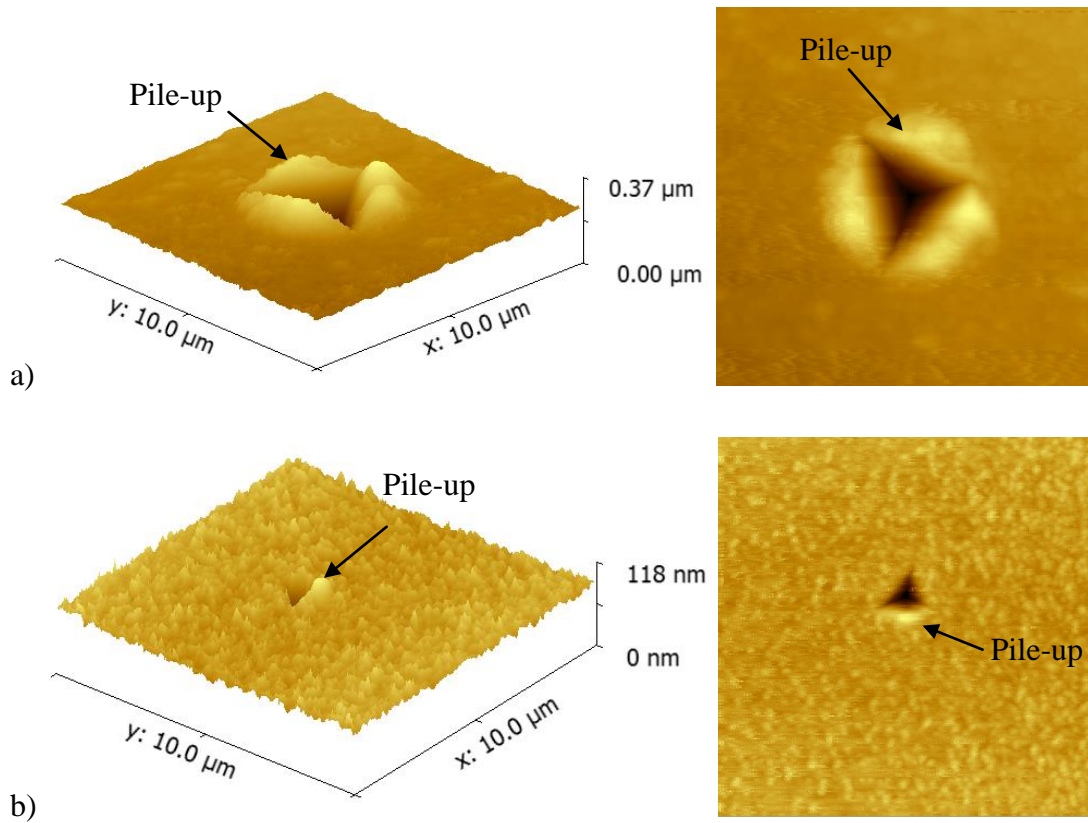


Figure 5. AFM images for typical indentations on Au thin film, three dimensional views and top-down views of (a) high load (10 mN), (b) lower load (1.5 mN).

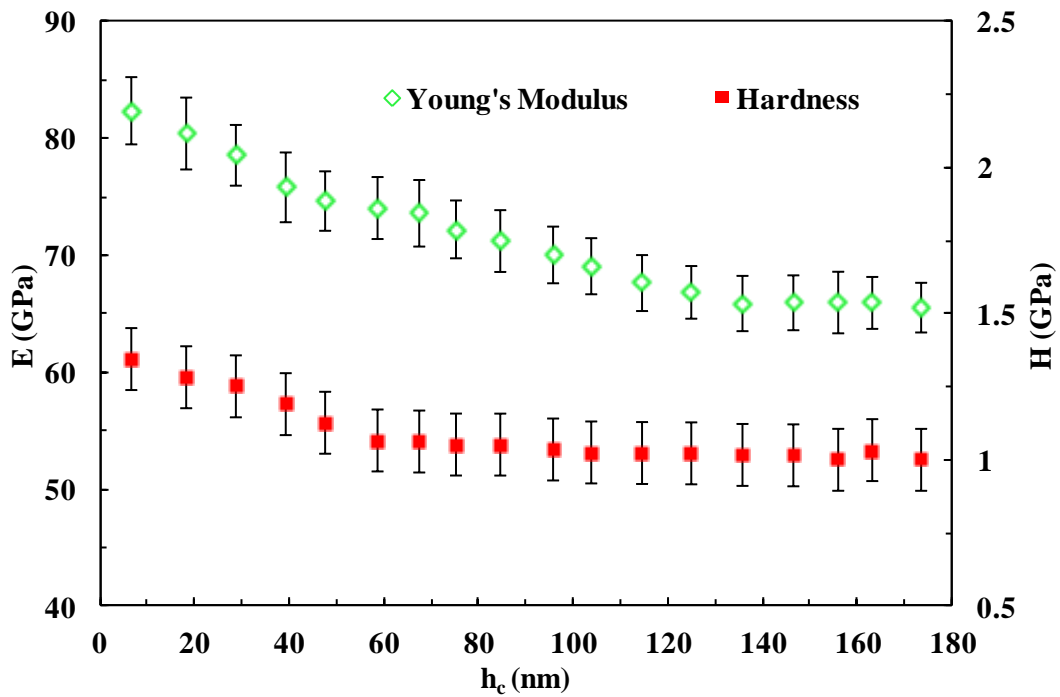


Figure 6. Obtained Young's modulus (left) and hardness (right) results for Au thin film after pile-up correction with standard deviations.

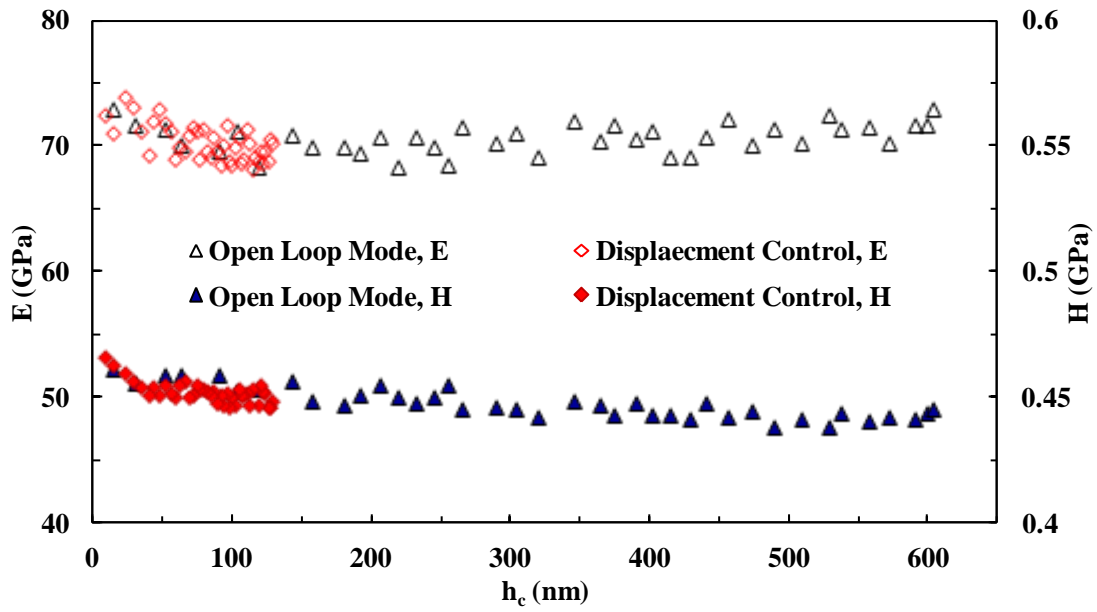
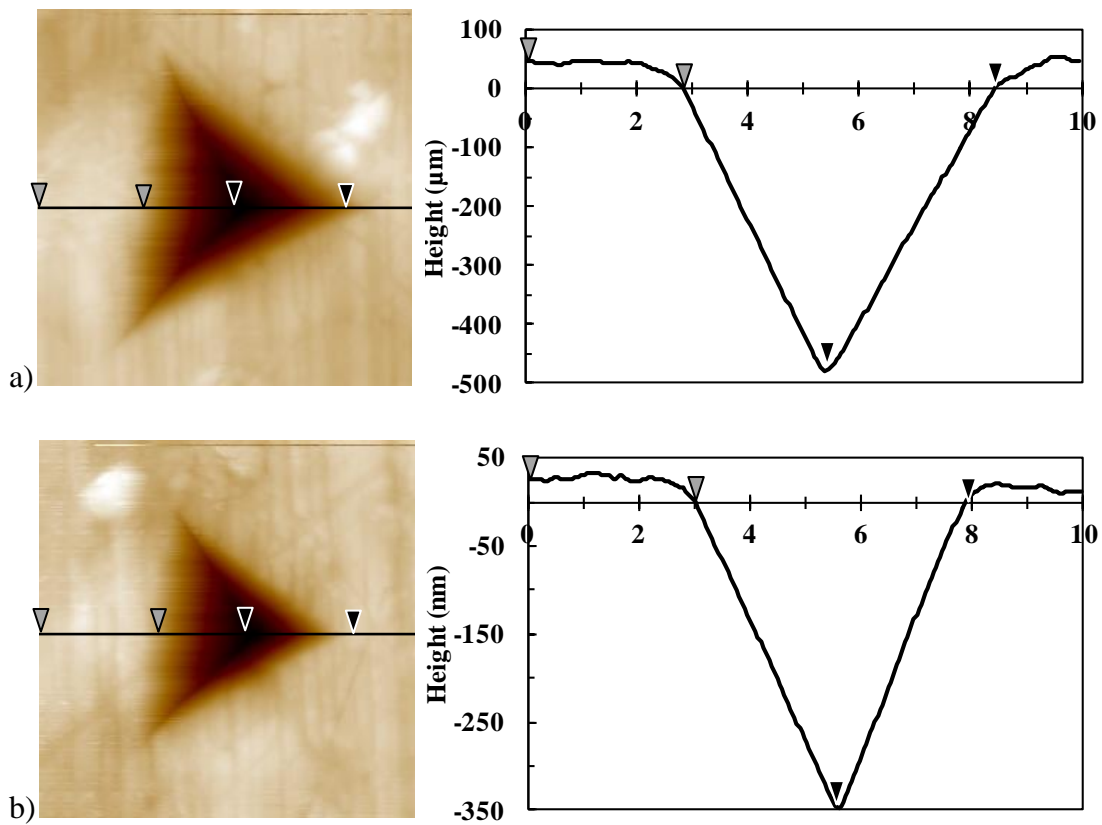


Figure 7. Young's modulus (left) and hardness (right) values of bulk Al obtained under open loop mode and displacement control.



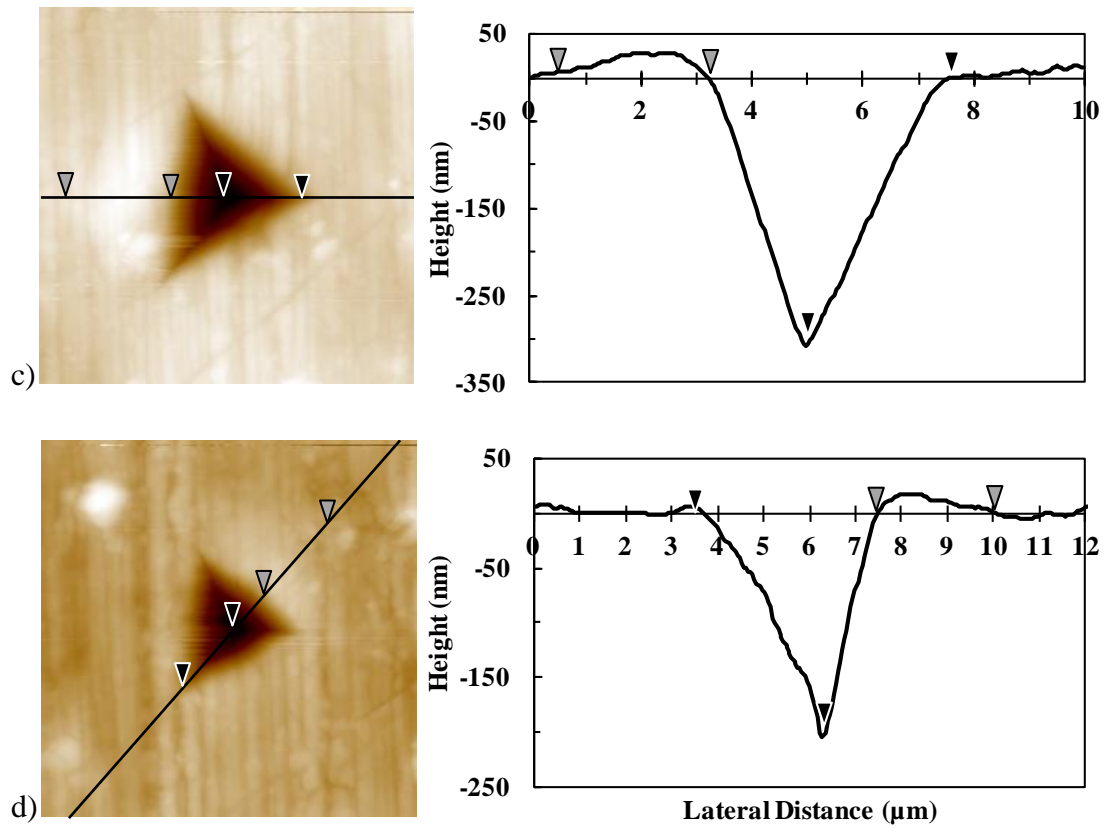


Figure 8. AFM images (left) and cross-sectional information (right) of the drawn line in the image obtained from bulk Al after indentation tests under (a) 10 (b) 8 (c) 5 and (d) 2 *mN* loads for open loop mode.

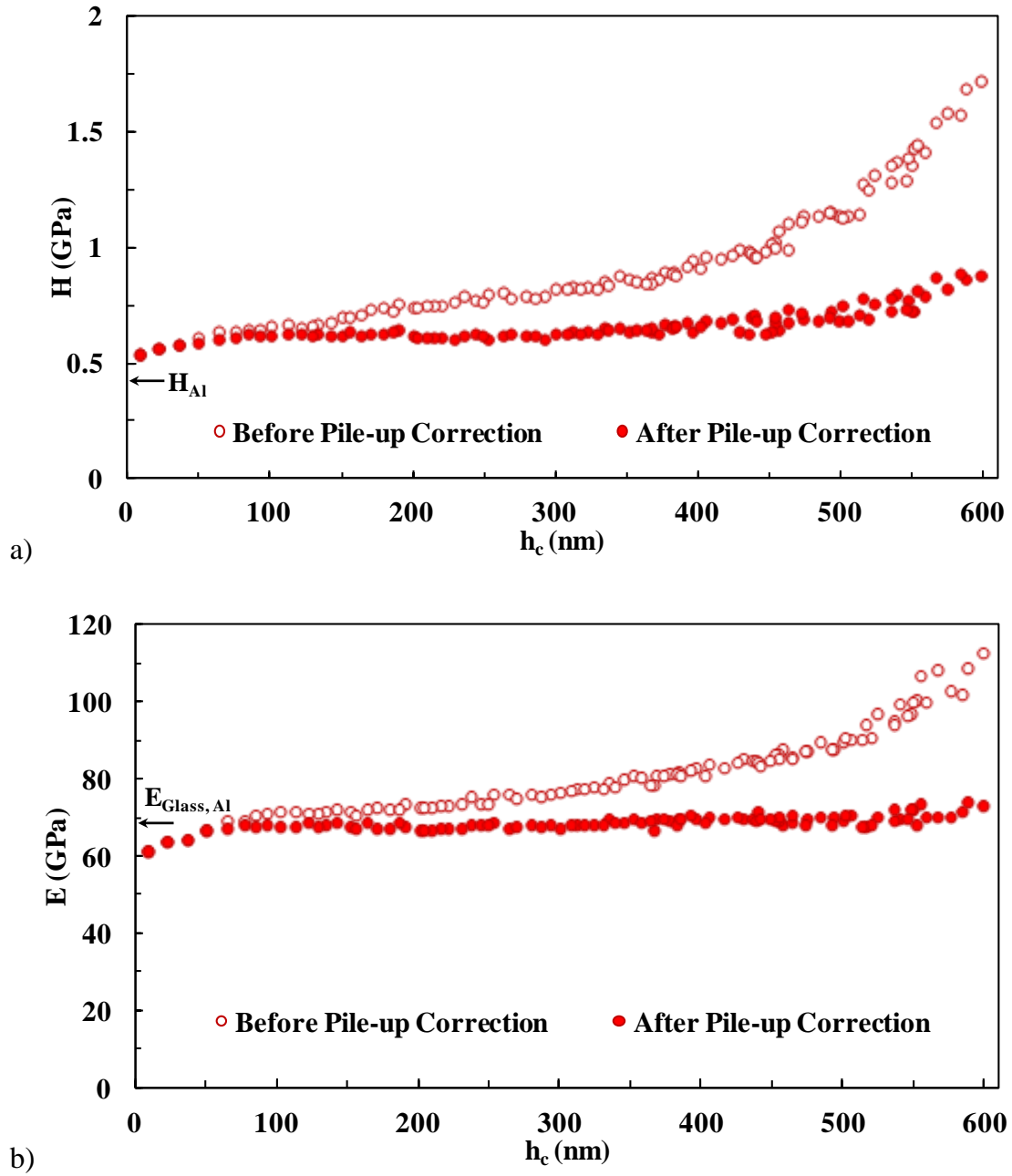


Figure 9. (a) hardness and (b) Young's modulus values of 1400 *nm* thick Al film for before pile-up correction (measured using the Oliver and Pharr method) and after pile-up correction (measured using the actual contact area).

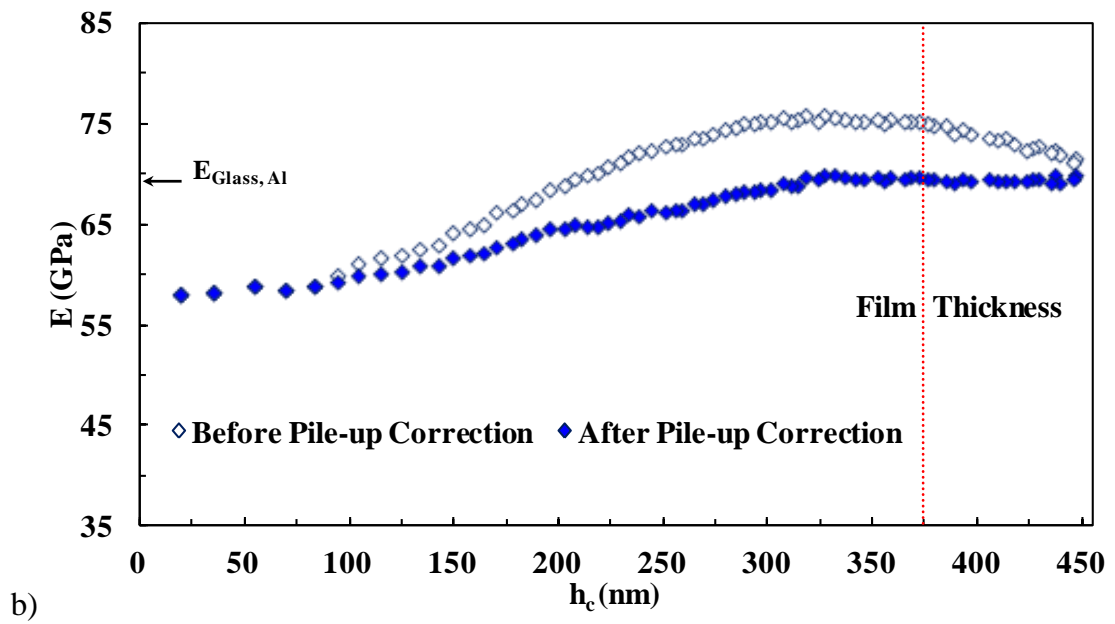
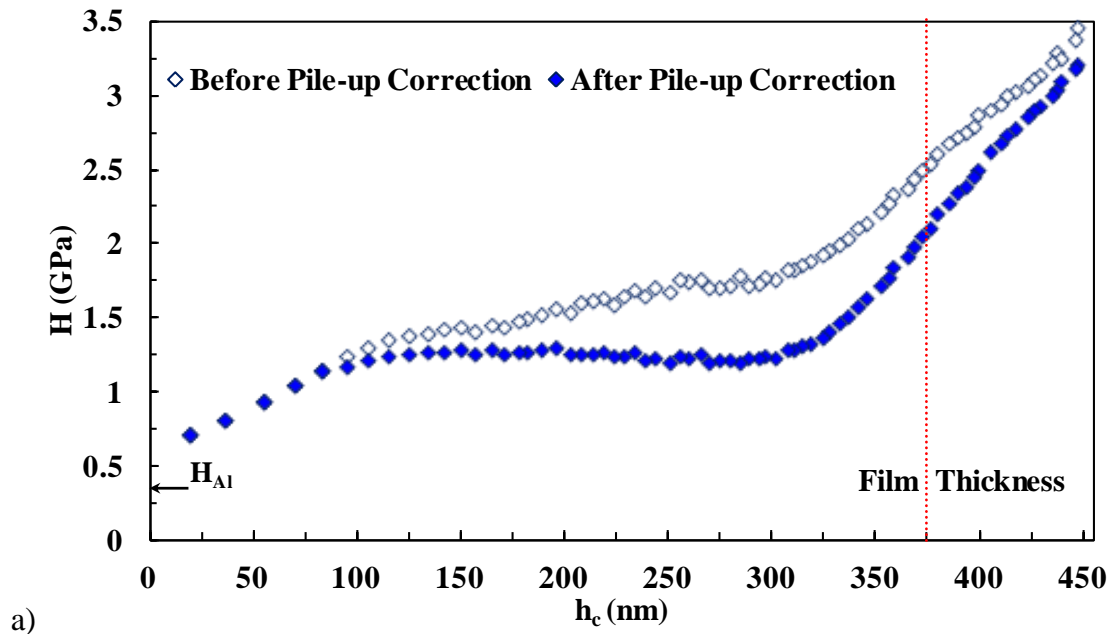


Figure 10. (a) hardness and (b) Young's modulus values of 375 nm thick Al film, before pile-up correction (measured using the Oliver and Pharr method) and after pile-up correction (measured using the actual contact area).

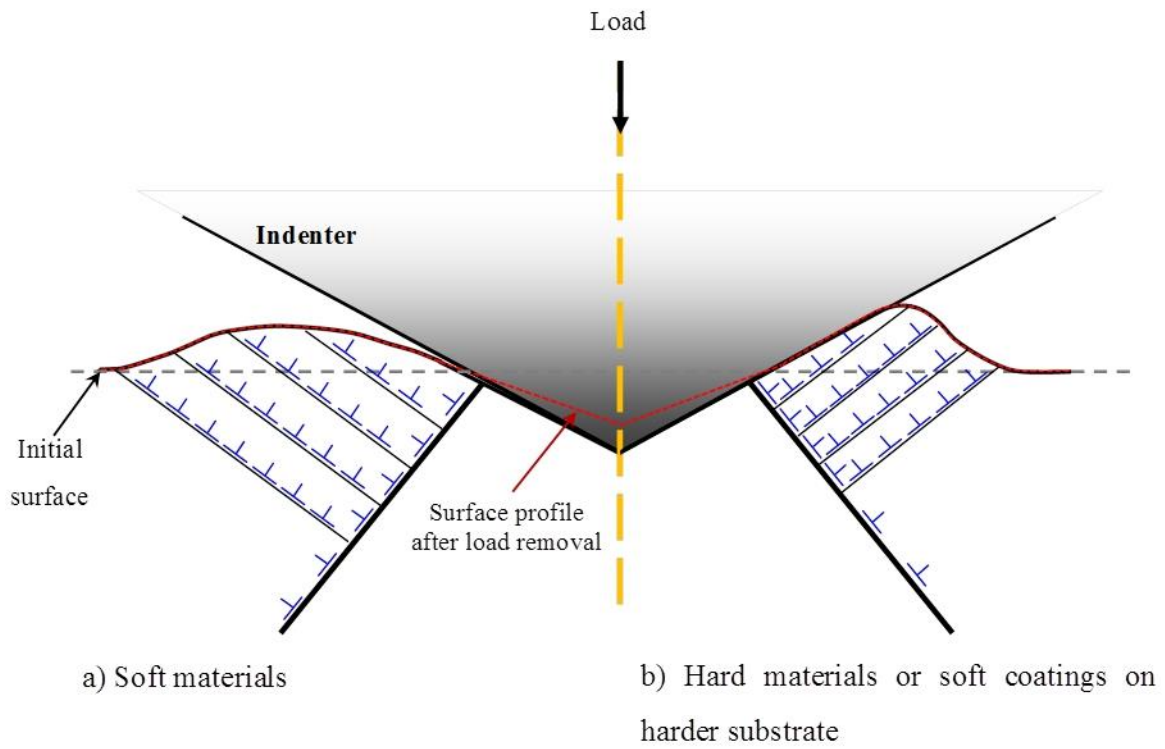


Figure 11. Schematic representations of the cross slip process during indentation: (a) softer materials with low H/E ratio and (b) harder materials with higher H/E ratio or soft coatings deposited on hard substrate.