Mesomechanics of multiple cracking of nanostructured gradient coatings under loading

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Abstract. The paper reports on theoretical and experimental investigations on deformation and fracture patterns of surface hardened materials. Simulation results on modeling processes of energy distribution and transformation at "coating-substrate" interface of a solid under loading by means of Stochastic Excitable Cellular Automata (SECA)-method are described. In doing so, deformable solid is considered as a multilevel system. The method allows taking into account self-organization of configuration perturbations for different levels of the system. An important role of stochastic modulation of an inelastic deformation at the interface in origin of inelastic deformation is discussed. It is shown experimentally that formation of toothed interface between nanostructured coating and substrate also results in formation of chessboard pattern of stress distribution there. This is manifested by multiple cracking of the coating with preferential crack orientation toward direction of maximum tangential stresses. This phenomenon could give rise to increase both strength and ductility of the surface hardened materials.

Introduction

According to [1], tension of flat specimens with hardly deformable coatings results in their multiple cracking of various kinds: i) spatial quasi-periodic transverse opening mode cracking being propagating along specimen gauge-length; ii) development of the system of self crossing transverse shear mode microcracks being oriented along conjugate directions of maximum shear stresses; iii) development of a grid of stochastically branched cracks. The character of multiple cracking depends on thickness of a coating, a relationship between mechanical characteristics (elastic moduli) of the coating and substrate, geometry of their interface. For instance, needle and toothed "coating - substrate" interface profiles ensures under loading stochastic distribution of stress meso-concentrators at the interface and nucleation of a grid of variously oriented microcracks. When coating thickness is small and the interface possesses needle profile multiple coating cracking can provide increasing of both strength and ductility (in contrast with non-coated specimens). In this paper a nature of mentioned difference has been revealed; mesomechanics of nucleation of cracks of both types is described; conditions for multiple cracking initiation and development are formulated.

Verification of the chessboard-like stress and strain distribution at "coating – substrate" interface

By analyzing results on one dimensional calculation [2, 3] where theoretical predictions about the spatial periodic change of normal and shear stresses at the interface of two dissimilar media were shown we put forward the idea that in three dimensional approximation the distribution of stresses and strains at a flat surface can possess a chessboard-like distribution [4]. The verification of this idea was undertaken in [5]. It was implemented in the stochastic model of excitable cellular automata. According to this model, the process of deformation of a solid is considered as a result of the redistribution of the input energy (under mechanical loading) over elementary mesoscopic scale volumes of a loaded specimen. The three-dimensional specimen being modeled is divided in a net of excitable cellular automata. Each of them is a mesoscopic-scale element (mesovolume) of the medium, which resides in one of four states featuring the elastic and plastic strains, as well as the deformation strengthening and pre-fracture of the material. The cellular automata are characterized by the internal energy of an element of the medium. Due to the stochastic distribution of the stress concentrators at the mesoscopic-scale level in the actual medium, the value of the internal thermal energy of automata is given randomly at the zero step of the algorithm. Each of the automaton states corresponds to a certain energy interval with illegible boundaries. They are threshold values for the transition of the mesovolume for the system being modeled in the next state.

While randomly transferring the external energy to cellular automata, the energy fractions increasing the thermal energy of the specimen and causing the volume variation for the given element of the medium are determined. In this case, the stochasticity is stipulated by the fact that stress-concentrator distribution is determined by the mesoscale level of the objects being modeled, i.e., obeys probabilistic principles. As a result, the values of both the thermal energy and the work spent for the variation of the volume and shape for each element of the medium are known at each time step of the algorithm. This makes it possible to calculate the components of the stress-strain tensor for each mesovolume and then to obtain the strain components for the entire specimen. In order to reveal the stress-distribution pattern in the form of the “chessboard” at the interface between the coating and the substrate, it is reasonable to analyze the initial loading stage when the original stress distribution is only slightly deformed by material inelastic flow. Therefore, all non-diagonal components of the stress tensor were taken to be zero, which made it possible to calculate by the Murnaghan formula [6] the distribution of only normal stresses at the interface and strains associated with them.

The conjugation of the coating and substrate in a loaded solid gives rise to two types of perturbations: 1) nanoconfigurational perturbations of the atomic structure at the interface of two dissimilar media which are of the type of atomic clusters of different configuration [5], and 2) sinusoidal field of elastic tensile and compressive stresses in the interface arising due to unequal elastic moduli of the coating and substrate. The self-organization of nanoconfigurational perturbations at the “coating-substrate” interface in the sinusoidal elastic field of varying normal tensile and compressive stresses governs the appearance of a chessboard-like distribution of stresses and inelastic strains at the interface.

Fig. 1,a illustrates the chessboard-like self-organization of atomic configurational perturbations at the “coating – substrate” interface subjected to biaxial tension. The dark zones in Fig. 1,a correspond to the zones of normal compressive stresses, the light ones - to the zones of normal tensile stresses.

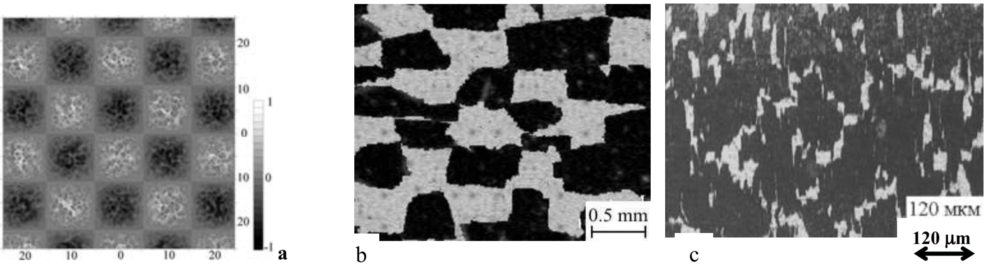

Fig. 1. Numerical simulation (a) and experimental proofs (b, c) of chessboard-like stress and strain distribution at the «nanostructured coating SiAlN - substrate» interface. b – thermal cycling, c – uniaxial tension

The most convincing evidence to the chessboard-like distribution of zones at the “thin film – substrate” interface subjected to hydrostatic compression and tension has been obtained at thermal cycling of a nanostructured SiAlN coating on a copper substrate (Fig. 1,b). The difference of thermal expansion coefficients of the coating and substrate causes the generation of biaxial stresses in the film owing to the necessity of deformation matching in the coating and substrate under thermal action. Since the thermal expansion coefficient of copper substrate considerably exceeds one of the SiAlN–coating, high tensile stresses are generated in the coating under heating whereas compressive stresses are generated under cooling. Under thermal cycling of such a specimen from 1 000°C with cooling in water to 20°C a rectangular network of cracks with spacing ~ 0.5 mm is formed in the coating. After 55 cycles under the action of normal tensile stresses individual cells of the coating flake off, thus making up a rectangular chessboard-like stress distribution.

Under uniaxial tension the necessity of deformation compatibility of the film and substrate having different elastic moduli also leads to the generation of high stresses at the “film – substrate” interface. Since the film is rigidly fixed to the substrate, under loading up to the beginning of fracture they should have the same strain degree. It follows from this condition that elastic stresses in the film σ_f are related with substrate stresses σ_s as:

$$\sigma_f = \frac{1-\nu_s^2}{1-\nu_f^2}\frac{E_f}{E_s}\sigma_s, \qquad (1)$$

where E_f and E_s, ν_f and ν_s are respectively Young’s modulus and Poisson’s ratio of the film and substrate. As Young's modulus of SiAlN–composition twice exceeds that of the copper one, the stress in the coating should be twice higher in comparison with the substrate. A necessity of deformation matching in the conjugated media gives rise to periodic spatial distribution of stresses and strains at the “coating-substrate” interface. Coating delaminating occurs in regions where normal tensile stress reach maximum value.

Effect of a chessboard-like stress and strain distribution at “coating – substrate” interface on multiple cracking of a coating

In the case of thin (according to current classification) coatings (less than 50 mn) the surface cracks formation and their further propagation is defined by chessboard-like pattern of the normal stress distribution at the coating-substrate interface. Periodic alternation of zones of hydrostatic tension and compression results in “channeling” effect of plastic shears in plastically deformed substrate to develop only within zones experiencing normal tensile stresses. As a result, cracking of the coated specimens develops in some stages: initially, a grid of the cracks being located at 45 degrees to the direction of loading is formed in the titanium nitride coating that decorate zones where maximal normal tensile stresses act (fig. 2). With increasing of strain the grid of opening mode cracking is formed. With further increments of strain and up to fracture the coating fragments change their orientation regarding direction of external load that testifies to the fact that rotational modes of deformation are being involved in plastic yielding of the loaded specimens. It should be noticed that such multiple cracking of the coating suppresses macro–localization of plastic deformation development in the specimen (Fig. 2, b) and provides increase of most its mechanical characteristics including ductility [7].

For thick (more than 50 microns) coating range the “chessboard” effect at the interface does not exert influence on the pattern of multiple cracking. Under loading of a specimen its plastic deformation initiates from the grips of testing machine propagating frontally only within ductile substrate accompanied by elastic loading of the coating. When the ultimate normal stress σ_{Un} is reached the coating failure manifests itself in the formation of transverse opening mode crack (Fig. 2,c). The front of plastic deformation in the substrate extends further along specimen axis causing primary opening mode multiple cracking of the coating. The distance Δl between neighboring primary cracks makes:

$$\Delta l = h\sigma_{Un}/\tau, \qquad (2)$$

where h - thickness of a coating, τ - shear stress on the interface “coating-substrate” [8]. The value Δl does not depend on thickness of a specimen and is proportional to thickness of the strengthened layer. Each primary transverse crack generates two mesobands of localized shears of a finite length in a substrate being oriented along conjugate directions of τ_{max} (Fig. 2,d) They cause indentation of a trihedral prism (arising between the neighboring cracks and conjugated mesobands) that is accompanied by secondary cracking of the coating regions being located between primary cracks.

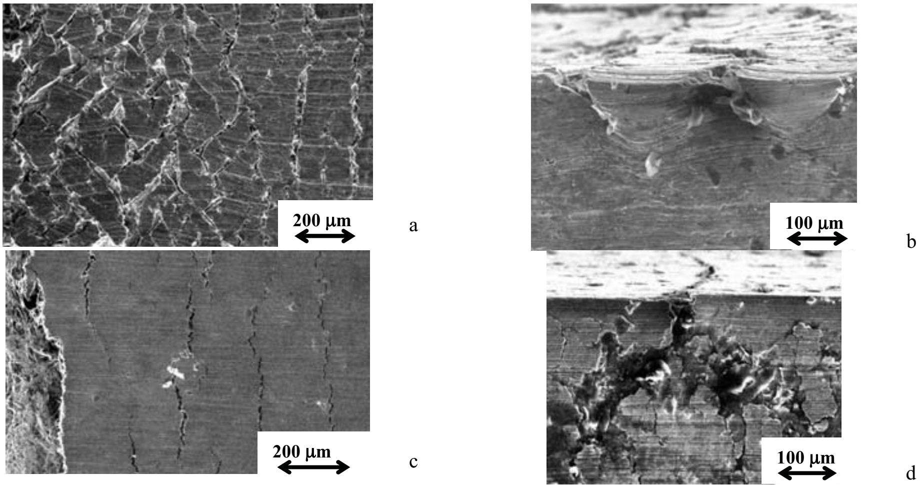

Fig. 2. SEM images of flat (a, c) and lateral faces (b, d) of a high-chromium steel specimen subjected to nitriding observed after tension tests. Time of nitriding made 5 (a, b) and 20 minutes (c, d); ε= 7 (a, b) и 5 % (c, d)

The calculations performed by a finite difference method [9] have shown that the intercrack distance (spatial period of cracking) growths with the enhancing coating thickness (Fig. 3). The regularity observed is also true for the case of increasing mechanical properties of the coating and substrate. (Fig. 5). In doing so, with enhancing spatial cracking period the strain localization degree becomes more pronounced (Fig. 4 and Fig. 6). It is the reason for substantial decrease of both strength and ductility of coated materials.

Methods for control geometry of the coating–substrate interface

The profile geometry of the “coating–substrate” interface exerts governing influence onto cracking pattern, and, hence, onto mechanical behaviour of materials with protective coatings under various external loadings. A number of recommendations to optimize formation modes of different coatings as well as surface hardening of structural and tool materials intended for application at various loadings conditions were reported in the paper [10]. In particular, it was shown that formation of coatings with needled or tooth-shaped structure as well as a gradient transient layer can give rise to increase of strength characteristics of the composition (adhesive, fatigue strength), and decreases both the level of stress concentration at internal interfaces and degree of strain localization related to acting of the cracks like structural notches.

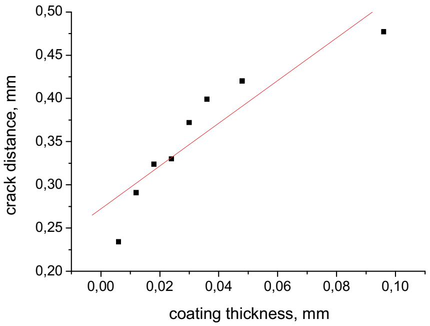

Fig. 3. Dependence of intercrack distance and the coating thickness. Numerical simulation.

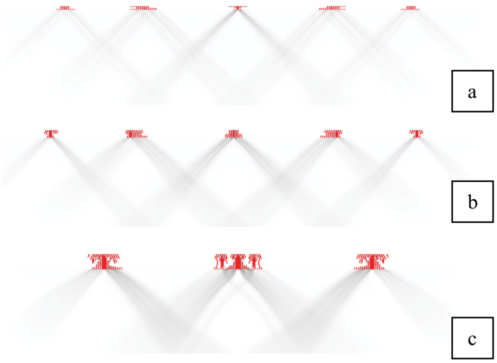

Fig. **4**. Coating cracking and propagation of mesobands of the localized deformation. Coating thickness made 12 (a), 24 (b) and 48 µm (c). Numerical simulation.

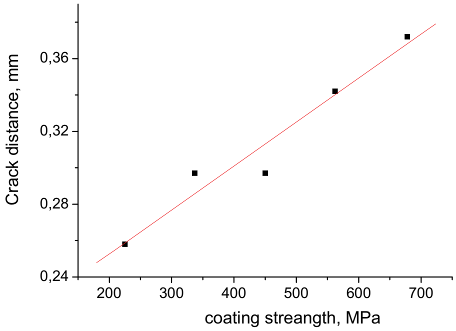

Fig. 5 Dependence of intercrack distance and the coating strength. Numerical simulation.

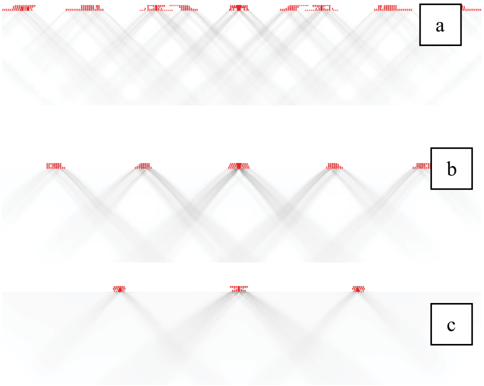

Fig. 6. Coating cracking and propagation of mesobands of the localized deformation in the coated material. The coating strength is 225 (a), 450 (b) and 678 MPa (c). Numerical simulation

For compositions intended for operation under cyclic loading a coatings with a smaller gradient of mechanical properties at the «coating-substrate» interface and higher fracture toughness are more preferable at keeping sufficient hardness of a working surface coating. One of ways to form the tooth-shaped “coating–substrate” interface is preliminary ultrasonic surface impact treatment. The ultrasonic treatment means straining subsurface layer of the substrate by a processing tool which oscillates with ultrasonic frequency. Ultrasonic treatment allows to form in the substrate surface layer a multilevel tooth-shaped relief with the maximal edge sizes of 1-1.5 microns (Fig. 7,a). Detailed investigations by means of a atomic-force microscope (AFM) have shown that large folds are consist of finer edges of 300 nanometers in height (Fig. 7,b). Alongside with the tooth-shaped interface formation the ultrasonic treatment leads to essential grains refinement up to 100-200 nanometers (Fig. 7,c).

One can form tooth-shaped “coating-substrate” interface both before and after deposition of strengthening coating. It is seen from Fig. 8, that ultrasonic treatment of nitrided surface layers of high-chromium steel the pattern of multilevel cracking is substantially changed.

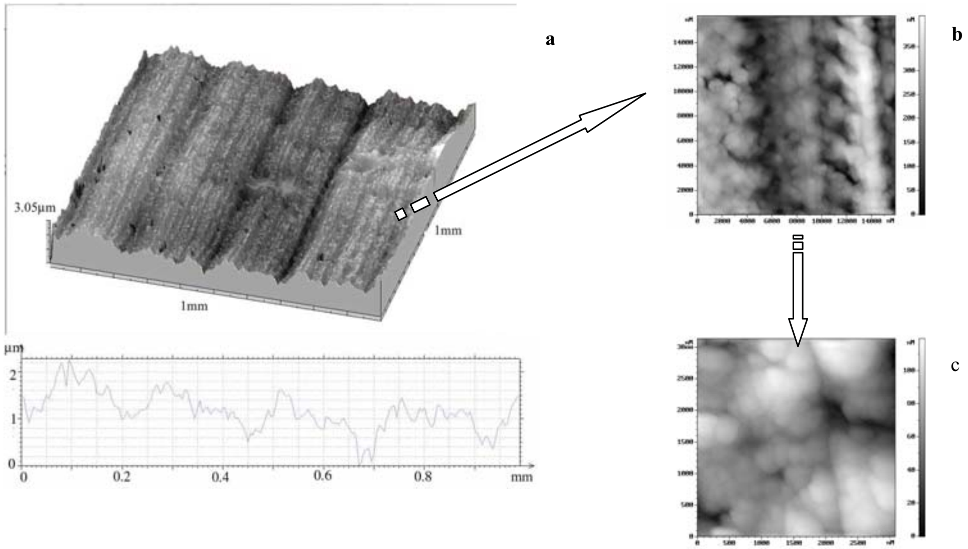

Fig. 7. Morphology (a) и AFM-images (b, c) of the technical titanium surface VT1-0, subjected to the ultrasonic treatment.

At low strains spatially periodically located cracks of opening fracture mode are formed in nitrided surface layers being (Fig. 8b) and not subjected to the ultrasonic treatment (Fig. 8a). However, in the ultrasonic treated specimens formation of the primary opening fracture mode cracks is stopped at $\varepsilon \sim 1\%$ with subsequent formation of secondary cracks oriented at the angle of 45° to the longitudinal axis of specimen (Fig. 8, b). Such multiple cracking ensures intensive propagation of pairs of conjugate plastic deformation mesobands into the substrate. They are oriented at the angle of 45 degrees to crack opening direction and give rise to rotation of mesovolumes in the subsurface layer.

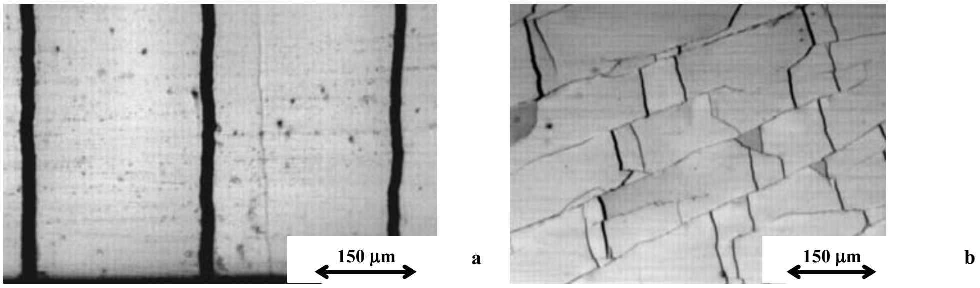

Fig. 8. Optical images of flat surfaces of a high-chromium steel specimen subjected to азотированию (a) followed by ultrasonic treatment (b). tension. $\varepsilon = 1$ %

It is evident from a plot on Fig. 9, that multiple cracking due to crack propagation in direction of maximum tangential stresses ensures simultaneous increase of ultimate strength and ductility of the nitrided specimen of high-chromium steel in contrast with surface strengthened ones non subjected to ultrasonic treatment.

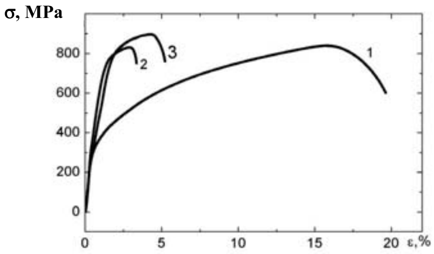

Рис. 9. Stress-strain curves of high-chromium steel specimen (1) subjected to nitriding (2) followed by ultrasonic treatment (3)

Summary

1. Ordered (controllable) multiple cracking of the coatings can be driven only when cracks formation is governed by propagation of localized shear bands in the substrate. This is based on “chess-board” like distribution of normal and tangential stresses at “coating - substrate” interface.
2. The type of multiple cracking exerts essential influence onto mechanical characteristics of a coated material. Cracks of opening fracture mode always decrease composition ductility. The crack of transverse shear can give rise to ductility increasing if they propagate along direction of maximum tangential stresses.
3. Chess-board like distribution of stresses and irreversible deformation at “thin nanostructured coating - substrate” interface plays important functional role at initiation of plastic shears and nucleation of microcracks along direction of maximum tangential stresses in the coating.
4. Multiple cracking of the coating along conjugate direction of maximum tangential stresses ensures more uniform deformation of a solid which can be a reason for increasing fatigue strength and ductility.
5. Multiple cracking phenomenon decorates zones to experience normal stresses that is illustrated by testing of thin nanostructured films, hydrogen saturated and nitrided surface layers.

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