Materials Physics and Mechanics 47 (2021) 633-638 Received: October 1, 2021
Research article

MIGRATION OF NONEQUILIBRIUM GRAIN BOUNDARIES IN METAL-GRAPHENE COMPOSITES WITH ULTRAFINE-GRAINED MATRIX

S.V. Bobylev*

Institute of Problems of Mechanical Engineering, Russian Academy of Sciences, St. Petersburg 199178, Russia

Peter the Great St. Petersburg Polytechnic University, St. Petersburg 195251, Russia

*e-mail: bobylev.s@gmail.com

Abstract. We propose a theoretical model describing stress-induced migration of nonequilibrium grain boundaries constrained by the presence of second phase inclusions in metal-graphene composites with the ultrafine-grained matrix. Within the framework of the model, the structure of nonequilibrium boundaries is represented as a combination of dislocation structure of equilibrium boundary and dislocations introduced from outside, trapped by the boundary during deformation. Using the disclination theory approach, a change in the energy of the system associated with such migration under the action of applied shear stress was found. It is theoretically shown that the presence of inclusions leads to the strengthening of the material and the suppression of grain growth.

Keywords: metal-graphene composites, ultrafine-grained materials, nonequilibrium grain boundaries, grain boundary migration, disclinations

1. Introduction

Stress-induced grain boundary (GB) migration is a special mechanism of plastic deformation and grain growth in nanocrystalline (NC) and ultrafine-grained (UFG) materials, and also plays an important role in fracture processes [1-10]. Most of the theoretical and computer models [3,4,6-10] focus on the study of ordinary equilibrium GB structures, however, their structure is often significantly distorted in the process of plastic deformation of the material. This is most typical for UFG metals and alloys produced by the methods of severe plastic deformation [11], where GBs capture a large number of lattice dislocations [4]. Such GBs are usually called nonequilibrium GBs. Migration of GBs, in the general case, leads to grain growth and destruction of the NC/UFG structure, which, in turn, leads to degradation of the mechanical and functional properties of the material. Grain growth is one of the main negative factors that must be dealt with during the synthesis of NC/UFG metals and alloys. One of the effective methods for suppressing grain growth is the synthesis of heterogeneous materials reinforced with second phase inclusions serving as obstacles to GB migration [4,12,13]. For example, significant progress has recently been made in the production of metallic materials reinforced with graphene platelets [14,15] or nanoribbons [16] demonstrating very high strength. Most fabrication methods of metal-graphene composites utilize plastic deformation processing similar to severe plastic deformation methods used for UFG single-phase metals synthesis. Thus, nonequilibrium GBs are typical in these composites as well. It is important to investigate the interaction between GB structure and reinforcing inclusions to better control the mechanical properties of metal-graphene composites.

http://dx.doi.org/10.18149/MPM.4742021_11

In this paper, we propose a theoretical model, which describes the features of the stress-induced migration of nonequilibrium GBs, constrained by the presence of second phase inclusions, in metal-graphene composites with UFG matrix.

2. Theoretical model

Let us consider the process of stress-induced migration of nonequilibrium GB in a composite material containing graphene inclusions (Fig. 1). For definiteness, we consider a fragment of the structure containing the tilt boundary AB with a structure traditionally represented as a wall of edge dislocations (Fig. 1a). A nonequilibrium GB, by definition, consists of intrinsic GB dislocations, which form an equilibrium GB structure (in Fig. 1, they are shown by solid symbols), and extrinsic dislocations (open symbols), trapped by the GB in the process of severe plastic deformation. In the general case, the capture of dislocations leads to a change in the GB misorientation angle. Let the initial equilibrium GB be characterized by a misorientation angle ω_0, and the capture of additional dislocations leads to a change in this angle by $\Delta\omega$. Following the approach developed in the models [3,5,6], a change in the GB misorientation angle leads to a disturbance in the balance of the misorientation angles in the triple junctions A and B, as a result of which wedge disclinations with the power $\pm\Delta\omega$ are formed in them. For definiteness, let us assume that in the initial configuration the GB AB is located close to the graphene inclusion (shown by a red rectangle), having a size *a* in the direction parallel to the GB plane. The size in the direction of migration of the GB is irrelevant in the framework of this model. For simplicity, we consider the case when the inclusion is located symmetrically relative to junctions A and B.

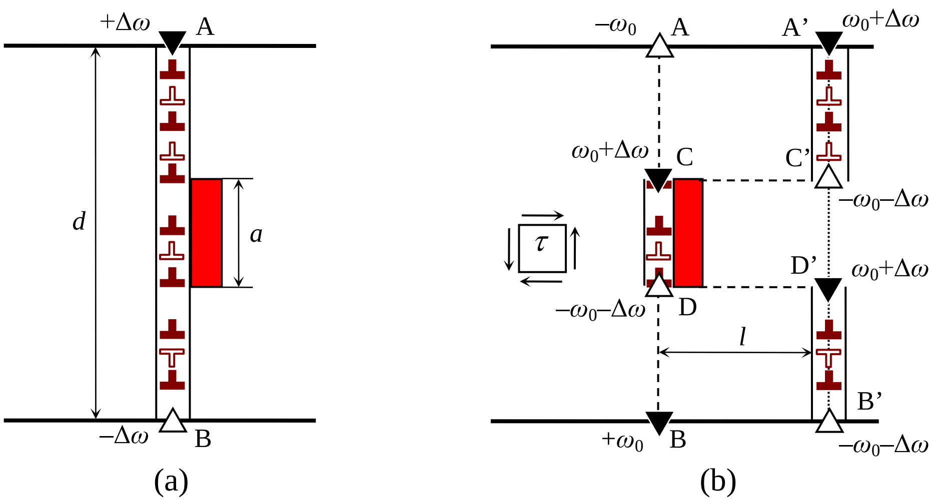

Fig. 1. The model of migration of a nonequilibrium tilt boundary is constrained by the presence of an inclusion (red rectangle). Solid dislocation signs denote intrinsic GB dislocations that form an equilibrium structure, and open dislocation signs correspond to extrinsic dislocations trapped by the boundary. (a) Initial state. (b) The boundary AB migrates to a new position, which leads to its fragmentation into three GBs (A'C ', CD and D'B') and the formation of an ensemble of disclination defects (see text)

Let the GB AB come into motion under the action of the applied shear stress τ (Fig. 1b). Within the framework of the proposed model, we consider only incoherent inclusions impermeable to dislocations. As a consequence, the migration of GB AB leads to the fragmentation of its structure. GB AB is divided into three fragments, namely: a fixed fragment CD, stopped by the inclusion and remaining in its original position, and two fragments A'C 'and D'B' migrating at the same distance *l* (Fig. 1b). Each of these fragments represents a tilt boundary cut off on both sides, which, according to the theory of disclinations [17], can be equivalently represented as a dipole of wedge disclinations with powers equal in magnitude to its misorientation angle. In this case, all three fragments have a misorientation $\omega_0 + \Delta\omega$; therefore, in the configuration shown in Fig. 1b, three disclination dipoles CD, A'C', and D'B' with corresponding powers are formed. In addition, after the initial disclination dipole AB shown in Fig. 1a moves away disclinations with powers $\pm\omega_0$ are formed at triple junctions A and B (see Fig. 1b). The ensemble of disclination defects shown in Fig. 1b is an equivalent representation of the dislocation structure of GBs. In the proposed model, we will characterize the degree of non-uniformity of GBs using a parameter $\Delta\omega$, without consideration of the details of dislocation structure inside the GBs.

Using the theory of disclinations [17], we calculated the change in the energy of the system associated with the migration of a nonequilibrium GB, characterized by a deviation $\Delta\omega$ of the misorientation angle from the equilibrium configuration, in the presence of inclusion on the migration path:

$$\Delta W = \frac{D\omega_0^2 d^2(1+\Delta\omega')}{4}\Big\{-4x(1-a')\tau/(D\omega_0) + (1+2\Delta\omega')\big[(4x^2+(1+a')^2)\ln(4x^2+(1+a')^2)$$
$$-(4x^2+(1-a')^2)\ln(4x^2+(1-a')^2) + (1-a')^2\ln(1-a')^2 - (1+a')^2\ln(1+a')^2\big] \qquad (1)$$
$$+2(1+\Delta\omega')\big[(x^2+a'^2)\ln(x^2+a'^2) - x^2\ln x^2 - a'^2\ln a'^2\big] + 2(x^2+1)\ln(x^2+1) - 2x^2\ln x^2.$$

Here $D = G/[2\pi(1-\nu)]$, G is the shear modulus, ν is Poisson's ratio, $\Delta\omega' = \Delta\omega/\omega_0$, $x = l/d$, $a' = a/d$, d is the length of migrating GB (see Fig. 1a). When $\Delta\omega' = 0$ and $a' = 0$ expression (1) turns into the similar expression for the energy change associated with migration of equilibrium GB found in [3].

3. Results and discussion

Examples of dependencies $\Delta W(x)$ are shown in Fig. 2 for values $\tau = 0.8D\omega_0$, $\Delta\omega' = 0.1$, and different values of the size a of inclusion. The nature of the dependences is completely similar to those obtained earlier for the equilibrium GB [3]. It is clearly seen that an increase in the size of the inclusion leads to a sharp increase in energy $\Delta W(x)$, which complicates the migration process. As before, it is possible to distinguish two modes of migration: stable (limited) and unstable, as well as to introduce into consideration two critical stresses: the critical stress τ_{c1} of the migration onset and the critical stress τ_{c2} of the transition to unstable migration, in which the GB loses its stability and migrates indefinitely.

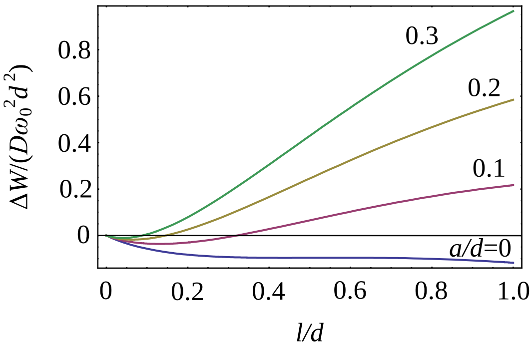

Fig. 2. Dependence of the change in the energy of the system as a result of migration of a nonequilibrium GB in the presence of inclusion on the normalized migration distance *l*/*d*, calculated for $\tau = 0.8D\omega_0$, $\Delta\omega' = 0.1$, and different values of the inclusion size *a*

Critical stress τ_{c1} is found using the condition $\Delta W(x=b/d)=0$, where b is the Burgers vector magnitude of the lattice dislocation:

$$\tau_{c1} \approx \frac{D\omega_0(2+\Delta\omega')b}{2(1-a')d}\ln\frac{d}{b}. \qquad (2)$$

For comparison, a similar expression for the critical stress τ_{c1}^e of an equilibrium GB [3] with a misorientation angle ω_0: $\tau_{c1}^e = D\omega_0(b/d)\ln(d/b)$. It can be seen from formula (2) that the nonequilibrium character of the GB, expressed by the parameter $\Delta\omega'$, leads to a proportional increase in the critical stress τ_{c1}. The presence of inclusion in the migration path also leads to the growth of τ_{c1}. The dependence of the critical stress on the grain size (for which the length d of the GB can be taken) does not change from the presence of nonequilibrium and inclusions and is expressed in the same way as in the case of migration of an equilibrium GB: $\tau_{c1} \sim d^{-1}\ln d$.

We estimated the stress τ_{c1} using the example of UFG Al–graphene composite with a shear modulus (of an aluminum matrix) $G=27$ GPa and Poisson's ratio $\nu=0.31$, $b=0.25$ nm, and grain size $d=100$ nm. Figures 3a,b show the dependences $\tau_{c1}(a/d)$ calculated for $\omega_0=5°$ and $\omega_0=30°$, respectively, and various values of $\Delta\omega'$. It is clearly seen that an increase in the inclusion size noticeably increases the stress required for migration onset. The found values of stresses are easily achievable in experiments. Accordingly, GB migration can make a significant contribution to the dislocation activity in the plastic deformation of UFG metal–graphene composites. The change in τ_{c1} due to the formation of nonequilibrium GBs and the introduction of inclusions of the second phase into the material is reflected in the corresponding change in the yield stress of the material. The presence of inclusions leads to an increase in the yield point, i.e., hardening of the material. However, to carry out a numerical assessment of this change, it is necessary to analyze the real structures of nonequilibrium GBs and their distribution in the bulk of the material. This is beyond the scope of this work.

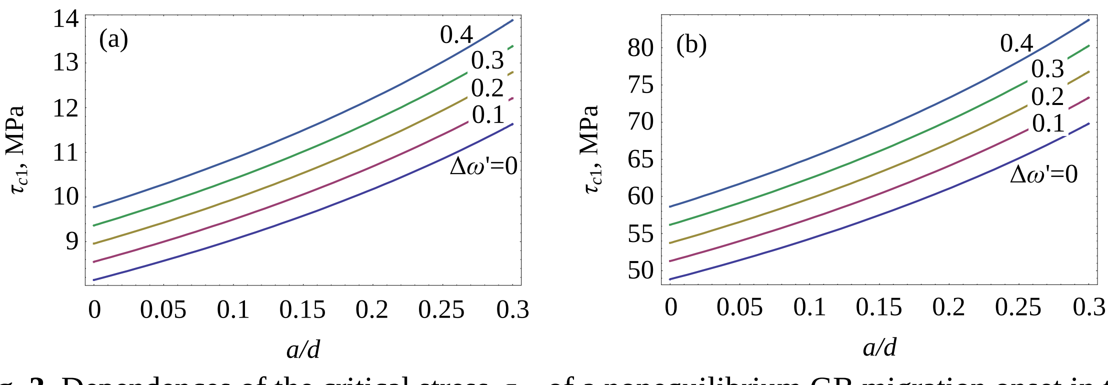

Fig. 3. Dependences of the critical stress τ_{c1} of a nonequilibrium GB migration onset in the presence of inclusion on the normalized size of the inclusion *a*/*d*, calculated for different values of the nonequilibrium parameter $\Delta\omega'$, $d=100$ nm and misorientation angles (a) $\omega_0=5°$; (b) $\omega_0=30°$

The critical stress τ_{c2} of the transition to unstable migration is defined as the minimum stress at which the dependence $\Delta W(x)$ becomes monotonically decreasing. The analytical expression for τ_{c2} cannot be obtained but it can be easily calculated numerically from (1).

Figure 4 shows an example of the dependence $\tau_{c2}(a/d)$ for several values of the parameter $\Delta\omega'$. We can see that critical stress τ_{c2} significantly increases with the introduction of inclusions. As the unstable migration mode is generally associated with strain-induced grain growth the increase of the critical stress τ_{c2} (Fig. 4) suggests that grain growth mechanisms are significantly hampered in composites reinforced by second phase inclusions (graphene platelets in our case).

In Ref. [3], it was shown that the second critical stress τ^e_{c2} for the equilibrium tilt boundary in the absence of inclusions depends only on the misorientation angle and is equal to $\tau^e_{c2} \approx 0.8D\omega_0$ (this value is shown with a dashed line in Fig. 4). However, in the case of migration constrained by the presence of inclusions, it can be seen from Fig. 4 that the critical stress τ_{c2} depends on both the misorientation angle $\omega_0 + \Delta\omega$ and the size of the inclusion *a*. At the same time in the case of nonequilibrium GB migration in the absence of inclusions (partial case of formula (1) with $a' = 0$) second critical stress τ_{c2} is exactly the same as in the case of equilibrium GB [3], i.e. $\tau_{c2} = \tau^e_{c2} \approx 0.8D\omega_0$.

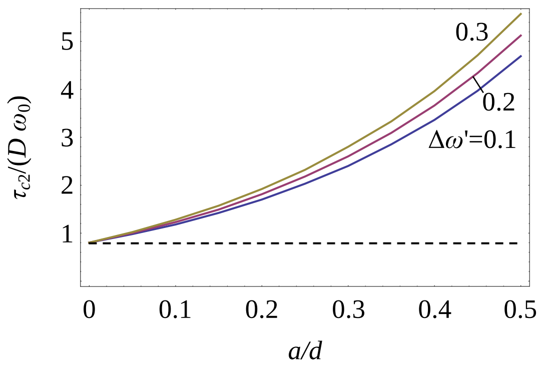

Fig. 4. Dependence of the normalized critical stress $\tau_{c2}/(D\omega_0)$ of the transition to unstable migration of a nonequilibrium GB, constrained by the presence of inclusions, on the normalized size of the inclusion *a*/*d* for different values of the nonequilibrium parameter $\Delta\omega'$. The dashed line shows the level of critical stress for migration of GBs in the absence of inclusions

4. Concluding remarks

Thus, in this work, we proposed a theoretical model describing the stress-induced migration of nonequilibrium GBs in the presence of second phase inclusions that prevent migration. Within the framework of the model, the structure of a nonequilibrium GB is represented as a combination of the dislocation structure of equilibrium GB and dislocations introduced from outside, trapped by the boundary during deformation. The degree of GB nonequilibrium is described by the deviation of its misorientation angle from the misorientation angle of the initial equilibrium structure, from which the nonequilibrium GB was formed. The presence of inclusion on the migration path leads to the fragmentation of the migrating GB. Using the disclination theory approach, a change in the energy of the system associated with such migration under the action of applied shear stress was found. As a result of energy analysis, the critical stresses of the migration onset and the transition to the unstable migration mode, when the GB migrates indefinitely, were found. The dependences of both critical stresses are calculated depending on the size of the inclusion that prevents GB migration and the degree of non-equilibrium of GBs. It is theoretically shown that the presence of inclusions leads to the strengthening of the material and the suppression of grain growth.

Acknowledgement. *The work is supported by the Russian Ministry of Science and Higher Education within the World-Class Research Center Program "Advanced Digital Technologies" (contract 075-15-2020-934 of 17.11.2020).*

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