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DIRECT MEASUREMENT OF RESIDUAL CONTACT AREA AND VOLUME DURING THE NANOINDENTATION OF COATED MATERIALS AS AN ALTERNATIVE METHOD OF CALCULATING HARDNESS

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Abstract - The mechanical properties of thin films can be measured by a variety of different techniques, with nanoindentation being one of the most recent developments in this growing field. By using a depth-sensing indentation method it is possible to obtain quantitative values for the hardness and modulus, and thus gain better insight into the response of a material to controlled deformation at such small scales. However, previous work [1] has shown that the effects of pile-up, particularly in soft films deposited on hard substrates, can produce significant overestimation of the hardness and modulus due to an underestimation of the true contact area by common nanoindentation analysis procedures. By measuring the topography of the residual indent using Scanning Force Microscopy (SFM) and combining this information with the indentation data, it is possible to gain a fuller understanding of the indentation method and its effects on the material being tested. In addition, the true contact area can be directly measured from the SFM images and subsequently used to recalculate the hardness of the material more accurately. Moreover, the SFM allows the plastic volume of indentation to be measured, from which hardness can also be calculated in terms of plastic work. Experimental results are presented for two types of thin film deposited on hard substrates where SFM analysis of indentations at varying depths gives significant additional information concerning the true response of the system to instrumented indentation at a nanometric scale. Pile-up effects can be precisely monitored as a function of depth and correlated to hardness variations encountered across the coating/substrate interface.

Keywords: Nanoindentation, Contact Area, Hardness

1. INTRODUCTION

The nanoindentation method for assessing mechanical properties at low loads and shallow depths is already well established for the characterisation of thin films as well as bulk materials. The depth-sensing indentation method produces a load-displacement curve from which quantitative property values can be calculated using a variety of approaches [2-6]. Although such approaches give significant information concerning the mechanical response to indentation, it has already been shown [7] that the effects of pile-up and sink-in, particularly in the case of soft films on hard substrates, can drastically affect the values calculated if the residual contact area is required in the calculation. This is especially the case when measuring important material parameters such as hardness, H, and modulus, E, as both are extracted from a load-displacement curve using the following equations:

$$H = \frac{P}{A} \tag{1}$$

$$E_{eff} = \frac{E}{1-\nu^2} = \frac{\sqrt{\pi}}{2}\frac{S}{\sqrt{A}} \tag{2}$$

where P is the load, A is the projected contact area, E_{eff} is the effective elastic modulus defined in terms of Young's modulus, E, and Poissons ratio, ν, and S is the experimentally measured contact stiffness, e.g., using the Oliver and Pharr method [3], the value of S corresponds to the gradient of the tangent to the unloading curve. Considering the fact that both H and E_{eff} are dependent on the contact area, and that the load-displacement curve from which A is calculated does not account for pile-up and sink-in effects, significant errors can occur if the true residual contact area is not measured by some other method.

The use of scanning force microscopy (SFM) for imaging residual indentations has already proved to be one of the only methods presently available for obtaining accurate dimensional information from an image area of only a few microns [8-9]. The true residual contact area of an indentation impression can therefore be measured directly from a SFM image and used to calculate hardness using equation (1). This method enables the greater or lesser contact area (due to *pile-up* or *sink-in* effects) to be taken into account and corresponds more closely with measurement of hardness at the micro- and macro-scales, where the residual contact area is measured directly by using optical microscopy.

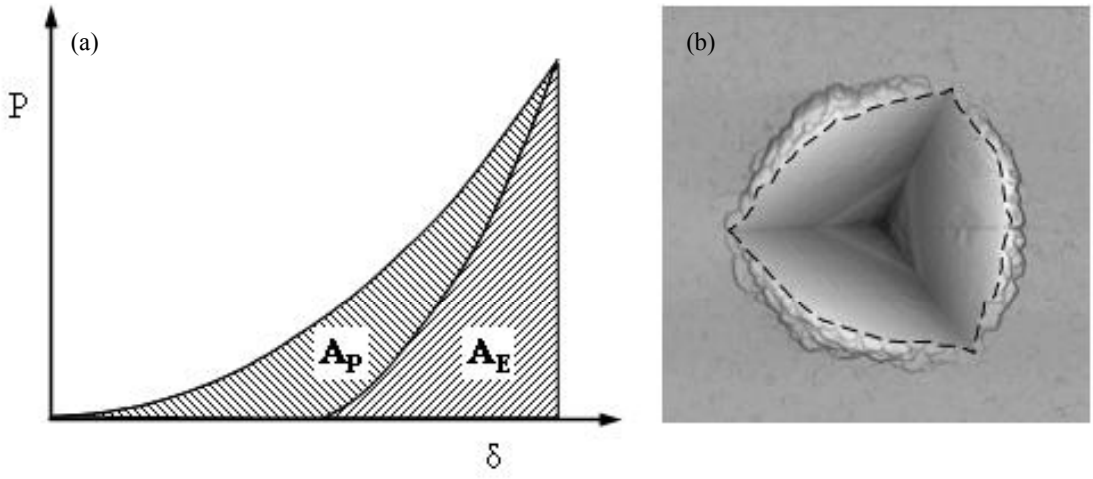

Fig. 1. Typical load versus depth plot (a) showing the elastic (A_E) and plastic (A_P) areas. The SFM image in (b) shows the selected boundary (dashed line) around the residual indent from which the projected contact area and plastic volume can be calculated.

Hardness can also be calculated by using an *indentation energy* approach [10-11] where the plastic work of indentation (the area enclosed by the loading and unloading curves) is divided by the residual volume of the indentation itself. This can also be interpreted as the irreversible energy required to create a unit volume of indentation at the maximum load during elasto-plastic surface deformation. The residual volume, V, can be measured directly from a SFM image or calculated from the load-depth curve and the indenter geometry using

$$V = \frac{1}{3}\left(A_P \times h_f\right) \qquad (3)$$

where A_P is the plastic contact area and h_f is the final depth of the residual impression after unloading. However, estimating A_P from the load-depth curve can be inaccurate as this method assumes that the unloading curve can simply be described by a power-law fit. Significant error in the calculated value of A_P also occurs if it is estimated from the indenter area function and the residual penetration depth, h_f, as any elastic recovery of the surface will not be taken into account, nor any *pile-up* or *sink-in* effects. The only accurate method is by direct imaging of the residual indentation impression.

This paper is aimed at providing a comparison between hardness calculated by conventional nanoindentation with that calculated using a direct SFM image of the residual impression.

2 EXPERIMENTAL

2.1 Instrumental set-up

The apparatus consists of a Nano Hardness Tester (NHT) with integrated SFM objective [7, 12]. This system, developed by CSM Instruments, comprises two distinct components; a measuring head for performing nanoindentations and an optical microscope for selecting a specific sample site prior to indentation and for checking the location of the imprint after indentation. Both components are directly linked by an electro-mechanical positioning system which allows movement along two perpendicular horizontal axes with a lateral displacement resolution of 1 μm. The measuring head applies load via an electromagnet assembly to a vertical rod, the end of which houses a diamond indenter of either Vickers or Berkovich geometry. Displacement of the rod is measured by a capacitive detecter and the rod is supported by two guide springs. The system has load and displacement resolutions of 1 μN and 0.3 nm respectively.

The NHT has several advantageous features, in particular its differential measurement of the indentation depth, made possible by a sapphire reference ring which remains in contact with the sample during the loading/unloading cycle, giving exact positioning of the indenter tip relative to the sample surface. Thus the overall elastic recoil of the sample and partial frame compliance are compensated, as is most of the associated thermal drift during measurement.

Having performed an indentation at a specific site, the sample is switched to the optical microscope/SFM objective and the residual imprint located within the 20 μm scan range of the SFM.

2.2 Sample materials and methodology

Two coating-substrate combinations were chosen having different mechanical properties. The first consisted of an aluminium film with a thickness, t_f,of 800 nm, the second a TiW film of thickness 320 nm, both sputtered onto [100] silicon wafers (p-type). The two samples were indented with maximum applied loads in the range 0.5 – 300 mN (24 load intervals) in order to utilise the full range of the instrument. The loading rate was maintained at a value double that of the maximum applied load so that each measurement load-depth cycle required the same amount of time. This was to minimise thermal drift variations and

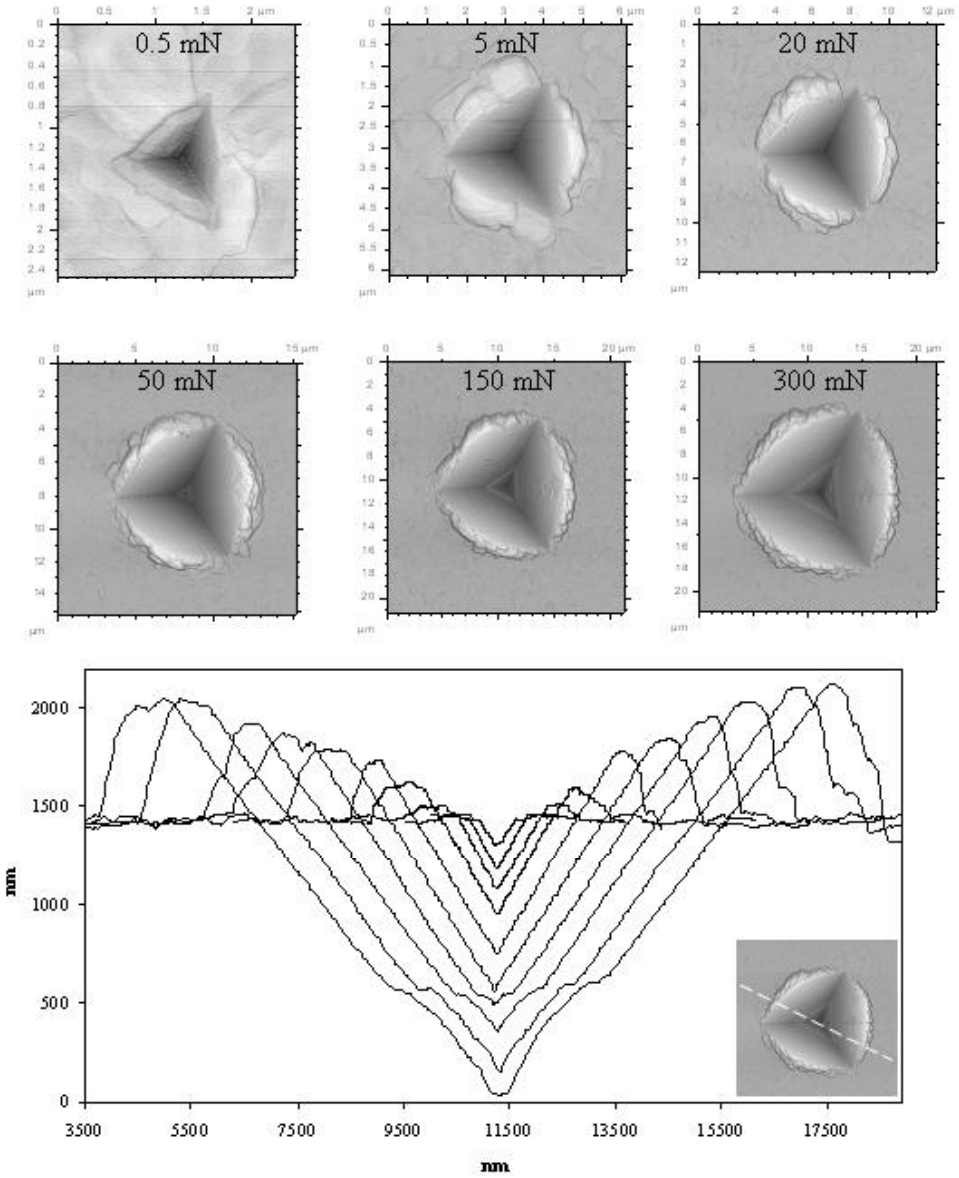

Fig. 2. Series of SFM images of residual Berkovich imprints for applied loads from 0.5 mN up to 300 mN with corresponding cross-sectional profiles. The sample is an aluminium film (thickness = 800 nm) sputtered onto a Si [100] substrate. Note the evolution of pile-up with increasing load.

time-dependent effects. At each value of applied load, a series of five indentations was made and a Berkovich diamond indenter was used for all measurements.

After each set of indentations, one or two of the residual imprints was positioned under the SFM objective and imaged in contact mode using a silicon cantilever of spring constant ~0.3 N m^{-1}. All images were made using the maximum possible resolution, i.e., 512 lines with 512 pixels per line. The images were then treated in a separate software package which allows the exact residual contact area and plastic volume to be computed. The hardness of each coating-substrate system was then calculated at each load using the following methods and nomenclature:

(i) H_{CALC}: Hardness calculated directly from the load-depth curve using the Oliver and Pharr method [3]. An average value was compiled from the five indentations made at each load interval.

(ii) H_{WORK}: Hardness calculated by dividing the plastic work of indentation (the area A_P of the load-depth curve, as shown in Fig. 1 (a)) by the residual plastic volume as measured directly from the corresponding SFM image.

(iii) H_{SFM}: Hardness calculated by dividing the maximum load at each interval by the projected contact area as measured directly from the corresponding SFM image.

The SFM image shown in Fig. 1 (b) shows the selected boundary for a typical residual indent, from which the projected contact area and plastic volume were calculated. Note that the boundary includes any pile-up effects.

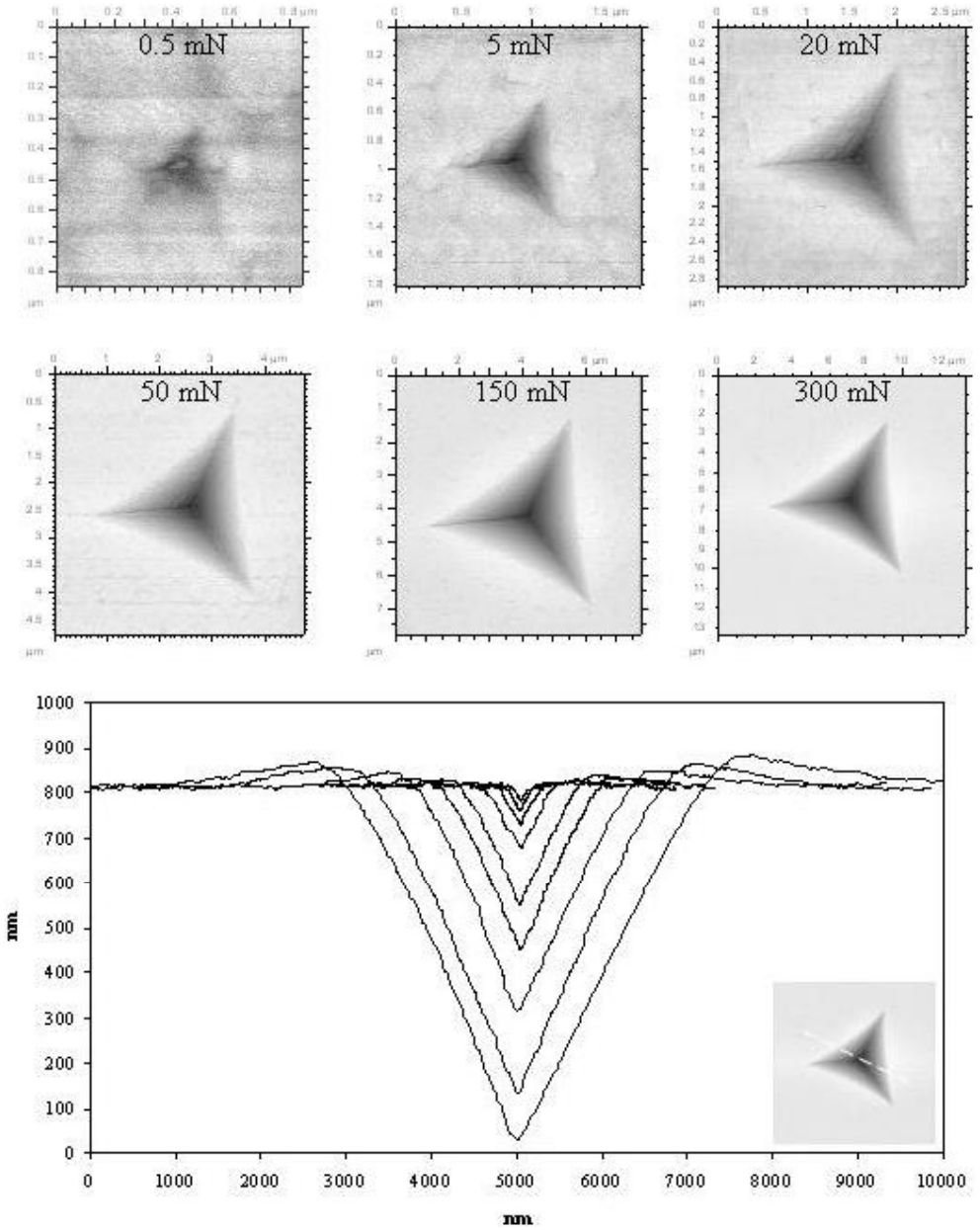

Fig. 3. Series of SFM images of residual Berkovich imprints for applied loads from 0.5 mN up to 300 mN with corresponding cross-sectional profiles. The sample is a TiW film (thickness = 320 nm) sputtered onto a Si [100] substrate.

3 RESULTS AND DISCUSSION

Fig. 2 shows a selection of SFM images of residual imprints on the aluminium sample over the applied load, P, and maximum depth, h_{max}, ranges $0.5 \leq P \leq 300$ mN and $157 \leq h_{max} \leq 2020$ nm respectively. Cross-sectional profiles through SFM images for a wider selection of imprints have been superimposed on the same axes in order to clearly show the evolution of pile-up with depth, as well as the change in pile-up volume with respect to indentation volume as the indenter reaches the coating-substrate interface and continues down into the Si substrate. At low loads, and where $h_{max} < 10\%\ t_f$, no pile-up is present, thus confirming a response typical of bulk aluminium. It is interesting to note that the substrate relaxes far more than the soft plastic coating deposited onto it, this being evident from the profiles where $h_{max} > t_f$.

Fig. 3 shows a selection of SFM images of residual imprints on the TiW sample over the ranges $0.5 \leq P \leq 300$ mN and $33 \leq h_{max} \leq 1297$ nm respectively. It can be seen, especially from the cross-sectional profiles, that pile-up is much less evident than for the Al-on-Si system and that the coating-substrate transition cannot be observed.

The hardness results are plotted in Fig. 4 for the two measured samples and discrepancies are apparent between H_{CALC}, H_{SFM} and H_{WORK}. In the case of the TiW (Fig. 4 (a)), all three calculation methods begin with an

overestimation of the coating hardness, after which the H_{CALC} values flatten off around a value of 12 GPa which corresponds with the known value of TiW. The initial overestimation is probably due to surface effects (surface roughness, oxide layers, tip calibration discrepancies, etc.) which are dominant at low loads. The values of H_{SFM} seem to follow roughly the same pattern as for H_{CALC} but with a consistent increase of about 2 GPa. Such a phenomenon can be attributed directly to the measurement of the projected contact area, as the values of P are the same in both cases, and signifies that this area is smaller when measured by SFM than when estimated from the load-depth curve. The exact reasons for this are not clear, especially given the fact that pile-up effects are minimal for TiW-on-Si and that both coating and substrate can be considered as having similar mechanical properties. The values of H_{WORK} are rather interesting as they seem low where $h_{max} < t_f$, reasonable (12 – 13 GPa) in the range $1 \leq h_{max}/t_f \leq 1.5$ and then rise gradually up to a value of 19 GPa which is obviously an overestimation if it is considered that the Si substrate has a hardness of ~9 GPa. The behaviour of H_{WORK} could perhaps be explained in terms of indentation energy effects; cracking in Si has already been observed [12] when indenting with a Berkovich indenter at loads above approximately 100 mN (in the case of TiW this value corresponds to a h_{max}/t_f ratio of 2.3).

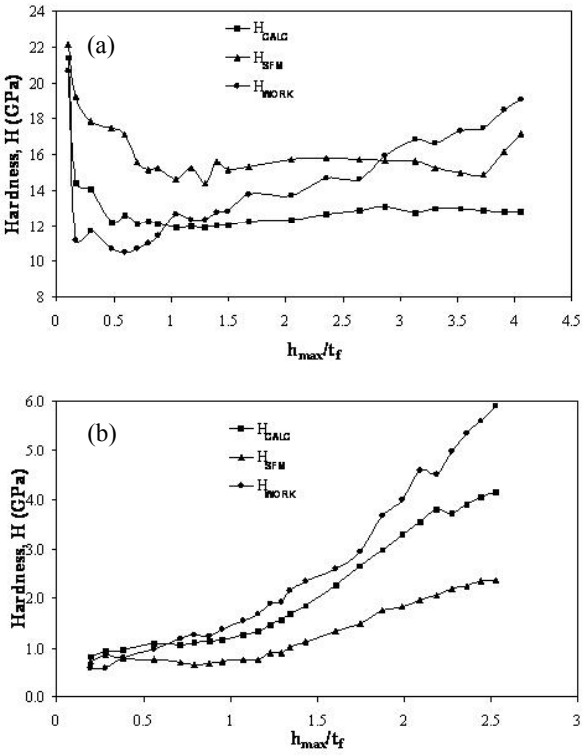

Fig. 4. Variation of hardness as a function of normalised depth (h_{max}/t_f) for TiW (a) and Al (b) thin films deposited onto a Si [100] substrate. H_{CALC} denotes the hardness calculated from the load-depth curves, H_{SFM} denotes hardness calculated with the true projected contact areas measured by SFM, and H_{WORK} denotes hardness calculated with the plastic volume measured by SFM.

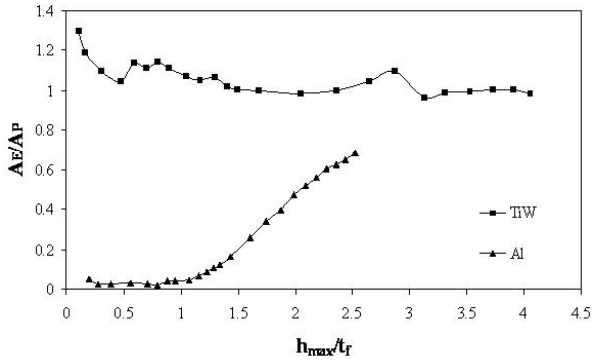

Fig. 5. Ratio of elastic and plastic work (A_E/A_P) plotted as a function of normalised depth (h_{max}/t_f) for TiW and Al thin films deposited onto a Si [100] substrate.

This factor would increase the plastic energy measured from the load-depth curve whilst not affecting the measured volume. However, no evidence of substrate cracking was observed from the SFM images, although such effects could be masked by the TiW coating.

For the case of the Al (Fig. 4 (b)), all three calculation methods provide an accurate representation of the coating hardness (~0.6 - 0.8 GPa) at low loads, after which they all tend towards that of the Si substrate (~9 GPa). This correlates well with previous work on the same coating-substrate system [1, 9]. The values of H_{SFM} are, in this case, consistently lower than the values of H_{CALC} and H_{WORK} due to pile-up effects which result in a larger projected contact area than that extrapolated from the load-depth curve. In contrast, the values of H_{WORK} are significantly higher than H_{CALC} at $h_{max}/t_f > 1.7$, perhaps due to substrate cracking as for TiW. It should also be noted that H_{WORK} tends towards the substrate hardness to a greater extent than H_{SFM} and H_{CALC} over the measured depth range.

Such data sets calculated by different methods beg the question of which method is the most valid for a particular coating-substrate system? Additional evidence can be gained by plotting variations in elastic and plastic contribution as the indentation depth increases through and beyond the coating-substrate interface. Fig. 5 shows the ratio of elastic and plastic work (A_E/A_P) plotted as a function of normalised depth for the two measured material systems, where A_E and A_P correspond to the curve areas described in Fig. 1. The TiW values show little variation, suggesting that the TiW constrains the Si and that the elastic properties of these two materials match quite closely (this has been confirmed by separate elastic modulus, E, measurements which give values of 155 GPa for TiW and 130 GPa for Si). In contrast, the Al values remain very low up to the coating-substrate interface ($h_{max}/t_f = 1$), a response representative of the dominant plasticity in aluminium. For depths greater than the interface, the values increase linearly as the substrate elasticity contributes to a growing extent to the total indentation energy.

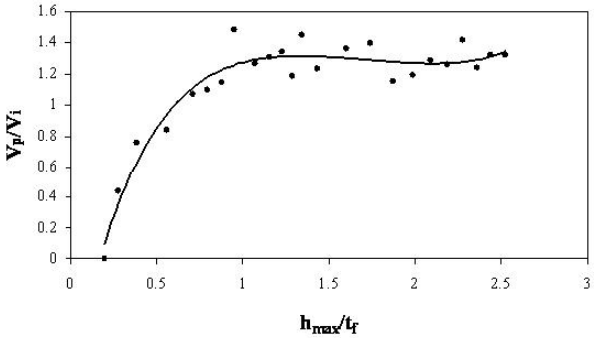

Fig. 6. Ratio of pile-up volume to indent volume (V_p/V_i) plotted as a function of normalised depth (h_{max}/t_f) for the Al film.

These observations for Al can be correlated with the ratio of pile-up volume, V_p, and indent volume, V_i, as shown in Fig. 6. Such volumes were calculated directly from the SFM images by defining the plane height of the material surrounding the residual indentation site and then calculating the volume of material above (V_p) and below (V_i) this reference plane. This ratio accurately describes the influence of pile-up through the range $0.2 \leq h_{max}/t_f \leq 2.5$, i.e, negligible at low load, but then becoming asymptotic at a V_p/V_i ratio of ~1.3. It is believed that this ratio would decrease at greater depths, as the thickness of the Al coating becomes negligible relative to the indentation depth, but such a study is beyond the range of the instrument used.

4 CONCLUSIONS

Although this study has only covered two different coating-substrate systems, the results have shown that useful hardness data can be calculated from post facto imaging of the residual indentation site with an integrated SFM. Advances in image-processing software allow both areas and volumes to be accurately measured, provided that pixellation and other error sources are taken into account [13]. For material systems where pile-up effects are dominant or exaggerated (as in the case of a soft coating deposited on a hard substrate), then H_{SFM} will give a more accurate representation of the composite hardness than H_{CALC} which does not account for pile-up effects. H_{WORK} can provide information concerning the energetic response of the system although further work is required to establish whether the error encountered when measuring a residual volume from an SFM image is significant. However, H_{WORK} can give a more representative hardness value for brittle materials where cracking is encountered.

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