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Comments on the paper "Modification of composite hardness models to incorporate indentation size effects in thin films", D. Beegan, S. Chowdhury and M.T. Laugier, Thin Solid Films 516 (2008), 3813–3817

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A B S T R A C T

Since the original work of Bückle concerning the substrate influence on the hardness measurement of thin film, more than 20 models were proposed to separate the contribution of the substrate. Subsequently to the development of these numerous models, a question arises: Which is the most relevant models among them? Indeed, the authors usually consider that their proposed model leads to the best prediction of the film hardness which is probably correct for a given experimental condition applied to a particular material. In addition, the authors also assume that the other models are not so relevant. But to have a sound discussion about the existing models, it is necessary to correctly apply them according to the author statement. In this paper, we better specified the application of the Jönsson and Hogmark model and that of Chicot and Lesage applied to the results obtained on copper films by Beegan et al. Contrarily to these authors, we show that the above-mentioned models lead to a good representation of the experimental data and a good predicted value of the film hardness.

1. Introduction

In a recent paper [1], Beegan et al. have studied the indentation behaviour of copper films deposited on oxidised silicon substrates in various conditions of deposition leading to film thickness ranging from 25 to 1400 nm. The authors have compared Jönsson and Hogmark (J–H) [2], Burnett and Rickerby (B–R) [3], Chicot and Lesage (C–L) [4], Korsunsky (K) [5] and Puchi-Cabrera (P-C) [6] models with the objective to fit the entire range of the experimental data. The authors concluded that both Korsunsky and Puchi-Cabrera models allow well predicting the hardness of the film since they give an adequate fitting of the composite hardness data variation. On the other hand, the Jönsson–Hogmark and Chicot–Lesage models are both obviously not able to fit the composite hardness data and consequently are not usable to predict the film hardness.

The aim of the present work is to demonstrate that author's conclusions concerning applicability of the Jönsson–Hogmark and Chicot–Lesage models are erroneous. For that, we show that the difference between the fitted curves deduced from application of the two models and the corresponding experimental data is due to a wrong application of these models by Beegan et al.

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Furthermore, additional problem often occurs when studying hardness evolution, *i.e.* the indentation size effect (ISE) which traduces the load-dependence of the nano and micro-hardness. This phenomenon has been associated with various causes such as work hardening, roughness, piling-up, sinking-in, shape of indenter, surface energy, varying composition and crystal anisotropy, which have been all discussed extensively by Cheng and Cheng [7]. We show that both the J–H and C–L models may be also improved by incorporating the ISE effect taking into account a linear relation between hardness and the reciprocal indentation depth.

2. Hardness models

The models applied by Beegan et al. to represent the composite hardness in relation with the film hardness (H_f) and the substrate hardness (H_s) are described by these authors in [1,8]. All these models may be stated as:

$$\frac{H_c - H_s}{H_f - H_s} = a \tag{1}$$

with:

$$a = \frac{A_f}{A} = 1 - \frac{A_s}{A} = 2\frac{C}{\beta} - \frac{C^2}{\beta^2} \tag{2}$$

for the J–H model, where A_f and A_s represent respectively the area on which the mean pressure H_f or H_s acts, $A_s+A_f=A$, C is a constant depending on the indenter geometry and the way that the coating deforms itself under the indenter, and β is the relative indentation depth h/t,

$$a = \frac{3ct}{2h}\left[\left(\frac{H_f}{E_f}\right)^{1/2} + \left(\frac{H_s}{E_s}\right)^{1/2}\right]\tan^{1/3}\phi \tag{3}$$

for the C–L model, where E_f and E_s are respectively the Young modulus of the film and of the substrate, c a constant and ϕ is the indenter semi-angle,

$$a = 1 + k\beta^2 \tag{4}$$

for the K model, where k is a constant related to the film thickness

$$a = \exp(-k\beta^n) \tag{5}$$

for the P-C model, where k and n are empirical material parameters ($n=1$ for the specimen under study).

3. Experimental results

3.1. Jönsson–Hogmark model

Beegan et al. [1] applied the Jönsson–Hogmark model to various copper film thickness ranging from 25 to 500 nm, deposited on oxidised silicon substrates. They obtained the fitting curve presented on Fig. 1 (thick grey line) to minimise the root mean square deviation (rms) between modelled and measured hardnesses. From this figure, the authors have concluded that this model is unable to adequately describe the hardness evolution of the coated materials.

Regarding Fig. 1, this conclusion seems obvious, but the curve plotted by Beegan et al. does not respect the physical signification of the J–H model. From Eq. (2) the parameter, a, varies from 1, when only the film hardness is measured ($A_f=A$; $A_s=0$), to 0, when the thickness of the film is negligible compared to the indentation depth ($A_f=0, A_s=A$) as reported by [8]. As a consequence, it must be stated that, $C/\beta=Ct/h$ from Eq. (2) must vary between 0 and 1. That is to say that C/β higher than 1 implies that $A_f=A$. Then for an indentation depth, h, lower than the product Ct, the composite (or experimental hardness) is equal to the film hardness, *i.e.* $H_c=H_f$ if $h\leq Ct$.

In order to verify the manner with which the model is applied by [1], the ratios, A_f/A and A_s/A, of the J–H area law of mixture are calculated and the corresponding functions are represented in Fig. 2 as a function of the normalised indentation depth.

In Beegan et al. approach, the influence of the substrate on the composite hardness represented by A_s/A (the dotted grey line in Fig. 2) decreases as the normalised depth decreases, and reaches a minimum (0) as $\beta=C$. At this point $A_f/A=1$ and the indentation pressure is completely supported by the copper film. Then, when the indentation depth decreases again, A_s/A increases and the substrate influence becomes more and more predominant. Furthermore, the film area which supports the indentation pressure becomes negative for indentation depth lower than $Ct/2$ (*i.e.* $\beta<C/2$). Obviously these variations have no physical meaning.

The correct representation of the J–H model consists in using parameter having physical signification as earlier highlighted by Iost and Bigot in [9] and may be considered as:

$$\begin{cases} \text{if } \beta\geq C \; a = \dfrac{A_f}{A} = 1-\dfrac{A_s}{A} = 2\dfrac{C}{\beta}-\dfrac{C^2}{\beta^2} \\ \text{if } \beta<C \; a = 1 \;\; \text{then} \;\; H_c = H_f \end{cases} \tag{6}$$

The results corresponding to the J–H model, applied while respecting the physical signification of the area law of mixture, are shown on Fig. 1 (black curves) for the composite hardness and in Fig. 2 for the two area ratios, *i.e.* A_f/A and A_s/A. The constant C and the predicted values for the film and the substrate hardnesses are also collected in Table 1. In practice, the parameter C is considered as a constant for the different film thicknesses since it only depends on the nature of the film. This constant is 0.1746 for a Berkovich diamond indenter for a hard film plastically deformed under indentation, deposited on a soft substrate. The physical signification is that the substrate begins to have influence on the composite hardness when the normalised depth reaches 0.1746 that is less restrictive that the 10% rule from Bückle [10,11]. A greater value for this constant suggests that the substrate influences the hardness measurement only for higher indentation depth as it was observed earlier for paint [12] or porous [13] coatings. This phenomenon only occurs for soft films deposited on hard substrates. As an example, for very soft films comparatively to the substrate (*e.g.* Al/Si), the plastic deformation may totally occur in the film when the indentation depth is lower than the film thickness (*i.e.* $\beta<1$) and the load required for the substrate to yield is reached only when the indenter tip is located at the film/substrate interface [14]. On the other hand, by applying the constant modulus assumption for determining film hardness of Al films deposited on glass, the same authors observed that the hardness is approximately constant as a function of depth until the indentation depth is equal to 75% of the film

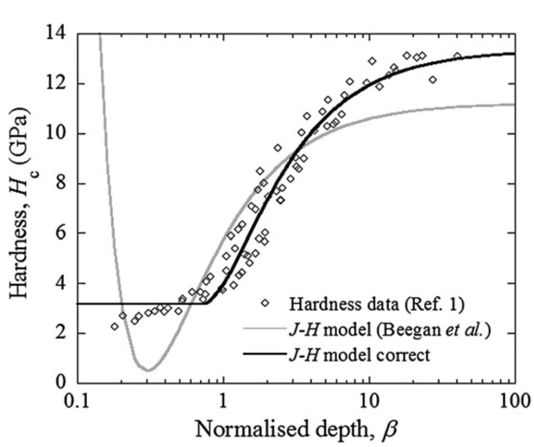

Fig. 1. Variation of hardness with normalised depth for copper films 25 to 500 nm thick. The thick grey line illustrates the JH model as applied by Beegan et al. [1] and the thick black line illustrates the JH model accurately applied.

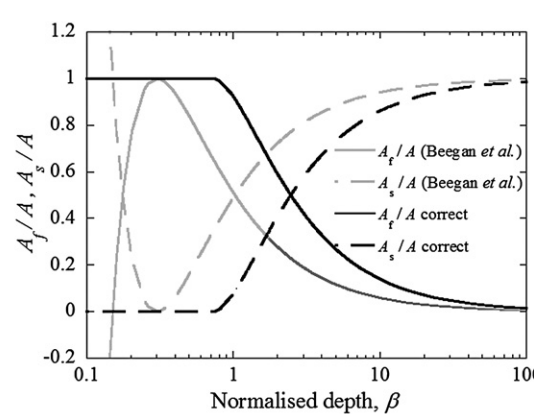

Fig. 2. Area ratios evolutions in the J–H model according to Beegan et al. [1] (grey lines) and Jönsson–Hogmark model correctly applied (black lines) for copper films 25 to 500 nm thick.

Table 1
Predicted values for copper films 25 to 500 nm and substrate hardnesses from the fitting of composite hardness models to the hardness data obtained.

Parameters	JH (Ref. 1)	JH (This work)	C–L (Ref. 1)	C–L (This work)	K (Ref. 1)	P-C (Ref. 1)
H_s (GPa)	11.23	13.36	9.32	12.77	12.64	12.73
H_f (GPa)	0.49	3.18	3.04	3.38	2.76	1.46
Constant	0.30	0.72	0.34	1.075	0.19	0.34
rms (GPa)	1.51	0.71	2.37	0.84	0.69	0.69

Ref. 1 referred to the results obtained by Beegan et al. [1], and "This work" to the correct application of the J–H and C–L models.

thickness (*i.e.* $\beta = 0.75$). Afterwards, the hardness starts to increase when the depth of penetration increases. Chen et al. [15] using the finite element method demonstrate that when the indentation depth is less than 50% of the film thickness (*i.e.* $\beta < 0.5$) for a soft film deposited on a hard substrate, one may directly obtain the film hardness. For these reasons we consider that 0.72 obtained for the constant β is of the same order of magnitude.

The hardness obtained for the thin copper film is higher than the value obtained by Beegan et al. [1] using K or P-C model (Table 1) but it is close to the value of 3 GPa reported by the same authors in previous works related to the same experiments [16,17]. Moreover the effectiveness of the model deduced of the root mean square is of the same order of magnitude than that obtained by the K and P-C methods.

3.2. Chicot–Lesage model

Beegan et al. [1] have also applied the C–L model (Eq. (3)) to determine the film hardness of the same various copper films thicknesses following the same methodology when applying the model of Jönsson and Hogmark. The authors obtained the fitting curve presented on Fig. 3 (thick grey line). This result is clearly not in favour of the model of Chicot and Lesage since the error associated to the application of this model is very consequent (Table 1).

Regarding Fig. 3, the model of Chicot and Lesage seems to be unable to reproduce the hardness evolution of this coated material. We propose to apply the model considering that the composite hardness is equal to the film hardness when the parameter a, in Eq. (3) is equal or is higher than 1.

Fig. 4 represents a and $1{-}a$ evolutions as a function of the normalised indentation depth. This figure clearly shows that these values were

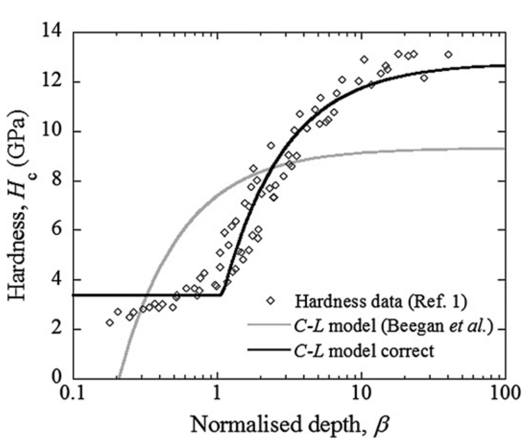

Fig. 3. Variation of hardness with normalised depth for copper films 25 to 500 nm thick. The thick grey line illustrates the C–L model as applied by Beegan et al. [1] and the thick black line the C–L model accurately applied.

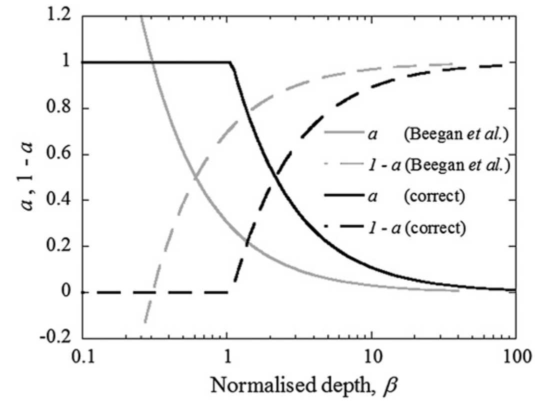

Fig. 4. a and $1{-}a$ evolutions in the C–L model according to Beegan et al. [1] (grey lines) and correctly applied (black lines) for copper films 25 to 500 nm thick.

equal to 1 and 0 for the film and the substrate, respectively, when the model is adequately applied. As a direct consequence, Fig. 3 confirms the good representation of the experimental composite hardness by the C–L model. Therefore the C–L (thick black line) model is clearly validated to predict both film and substrate hardness that it would be expected for the data supplied by Beegan et al. [1].

4. Indentation size effect

For the experimental data reported here-above, a strong indentation size effect (ISE) is observed for the copper film hardness at low indentation depth. Beegan et al. [1] interpret the ISE using the strain gradient plasticity theory (SGP) [18,19] which links the micro-scale dislocations theory to the meso-scale plastic strain gradient:

$$H = H_{0f}\sqrt{1 + \frac{h_f^*}{h_c}} \tag{7}$$

where H_{0f} is the true hardness corresponding to an infinite penetration depth, h^* a material characteristic length and h_c the contact depth.

By incorporating Eq. (7) in the two K and P-C composite hardness models they obtained:

$$H_c(K) = H_{0s} + \frac{H_{0f}\sqrt{1 + \frac{h_f^*}{\beta t}} - H_{0s}}{1 + k\beta^2} \tag{8}$$

$$H_c(P{-}C) = H_{0s} + \left(H_{0f}\sqrt{1 + \frac{h_f^*}{\beta t}} - H_{0s}\right)\exp(-k\beta) \tag{9}$$

We think that the strain gradient plasticity theory is not sufficient to explain the ISE for the experiments into consideration. The main reason is given by Beegan et al. [16,20] mentioning a piling-up around the indent. Indeed the Oliver and Pharr [21] model used by the authors to compute the indentation hardness can only be used when the indentation stress does not produce plastic pile-up. If the pile-up is not accounted in the contact area calculation, the hardness is overestimated. Careful observations of the true contact area supporting the applied load (included the pile-up) were performed by Beegan et al. [16,20]. When the copper film thicknesses are equal to 950 nm and 1400 nm these authors observed that the indentation hardness decreases as a function of the indentation depth before the substrate interferes in the hardness measurement. The size of these pile-up increases proportionally with increasing the penetration depth and the true hardness is constant for loads higher than 20 mN. Since pile-up modifies the contact area between the indenter and the copper film, we propose then to model the hardness evolution of the copper film, H_f, using a usual linear evolution:

$$H_f = H_{0f} + \frac{B_f}{h} \tag{10}$$

The physical basis of such a variation of indentation hardness with the applied load (and consequently with the indentation depth) is related to the previous work from Chaudhri and Winter [22]. The authors have shown that the piled-up region of a hardness indentation in metals supports the indenter load, and that the normal pressure is distributed uniformly over the projected area of the indentation. Eq. (10) was first empirically introduced by Bernhardt [23] and afterwards by Vingsbo et al. [24] to improve the J–H model by taking into account the indentation size effect for both the coating and the substrate. This empirical relation was justified by a simple geometrical model of the deformation of the material around the impression which is connected to the diagonal correction [25]. This is verified for steels, copper, thick hard chromium [25] or titanium and aluminium alloys [26] for Vickers indentation. Eq. (10) may also be obtained by considering the proportional specimen resistance of Li and Bradt [27] or the work of indentation [28]. Furthermore, the ISE of the copper coating cannot be explained only by Eq. (7) since the amount of pile-up is enhanced by the presence of a hard substrate as shown by Chen et al. by finite element analysis [15] and therefore the length h^* in Eq. (7) is not a material characteristic length. On the other hand, Chicot [29] suggests applying the hardness length-scale (HLS) factor to describe nano and microindentation size effect since the absolute hardness and the characteristic length of SGP theory are both dependent on the indentation scale of measurement. Indeed, it is shown that both nanoindentation data (h_c<100 nm) and micro-indentation data (h_c>100 nm) may be mathematically represented by the SGP theory but is physically interpreted by the HLS factor connected to the shear modulus and the Burgers vector of the material.

Data of Beegan et al. (Fig. 6 in [1]) corresponding to hardness measurements performed on 950 nm and 1400 nm thick copper film are fitted using the linear model. Purposely, we do not consider the value corresponding to depths higher than the threshold value $C\,t$ for the studied film since for higher indentation depth the substrate influences the measured hardness when considering the J–H model. Fig. 5 represents the hardness evolution and the best linear fit calculated independently of the film thickness, and the constants found for Eqs.(7) and (10) are reported in Table 2.

Fig. 6 shows a comparison between fitting data by Eq. (7) (Beegan et al.) and fitting data by Eq. (10) (this work). In both cases the models well fit the data and have low errors. From these results it is not possible to state if a model is better than the other one. In this figure, the data included in a square box are not considered in the pile-up fit since the indentation depth is higher than the product $C\,t$ and because the substrate influences the measured value. It is also noticed that the true hardness H_{0f} found with the pile-up hypothesis for the 950–1400 nm thick copper films (3.10) is very close to those obtained by the best fit of the 25–750 nm data by the J–H model in Fig. 1 (*i.e.* 3.18).

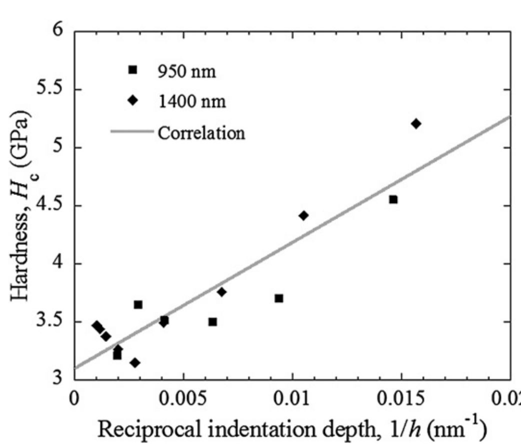

Fig. 5. Plot of hardness versus the reciprocal indentation depth for copper 950 and 1400 nm films on oxidised silicon substrate based on Beegan et al. data (Fig. 6 of Ref. 1).

Table 2
Values of H_{0f} and h^* or B_f obtained from the fitting of the 950 and 1400 nm copper films by the SGP (from Ref. 1) and by the linear methods (this work).

Sample	H_{0f} (GPa)/h^*(nm)	H_{0f} (GPa)	B_f (GPa nm)
Method	SGP, Eq. (6)	Linear, Eq. (10)	Linear, Eq. (10)
1400 nm	3.15/95.37		
950 nm	3.01/77.57		
950–1400 nm		3.10	108.6

The linear fit to represent the indentation size effect of the films can be introduced into the general relation of the J–H model (Eqs. (1) and (2)). It enables us to predict the film hardness and to adequately represent the hardness evolution of the coated material

$$\begin{cases} \beta \geq C\,H_c = \left(\frac{2C}{\beta} - \left(\frac{C}{\beta}\right)^2\right)\left(H_{0f} + \frac{B_f}{h}\right) + \left(1 - \frac{2C}{\beta} + \left(\frac{C}{\beta}\right)^2\right)H_s \\ \beta < C\,H_c = H_{0f} + \frac{B_f}{h} \end{cases} \tag{11}$$

Eq. (11) is used to fit the entire range of data for copper films (25 to 1400 nm). The best fit is represented on Fig. 7 (thick black line) and compared with the K (thick grey line) and P-C (dotted grey line) with ISE correction made by Beegan et al. [1]. The values obtained by the fit are presented in Table 3. It is obvious that we obtained an excellent correlation between the J–H model and the experimental data from Beegan et al. Furthermore this model correctly predicts both film and substrate hardnesses. The effectiveness of this model is enhanced when the indentation size effect is taken into account through a correction of the pile-up high. Similarly we show that the C–L model is improved when taking into consideration the coating ISE through Eq. (10) (Table 3, Fig. 7).

5. Conclusion

Hardness measurements of copper thin films deposited on oxidised silicon substrates were previously analysed by Beegan et al.

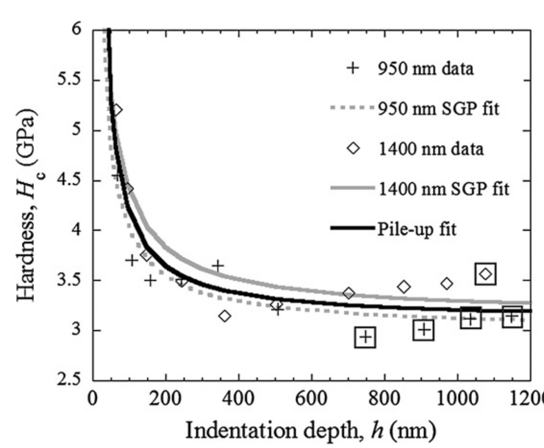

Fig. 6. Plot of hardness versus the contact depth for copper 950 and 1400 nm films on oxidised silicon substrate from Beegan et al. data (Fig. 6 of Ref. 1). The lines represent the fit obtained by Eq. (7) (grey lines, Beegan et al. interpretation) and Eq. (10) (thick black line, this work).

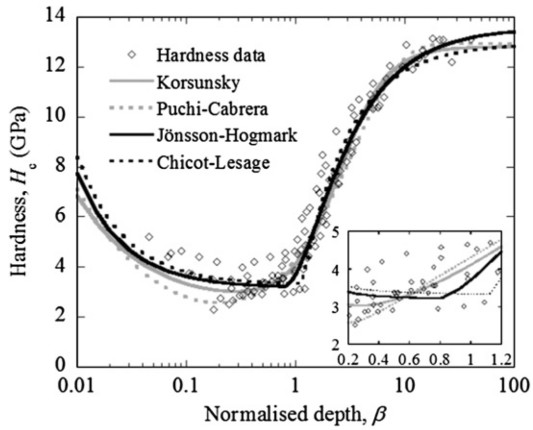

Fig. 7. Plot of hardness versus normalised depth for various copper film thicknesses ranging from 25 nm to 1400 nm fitting by K model (thick grey line), P-C model (dotted grey line), J–H model (thick black line) and C–L (dotted black line). The experimental data are from Beegan et al [1], the ISE effect is incorporated into composite hardness by introducing the strain gradient plasticity model (Eq. (7)) in the K and P-C models (the work from Beegan et al [1]), or the relation (Eq. (10)) in the J–H or C–L models (this work).

[1] using the models of Korsunsky and Puchi-Cabrera. These models lead to an adequate representation of the experimental data and to a good prediction of the film hardness. Nevertheless, the authors definitely reject Jönsson–Hogmark and Chicot–Lesage models which do not allow well fitting the hardness data. We clearly show that this negative response is due to an erroneous application of these models that is in contradiction with their physical signification. We show here that a correct application of these models allows to adequately represent the experimental hardness data and lead to a good predictive value for the film hardness, as well as the descriptive models of Korsunsky and Puchi-Cabrera.

In addition, the models must be improved by considering the indentation size effect for the film hardness behaviour as mentioned by Beegan et al. whom applied the strain gradient plasticity theory at the small indentation depths. However we propose to consider the linear model which is easier to introduce into the models and has a more pronounced physical meaning for the experimentation under study due to the presence of the piling-up. It is noticeable that the two ISE representations are similar to represent the film hardness variation.

Finally, the Chicot–Lesage and Jönsson–Hogmark models are able to predict hardness of composite material. It seems that several authors usually reject these last ones due to a bad appreciation of their physical meaning.

Table 3
Predicted values for copper films 25 to 1400 nm and substrate hardnesses from the fitting of composite hardness models to the hardness data obtained.

Parameters	JH (This work)	C–L (This work)	K (Ref. 1)	P-C (Ref. 1)
H_{0s} (GPa)	13.58	12.94	12.82	12.95
H_{0f} (GPa)	3.17	3.29	2.65	1.15
Constant	0.78	1.13	0.1599	0.2952
h^* (nm) or B_f (GPa nm)	$B_f=63.85$	$B_f=71.34$	$h^*=79.065$	$h^*=518.551$
rms (GPa)	0.689	0.780	0.702	0.810

Ref. 1 referred to the results obtained by Beegan et al. [1], and "This work" to the correct application of the JH and C–L models with an ISE as Eq. (10).

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