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# EFFECT OF GRAIN SIZE DISTRIBUTION ON THE YIELD STRENGTH OF METAL-GRAPHENE COMPOSITES

**S.V. Bobylev**✉**, A.G. Sheinerman**

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

✉ bobylev.s@gmail.com

**Abstract.** A theoretical model describing the effect of grain size distribution in the metal matrix of metal-graphene composite on its yield strength. The model describes the initial stage of plastic deformation in metal-graphene composites and takes into account both dislocation mechanisms of plasticity and the process of grain boundary sliding. The yield strength of the aluminum-graphene composite was calculated depending on the average grain size of the matrix and the volume fraction of graphene. Our analysis shows that the presence of dispersion in the grain size distribution of the metal matrix decreases the yield stress so special attention should be paid to reducing this dispersion during the synthesis of the composite.

**Keywords***:* metal-graphene composites, ultrafine-grained materials, yield strength, grain boundary sliding

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## 1. Introduction

Composites based on metals with reinforcing two-dimensional inclusions are attracting more and more attention as new generation materials for a wide range of engineering applications [1-6]. A large number of works in the field of composite science show that the incorporation of graphene or its derivatives into metal matrices leads to a significant increase in the mechanical properties of composites in comparison with unreinforced materials. The two-dimensional geometry of graphene particles makes it possible to effectively (in comparison with other classes of reinforcing materials) block the movement of dislocations, which opens up new possibilities for controlling the growth of grains in metal matrices, their mechanical characteristics, and thermal stability.

Recently, significant progress has been achieved in the development of metallic materials reinforced with graphene platelets [6-8] or nanoribbons [9], demonstrating both high strength and good ductility. The nature of this simultaneous improvement in strength and

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ductility was explained [8] to be the capture of dislocations by graphene platelets during plastic deformation. Due to the large total surface area, graphene plates are capable of trapping a large number of dislocations, which creates high internal stresses, increasing strain hardening and ductility. The latter was confirmed in a theoretical model [10] describing stress-strain curves of metal-graphene composites with bimodal matrix. However, the model [10] considered only dislocation mechanisms of plasticity. At the same time, grain-boundary (GB) sliding is often observed in metal-graphene composites, which, as was theoretically shown in [6], makes it possible to obtain more realistic behavior of the yield stress dependences on the grain size. At the same time, both works [10,11] did not take into account the possible inhomogeneity in the distribution of grain sizes within the ultrafine-grained (UFG) metallic phase. It is known [12,13] that in ordinary metals such inhomogeneity affects, for example, the yield stress of the material, therefore, taking this factor into account will provide better accuracy of modeling the mechanical properties of the material.

We propose a theoretical model that allows calculating the yield stress of metal-graphene composites with an inhomogeneous metal matrix characterized by a certain dispersion of grain sizes.

## 2. Model

Let us consider a sample of a composite material with an inhomogeneous UFG matrix and inclusions in the form of graphene platelets under the action of uniaxial tensile stress $\sigma$ (Fig. 1a). In this work, we assume that all graphene platelets lie along the GBs (which is usually the case in composites with small grain sizes) in the metal matrix (shown by green rectangles in Fig. 1), and their average size is much smaller than the typical GB length. For definiteness, we treat graphene platelets as disks with a thickness $H$ and a diameter $L$. Then we assume that plastic deformation in this composite occurs through two main mechanisms: 1) emission of lattice dislocations from GBs, their sliding through grain, and subsequent absorption by neighboring GBs; and 2) GB sliding, which is realized by sliding of monolayers of graphene plates located in GBs, and subsequent sliding over the regions of GBs free of graphene.

In this work, we will consider only the case of graphene platelets with a length less than the GB length in the UFG phase. We make the following model assumptions: (1) the presence of graphene platelets at the GBs decreases the adhesion of neighboring grains and makes sliding along the GBs with graphene easier than along the GBs without it; (2) the critical GB sliding stress is independent of the fraction of the GB area occupied by graphene, i.e. the presence of even one graphene platelet decreases the critical value stress for the activation of intergranular slip; (3) the local yield stress for each of the GB in the UFG phase is determined by the minimum of the yield stress of the UFG phase without graphene and the critical slip stress; (4) the macroscopic yield stress is calculated as the average value of the local yield points over all GBs in the UFG phase.

The mechanism of GB sliding is described in detail in [11], according to which, clusters of non-crystallographic edge dislocations are formed as a result of the sliding of graphene monolayers at the edges of the plates. These dislocations have arbitrarily small Burgers vectors equal in magnitude to the jump in displacements produced by sliding graphene monolayers. To calculate the yield stress of metal-graphene composites, let us consider the very beginning of this process when the first dislocation dipoles nucleate at the edges of graphene plates. Let us write down the condition for the onset of GB sliding in the form: $\tau > \tau_{c2}$, where $\tau_{c2}$ is the critical shear stress for the sliding of graphene monolayers. We will also assume that along with GB sliding in the considered grain, intragranular plastic deformation also occurs via emission of lattice dislocations (see the emission of a dislocation from GB FG to GB DE shown in Figs. 1b–d). The process of dislocation emission from GBs is also activated when the tensile load exceeds certain critical stress.

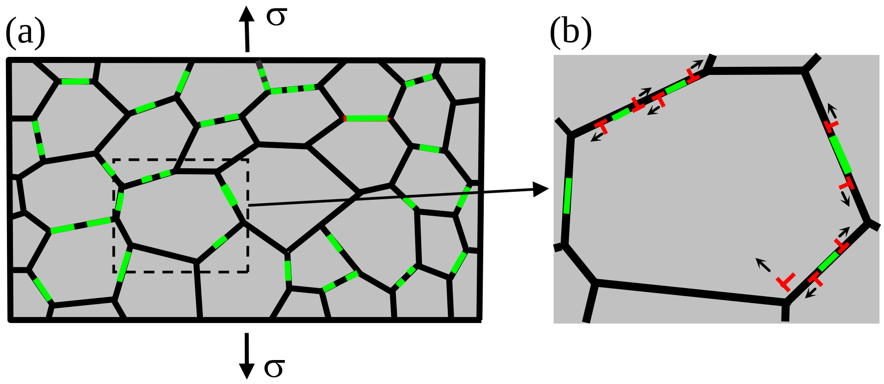


**Fig. 1.** Scheme of deformation of the composite "inhomogeneous UFG metal - graphene". (a) Composite sample under applied tensile load. (b) Plastic deformation in a grain of the composite material. Sliding along monolayers of graphene platelets (green rectangles) in GBs leads to the nucleation of edge dislocation dipoles with small Burgers vectors at the edges of such plates. Simultaneously, GBs can emit lattice dislocation into the grain (see dislocation in the grain interior). Dislocations slide towards the nearest triple junctions or towards each other

Let the grain sizes $d$ in the metal matrix be distributed according to the log-normal law [12,13]:

$$f(d,s)=\frac{1}{\sqrt{2\pi}d\,s}\exp\left[-\frac{1}{2}\left(\frac{\ln(d/d_m)}{s_d}\right)^2\right], \quad (1)$$

where $s_d$ is the dispersion of the distribution, $d_m$ is the median grain size. The average grain size $\bar{d}$ is related to the median as $\bar{d}=d_m\exp(s_d{}^2/2)$ [12]. Let the critical stress $\sigma_{c0}$ be the minimal stress initiating intragranular plastic deformation in pure metal (without graphene). Stress $\sigma_{c0}$ depends on the grain size according to the Hall–Petch relation, which in the case of the structure with grain sizes obeying log-normal distribution (1) has the form [12]:

$$\sigma_{c0}=\sigma_0+k_{\mathrm{HP}}\exp(-9s_d{}^2/8)\bar{d}^{-1/2} \quad (2)$$

where $\sigma_0$ and $k_{\mathrm{HP}}$ are the material parameters independent of the grain size.

Following [10], we assume that the main contribution to the yield stress increase is made by Orowan strengthening mechanism [14]. We also assume that graphene is present in a significant fraction of GBs, so that plastic deformation in the UFG phase occurs mainly due to the movement of dislocations between GBs containing graphene platelets. This means that graphene in the UFG phase should affect the motion of lattice dislocations by pinning dislocation segments emitted from GBs. In an elastically isotropic material characterized by a shear modulus $G$ and Poisson's ratio $\nu$, the stress $\sigma_{em}^{GB}$ required to unpin a dislocation segment initially pinned by obstacles situated at a distance $l$ from each other is written as [14]

$$\sigma_{em}^{GB}=\frac{MGb}{4\pi(1-\nu)}\left\{\left[2-\nu\left(3-4\cos^2\gamma_0\right)\ln\frac{l}{b}-2+\nu\right]\right\}, \quad (3)$$

where $M$ is the Taylor factor and $\gamma_0$ is the angle between the dislocation line and its Burgers vector. For metals with a face-centered crystal lattice: $M\approx 3.06$ and $\gamma_0=\pi/3$. In our case, graphene platelets serve as obstacles, so $l$ is the average distance between the nearest points of neighboring graphene platelets lying in the same GB. The expression for the distance $l$ is found in Ref. [10]:

$$l = \frac{\left(\pi^2 - 8 f_{GB}\right) L}{4\pi f_{GB}}, \tag{4}$$

where $f_{GB}$ is the fraction of the GB area occupied by graphene platelets. The parameter $f_{GB}$ can be expressed via the volume fraction $f_V$ of graphene in the composite as $f_{GB} = f_V \bar{d}/(3H)$, where $\bar{d}$ is the average grain size of the metal matrix. Introducing critical stress $\sigma_{c1}$ for the onset of intragranular plastic deformation of the composite, we can write: $\sigma_{c1} = \sigma_{c0} + \sigma_{em}^{GB}$.

Now we introduce the critical stress $\sigma_{c2}$ for the onset of GB sliding in GBs containing graphene platelets. This stress is related to the critical shear stress $\tau_{c2}$ as $\sigma_{c2} = M_1 \tau_{c2}$, where $M_1$ is some geometric factor similar to the Taylor factor *M*. We assume that if the critical stress $\sigma_{c1}$ for the emission of lattice dislocations from GBs is less than the critical stress $\sigma_{c2}$ of activation of slipping along monolayers of graphene platelets ($\sigma_{c1} < \sigma_{c2}$), then plastic deformation in the composite occurs through the movement of lattice dislocations through grain interior. If $\sigma_{c1} > \sigma_{c2}$, then in GBs containing graphene plates, GB sliding is activated, and GBs without graphene initiate deformation via the emission of lattice dislocations. Consequently, the critical stress $\sigma_c$ for the activation of plastic deformation in GBs is found from the following conditions: $\sigma_c = \sigma_{c1}$ for GBs without graphene platelets, and $\sigma_c = \min(\sigma_{c1}, \sigma_{c2})$ for GBs containing at least one graphene platelet.

Now, the yield stress $\sigma_y^{ufg}$ of the composite can be defined as the average value of the critical stress $\sigma_c$ [8,9]. This gives:

$$\sigma_y^{ufg} = f_{gr} \min(\sigma_{c1}, \sigma_{c2}) + (1 - f_{gr})\sigma_{c1}, \tag{5}$$

where $f_{gr}$ is the fraction of GBs containing at least one graphene platelets. The character of the graphene distribution over GBs is largely determined by the sintering technique used to manufacture the composite which can be quite different. Because of that, we can treat the parameter $f_{gr}$ as a random variable obeying a certain random distribution law. However, for our purposes, it is more convenient to work with another random parameter, namely, the fraction of the area of the random GB occupied by graphene (we denote it as $s_f$). Because values of $s_f$ range from 0 to 1 (by definition), it is natural to assume that $s_f$ obeys the beta distribution law, which is usually used for random quantities lying in the range of values from 0 to 1. Then the original parameter $f_{gr}$ can be expressed in terms of $s_f$ as

$$f_{gr} = 1 - \int_0^{f_s} \rho(s_f)\, ds_f, \tag{6}$$

where $\rho(s_f) = s_f^{\alpha-1}(1 - s_f)^{\beta-1}/\mathrm{B}(\alpha,\beta)$ is the probability density function of the beta distribution, $\mathrm{B}(\alpha,\beta)$ is the beta function, $\alpha$ and $\beta$ are the parameters of the beta distribution, $f_s$ is the average ratio of the area of one graphene platelet to the total area of one GB. We also assume that the GBs with $s_f < f_s$ do not contain graphene at all. In the case of a cubic grain shape, the value $f_s$ can be written in the form $f_s = 3\pi L^2/(4\bar{d}^2)$.

In our case, the average value of the fraction $s_f$ is known. It is equal to the fraction of the area of all GBs containing graphene platelets: $f_{GB} = f_V \bar{d}/(3H)$. And the average value of a random variable controlled by beta distribution law is equal to $\alpha/(\alpha+\beta)$. Thus, we have $\alpha/(\alpha+\beta) = f_{GB}$, and from here we obtain:

$$\beta = \alpha(1 - f_{GB})/f_{GB}. \tag{7}$$

Thus, we can specify the character of the graphene distribution over GBs by a single parameter $\alpha$.

## 3. Results and discussion

Thus, formulas (1)-(7) determine the yield stress of metal-graphene composites, which takes into account the inhomogeneous distribution of grain sizes in the metal matrix and the action of both dislocation mechanisms of plasticity and GB sliding along GBs containing graphene platelets. Using these formulas, we calculated the yield stress of the aluminum-graphene composite, characterized by the values of the parameters $G = 26.5$ GPa, $\nu = 0.34$, $\sigma_0 = 25$ MPa, $k_{\mathrm{HP}} = 0.04$ MPa·m$^{1/2}$, $b = 0.29$ nm. We also put $M_1 = M$, $L = 100$ nm and $\alpha = 3$. Figure 2 shows the dependences of the yield stress $\sigma_y^{ufg}$ of the aluminum-graphene composite on the average grain size $\bar{d}$ for various fixed values of the volume fraction $f_V$ of graphene (Figs. 2a,b) and different values of the dispersion $s_d$ of the grain size distribution. It is clearly seen that an increase in the dispersion of the grain size distribution leads to a significant yield stress decrease (in comparison with the structure with fixed grain size): with a dispersion $s_d = 0.6$, the decrease is ~ 15%. Thus, it can be concluded that the best mechanical properties are achieved with the smallest possible variation in the grain size of the metal matrix.

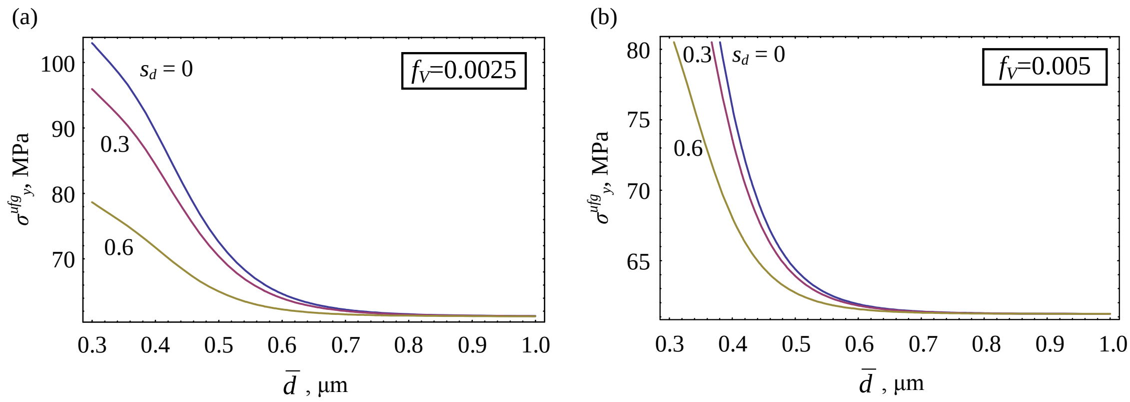


**Fig. 2.** Dependences of the yield stress $\sigma_y^{ufg}$ of the aluminum-graphene composite on the average grain size for various values of the volume fraction $f_V$ of graphene and the dispersion $s_d$ of the grain size distribution

Figure 3 shows the dependences of the yield stress $\sigma_y^{ufg}$ of aluminum-graphene composite on the volume fraction $f_V$ of graphene for grain sizes $\bar{d} = 300$ nm and 500 nm and various values of dispersion $s_d$ of distribution (1). As follows from Fig. 3, the yield stress grows only up to a certain critical value of graphene content and then starts decreasing. Such a dependence of the mechanical properties of the "metal-graphene" and "ceramics-graphene" composites is quite typical [4] and more realistic than the monotonic increase in the yield stress, which is obtained without taking into account GB sliding. This behavior is explained that, although an increase in the graphene volume fraction hampers the emission of dislocations from GBs, at the same time it enhances the negative contribution (decreasing yield strength) of intergranular slip to the yield stress. As a consequence, the presence of maxima on the curves is a consequence of the competition between two deformation mechanisms: intragranular dislocation slip and GB sliding. It is also seen (Fig. 3b) that the dependences tend to reach saturation with an increase in the graphene content, which occurs when all GBs contain graphene platelets.

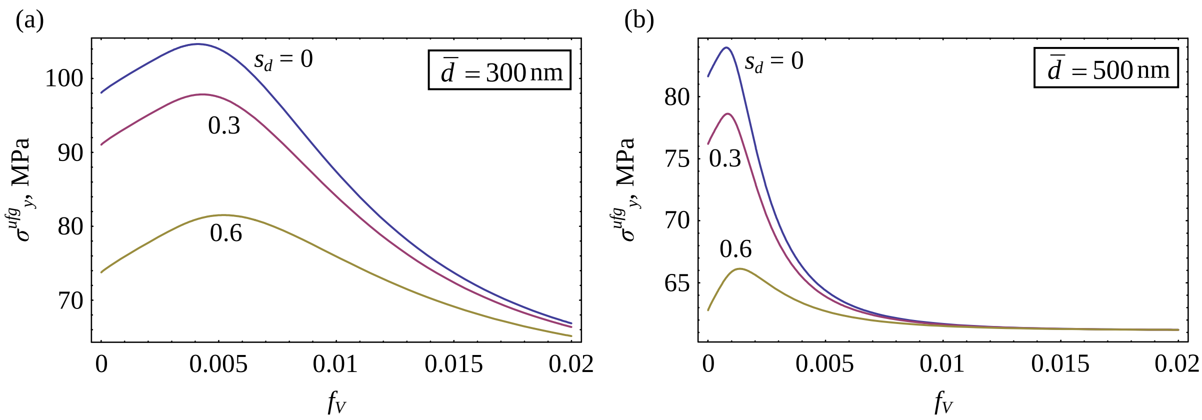


**Fig. 3.** Dependences of the yield stress of the aluminum-graphene composite on the volume fraction $f_V$ of graphene for average grain sizes $\bar{d} = 300$ nm and 500 nm and different dispersion values $s_d$ of the grain size distribution

## 4. Conclusions

Thus, our analysis shows that the presence of dispersion in the grain size distribution of the metal matrix leads to a decrease in the yield stress. This means that during the synthesis of composites, special attention should be paid to reducing this dispersion. It was also shown that taking into account GB sliding leads to a more realistic (qualitatively consistent with experimental data [4]) dependence of yield strength on the graphene content in the composite, which manifests itself in a decrease of the yield strength when a certain critical value of the graphene volume content is exceeded.

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## THE AUTHORS

**Bobylev S.V.**
e-mail: bobylev.s@gmail.com
ORCID: 0000-0003-0894-4308

**Sheinerman A.G.**
e-mail: asheinerman@gmail.com
ORCID: 0000-0001-9909-2950