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FORMATION AND CONVERGENCE OF NANOCRACKS IN MECHANICALLY LOADED NANOCRYSTALLINE SOLIDS

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Abstract. A theoretical model is suggested which describes formation and convergence of nano-scale cracks along grain boundaries in nanocrystalline materials under quasistatic mechanical loading. In the framework of the suggested model, the convergence of cracks is described as a percolation process consisting of independent elementary events each being the formation of a nano-scale crack along one grain boundary. The effect of a distribution in geometric parameters (length, orientation) of grain boundaries in a mechanically loaded nanocrystalline solid on the convergence of nano-scale cracks, leading to the macroscopic crack formation, is analysed.

1. INTRODUCTION

Nanostructured solids showing unique physical, chemical and mechanical properties represent the subject of intensive experimental and theoretical study; see, e.g., [1-3]. Of particular interest are outstanding mechanical characteristics exhibited by nanocrystalline (nano-grained) solids due to the nano-scale and interface effects [4-6]. In particular, such mechanisms of plastic and superplastic flow in nanocrystalline materials as the grain boundary sliding [7-9], rotational deformation mode [10-12], grain boundary diffusional creep (Coble creep) [13-15] and triple junction diffusional creep [16] are associated with the active role of grain boundaries (for a review, see [17]). A very high strength and superhardness of nanocrystalline coatings are also related to the specific behavioral peculiarities of ensembles of grain boundaries in the nanocrystalline matter under mechanical load [18,19]. For instance, cracks in mechanically loaded nanocrystalline solids are commonly formed at grain boundaries whose nanoscopic dimensions cause very high values of the critical stress for their formation (see [18] and references therein). Following [18], this specific feature of the formation of nano-scale cracks, which hereinafter will be denoted as nanocracks, is the key factor responsible for extremely high strength characteristics of nanocrystalline bulk materials and coatings. However, together with the formation of nanocracks, the convergence of nanocracks, resulting in catastrophic failure, crucially influences the strength of nanocrystalline solids. The main aim of this paper is to suggest a theoretical model describing both the formation and convergence of nanocracks in a nanocrystalline solid under quasistatic tensile deformation. In the framework of the model, the formation of a macroscopic crack (leading to failure of the solid as a whole) is described as a percolation process consisting of the elementary events of the formation and convergence of isolated nanocracks.

2. FORMATION OF MACROSCOPIC CRACK AS A PERCOLATION PROCESS ASSOCIATED WITH CONVERGENCE OF NANOCRACKS

Let us consider a nanocrystalline solid under tensile stress $\boldsymbol{\sigma}_0=\sigma_0\boldsymbol{e}_x\boldsymbol{e}_x$ (Fig. 1). The stress causes

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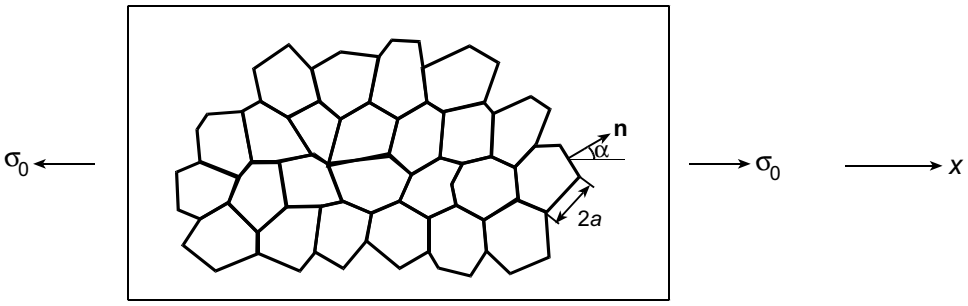

Fig. 1. Nanocrystalline solid under uniaxial tensile stress.

nanocracks to be formed in the mechanically loaded nanocrystalline solid. Following [18], we assume that nanocracks are only formed along grain boundaries. In doing so, with nanoscopic scales of nanocracks at grain boundaries and large angles made by adjacent grain boundary planes at triple junctions, we assume that the formation of a nanocrack at a grain boundary is independent on the events of the formation of nanocracks at any other grain boundaries. As a corollary, the formation of a macroscopic crack – a carrier of the catastrophic failure – results from elementary independent events of the formation of nanocracks at grain boundaries of a quasistatically loaded nanocrystalline solid.

The macroscopic crack formation under consideration is a partial case of percolation described by the standard mathematical methods of the theory [20-23] of percolation in physical systems. This allows us to use these methods in a theoretical description of evolution of the nanocrack ensemble in a deformed nanocrystalline solid. In this context, according to the general representations of the percolation theory [20-23], the macroscopic crack is formed when the concentration n of nanocracks reaches some critical value n_c. (The nanocrack concentration by definition is the ratio of the number of grain boundaries at which nanocracks are formed to the total number of grain boundaries.)

A stable nanocrack of length $2a$ is formed at a grain boundary with length $2a$ and normal $\boldsymbol{n}$ to the grain boundary plane, if $\sigma_{nn} > \sigma_c(a)$. Here σ_{nn} is the stress tensor component at the boundary, and $\sigma_c(a)$ is the critical normal stress characterizing the nanocrack formation. The critical stress $\sigma_c(a)$ in the first approximation is given as [18, 24]:

$$\sigma_c(a) = k\sqrt{\frac{\gamma E}{a}}, \qquad (1)$$

Here γ denotes the specific surface energy of the solid, E the Young modulus, and k the factor taking into account the nanocrack geometry. For a grain boundary (and, therefore, a nanocrack formed at this boundary) whose plane has the normal $\boldsymbol{n}$ making the angle α with axis x, we have: $\sigma_{nn}=\sigma_0\cos^2\alpha$. In this situation, the condition $\sigma_{nn}>\sigma_c(a)$ can be re-written in the following form: $\sqrt{a}\cos^2\alpha > M$, where $M=k\sqrt{\gamma E}/\sigma_0$. (Here, for simplicity, we consider only the normal failure mode I, neglecting the shear failure mode II. Analysis of the shear mode contribution to the macroscopic crack formation in strained nanocrystalline materials is the subject of further investigations of authors.

3. DISTRIBUTIONS OF GEOMETRIC PARAMETERS OF GRAIN BOUNDARIES AND THEIR EFFECT ON FORMATION AND CONVERGENCE OF NANOCRACKS

Let us calculate the nanocrack concentration, using the criterion (1) for their formation. To do so, let us consider distributions in length and orientation of grain boundaries (at which nanocracks are formed). First, let us assume that orientation of grain boundary is random. That is, the distribution in grain boundary orientations is described by the distribution function $\rho_\alpha(\alpha)=1/(2\pi)$, $-\pi < \alpha \le \pi$. With designation $t=\cos^2\alpha$, the function $\rho_\alpha(\alpha)$ can be re-written in terms of t as follows:

$$\rho_t(t) = \frac{4\rho_\alpha(\alpha)}{|dt\ /\ d\alpha|} = \frac{1}{\pi\sqrt{t(1-t)}}. \qquad (2)$$

Here we take into account the fact that one value of t in the range $0<t<1$ corresponds to four values of α in the range $-\pi < \alpha \le \pi$.

Following [13], grain boundary length is assumed to be log-normally distributed:

$$\rho_a(\alpha) = \frac{1}{a\sqrt{2\pi s^2}} \exp\left(-\frac{(\ln a - \overline{\ln a})^2}{2s^2} \right). \quad (3)$$

Here $\overline{\ln a}$ and s are the mean value and standard deviation of ln a, respectively.

With $u=\sqrt{a}$, we consider the following distribution function $\rho_u(u)$:

$$\rho_\alpha(u) = \frac{\rho_a(a)}{du\,/\,da} = \frac{\sqrt{2}}{u\sqrt{\pi s^2}} \exp\left(-\frac{(2\ln u - \overline{\ln a})^2}{2s^2} \right). \quad (4)$$

With the equation:

$$\bar{a} = \int_0^\infty a\rho_a(a)da = \exp\left(\overline{\ln a} + \frac{s^2}{2} \right) \quad (5)$$

formula (4) can be re-written as follows:

$$\rho_u(u) = \frac{\sqrt{2}}{u\sqrt{\pi s^2}} \exp\left(-\frac{(2\ln u - \ln \bar{a} + s^2/2)^2}{2s^2} \right). \quad (6)$$

In these circumstances, the nanocrack concentration is given as:

$$n = \int_0^\infty \rho_u(u) P(t > M/u) du, \quad (7)$$

where $P(t>M/u)$ is the probability of the fact that $t>M/u$. This probability by definition is

$$P(t > M/u) = \Theta(1 - M/u) \int_{M/u}^1 \rho_t(t)dt = \frac{2}{\pi}\Theta(1 - M/u) \arccos\sqrt{\frac{M}{u}}. \quad (8)$$

Here $\Theta(p)$ is the Heaviside function ($\Theta(p)=1$, if $p \geq 0$, and $=0$, if $p<0$).

With $u=M/v^2$ and expressions (6) and (8) substituted into formula (7), we have:

$$n = \sqrt{\frac{32}{\pi^3 s^2}} \times \int_0^1 \frac{1}{v} \exp\left(-\frac{(4\ln v + 2\ln[\sigma_0/\sigma_c(\bar{a})] - s^2/2)^2}{2s^2} \right) \times \arccos v\, dv. \quad (9)$$

In formula (9) it is taken into account that $\sqrt{M}/\bar{a} = \sigma_0/\sigma_c(\bar{a})$.

Thus, we have the nanocrack concentration n given by formula (9). The condition that the macroscopic crack, a carrier of the catastrophic failure, is formed represents the following inequality: $n>n_c$. Here n_c denotes the critical nanocrack concentration determined by geometry of the system [20-23]. After some analysis based on the percolation theory [20-23,25], in the discussed situation with nanocrystalline solids, we have: $n_c \approx 0.125$.

Dependences of n on $\sigma_0/\sigma_c(\bar{a})$ at different values of s, given by formula (9), are shown in Fig. 2. The dashed horizontal line in Fig. 2 shows value of n_c ($n_c \approx 0.125$). The macroscopic crack formation occurs in the range of parameters corresponding to the region above the horizontal line $n=n_c$ in Fig. 2. As it follows from Fig. 2, when parameter s increases at fixed value of the mean grain size $\bar{a}$, the critical (minimum) external stress necessary for the formation of a macroscopic crack just slightly increases. This critical external stress is larger by only a small value than the stress $\sigma_c(\bar{a})$. As a corollary, the criterion for the macroscopic crack formation in a nanocrystalline solid characterized by conventional distributions (say, given by formulas (2) and (3)) in both grain size and grain boundary plane orientation is close to that for the formation of an isolated nanocrack at one grain boundary with favoured orientation of its plane relative to the normal tensile stress.

Let us consider the situation with two types of grain boundaries: conventional and highly failure-resistant boundaries. A high resistance to the nanocrack formation at a part of grain boundaries

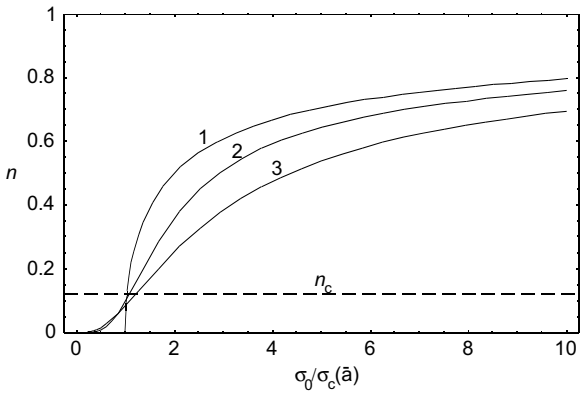

Fig. 2. Dependences of the concentration n of grain boundaries with nanocracks on parameter $\sigma_0/\sigma_c(\bar{a})$, for s=0, 1 and 1.5 (curves 1, 2 and 3, respectively). Dashed horizontal line shows value of the critical concentration $n_c \approx 0.125$.

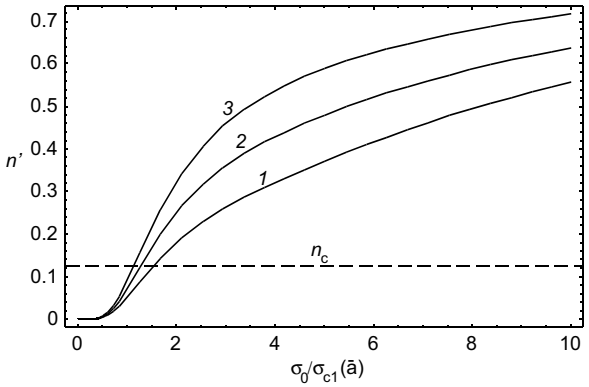

Fig. 3. Dependences of the concentration n' of grain boundaries with nanocracks on parameter $\sigma_0/\sigma_c(\bar{a})$, for s=1, k_2/k_1=5, and c_1=0.5, 0.7 and 0.9 (curves 1, 2 and 3, respectively). Dashed horizontal line shows value of the critical concentration $n_c \approx 0.125$.

can occur, for instance, due to compositional inhomogeneities of a nanocrystalline solid containing grain boundaries with two sorts of chemical composition. Such compositional inhomogeneities of the grain boundary phase, leading to different failure resistances of grain boundaries with different compositions, can be formed in nanocrystalline coatings deposited at a low current plasma density and subjected to energetic ion bombardment [19].

In the framework of our model, the existence of conventional and highly failure-resistant grain boundaries in a nanocrystalline solid is described as the existence of two types of grain boundaries characterized by two values of coefficient k. More precisely, we assume that grain boundaries whose total volume fraction is c_1 (c_2, respectively) are characterized by parameter k=k_1 (k=k_2, respectively). In these circumstances, the critical normal stress for the formation of nanocracks of the first type is $\sigma_{c1}(a)=k_1\sqrt{\gamma E/a}$, while the critical normal stress for the formation of nanocracks of the second type is is $\sigma_{c2}(a)=k_2\sqrt{\gamma E/a}$. Then the nanocrack concentration is given by the following formula:

$$n' = c_1 n(k = k_1) + (1 - c_1) n(k = k_2). \qquad (10)$$

Dependences of n' on $\sigma_0/\sigma_{c1}(\bar{a})$, for s=1, k_2/k_1=5 and different values of c_1, are presented in Fig. 3. As it follows from Fig. 3, the existence of a part of grain boundaries with a high failure resistance in a nanocrystalline solid gives rise to enhancement of the critical applied stress for the formation of a macroscopic crack. However, this critical stress does not run parallel with the volume fraction (c_1) of highly failure-resistant grain boundaries. For instance, for c_1=0.5 (when the number of grain boundaries with high failure resistance is equal to that of conventional grain boundaries), the critical value of the ratio $\sigma_0/\sigma_{c1}(a) \approx 1.8$. For c_1=0, we have $\sigma_0/\sigma_{c1}(\bar{a}) \approx 1.05$. That is, the strength of a nanocrystalline solid with grain boundaries of two types increases. The above conclusion is indirectly supported by experimental data [19] indicating a tentatively two-fold increase of the microhardness of a nanocrystalline coating, occurring due to compositional inhomogeneities in its grain boundary phase [19].

4. CONCLUDING REMARKS

To summarize, conventional distributions of grains in size and boundary orientation, inherent to a real nanocrystalline solid, do not essentially influence the conditions for the microscopic crack formation in the solid. At the same time, if a part of grain boundaries becomes highly failure-resistant (due to changes in composition), the failure resistance of a nanocrystalline solid increases. In doing so, the critical applied stress for the formation of a macroscopic crack does not run parallel with the content of highly failure-resistant grain boundaries. It reflects the percolation nature of evolution of nanocracks in mechanically loaded nanocrystalline solids.

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