Tensile Stress as a Factor of Deformation and Failure of Thin Films and Coatings

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Abstract. The paper provides analysis of literature data regarding reasons for development of tensile stresses in continuous thin films and coatings, in the process of their formation and under various external influences, as well as study of main mechanisms of their deformation and failure under tensile stresses. Special focus is drawn to the role of film/substrate interface in failure of thin-film structures. Factors controlling initiation and propagation of failure throughout film-substrate system are considered. Main reasons for emergence of the most common models of failure of thin-film structures – edge delamination from the base and film cracking – are demonstrated. In the process of film failure, each of these mechanisms can contribute to the development of the other one, and on the other hand, they both provide relaxation of tensile stresses, thereby competing and suppressing the development of each other. Study of stress relaxation is important for obtaining new structures and certain physical and mechanical properties of coatings. Determining the critical values of stresses that lead to failure is an urgent task, regardless of the method of obtaining coatings and thin films. Development of theoretical basis for predicting and regulating values and sign of stresses that occur during the formation of nanostructures in a plasma environment complement existing models for obtaining these structures in order to further study the plasma methods of growing nanostructures.

Keywords: Tensile stress · Stress-strain state · Thin films and coatings · Nanotechnology · Coating failure

1 Introduction

Carbon and oxide nanostructures are promising materials for use in nanoelectronics and other brunches of industry. Silicon-based nanostructures are considered as potential materials for the production of the next generation of anodes of lithium-ion batteries due to their ultra-high specific capacities [1–3]. Due to their properties, vertically oriented structures of graphene can be used for accumulation and storage of electrochemical energy (in high-performance supercapacitors, fuel cell catalysts, etc.) [4, 5]. In the manufacture of modern semiconductors, field-effect transistors, sensors, oxide nanostructures are widely used, the properties of which largely depend on the methods and conditions of their formation [6, 7]. It should be mentioned that the presence of a boundary between

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the main material and a layer formed on its surface is a principal feature of all technologies that use the particles (atoms, molecules, or clusters) with energy less than a few keV [8]. A lot of experiments are dedicated to control the parameters of the treating flux to improve the performance of the coatings and thin films [9, 10]. Failure of thin films and coatings is one of the main causes of failure of parts and products on which they are applied caused by combined action of internal mechanical stresses (hereinafter referred to as stresses), stresses arising in the process of their formation (residual), and stresses arising during operation. Failure of films and coatings can be manifested in the form of cracking under tensile stresses [11, 12] or delamination, curvature or swelling at compressive stresses [13–15]. Residual stresses arising in coatings and films affect almost all major performance characteristics of parts. In addition to the possibility of cracking and delamination, residual stresses affect adhesion and cohesion, static and multi-cycle strength, wear resistance, heat resistance [16–18], and can positively affect physical properties of thin layers and nanostructures, such as electrical conductivity [19], dielectric constant [20], piezoelectricity [21], etc.

The reasons for occurrence and development of stresses in coatings and thin films can be as follows:

1) stresses associated with growth of coatings and films. In this case, it is necessary to distinguish between internal stresses due to increasing coverage and stresses caused by geometric characteristics of the surface. The increase in internal stresses is due to chemical reactions, phase transformations, particle bombardment [22–24], etc.;
2) thermal stresses that occur as a result of changes in temperature and various coefficients of thermal expansion of coating materials and substrate [25];
3) external stresses arising from deformation of coating-substrate system [26].

   The objective of this article is to analyze the existing models of deformations and failures of thin films and coatings caused by tensile stresses. Understanding the nature of these stresses in thin films and coatings, as well as the possibility to control the stress-strain state of the coating-substrate system is an urgent task, as it directly affects formation, processing and service life of new materials and products.

2 Edge Delamination of Films from Substrate

Under tensile stresses in the film, the effects associated with the stress concentration arising near the free film edge are of particular importance. Figure 1 shows a diagram illustrating the development of shear stresses at film substrate interface as a result of load transfer from the substrate to the film.

If the film is separated from the substrate, then the elastic relaxation of tensile stresses in free state would lead to a decrease in its transverse dimensions (see Fig. 1a). However, under conditions of rigid connection with the substrate, film dimensions need to correspond to substrate dimensions. This situation is equivalent to the application of external forces to unstressed film, which stretch the film to substrate dimensions, leading to its biaxial deformation (see Fig. 1b). If the stretched film is now rigidly fixed on the substrate again, then in order to remove applied external forces in accordance with the principle of superposition, it is necessary to apply forces of the same magnitude – but with the opposite sign – to free edges of the film.

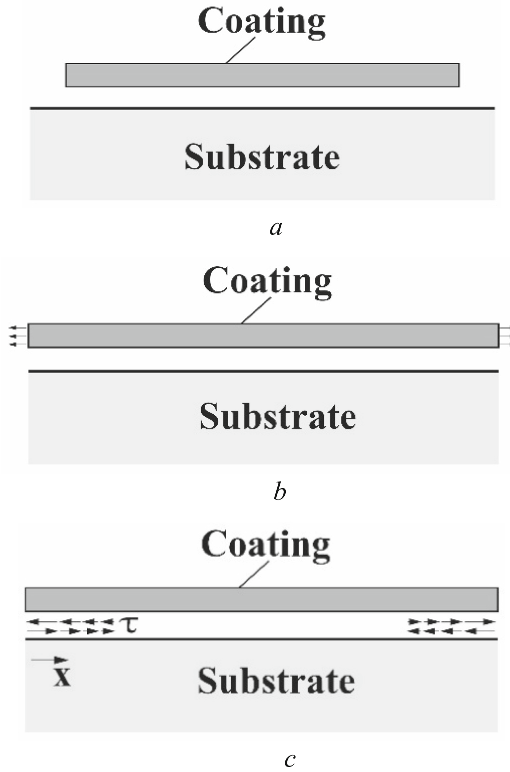

Fig. 1. Schematic diagram of occurrence of shear stresses at film/substrate interface: *a* – unstressed film in a free state; *b* – tensile stresses are applied to the film so that its transverse dimensions correspond to substrate dimensions; *c* – after fixing the film on the substrate, removal of external forces leads to development of shear stresses near the free edge of film.

As a result of the application of these compensating forces at the film edges, normal stresses become equal to zero, and shear stresses appear at film/substrate interface near free edges of the film (see Fig. 1c). These shear stresses provide biaxial deformation of the film, and, thereby, maintain biaxial tensile stresses therein.

In the membrane approximation, when the film is absolutely flexible, and it has no resistance to bending deformation, which makes it possible to ignore the influence of bending moment on the stress-strain state, distribution of shear stresses at the interface $\tau(x)$ is described by the integral equation [27]:

$$\frac{1}{h_f \overline{E}_f} \int_0^x \tau(\xi) d\xi - \frac{2}{\pi \overline{E}_s} \int_0^\infty \frac{\tau(\xi)}{\xi - x} d\xi = \left(1 + \nu_f\right) \varepsilon_m, \; 0 < x < \infty \tag{1}$$

where h_f is film thickness, $\overline{E}_f = \frac{E_f}{\left(1 - \nu_f^2\right)}$ and $\overline{E}_s = \frac{E_s}{\left(1 - \nu_s^2\right)}$ are the moduli of plane deformations, and E_f, E_s, ν_f, and ν_s are Young's moduli and Poisson's ratios of film

and substrate, respectively, ε_m is biaxial deformation of film at a distance from the edge $(x \to \infty)$.

The exact analytical solution of Eq. 1 is unknown. However, it was shown that, asymptotically at $x \to 0$ shear stresses change as follows:

$$\tau(x) \sim \sigma_m \sqrt{\frac{kh_f}{2\pi x}}, \tag{2}$$

where

$$k = \frac{\overline{E}_s}{E_f} = \frac{E_s}{E_f}\frac{1 - \nu_f}{1 - \nu_s}. \tag{3}$$

Thus, the distribution of shear stresses has a singularity in the limit at $x \to 0$, where the magnitude of these stresses tends to infinity.

Film stiffness, which determines its resistance to bending, leads to the need to consider additional effects arising from occurrence of bending moment. For instance, within the Kirchhoff plate model, the tendency to compress the surface layer of a stretched film causes an internal bending moment M and normal stresses to the interface p near its free edge, which tend to delaminate the film from the substrate (see Fig. 2) [28].

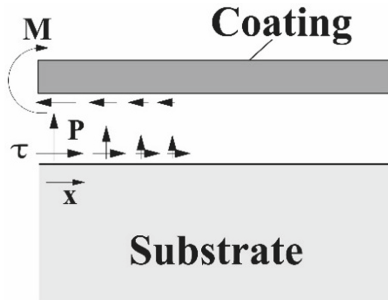

Fig. 2. Diagram of stress distribution near the free edge of film in the framework of Kirchhoff model.

To determine the dependences $\tau(x)$ и $p(x)$ which, as well as the dependence $\tau(x)$ in the model of the membrane film, have a singularity at the edge of the film at $x = 0$, it is necessary to solve the system of integral equations. The results of its numerical solution for the case when the film and the substrate have the same elastic properties $(k = 1)$ were obtained in [27].

Occurrence of concentration areas of shear τ and normal stresses p near the free edges of film leads to the fact that one of the most frequent types of failure of thin films and coatings under stress is their edge delamination (see Fig. 3). This mechanism of failure of thin-film structures was observed in many experimental studies [13, 28].

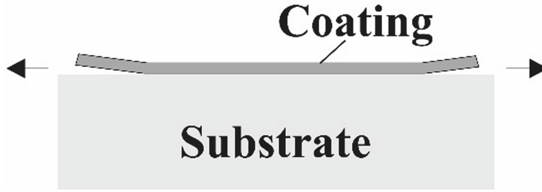

Fig. 3. Film edge delamination from the substrate under tensile stresses.

In theoretical studies, the problem of edge delamination of films and coatings is considered within the framework of two approaches to failure mechanics. The first one assumes that initially the interface between film and substrate is defect-free and delamination starts from the free film edge [29]. Various models are used to study distribution of shear and normal stresses in the immediate vicinity of the film edge and their role in the formation of delamination area. In the general case, the occurrence of edge delamination – failure initiation along the interface – is the result of their joint action, i.e. is a mixed form of failure, which is controlled by a combination of deformation modes I and II. However, limiting cases are also considered when edge delamination develops as a result of only normal [30] or only shear stresses [29] on the interface. In fact, this approach is aimed at developing recommendations for reducing the magnitude of stresses at the interface by selecting the optimal ratio of thicknesses, elastic, thermal, and other characteristics of film and substrate or individual layers in multilayer structures [30]. However, to date, it does not provide an unambiguous criterion to accurately predict the conditions under which edge delamination occurs.

Another approach is based on the assumption that delamination originates in the area of an already existing failure with a sharp tip at the edge of film/substrate interface [31]. This failure can be both the result of separation of the film edge from the substrate under stresses at the interface, which are considered within the first approach, and the initial defect that arose during the film deposition. Stresses at the film/substrate interface tend to cause propagation of the initial failure, while the strength of the material prevents this. This situation can be described using the Griffiths energy failure criterion, according to which the failure propagates if the resulting decrease in potential energy (elastic deformation energy) is greater than or equal to the work required for failure of the material per unit area resulting from this free surface.

The change in the elastic deformation energy of a body ∂U as a result of the crack propagation of length a to an infinitely small distance per unit increment of the crack length is characterized in failure mechanics by the value G, which is called the rate of deformation energy release:

$$G(a) = -\frac{\partial U(a)}{\partial a}. \tag{4}$$

In turn, the energy required for breaking bonds and failure increase, per unit increment of its length, is the specific fracture work Γ. Thus, the Griffiths criterion can be written as follows:

$$G \geq \Gamma. \tag{5}$$

If the film is in a plane deformed state, and the failure front at the interface is a straight line, then the rate of deformation energy release is determined by the elastic deformation energy per unit area of the stressed film. At the stage of stable failure growth, the rate of energy release does not depend on the length of the initial crack:

$$G_s = \frac{\sigma_m^2 h_f}{2\overline{E}_f}, \tag{6}$$

where σ_m is the value of biaxial stresses in the film, h_f is film thickness, and E_f is the modulus of plane deformation. Therefore, from Eq. 5 and Eq. 6 it can be obtained that the film spontaneously delaminates from the substrate under the condition

$$\frac{\sigma_m^2 h_f}{2\overline{E}_f} \geq \Gamma. \tag{7}$$

Taking into consideration that the failure at the interface is constant for a specific film-substrate composition under certain loading conditions, it follows from Eq. 7 that there is a critical value of stresses at which the film edge delaminates from the substrate:

$$\sigma_c = \sqrt{\frac{2\Gamma\overline{E}_f}{h_f}}. \tag{8}$$

As seen from Eq. 8, this critical stress decreases with increasing film thickness, i.e. the thicker the film, the less stress is required to delaminate its edge from the substrate. This also means that a certain stress level corresponds to the critical film thickness at which it delaminates.

It should also be added that the rate of energy release depends significantly on whether the edge of the film coincides with the edge of the substrate or not. The simulation carried out in [31] showed that if film and substrate edges do not coincide, i.e. the film does not cover the entire substrate, the rate of energy release G during failure propagation reaches G_s already at the initial failure length a approximately equal to the film thickness hf. Meanwhile, when film and substrate edges coincide, G approaches G_s only for a failure length of the order of 40 h_f and higher. Moreover, in the latter case, a strong dependence of G on the ratio of the elastic characteristics of film and substrate can be observed.

3 Film Cracking

The second main mechanism that provides partial relaxation of tensile stresses in films and coatings is the formation of transverse cracks therein, i.e. failures normally directed to the film/substrate interface (see Fig. 4).

As in the case of edge delamination of films, the driving force of the cracking process is provided by the elastic energy accumulated in the film. The possibility of forming a transverse failure in a film depends on the ratio of the energy released during its prop-agation per unit length along the film/substrate interface:

$$W_m = \int_0^a G(z)dz, \tag{9}$$

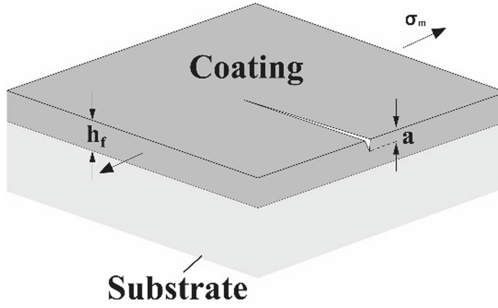

Fig. 4. Generation of a transverse cracks in a thin film under tensile stresses

where.

$$G(z) \approx 1{,}258\pi \frac{\sigma_m^2}{\overline{E}_f} z, \tag{10}$$

is the rate of energy release,

$$W_c = a\Gamma_f. \tag{11}$$

is the energy required for film failure, and a is the depth of crack penetration into the film perpendicular to the film/substrate interface [32].

Freund *et al.* concluded that the failure in the course of its propagation deepens tending to approach the film/substrate interface. If the condition $W_m > W_c$ is still satisfied for $a > h_f$, then the failure penetrates into the substrate and deepens into it until W_m becomes less than W_c. If the substrate has a substantially higher rigidity than the film, then it inhibits the release of elastic energy accumulated in the film, i.e. complicates the failure propagation therein. On the contrary, a pliable substrate easily undergoes elastic deformation and allows a greater release of elastic energy in the film, which results in the easier failure propagation in the latter.

As noted above, the penetration depth of a stable transverse failure into the film/substrate composition, as a rule, reaches the film thickness or overcomes it. The formation of this failure actually leads to the occurrence of new free film edges, where the tensile stresses acting in the film plane are equal to zero. In the areas adjacent to the failure edges, a partial relaxation of these stresses also occurs. However, with distance from the crack, the rigid bond with the substrate prevents the relaxation of tensile stresses in the film, and they increase, approaching the initial level at a distance of several film thicknesses. This contributes to the emergence of new cracks in areas where stresses remain unrelaxed with parallel propagation to the first one. Thus, a system of parallel cracks is formed in the film with each providing the same dissipation of elastic deformation energy in the areas adjacent thereto.

When considering the above mechanisms of failure of thin-film structures, it was assumed that in the case of film delamination, the crack propagates exclusively along the film/substrate interface, while transverse failure occurs only perpendicular to the interface. However, in reality, a crack propagating along the interface can deviate from

it, penetrating into the substrate or into the film, and even emerging onto the free edge of the film, leading to shearing of its fragments. In turn, a crack moving normally to the interface can deviate along it, causing the film to delaminate from the substrate. Thus, on the one hand, in the process of film failure, each of these mechanisms can contribute to the development of another, and on the other hand, they both provide relaxation of tensile stresses, and therefore compete with each other and can suppress the development of each other [27, 33, 34].

4 Conclusion

Regardless of the mechanism of stress development, the main factor leading to their occurrence is the rigid connection between the film and the substrate along the interface, which necessitates the compatibility of their deformations. This imposes restrictions on the change in the size of films, and also causes the transfer of stresses from the substrate to the film. Therefore, the stress-strain state of the interface largely determines the nature of deformation and failure of film structures. In this case, one of the key factors influencing the development and distribution of stresses in thin films and coatings, and, consequently, generation and propagation of cracks in the film-substrate system is the interface curvature. Understanding its role in the processes of deformation and failure of thin films and coatings is of great importance in order to increase their reliability and durability.

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