Thin Solid Films 572 (2014) 189–199

Contents lists available at ScienceDirect

Thin Solid Films

journal homepage: www.elsevier.com/locate/tsf

A comparison of nanoindentation pile-up in bulk materials and thin films

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ARTICLE INFO

Available online 27 August 2014

Keywords:
Nanoindentation
Pile-up
Bulk materials
Thin films

ABSTRACT

During nanoindentation testing, there are many issues that need to be considered if high-quality data are 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. Hence, the volume of the pile-up is not always proportional to the indentation load and its shape can vary. Therefore, an 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 penetration of the indenter is greater than 10% of the thickness of the film, there are some errors, especially at greater depths, in the measured mechanical properties due to the influence of the substrate on the results; this consequently affects the load–displacement curve [16,18]. Furthermore, issues such as pile-up are significant when dealing with soft coatings on hard substrates, and the hardness and modulus 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 then sinks-in rather than piling-up. The appearance of pile-up and sink-in behaviour and its amount 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 results in an underestimation of the contact area causing an overestimation of the Young's modulus and hardness values [21]. Thus, 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 which supports the load.

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

http://dx.doi.org/10.1016/j.tsf.2014.06.060
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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 allow 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 apparent volume of the pile-ups is not always proportional to the indentation load, and the 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 measured 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 μm × 10 μm 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 roughness 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 roughness of the bulk samples tested in this work is less than 0.25 nm.

2.1.1. Copper

The 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 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 regard 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 (110) 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 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 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 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 in 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. Figs. 1(a) and (b) illustrate two AFM images (10 μm × 10 μm 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 Figs. 1(a) and (b) obtained from the three different sides of the indentation edges are shown in Figs. 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

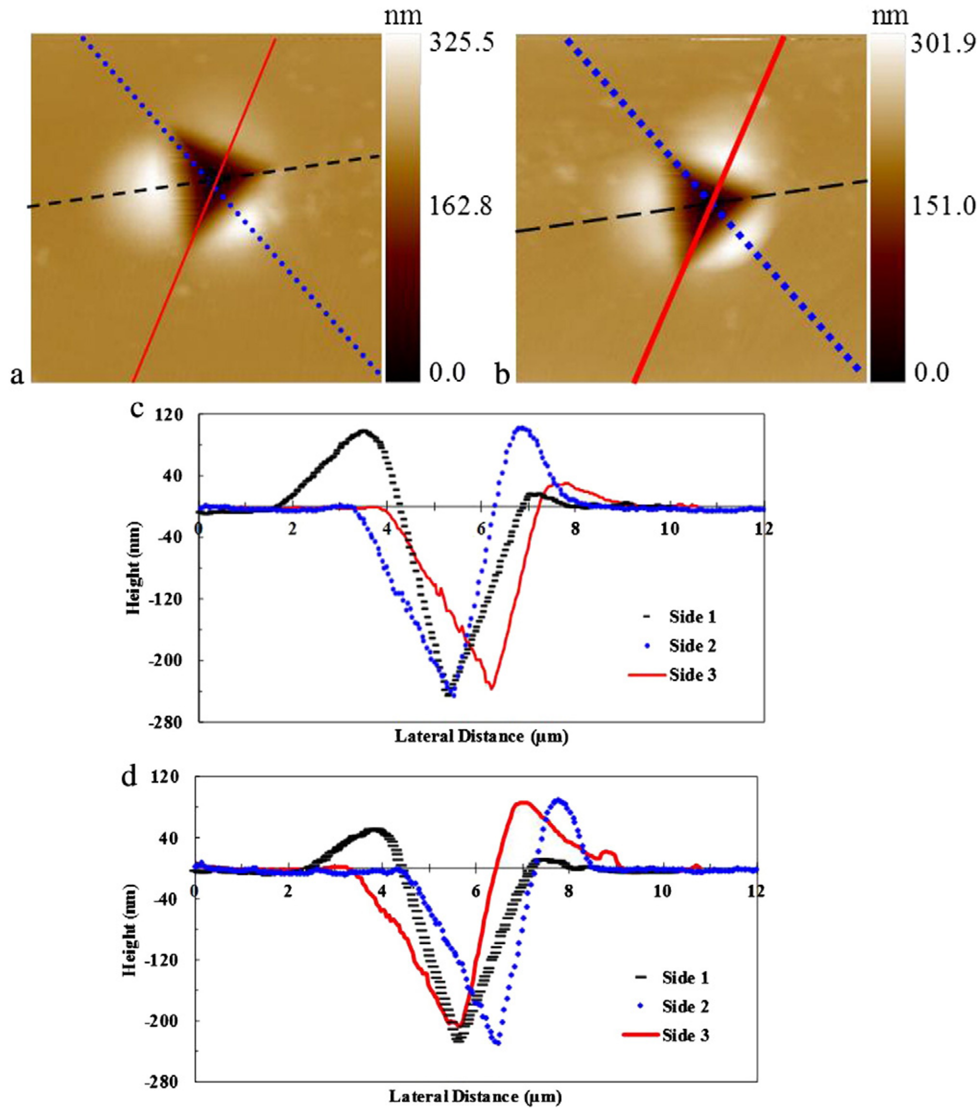

Fig. 1. (a and b) AFM images as well as respective (c and d) cross-sectional traces of the lines drawn in the images obtained from bulk Cu after indentation tests under 10 and 9 mN loads for open-loop mode.

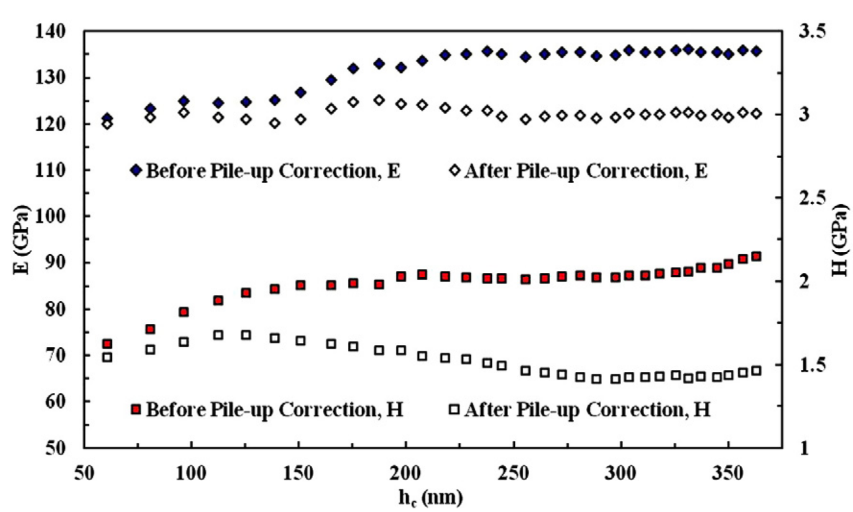

Fig. 2. Hardness and Young's modulus values obtained from high load indentation tests for bulk Cu using open-loop mode showing data before and after pile-up correction.

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 Fig. 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' 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 Figs. 3(a) and (b). As can be seen from Fig. 3, both the pile-up height and its width increase as the contact depth increases as expected as the width of the pile-up is related to the plastic radius zone around the indentation contact. For the indentations with contact depths of less than 50 nm, the pile-ups were extremely small and not observable. The effect of pile-up on the calculated hardness and modulus values at very small indentation depths is therefore 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. Surface roughness effects may also be significant.

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 (Fig. 4). 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 promotes pile-up formation from the thin film in a similar manner to a heavily work hardened material.

It should be noted that although the effect of pile-up in the hardness and modulus values is significant, 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 for the Cu samples. Au was chosen due to it

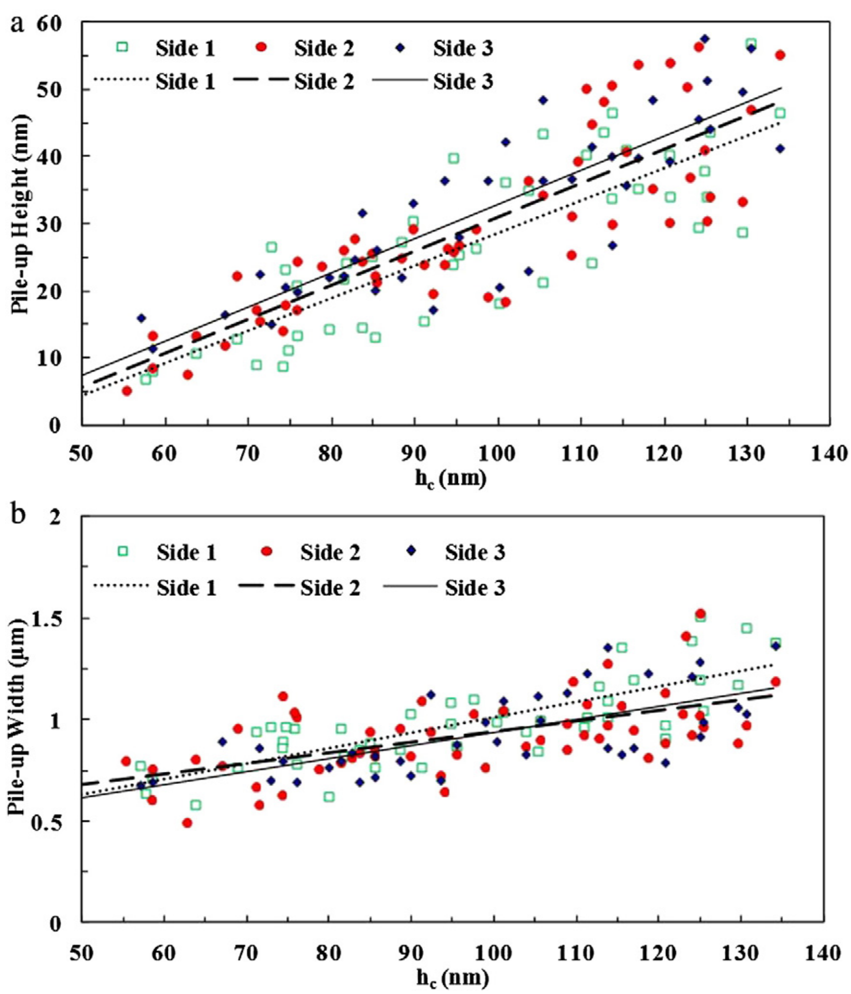

Fig. 3. (a) Height and (b) width of the pile-ups obtained from a single cycle nanoindentation test under open-loop mode for bulk Cu.

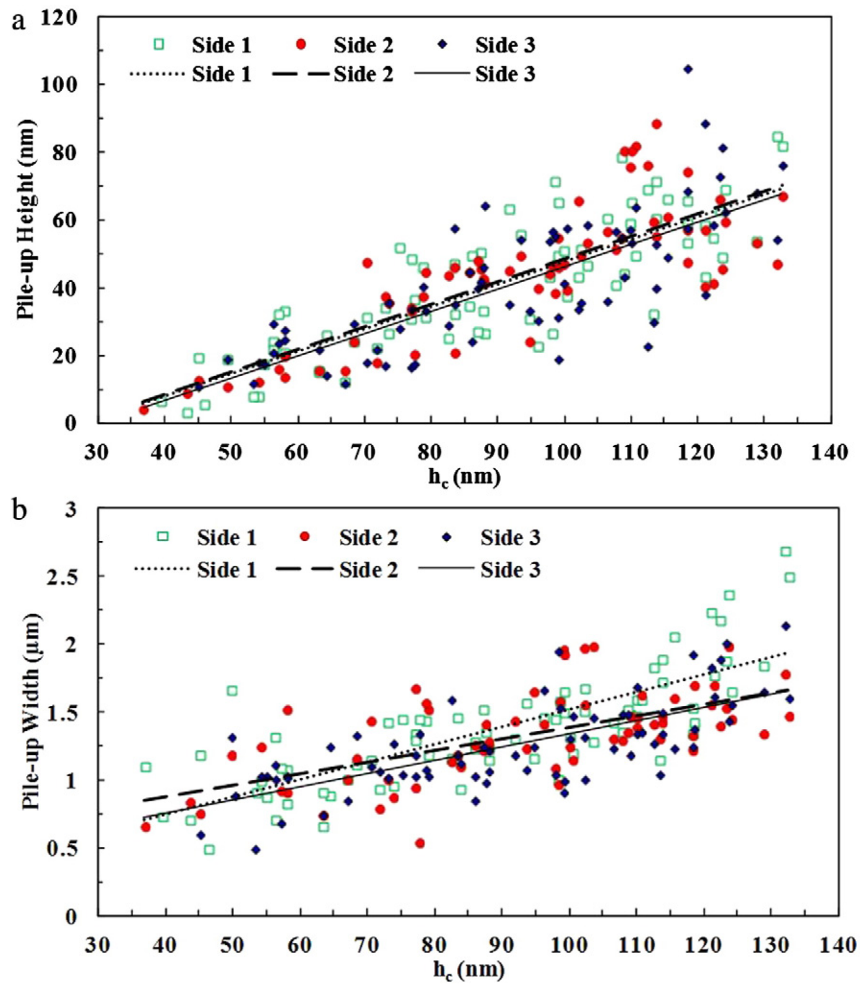

Fig. 4. (a) Height and (b) width of the pile-ups obtained from a single cycle nanoindentation test under open-loop mode for the Cu thin film.

having the same crystal structure as 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 the same indentation conditions. Fig. 5 shows two different AFM images (3D views and top-down views) obtained from typical indentation tests carried out on an Au thin film. The appearance of pile-up for Au films was clear from the AFM images under the full range of loads tested. All results from the indentation tests are therefore affected by pile-up.

An experiment was then carried out on the bulk Au single crystal under the same test conditions to assess the required pile-up correction. The average hardness and modulus values obtained from single crystal Au were 1.05 ± 0.2 and 84.91 ± 1 GPa, respectively. The corresponding AFM images confirmed the appearance of significant pile-up for bulk Au even at low loads; the pile-ups were more symmetrical, so 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 with the bulk Au, the thin films 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 Fig. 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/SiO_2 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 and texture) [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. The films showed a texture closer to (100), which probably explains the lower modulus values when compared to the (110) oriented single crystal.

In general, the values vary in a similar manner to that of thin Cu films and bulk Cu under similar indentation conditions. As with the Cu thin films, the hardness graphs shown in Fig. 6 can be divided in to 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 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 on 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

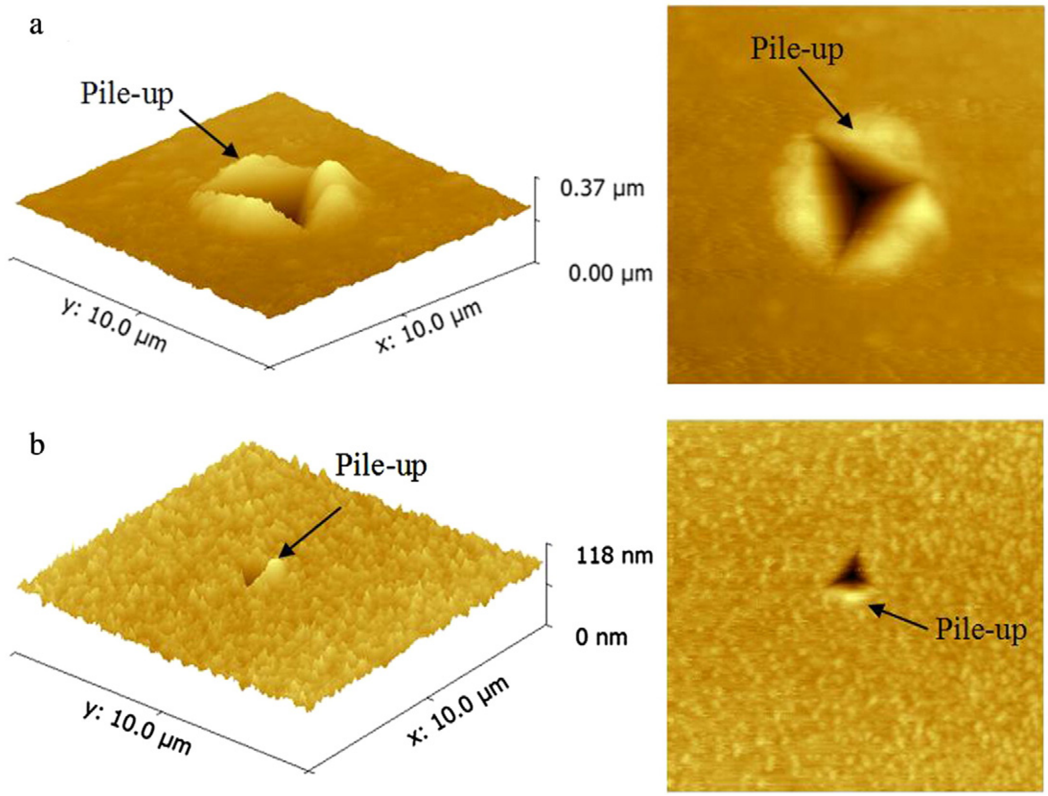

Fig. 5. AFM images for typical indentations on the Au thin film, three dimensional views and top-down views of (a) high load (10 mN), (b) lower load (1.5 mN) impressions.

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

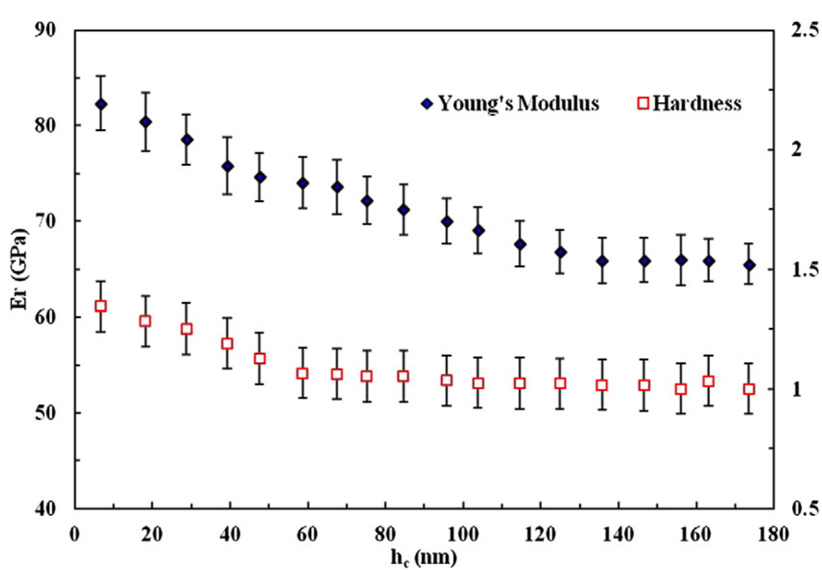

Fig. 6. (a) Young's modulus and (b) hardness results for Au thin film after pile-up correction.

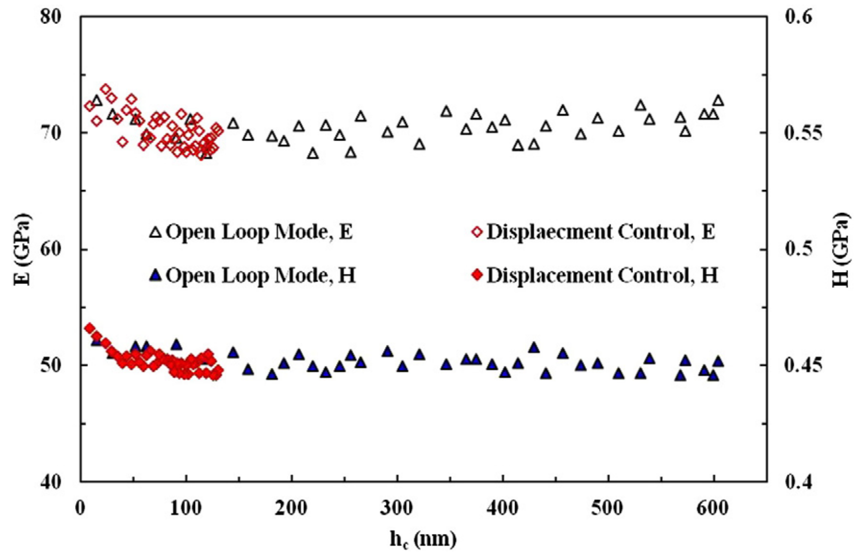

Fig. 7. (a) Young's modulus and (b) hardness values of bulk Al obtained under open-loop mode and displacement control.

displacement control for contact depths less than 130 nm. The hardness and modulus values for the bulk Al sample are shown in Fig. 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 regard 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 Fig. 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 presented previously. 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 have 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 Fig. 9 as a function of penetration depth.

The hardness values from the Oliver and Pharr analysis method (shown with open circles in Fig. 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 Fig. 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 from 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 were compared to that of the 375 nm Al film shown in Fig. 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 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 indenter displacement of 325 nm, the hardness increases rapidly while the pile-ups get slightly smaller and the effect on the

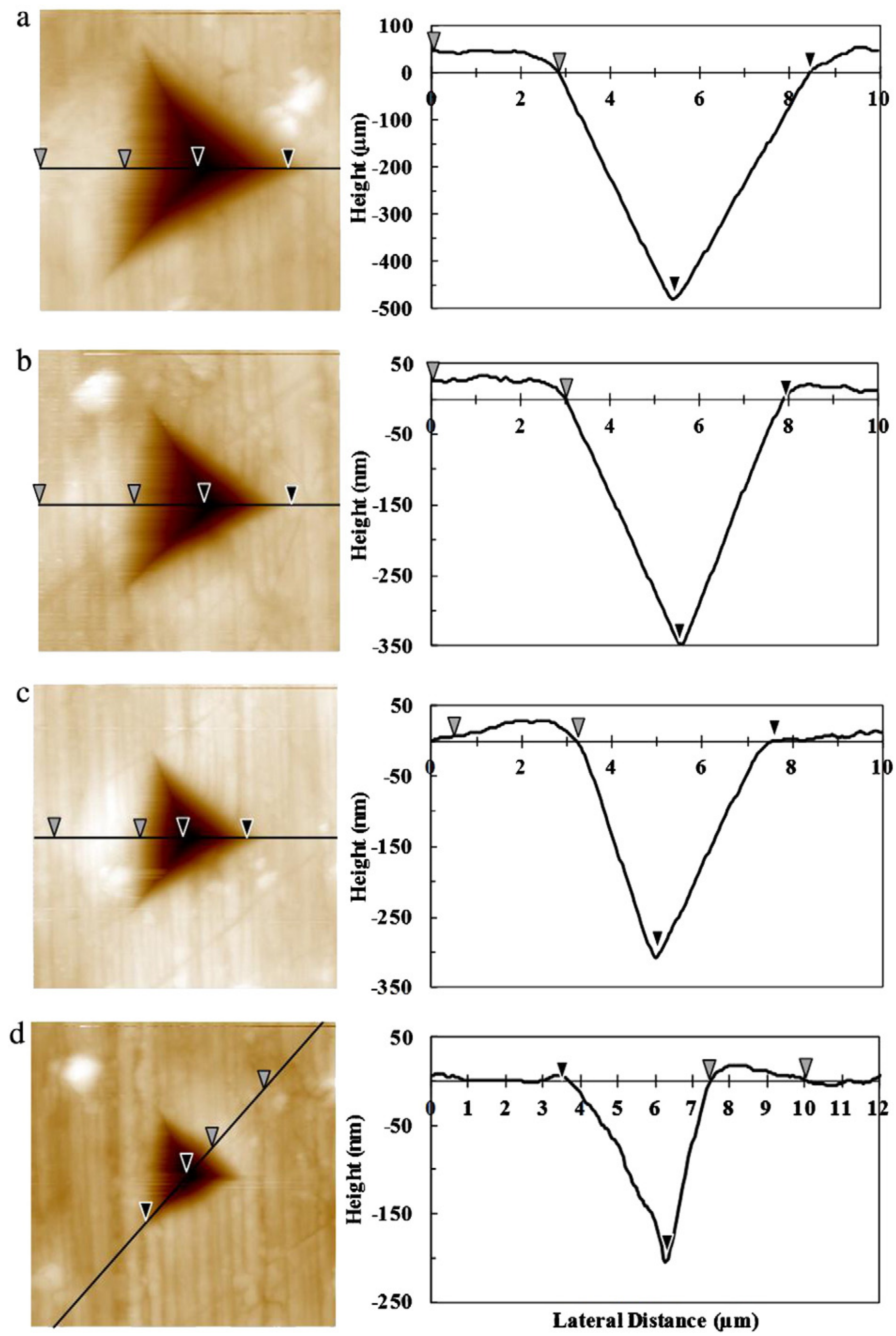

Fig. 8. (a) AFM images and (b) cross-sectional information from the line drawn in the image obtained from bulk Al after indentation tests under (i) 10, (ii) 8, (iii) 5, and (iv) 2 mN loads for open-loop mode.

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 increase with increasing contact depth and are 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 end of the unloading curve [18].

Although both the aluminium thin film and glass have similar Young's moduli, the glass has a higher hardness value, requiring a higher contact pressure for plastic deformation 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 amount of pile-up. When the indenter penetration is a reasonable fraction of the film thickness, the hardness and modulus

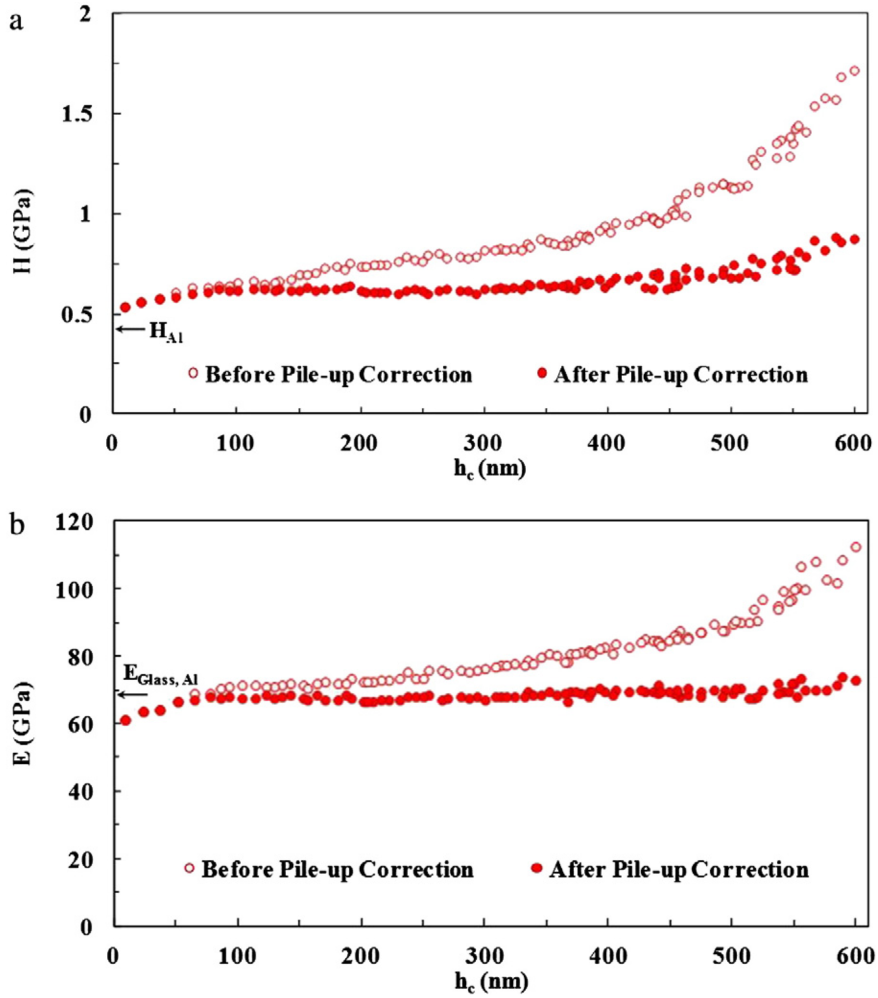

Fig. 9. (a) Hardness and (b) Young's modulus values from the 1400 nm thick Al film before pile-up correction (measured using the Oliver and Pharr method) and after pile-up correction (measured using the actual contact area).

values depend on the thin film's elastic recovery with influences from the hard substrate and the way it affects pile-up. However, in the scenario in which the indenter displacement is greater than the film thickness, both the film and the substrate affect the loading and unloading parts of the load–displacement curve. 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 measured mechanical properties are strongly dependent on the measured stiffness, any changes in the curve can have an effect on the hardness and modulus 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] that softer materials such as bulk Al with low H/E ratios (0.006) show a smaller pile-up effect than harder materials such as Cu with higher H/E ratios (0.012) where cross slip during indentation is more pronounced (Fig. 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 effect 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. Generally, for submicron metal films, the Oliver and Pharr method will overestimate these properties unless a direct measurement of contact area is obtained. Calibration methods 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. Conclusions

The purpose of this study was to perform a comparison between the pile-up appearance for various metals 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 using the Oliver and Pharr method was identified for both cases. When the appearance of pile-up for bulk Al was compared with Cu and Au, it was found that the effect of pile-up on the mechanical properties of bulk Al is much smaller 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 lower pile-ups were seen for bulk Al and 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

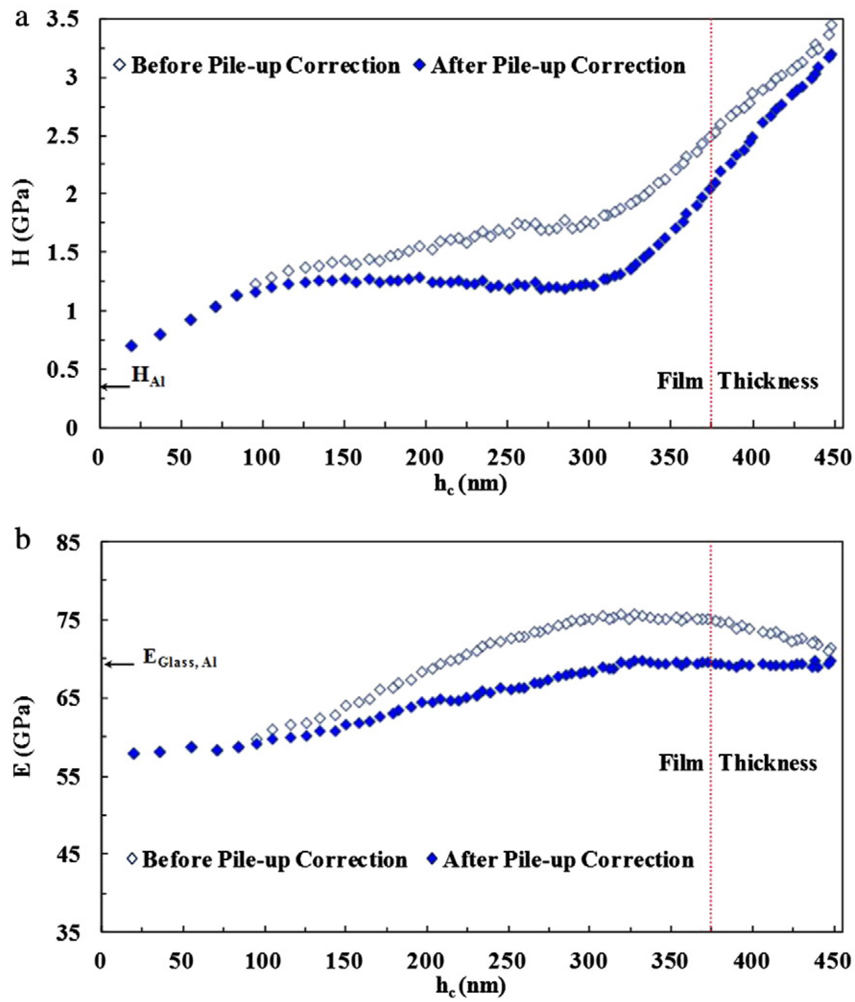

Fig. 10. (a) Hardness and (b) Young's modulus values from the 375 nm thick Al film before pile-up correction (measured using the Oliver and Pharr method) and after pile-up correction (measured using the actual contact area).

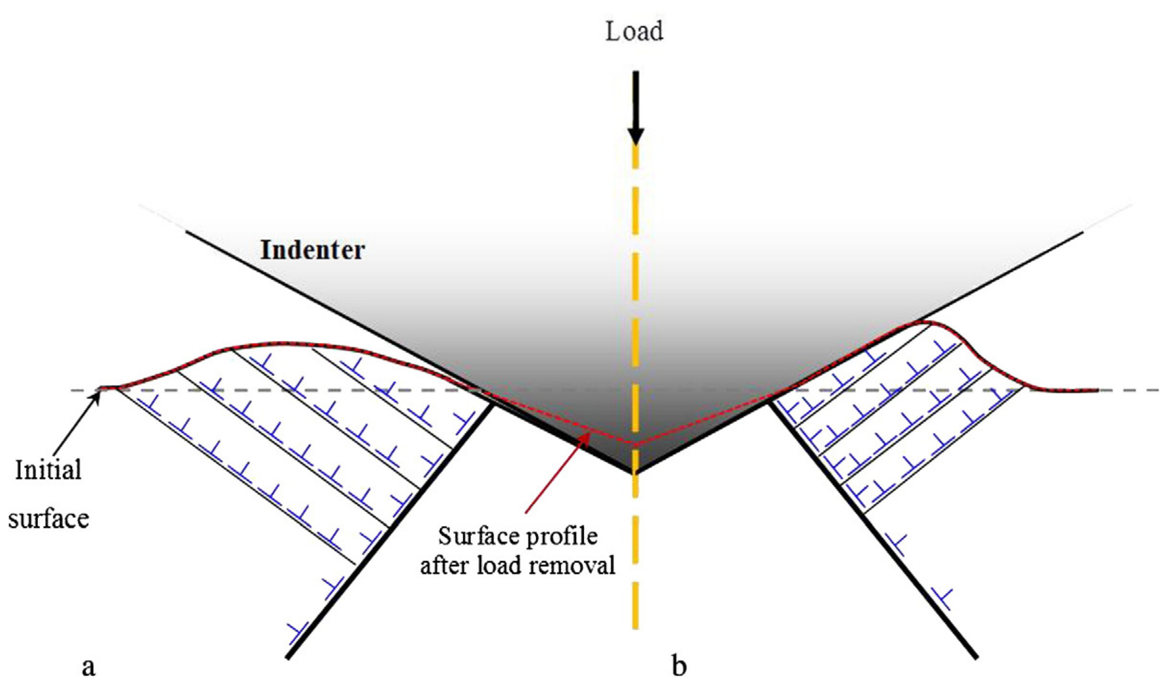

Fig. 11. Schematic representations of the cross slip process during indentation: (a) softer materials with low H/E ratio and (b) harder materials with higher H/E ratio or soft coatings deposited on a hard substrate.

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 shape of the pile-up formed, 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 in the data measured.

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.

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