ELSEVIER

Surface and Coatings Technology 131 (2000) 457–461

SURFACE & COATINGS TECHNOLOGY

www.elsevier.nl/locate/surfcoat

A model for coated surface hardness

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Abstract

The authors present a model that predicts the hardness of a film deposited on the surface of a substrate. The model is based on a simple mathematical relationship involving the hardness of the film and the hardness of the substrate, together with the hardness of the composite film/substrate at a given film thickness/penetration depth ratio. The parameters of the present model depend on the mechanical properties of the film/substrate pair and can be experimentally determined by using depth-sensing hardness instruments. In the models found in the literature, the equation parameters are constants typical for each model. The present model is proposed and validated for the case of a film (W–C–Co coating) harder than the substrate (various metallic materials) and it allows determination of the film hardness without the need to determine either the film thickness or the substrate hardness.

Keywords: Thin films; Indentation; Hardness; Modelling

1. Introduction

The characterization of the mechanical behaviour of thin coatings is often made using indentation hardness tests. If the ratio of the indentation depth to the thickness of the film is small enough, the observed behaviour is only due to the properties of the coating. In many cases, most experimental apparatus, including ultramicrohardness tester, are not sufficiently sensitive to allow the needed accuracy in the load and penetration measurements to assess only the behaviour of the coating. To overcome this, higher load hardness tests should be used, on the condition that the contributions of substrate and thin film in the final result can be conveniently separated.

A large number of models have been used for this purpose [1–19]. Most of them can be summed up by the following general equation:

$$\frac{H_c - H_s}{H_f - H_s} = F\left(\frac{t}{h}\right) \quad (1)$$

where H_c, H_s and H_f are the composite, substrate and film hardnesses, respectively, t is the thickness of the film and h is the depth of the indentation. F is a function that depends on the type of mathematical model; its parameters are related to the properties of the film and the substrate materials, size and geometry of the deformed region, etc.

In a recent work by Fernandes et al. [19], the kinematic model was applied to the case of a spherical cavity problem in order to undertake a theoretical study of the mechanical behaviour of a bilayer, during the hardness test. The authors concluded that the size of the plastically deformed region and its evolution during the hardness test strongly influences the behaviour of the composite, this being the reason why the former models fail, in general, to produce reasonable

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0257-8972/00/$ - see front matter
PII: S0257-8972(00)00839-2

results because they do not give any guarantee for the values of their characteristic parameters, which are not specific to each film/substrate pair. The formulation of an appropriate model must, directly or indirectly, take into consideration that, during the indentation test of a material, the size of the plastically deformed region depends on the elastic and plastic properties of the material under indentation.

In the present research, a model to predict the hardness of the film from the hardness value of the composite was developed and validated for the case of W–C–Co films on six different softer substrates. In the model, the values of the parameters are not imposed a priori, but they can be determined experimentally, from the results obtained using a depth sensing indentation apparatus.

2. A hardness model for the composite film / substrate

The formulation of the present model is based on the following two hypotheses.

The composite follows a simple behaviour described by the linear equation (the validity of such a linear relationship has been discussed elsewhere [20–22]):

$$\frac{H_c - H_s}{H_f - H_s} = A\left(\frac{t}{h_D}\right) + B \tag{2}$$

where H_c, H_s and H_f are the composite, substrate and film hardnesses, respectively, t is the thickness of the film and h_D is the plastic penetration depth of the indentation. A and B are equation parameters that depend on the film and substrate properties.

The condition $(H_c - H_s)/(H_f - H_s) = 1$ defines the critical value of the indentation depth $(h_D)_C$, above which the substrate starts to deform plastically, for a given thickness t of the film. For a given film/substrate pair, it is possible to determine the value of $(h_D)_C$, which is a necessary condition in the determination of the values of the parameters A and B.

Using Eq. (2) and handling three pairs of $(H_c;\ h_D)$ values, $(H_{c1};\ h_{D1})$, $(H_{c2};\ h_{D2})$ and $(H_c = H_f;\ h_D = (h_D)_C)$ (these values correspond to the equality $(t/h_D) = (t/h_D)_C$, which is verified for $(H_c - H_s)/(H_f - H_s) = 1$) which can be obtained experimentally by performing two indentation tests at two different loads, the value of H_f can be written as follows:

$$H_f = \frac{H_{c2}[h_{D1} - (h_D)_C]h_{D2} - H_{c1}[h_{D2} - (h_D)_C]h_{D1}}{[h_{D1} - (h_D)_C]h_{D2} - [h_{D2} - (h_D)_C]h_{D1}} \tag{3}$$

Similarly, the parameters A and B on Eq. (2) can be determined using the following equations:

$$\begin{cases} A\left(\dfrac{t}{h_D}\right)_C + B = 1 \\ \dfrac{A}{B} = \left(\dfrac{1}{t}\right)\dfrac{h_{D1}h_{D2}(H_{c1} - H_{c2})}{h_{D2}(H_{c2} - H_s) - h_{D2}(H_{c1} - H_s)} \end{cases} \tag{4}$$

The possibility given by Eq. (3) of determining the film hardness by carrying out indentation tests in the composite, using only two nominal load tests, without the need to know the film thickness and the substrate hardness is very useful from a practical point of view. The two nominal load values to be used must be sufficiently separated (for example, corresponding to the values of $(H_c - H_s)/(H_f - H_s)$ close to 0.05 and to 0.15).

3. Experimental details

Vickers hardness tests were performed on several coating/substrate systems. Six different substrates were used: 1-mm-thick sheet samples of copper in annealed condition (*ACu* samples), mild steel, rolled up to 80% reduction (*RS* samples), and *AISI M2* steel samples, submitted to four different heat treatments in order to obtain different hardness values (Table 1). The *AISI M2* sample *A (M2A)* was used in an as-received annealed condition. The other three samples *M2B*, *M2C* and *M2D* were water-quenched from 1220°C and triple tempered, respectively, at 550, 600 and 650°C for 2 h. The W–C–Co amorphous coating used was obtained by r.f. sputtering, from a WC + 15 wt.% Co target [23]. The thickness of the films measured by scanning electron microscopy (SEM) observations of the cross-section of the coated samples, is for all the *M2* samples 1.3, 2.5, 5.2 and 8.2 μm; whereas for the copper and deformed mild steel samples is 2.2, 4.4 and 6.6 μm. As previously stated [24], SEM examinations of indentation geometry did not show any fracture morphology.

Table 1
Hardness values H_s at maximum load (1 N) for all the studied substrate samples[a]

Material	ACu	RS	M2A	M2B	M2C	M2D
H_s (GPa)	1.00	2.25	3.50	5.40	7.60	9.00
H_f/H_s	18.50	8.22	5.29	3.43	2.43	2.05

[a] The corresponding ratios between the film and the substrate hardness (H_f/H_s) are also indicated.

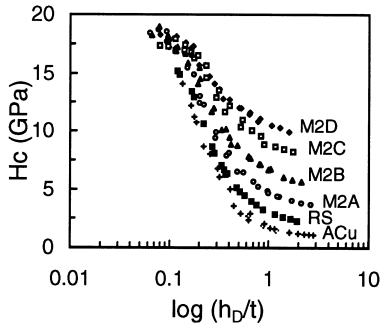

Fig. 1. The hardness of the composite as a function of the penetration depth normalized to the thickness (h_D/t).

Each hardness value is a result of at least five indentation tests performed in a Fisherscope H100 depth sensing ultramicrohardness testing system. When using a depth sensing indenter instrument, the determination of the contact area (or penetration depth) is obtained from the unloading part of the curve [25–29]. A calibration method based on the comparison between depth sensing results and the optical measurement of the diagonals (D) of the remaining indentations after unloading was applied to take into account the imperfect shape of the tip of the diamond Vickers pyramidal indenter, and the presence of an extruded lip adjacent to the indentation [30].

4. Results and validation

For each coating/substrate system, the data for the hardness of the composite, H_c, were obtained as a function of h_D, normalized to the unity of thickness t (Fig. 1). The hardness of the film is assumed to be 18.5 GPa by considering the value of the hardness for small indentations for which no contribution of the substrate to the hardness of the composite is expected. So, the ratio between the hardness of the film H_f and the hardness of the substrate H_s is within the range $2.05 < H_f/H_s < 18.5$ (Table 1).

The experimental results were analysed by plotting $(H_c - H_s)/(H_f - H_s)$ as a function of (t/h_D), as shown in Fig. 2. For each substrate, the behaviour can be approximately represented by a straight line for $(H_c - H_s)/(H_f - H_s)$ in the range 0.05 and 0.9, in agreement to the previously proposed Eq. (2).

The validation of the method is made by comparing the critical penetration depth, the parameters A and B, the hardness curves and the error in the calculation of the film hardness as determined from the experimental results and by using the model.

4.1. Determination of the critical penetration depth $(h_D)_C$

The application of the model requires the previous

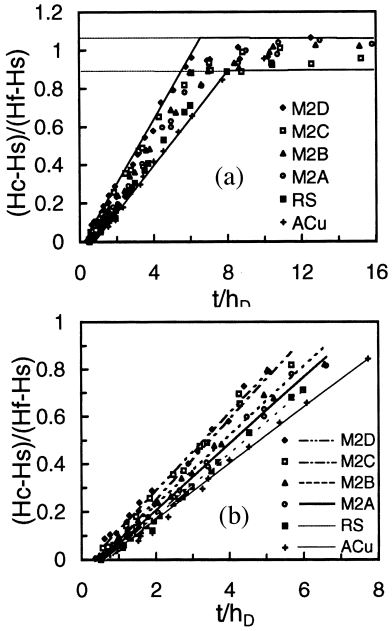

Fig. 2. The ratio $(H_c - H_s)/(H_f - H_s)$ as a function of (t/h_D) for all the composites studied in this work: (a) all the results are shown; (b) linear fitting of the data within the range $0.05 < (H_c - H_s)/(H_f - H_s) < 0.9$.

calculation of the critical penetration depth $(h_D)_C$. An expeditious method to determine its value from the loading part of the indentation curves is presented elsewhere [22]. Briefly, this method takes into account the Meyer type equation [25,26]: $P = kh^n$, where k and n are constants that depend on the elastic and plastic properties of the material. For Vickers indentation of composite materials such as thin films on a substrate, the values of k and n must evolve from those typical of the film, for low load values, to those characteristic of the substrate for very high loads [31]. The critical depth $(h_D)_C$ can be evaluated from the deviation to linearity on the loading part of either the curve $\log(P) = \log(h)$ or the plot $(dP/dh) = f(h)$ [32,33]. In both cases, (h_C) indicates the value for which plastic deformation starts

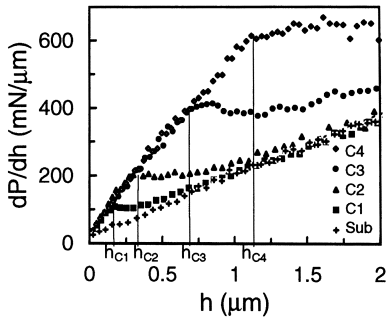

Fig. 3. Differential load $(dP/dh) \approx (P_{i+1} - P_i)/(h_{i+1} - h_i)$ as a function of $h = (h_{i+1} + h_i)/2$ for the thickness of 1.3 μm (h_{C1}), 2.5 μm (h_{C2}), 5.2 μm (h_{C3}) and 8.2 μm (h_{C4}). The case considered concerns the substrate *M2A*. The determination of the values of h_C is indicated.

Table 2
Values of $t/(h_D)_C$ considering that the critical value of the plastic depth $(h_D)_C$ was determined under the following conditions: fitting Eq. (2) to the experimental results and determining $(h_D)_C$ from h_C, after indentation geometry and elastic corrections

Material	ACu	RS	M2A	M2B	M2C	M2D
$t/(h_D)_C$ exp.	8.80	8.06	7.66	7.27	6.94	6.54
$t/(h_C)_C$	8.06	7.87	7.87	7.18	6.75	6.35

Table 3
Values of A and B determined under the following conditions: fitting Eq. (2) to the experimental results and determining $(h_D)_C$ from h_C, after indentation geometry and elastic corrections

Case	Material	ACu	RS	M2A	M2B	M2C	M2D
$(h_D)_C$	A	0.125	0.136	0.141	0.147	0.148	0.157
exp.	B	−0.095	−0.093	−0.076	−0.066	−0.030	−0.027
$(h_D)_C =$	A	0.129	0.136	0.132	0.146	0.152	0.158
$(h_C)_C$	B	−0.075	−0.070	−0.040	−0.050	−0.027	−0.005

in the substrate. Fig. 3 shows an example of determination of the critical depth from the plot $(dP/dh) = f(h)$. This value must be corrected by extracting the contribution of the elastic component h_c ([27,28], for example) and by taking into account the geometrical defects of the indenter and the formation of the extruded lip [25–27,30].

Table 2 presents $t/(h_D)_C$ values determined in different ways:

1. Directly, by fitting Eq. (2) to the experimental results for each film/substrate pair (Fig. 2) and calculating t/h_D for $(H_c - H_s)/(H_f - H_s) = 1$ (H_f = 18.5 GPa).
2. By determining the (h_C) value from the loading part of the loading depth curve, as above explained, and correcting this value to $(h_C)_C$.

Table 2 confirms a very good correlation between the critical depth values $t/(h_C)_C$ and $t/(h_D)_C$, calculated using both methods.

4.2. Determination of the parameters A and B of the model

Using Eq. (4) and two pairs of values $(H_{c1}; h_{D1})$ and $(H_{c2}; h_{D2})$, the parameters A and B can be calculated for each substrate case. As referred to above, among all the experimental values $(H_c; h_D)$, the two pairs were selected for values of $(H_c - H_s)/(H_f - H_s)$ close to zero and within the range (0.20–0.30). As shown in Table 3, there is a good agreement between the values of A and B calculated by this method and by plotting the experimental results, as shown in Fig. 2b, and fitting a straight line to each film/substrate pair $(h_D)_C$.

4.3. Comparison between the experimental and model hardness curves

In Fig. 4 the experimental results are compared with the predictions of the model obtained using two hardness tests (the same as mentioned above) and considering the values of $(h_C)_C$. A good correlation between the analytical values and the experimental results is obtained.

4.4. Evaluation of the error on the calculation of the hardness of the film

The error on the determination of the value of H_f was calculated, for values of $(H_c - H_s)/(H_f - H_s)$ close to 0.25 and 0.10. In the first case, the error was less than 5%, except for the two softest substrates (*ACu* and *RS*), for which the error can be as high as 10%, whereas in the other case, the error values are approximately 10% and 20%, respectively.

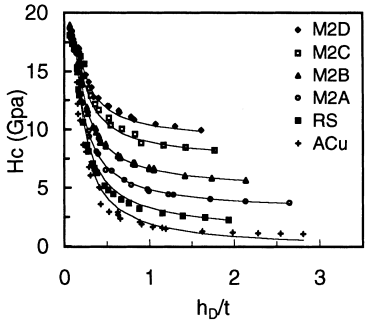

Fig. 4. Comparison between the experimental results and model predictions [Eq. (2)]. The constants A and B were determined, using Eq. (4), after indentation geometry and elastic corrections. See text for details.

5. Conclusion

A model to predict film hardness when performing hardness tests on a composite material is presented. This model allows the determination of an accurate value of the hardness of a hard film on a soft substrate, without the need to previously determine the thickness of the film and the hardness of the substrate. Using a depth sensing hardness instrument, the following procedure is used to apply the model:

1. To perform two different nominal indentation loads and determining the respective pairs (hardness, H_c; residual indentation depth, h_D) calculated from the experimental data: (H_{c1}; h_{D1}), (H_{c2}; h_{D2}).
2. To determine the critical indentation depth (h_C) from the loading part of either curves $\log(P) = \log(h)$ and/or $(dP/dh) = f(h)$. The deviation from linearity indicates the (h_C) value.
3. To extract the elastic contribution of (h_C) and perform the usual correction of the indenter geometry (as proposed by Oliver and Pharr [25] or using some other method, [30] for example) allowing to obtain the correspondent residual indentation depth.
4. To use Eq. (4) for determining the hardness of the film H_f.

Moreover, it is possible to determine the parameters A and B of the equation that formalize the model [Eq. (2)]. The correct determination of these parameters is the main difficulty in former models, which do not directly or indirectly take the mechanical properties of the substrate and the composite into account. When compared with other models, the present one can be easily adapted to different film/substrate pairs and a high level of accuracy can be obtained.

Acknowledgements

The authors are indebted to Fundação para a Ciência e a Tecnologia do Ministério da Ciência e Tecnologia and Program PRAXIS XXI, for financial support. A grant to A. Cavaleiro from Fundação Oriente is also acknowledged.

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