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Elastic relaxation of indented coated substrates using a coated cavity model

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Abstract

The elastic and plastic properties of materials can be measured very conveniently using depth-sensing nanoindenters, although some interpretation of the results is necessary in order to remove the effects of elastic sinking-in of the impression under load. The interpretation of the indentation of coated substrates, when the depth of indentation is comparable with the film thickness, is more difficult, however. The elastic unloading curve is non-linear, even for a constant contact area, reflecting the depth dependence of the elastic properties. This makes necessary a more careful calculation of the hardness, based on a model of the elastic properties. A simple model of these properties is proposed, derived for a coated spherical cavity geometry. However, the approach fails to account for the observed behaviour, since the elastic displacements in the real geometry are not radial. A more appropriate treatment of the problem is discussed.

1. Introduction

Depth-sensing nanoindenters are widely used as an efficient means of measuring the hardness and elastic properties of materials [1, 2]. These devices operate by monitoring the load on the indenter for a range of depths of penetration into the surface. Assuming the geometry of the indenter is known precisely (which is not always the case), the indentation depth can be used to calculate the area of the indentation, and therefore to obtain the hardness, which is the load per unit projected area of the indentation. This is much easier than imaging and measuring the indentation impression after the indenter has been completely withdrawn, which is the traditional method for determining hardness.

The main problem with the depth-monitoring procedure is that the deformation of a substrate by the nanoindenter is a sum of plastic and elastic contributions. In particular, the indentation impression is often depressed elastically below the original surface of the substrate, as shown in Fig. 1. If the total indentation depth z_{tot} were used to calculate the area of contact, then the hardness would be underestimated since the apparent area, calculated according to the indenter geometry, would be too large.

This problem can be overcome by monitoring the relaxation of the load as the indenter is withdrawn. The

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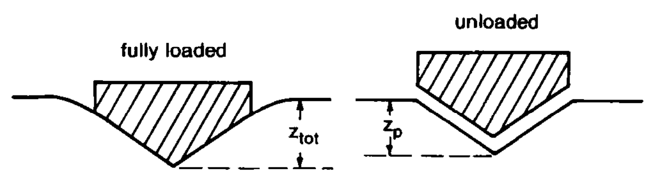

Fig. 1. An illustration of the problems introduced by the elastic sinking-in of an indentation impression under load.

deformation in this case is entirely elastic, and the relaxation can be interpreted [2] using a simple theoretical model derived by Sneddon [3] for a flat cylindrical punch indenting an elastic material. The gradient of the unloading curve in this idealized case is

$$\frac{dP}{dz} = 2\left(\frac{A}{\pi}\right)^{1/2} E_r \qquad (1)$$

where P is the load, A is the area of the punch and E_r is an effective elastic modulus given by

$$\frac{1}{E_r} = \frac{1-\nu^2}{E} + \frac{1-\nu_0^2}{E_0} \qquad (2)$$

where E and ν are Young's modulus and Poisson's ratio of the substrate, and E_0 and ν_0 the corresponding quantities for the punch. Since for homogeneous substrates the right-hand side of eqn. (1) is constant, the relaxation curve is linear for a flat punch.

In the case of a pointed indenter, however, the contact area can decrease as the indenter is withdrawn. This results in a non-linear relaxation curve. However, the flat punch analysis can then be applied locally, and the elastic depression, or "sinking-in", of the indentation can be identified by extrapolating to zero load along the tangent of the relaxation curve at the point of maximum penetration. This is illustrated in Fig. 2(a). The intercept of this line on the z axis is known as the plastic indentation depth z_p. This is precisely what is required to evaluate the contact area under load, and the calculation of the hardness then follows.

This procedure provides a method for obtaining the hardness corrected for the effect of elastic sinking-in of the indentation. In addition, the elastic properties of the substrate can be obtained, by interpreting the relaxation slope using eqn. (1) with the contact area A calculated from z_p. It is not correct to say, however, that the calculated hardness is then always representative of the plastic properties of the substrate. For example, if the indentation is elastic, such that no residual impression is produced, the procedure would still generate a "plastic depth" z_p, and a corresponding hardness. This is illustrated in Fig. 2(b), the correspondence of the loading and unloading curves reflecting the elasticity of the deformation. The unloading curve is entirely elastic relaxation, and the hardness resulting from the apparent value of z_p does not represent the plastic properties of the material. There is thus a danger of misinterpreting results obtained with depth-sensing nanoindenters for very hard materials, and experiments with diamond and diamond-like carbon films illustrate this point [4, 5].

Setting these difficulties aside, however, a further problem arises when using a depth-sensing nanoindenter to characterize the properties of a coated substrate, when the indentation depth is comparable with the thickness of the film. The elastic response of a coated substrate depends on the depth of penetration [6–8] just as the hardness depends on indentation depth [9, 10]. For large elastic depressions, relative to the coating thickness, E_r in eqn. (1) will be given by the properties of the substrate material, whilst for small sinking-in depths the appropriate properties will be those of the coating. The unloading curve procedure, however, assumes that the elastic properties are constant as the indenter is withdrawn, which is not now the case.

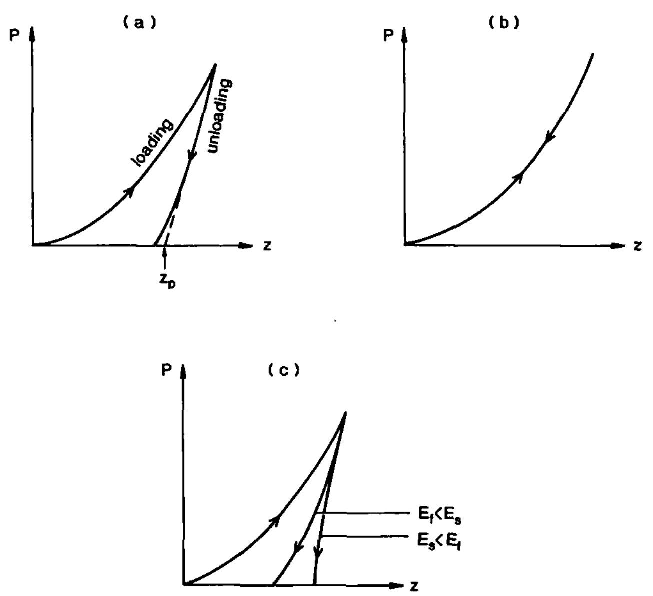

Fig. 2. Typical loading and unloading curves for (a) an elastoplastic material, (b) an elastic material, and (c) an elastoplastic coated substrate.

For a relatively compliant coating on a stiffer substrate, for example, the slope of the unloading curve will decrease during the relaxation, even if the contact area remains constant. Conversely, the slope would increase as the load is reduced for a stiff film on a more compliant substrate. This is illustrated in Fig. 2(c). In such cases, the plastic indentation depth z_p is equal to the actual intercept between the unloading curve and the zero-load axis (as long as the contact area remains constant), and extrapolating along a tangent to the unloading curve is not correct. If the contact area also varies, then the two non-linear effects become entangled.

These difficulties may be overcome if the Sneddon model [3] can be replaced by a model more appropriate to a coated substrate, so that instead of eqn. (1) a non-linear unloading curve is predicted which takes into account the variation in elastic properties. Given the properties of the materials, and the indentation depth, the elastic depression of the indentation might then be estimated more accurately.

The change in elastic behaviour was noted by Doerner and Nix [11], and modelled empirically using eqn. (1) with

$$\frac{1}{E_r} = \frac{1-\nu_f^2}{E_f}\left[1-\exp\left(-\frac{\alpha h}{z_p}\right)\right] + \frac{1-\nu_s^2}{E_s}\exp\left(-\frac{\alpha h}{z_p}\right) + \frac{1-\nu_0^2}{E_0} \tag{3}$$

where the suffices f and s denote substrate and film properties, h is the thickness of the coating and α is a constant chosen equal to 0.25 to obtain the best fit to data for tungsten films on silicon [11]. Using such a model, the elastic properties of the film can be deduced from load relaxation measurements, taking into account any effect of the underlying substrate. Doerner and Nix did not, however, use eqn. (3) to refine the procedure for calculating the hardness from load *vs.* depth curves: the unloading curve was modelled as a straight line [11].

The above empirical description of the elastic properties seems successful, but it would be better to compare it with a theoretically more rigorous model to assess its wider application, in particular, the universality of the coefficient α. King [6] presented an analysis of the indentation of an elastic layered medium which supports eqn. (3) as a representation of the behaviour, and suggests values of α for particular cases. The analysis was, however, complicated and it would be useful to have available a simpler approach which could be used more easily. A recent model of the elastoplastic indentation of coated substrates [10] provides an opportunity for such a development. This approach uses the known elastoplastic deformation in a spherically symmetric coated substrate geometry (a coated cavity) to model the indentation of a coated plane substrate, and predicts a depth-dependent hardness in good agreement with data [10]. An analysis of elastic relaxation in this geometry might similarly provide a model of unloading in the plane substrate situation, and this is addressed in this paper.

In Section 2, such a development is described, and the resulting model is compared with King's approach in Section 3. Conclusions of the study are given in Section 4.

2. Analysis of the coated cavity

We consider a spherical cavity with radius a surrounded by a shell of film material of thickness $t-a$ with Young's modulus E_f and Poisson's ratio ν_f. Outside the film is a substrate material with elastic constants E_s and ν_s. The cavity represents the indentation impression, and the shell thickness $t-a$ is related to the coating thickness in the real plane geometry. Such a geometry provides the framework for the hardness model of coated substrates proposed in ref. 10. We now consider the relaxation of the cavity from an elastoplastic stress state produced by indentation. The depth of the indentation fixes the size of the cavity, and the pressure within the cavity is related to the load placed on the indenter. We focus on the displacement of the inner surface of the cavity as the load is decreased. This will model the elastic relaxation in the plane substrate geometry as the indenter is withdrawn.

We consider the response to a (negative) increment in cavity pressure δp. The change in radial stress at radius r is given by

$$\delta\sigma_r = \frac{A}{r^3} + B \tag{4}$$

for $a < r < t$, and

$$\delta\sigma_r = \frac{D}{r^3} \tag{5}$$

in $r > t$, where A, B and D are constants. The elastic displacement in $a < r < t$ is

$$\delta u = -\frac{A(1+\nu_f)}{2E_f r^2} + \frac{Br(1-2\nu_f)}{E_f} \tag{6}$$

and in $r > t$ it is given by

$$\delta u = -\frac{D(1+\nu_s)}{2E_s r^2} \tag{7}$$

Demanding continuity in radial stress and displacement at $r = t$, and using the stress boundary condition at $r = a$, we obtain

$$-\delta p = \frac{A}{a^3} + B \tag{8}$$

$$\frac{A}{t^3} + B = \frac{D}{t^3} \tag{9}$$

and

$$-\frac{A(1+\nu_f)}{2E_f t^2} + \frac{Bt(1-2\nu_f)}{E_f} = -\frac{D(1+\nu_s)}{2E_s t^2} \tag{10}$$

Solving these equations, and using eqn. (6) to give δa, the incremental displacement at the inner surface of the cavity, we obtain

$$\delta a = a\delta p \frac{\begin{array}{l}(1+\nu_f)/2E_f\,[(1+\nu_s)/2E_s + (1-2\nu_f)/E_f] \\ \quad + [(1-2\nu_f)/E_f]\,(a/t)^3 \\ \quad \times [(1+\nu_s)/2E_s - (1+\nu_f)/2E_f]\end{array}}{\begin{array}{l}(1+\nu_s)/2E_s + (1-2\nu_f)/E_f \\ \quad - (a/t)^3\,[(1+\nu_s)/2E_s - (1+\nu_f)/2E_f]\end{array}} \tag{11}$$

which gives the correct limiting cases for $t = a$, $t \to \infty$, or $E_f = E_s$ and $\nu_f = \nu_s$ [12], corresponding to substrate only, film only, and identical film and substrate properties respectively.

3. Application of the model

The solution obtained for the coated cavity geometry must now be applied to the real situation of a coating on a plane substrate. At this stage, rigour is lost, since there are many ways of drawing an analogy between the two geometries. The assumption that the coated cavity solution is relevant to the real case will be tested by comparing the results with observed behaviour.

The parameters a and t in eqn. (11) must be related to corresponding dimensions in the plane substrate geometry. In the coated cavity hardness model [10], the cavity volume is equated with the indentation volume, so that

$$a = z_p\left(\frac{2}{\pi}\tan^2\phi\right)^{1/3} \tag{12}$$

for a pyramidal indenter, with z_p the depth of plastic indentation and ϕ the semi-included angle between faces of the pyramid. Also, the parameter t is given by

$$t = a + h \sin\phi \tag{13}$$

which completes the parametrization.

The best way to demonstrate the effect of the film on the elastic relaxation behaviour is to compare the slope of the unloading line in the coated case to the slope in the absence of the film, for a range of indentation depths. Defining $S = \mathrm{d}p/\mathrm{d}a$ we have

$$\frac{S}{S_0} = \frac{\begin{array}{l}(1+\nu_s)/2E_s + (1-2\nu_f)/E_f \\ \quad - (a/t)^3\,(1+\nu_s)/2E_s - (1+\nu_f)/2E_f\end{array}}{\begin{array}{l}[E_s(1+\nu_f)]/[E_f(1+\nu_s)] \\ \quad \times [(1-\nu_s)/2E_s] + [(1-2\nu_f)/E_f] \\ \quad + [2E_s(1-2\nu_f)]/[E_f(1+\nu_s)] \\ \quad \times (a/t)^3\,[(1+\nu_s)/2E_s - (1-\nu_f)/2E_f]\end{array}} \tag{14}$$

where S_0 is the unloading slope in the absence of the film. Inserting values for a and t according to eqns. (12) and (13) produces our final result.

This should be compared with the corresponding result derived from eqn. (1) and the empirical expression eqn. (3) suggested by Doerner and Nix [11]. They define S as $\mathrm{d}P/\mathrm{d}z$, but the ratio S/S_0 is directly comparable with eqn. (14). For $E_0 \to \infty$, corresponding to a rigid indenter, the result is

$$\frac{S}{S_0} = \frac{\dfrac{1-\nu_s^2}{E_s}}{\begin{array}{l}[(1-\nu_f^2)/E_f][1-\exp(-\alpha h/z_p)] \\ \quad + [(1-\nu_s^2)/E_s]\exp(-\alpha h/z_p)\end{array}} \tag{15}$$

which was shown by King [6] to be a good representation of the elastic behaviour of coated plane substrates indented by flat punches, obtained from calculations for the special case of $\nu_s = \nu_f = 0.3$.

The two expressions, eqns. (14) and (15), are compared for a range of E_f/E_s and various z_p/h in Fig. 3 for $\nu_s = \nu_f = 0.3$, using $\phi = 68°$. Unfortunately, the coated cavity solution seriously overestimates the unloading slope in all cases. This is not, however, due to the parametrization (eqns. (12) and (13)) unless these expressions were made to include an unlikely dependence on E_f/E_s. The coated cavity solution seems unable to describe the behaviour.

Why should the coated cavity solution for the elastic relaxation fail when the corresponding elastoplastic loading solution [10] is successful? Clearly, the spherical geometry must be inappropriate for a description of the elastic displacements in the plane substrate case. The description might have succeeded if the relaxing elastic displacements in the indented plane surface situation were largely radial relative to the centre of the indentation. However, the elastic deformation we are attempting to represent is in fact the relaxation of the sinking-in of the indented impression, as shown in Fig. 1, and these displacements are mainly normal to the surface of the film. We are forced to conclude that the cavity model should not be pursued too far in attempting to model the process of indentation, and it is unrealistic to expect it to model elastic relaxation.

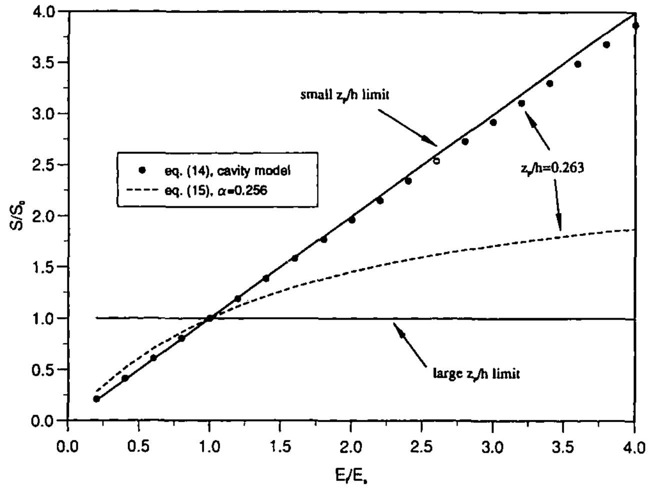

Fig. 3. Comparison of elastic unloading slopes according to the coated cavity model (eqn. (14)) and calculations by King [6] (represented by eqn. (15)), for a range of ratios E_f/E_s and various z_p/h values, for $\nu_s = \nu_f = 0.3$. S_0 is the slope in the absence of a film.

4. Conclusions

By monitoring the load–displacement curve for penetration into a substrate, depth-sending nanoindenters can measure both the hardness and elastic properties of the material. The total indentation depth, however, has to be corrected for the elastic sinking-in of the indentation impression, in order that the true area of contact under load can be deduced and the hardness calculated. The procedure for doing so uses the unloading curve, and a procedure for correcting for the elastic effect has been developed [2], based on a simple analysis for a flat punch indenting a homogeneous elastic substrate, which predicts a linear unloading curve. The unloading behaviour can also be used to determine the elastic properties. The method can be misleading, especially for very hard materials, but is successful in most applications, and is much more efficient than measuring the indentation impression visually.

Nanoindenters are very suitable for studying coated substrates, since the depth of penetration can be less than the film thickness, allowing the properties of a thin coating to be studied. The elastic response of coated substrates is depth dependent, however, which means that the correction of the indentation depth for elastic sinking-in must be accomplished using a non-linear unloading curve. It does not appear that a non-linear analysis is applied in practice, and this may introduce errors in the calculated hardness. Also, the interpretation of the unloading curve to obtain the elastic properties of the coating, correcting for any effect of the substrate, is made more difficult.

An empirical approach to modelling the variation in elastic properties exists [11], supported by a partly numerical elastic analysis [6], but in this paper a simpler theoretical model has been investigated, based on the stress analysis of a coated cavity in an elastic medium. The model is ultimately unsuccessful, but the approach might have yielded a reasonable description of the behaviour if the elastic displacements in the indented plane substrate geometry had been approximately radial. This is the presumed reason why a similar approach is more successful in describing large-strain elastoplastic indentation [10]. In the present case, it is probably a poor approximation, since the relevant elastic displacements are largely normal to the substrate surface. This is reflected in a disagreement between the slopes of the load relaxation curves predicted by the present model and a more accurate, though more complicated, analysis due to King [6]. The interpretation of relaxation curves for coated substrates is therefore probably best approached using the numerically generated solutions given by King, in the absence of a simpler model.

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