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Buckle depression as a signature of Young's modulus mismatch between a film and its substrate

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ABSTRACT

The buckling structures of rigid films on soft substrates exhibit a "mexican hat" shape characterized by a nanometer scale depression at both edges. Based on finite elements simulations, a mathematical formulation is proposed to extract the elastic modulus mismatch between the film and its substrate, from the fine characterization of the buckle morphology.

1. Introduction

Thin films and coatings are used in a wide range of technological applications, such as microelectronics, packaging or optics. They often develop high residual stresses during the deposition process, sometimes about few GPa in compression. Such large compressive stresses may cause the nucleation and growth of buckling structures [1–5] that generally result in the loss of functional properties that were initially conferred to such film/substrate composites. On the other hand, the fine investigation of the morphology of the buckling structures can be of great technological interest in order to qualitatively, or even quantitatively, extract some mechanical parameters of the films/substrate systems [6–9] such as the adhesion properties for instance.

Various methods are now available for determining the Young's moduli of film/substrate systems, the usual one being the well-known nanoindentation. One can also note the "strain-induced elastic buckling instability for mechanical measurement" [10–12] based on wavelength measurements of wrinkled structures appearing at free surface of stressed coated soft materials. In this case, the film stays bonded to its substrate, with no delamination at the interface. The method proposed in this paper is quite similar since it is also based on morphological investigations of surfaces under stress. However, it uses the buckling structures that appear at free surfaces of stressed films when the interface is debonded.

The buckling phenomenon has been widely investigated by the past, both analytically in the framework of the Foppl-Von Karman theory of thin plates [13–16] and numerically by finite elements simulations [17,18]. The influence of various mechanical parameters, such as thickness, stresses or Young's modulus of the film has been now clearly identified (see review in [19]). The effect of the elastic properties of the substrate has also been examined [20,21]. It was of particular importance for the development of stretchable devices for which there is a strong contrast between the Young's moduli of the metallic film and the polymeric substrate. It has been numerically shown that the critical stress for buckling to occur decreases with the decreasing stiffness of the substrate [21]. This mechanical response is associated with an enhanced maximum deflection (compared to the case of a rigid substrate) and the appearance of a nanometer scale depression at the edges of the buckles [21,22]. These depressions may consequently be a relevant signature of the elastic properties mismatch between the film and its substrate. In this context, finite elements simulations of buckles under stress have been carried out. The results are compared and discussed with experimental investigations carried out on various film/substrate systems.

2. Experiments

Polycarbonate substrates ($E_s = 2.5E_s = 2.5$ GPa, according to the manufacturer) of dimensions $10 \times 10x210 \times 10 \times 2\ \text{mm}^3$ were coated by physical vapour deposition with 100 and 200 nm thick nickel ($E_f = 200E_f = 200$ GPa) [23], 200 nm thick zirconium ($E_f = 90E_f = 90$ GPa) [24] and 200 nm thick tungsten ($E_f = 410E_f = 410$ GPa) [25] films. Coated samples were then deformed by compression at room temperature in order to induce buckling [26]. The straight-sided (SS)

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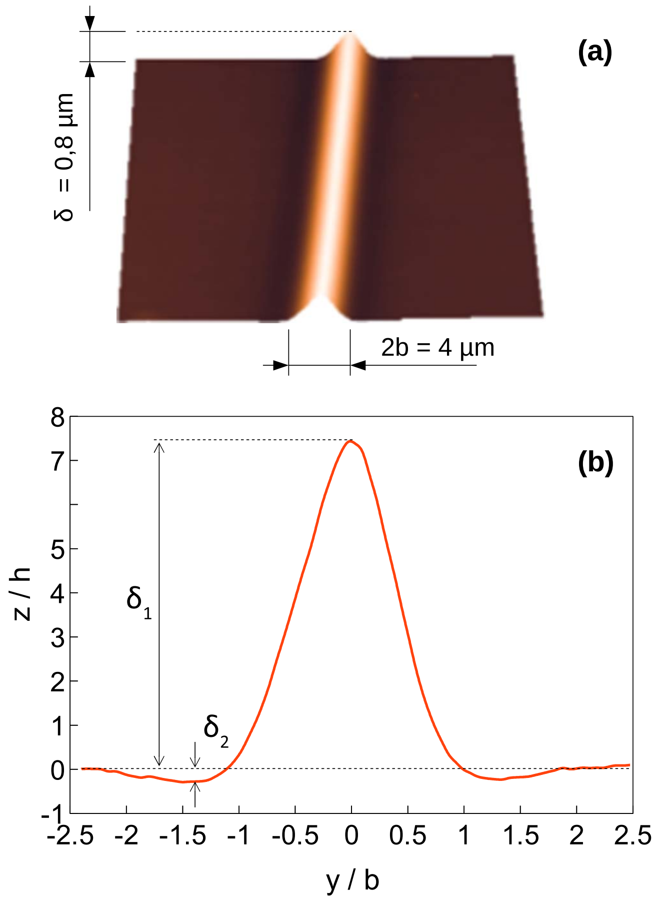

Fig. 1. Characteristic profile of a Ni/PC buckle ($\alpha = +0.97$) showing a nanometer scale depression at the buckle edges. a) AFM image of a straight-sided buckle of width $2b$ = 4 µm and h = 100 nm. b) Normalized out-of-plane displacement *vs.* normalized position. δ_1 and δ_2 correspond to the maximum deflection of the buckle and the depression, respectively.

buckles are lying in this case perpendicularly to the compression axis [1,26]. The morphological parameters of the SS buckles have been extracted from atomic force microscopy (AFM) measurements. A characteristic buckle morphology is presented in Fig. 1, with δ_1, δ_2 and $2b$, the maximum deflection, depression depth and width of the SS buckle, respectively. As expected, due to the substrate compliance, the profile exhibits a depression on both sides, of only a few nanometer depth.

Gold (E_f= 80E_f = 80 GPa) [27] films of thickness 400 and 630 nm were also deposited on silicon wafers (E_s = 100E_s = 100 GPa) [28]. For thesesthese systems, adhesion properties are altered by putting the coated materials in water for a few seconds to induce buckling.

In the following, the elastic contrast of the different film/substrate systems is characterized by their Dundur's coefficient α given by [29]:

$$\alpha = \frac{\overline{E_f} - \overline{E_s}}{\overline{E_f} + \overline{E_s}} \tag{1}$$

with $\overline{E_i} = E_i/(1-\nu_i^2)$ the reduced Young's modulus. α ranges from -1 −1(for a rigid substrate) to +1 (for a soft substrate). The α values for our Au/Si, Zr/PC, Ni/PC, and W/PC experimental cases are thus equal to -0.07 −0.07, +0.95, +0.97 and +0.99 respectively.

3. Finite elements modeling

Finite element simulations (FEM) were performed using the ABAQUS software [30] with an explicit solver used in a quasi-static context. The studied system is composed of three different regions (Fig. 2). The film is modeled by a non-linear plate, the substrate by a homogeneous three-dimensional solid. Both materials are assumed linear elastic isotropic. A thickness ratio of h/H=100h/H = 100 is taken, with h (*resp.* H) the film (*resp.* the substrate) thickness. Finally, the interface consists in a mixed-mode cohesive zone (CZ), which is located everywhere at the interface. Standard cohesive elements have been used to manage the film debonding from its substrate (over a width $2b$ in Fig. 2). A bi-linear traction/separation law is used to pilot the cohesive zone behavior [8]. The interface traction vector $\vec{T}$ (T_n, T_t) is related to the separation vector $\vec{\delta}$ (δ_n, δ_t) representing the relative displacement between the two faces at the crack front. Both traction and separation vectors can be written as their normal (mode I opening) and tangential (mode II shearing) contributions, (T_n, δ_n) and (T_t, δ_t) respectively. The (Oy) direction is lying along the width of the SS buckle, while axis (Ox) is oriented along the buckle axis. The (Oz) direction is normal to the initial film/substrate interface plane. Symmetric boundary conditions are imposed on both sides of the box, at $y = \pm L$, with $L > > b$. The bottom face is pinned. The calculation is carried out using the Green-Lagrange strain tensor to take into account large displacements. A bi-axial compressive stress is induced in the film by a thermal loading, such that $\sigma_{xx} = \sigma_{yy} = -\sigma_0$ and $\sigma_{xy} = 0$, with $\sigma_0 > 0$. An initially delaminated zone (*i.e.* with no cohesive interaction) of width $2b_0$ is initially introduced for the SS buckle nucleation. The quantity b_0 is chosen just above the minimum width necessary for buckling at the given stress σ_0 [19]. The growth of the SS buckle is then controlled by the increase of σ_0, which results in particular in the increase of the delamination width $2b$. This numerical procedure ensures that the extracted buckled shape is in equilibrium with the crack front at the interface (for more details, see [22]). In the following, different elastic mismatches between the film and its substrate have been investigated and characterized by the Dundur's coefficient α. It is assumed that the influence of the second Dundur's coefficient β is negligible. For all the calculations, the value of β has been chosen equal to $\alpha/4$, as used in [19,22].

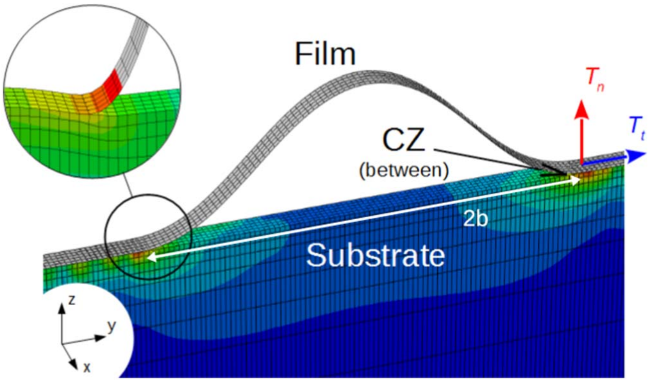

Fig. 2. FEM configuration showing the SS buckle at its equilibrium state. $2b$ correspond to the width of the buckle. Inset: zoom at the crack front that exhibits a nanometer scale depression characteristic of the "mexican hat" shape.

4. Results

In Fig. 3 is presented the evolution of the δ_2/δ_1 ratio as a function of an adimensional parameter B proposed by Zhang et al. in [31] to describe this morphological feature and defined as:

$$B = 1.2\left(\frac{b}{h}\right)\left(\frac{1-\alpha}{1+\alpha}\right)^{1/3} \tag{2}$$

Low values of B thus correspond to the case of soft substrates (with respect to the deposited film), while high values correspond to rigid ones. As expected, it is observed in Fig. 3 that δ_2/δ_1 tends towards 0 for an infinitely rigid substrate. In this case, the equilibrium shape of a SS buckle is given by the Euler column for which no depression is observed