Continuum mesoscale modelling of nanocrystalline fcc metals under shock-loading using an spectral formulation fed by molecular dynamics results

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Abstract. In this paper we present a micromechanical approach based on Fast Fourier Transforms to study the role played by dislocation glide and grain boundary (GB) accommodation in the determination of the yield strength of nanostructured materials under shock. For this, we construct unit cells representing self-similar polycrystals with different grain sizes in the nanometer range and use local constitutive equations for slip and grain boundary accommodation. We study the effect of grain size and shock pressure on the local and effective behavior of nanostructured fcc materials with parameters obtained from experiments and atomistic simulations. Predictions of a previous pressure-sensitive model for the effective yield strength behind a shock front are improved by considering strain partition between slip and GB activity.

1. INTRODUCTION

The yield strength of polycrystalline materials depends strongly on their grain size. For aggregates with grains in the micron range and above, the yield stress increases inversely with the square root of the grain size. This dependence, known as Hall-Petch effect, has been explained in terms of dislocation interaction with the crystal lattice, with other dislocations, and with grain boundaries (GBs). For coarse-grained (CG) aggregates, the GBs main role is to act as barriers to dislocation motion and promote the formation of pile-ups in the bulk of the grains, (e.g. see [1]). In spite of the importance that this barrier effect has in the determination of the mechanical properties of the aggregate, in polycrystals with grains in the micron range and above, the highly disordered GB regions represent only a negligible volume fraction of the material. On the contrary, when the grain dimensions approach nanometer sizes, the volume fraction of GBs become significant. Then, the GB regions start to play an active role in accommodating deformation (e.g. [2]), not only at very low deformation rates when diffusional flow at GBs becomes significant regardless of the grain size, but at any applied strain rate. This GB accommodation may involve local shear shuffling of atoms, GB sliding, and/or diffusive processes that happen near the GBs. Regardless of the specific atomic displacements involved in GB accommodation, given the highly disordered character of the GB regions, the microscopic mechanisms associated with GB activity are intrinsically different from the ones associated with plastic deformation by dislocation motion that takes place in the bulk of the grains. A sound constitutive equation of GB deformation would not involve directionality, as it is indeed the case of the usual constitutive description of crystal plasticity. Also, due to the short range of the interactions involved in GB accommodation, the stress required to activate this mechanism should be essentially independent of the grain size of the aggregate. Furthermore, the constitutive equations that describe dislocation glide in the bulk of the grains and accommodation at GBs will have, in general, different sensitivities to temperature, strain-rate and pressure. Regarding pressure sensitivity, given that both GB accommodation (e.g. [3]) and dislocation glide (e.g. [4]) are likely to become more difficult as hydrostatic pressure increases, the overall effect of pressure on plastic deformation of materials with nano-sized grains is going to be determined by the relative values of the corresponding sensitivity factors.

In a recent contribution, Bringa et al. [5] interpreted the stress profiles obtained by Molecular Dynamics (MD) simulations of shocks in nanocrystalline Cu in terms of different shock pressure

dependences of slip and GB accommodation. These authors assumed a linear dependence with shock pressure of the flow stresses behind the shock front associated with both dislocation slip (i.e. Steinberg-Guinan model for shock-induced dislocation plasticity [4]), and GB accommodation (inspired in Mohr-Coulomb's law for plasticity of amorphous materials [3]). Adjusting the corresponding pre-factors using MD results, they proposed that the yield strength is given by the minimum between the flow stresses associated with dislocation slip and GB accommodation. Both mechanisms depended on grain size and applied pressure. This simple approach proved to be compatible with the reported Cu MD results and Ni experiments, suggesting that, due to the apparent suppression of the softening associated with GB accommodation, the flow stress can reach ultrahigh values, at the high pressures produced by shock-loading. However, as it was already acknowledged in [5], the use of a simple minimum criterion to decide whether the macroscopic yield strength is equal to the flow stress associated with either slip or GB accommodation, does not consider the likely occurrence of strain partition between both mechanisms.

Based on the above model, in this work we report a new micromechanical formulation to study the effect of grain size and shock pressure on the yield strength of fcc nanocrystalline materials. The proposed model is a full-field approximation that takes into account the actual topology of the aggregate consisting in bulk crystalline regions surrounded by a GB percolating phase. Both regions co-deform plastically according the following constitutive behaviors: a) for grain interiors: crystal viscoplasticity, with Hall-Petch grain size dependence and Steinberg-Guinan pressure dependence of the flow stress for slip activation; b) for grain boundaries: isotropic viscoplasticity with flow stress independent of grain size, and Mohr-Coulomb pressure dependence. The length-scale of the problem is given by the GB thickness, taken to be 1 nm. Instead of using Finite Element analysis, we use here a very efficient approach based on the Fast Fourier Transform (FFT) algorithm. This FFT-based solution of a unit cell problem for a representative volume element (RVE) gives the local mechanical fields that develop inside heterogeneous materials in great detail [6,7]. Periodic boundary conditions, required for this type of spectral approximation, are sufficient for the kind of parametric study that we want to perform. It is worth noting that, given the viscoplastic character of the present approach, it is not intended to describe elastic effects. In particular, no attempt is made here to develop a micromechanical modeling of the complex problem of propagation of elastoplastic waves in shocked materials. Instead, the present model aims at considering the yield strength resulting from the microstructural changes left in the material after the passage of a shock front.

2. CONSTITUTIVE MODEL

The FFT-based formulation is based on the solution of a unit problem for an RVE with periodic boundary conditions. Briefly, it consists in finding a strain rate field, associated with a kinematically admissible velocity field, that minimizes the average of local work rate, under the constraint imposed by the strain compatibility condition (for details, see [6]). In the present case, the unit cell represents an aggregate of single crystal grains with prescribed orientations, surrounded by outer layers of percolating grain boundary regions. The particular orientation of each single crystal determines different anisotropic plastic properties from grain to grain, while due to their intrinsic disordered character the GBs are assumed to have a homogeneous isotropic behavior throughout the interconnected "GB phase". A 3-D implementation of the FFT-based method requires to discretize a cubic unit cell using a regular grid of NxNxN Fourier points (FPs). In this work we adopted N = 128, resulting in 2097152 discretization points. Each FP belongs either to a grain interior, or to the GB phase. The length-scale associated with the RVE is determined by the GB thickness, taken to be 1 nm. Self-similar RVEs were generated as follows: a) the 3-D unit cell is first partitioned into grains by Voronoi tessellation. This discrete Voronoi procedure consists in randomly distributing 27 grain nuclei in the cubic unit cell and assigning each FP to its nearest nucleus, accounting for periodic boundary conditions across the RVE limits; b) random crystallographic orientations are assigned to each grain; c) in order to determine the FPs belonging to the GB phase, the sets of Fourier points with up to first, second and third neighbours belonging to a
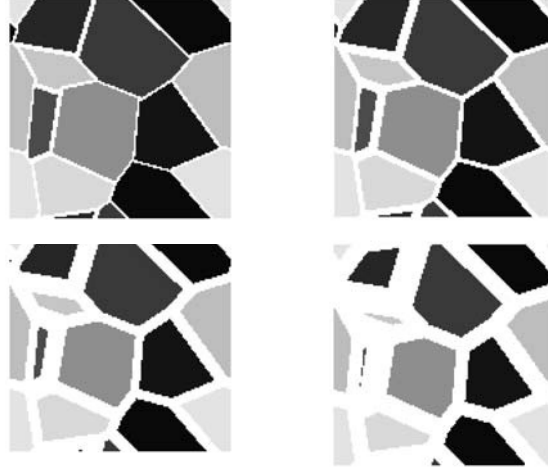
Figure 1. 2-D sections (YZ plane at X = 64) of to the four self-similar 3-D RVEs. In white: grain boundaries; in gray: grains. Lighter (darker) shades represent softer (harder) grains under uniaxial tension or compression.

different grain are determined. A fourth set can be obtained by choosing one of the two points of each pair of first-neighbour FPs belonging to different grains. Next, by assigning these four sets of Fourier points to the GB phase, four self-similar RVE are determined, with the same number of grains, and the same topology and crystallographic texture, but differing in the volume fraction of GB phase and in grain size (the resulting RVEs have 29, 15, 7 and 5 nm average grain size).

The local constitutive equation for a point $\mathbf{x}$ belonging to a grain interior is given by the following power law for single crystal viscoplasticity:

$$\dot{\varepsilon}(\mathbf{x}) = \dot{\gamma}_o \sum_s \boldsymbol{\mu}^s(\mathbf{x}) \left(\frac{|\boldsymbol{\mu}^s(\mathbf{x}) : \boldsymbol{\sigma}(\mathbf{x})|}{\tau_o^s} \right)^{n_{GI}} \times \text{sign} \left[\boldsymbol{\mu}^s(\mathbf{x}) : \boldsymbol{\sigma}(\mathbf{x}) \right] \tag{1}$$

where $\dot{\varepsilon}(\mathbf{x})$ and $\boldsymbol{\sigma}(\mathbf{x})$ are the local strain rate and stress tensors and $\boldsymbol{\mu}^s(\mathbf{x})$, $\dot{\gamma}^s(\mathbf{x})$, $\tau^s(\mathbf{x})$ and τ_o^s are, respectively, the orientation-dependent Schmid tensor, the shear rate, the resolved shear stress, and a reference shear stress, of slip system (s). The pre-factor $\dot{\gamma}_o$ is a reference shear rate, and n_{GI} is the power-law exponent. The local constitutive behavior for a point $\mathbf{x}$ belonging GB phase is here assumed to be given by the following J_2- isotropic power law for nonlinear isotropic materials:

$$\dot{\varepsilon}(\mathbf{x}) = \frac{3\dot{\varepsilon}_o}{2\sigma_{eq}(\mathbf{x})} \left(\frac{\sigma_{eq}(\mathbf{x})}{\sigma_o} \right)^{n_{GB}} \boldsymbol{\sigma}(\mathbf{x}) \tag{2}$$

where $\sigma_{eq}(\mathbf{x})$ and σ_o are the von Mises equivalent of the stress and a reference equivalent stress, respectively, $\dot{\varepsilon}_o$ is a reference strain rate, and n_{GB} is a power-law exponent. In what concerns the explicit pressure and grain size dependence of our state variables τ_o^s and σ_o, we adopt here Bringa et al [24] approach, with slight modifications:

$$\tau_o^s = \frac{C\,(G_o + \beta P)(d/d_o)^{-0.5}}{\overline{M}_{CG}}; \quad \sigma_o = \sigma_a + \alpha P \tag{3}$$

where P is the shock pressure, G_o is the shear modulus at zero pressure, C is material-dependent adjustable parameter, σ_a is the flow stress of the amorphous material at zero pressure, α and β are pressure-sensitivity factors, d_o is a reference grain size and $\overline{M}_{CG}$ is the average Taylor factor. The value of σ_a has been estimated to be 0.9 GPa for Cu. Using Molecular Dynamics, the resulting elastic

constants for Cu as a function of pressure (at $T = 0$ K) gives $G_o = 45$GPa, and $\beta \approx 1$ for P < 60 MPa, in agreement with experiments. The value of the pressure-sensitivity factor for GB accommodation was assumed to be $\alpha = 0.04$. The adopted reference grain size was $d_o = 30$ nm and, by assuming that the flow stresses of slip and GB accommodation are equal when the grain size is d_o, a value of C = 0.04 was obtained.

3. RESULTS

3.1 Grain size dependence of local deformation fields

Figure 2a shows, for the four self-similar RVEs, the 2-D section maps of local von Mises equivalent strains, relative to the applied macroscopic von Mises equivalent strain, for the case of axisymmetric compression along the z-axis, with no superimposed pressure (P = 0 in Eqs. 3). The 2-D sections shown in Fig. 2 correspond to the same YZ cuts as the ones displayed in Fig 1 (note that in these plots the compression axis lies in the vertical direction). The main observation is that the strain is concentrated at GBs, reaching 10 times the macroscopic strain, while significant portions of the grain interiors undergo local strains which are less than the applied macrostrain. This trend is more marked as grain size decreases, e.g. see the grain in the center (marked "A"). Another interesting feature is the grains that deform the most (e.g. grain "A") are not necessarily the softest (grain "A" has an intermediate yield strength, see Fig. 1) but the most ubiquitous. For instance, in the 29 and 15 nm cases, grain "A" provides a link between GBs well oriented for the propagation of a transgranular deformation band at approximately 50 degrees with respect to the compression direction. At smaller grain sizes, the fine structure of the strain field inside the GBs can be observed. Interestingly, the strain seems to concentrate near the transition zones between GB and the bulk of the grains, especially near triple junctions. This may indicate the occurrence of local shears consistent with grain boundary sliding and grain rotation.

Figure 2b shows the relative activities (strain partition) in the grain interiors and the GB phase, along with the corresponding volume fractions, as a function of grain size, corresponding to the cases shown in Fig. 2a. The relative activities are defined as the averages of local strains over every Fourier point belonging to each region, normalized by the overall strain. Consistent with the results of Fig. 2a, the
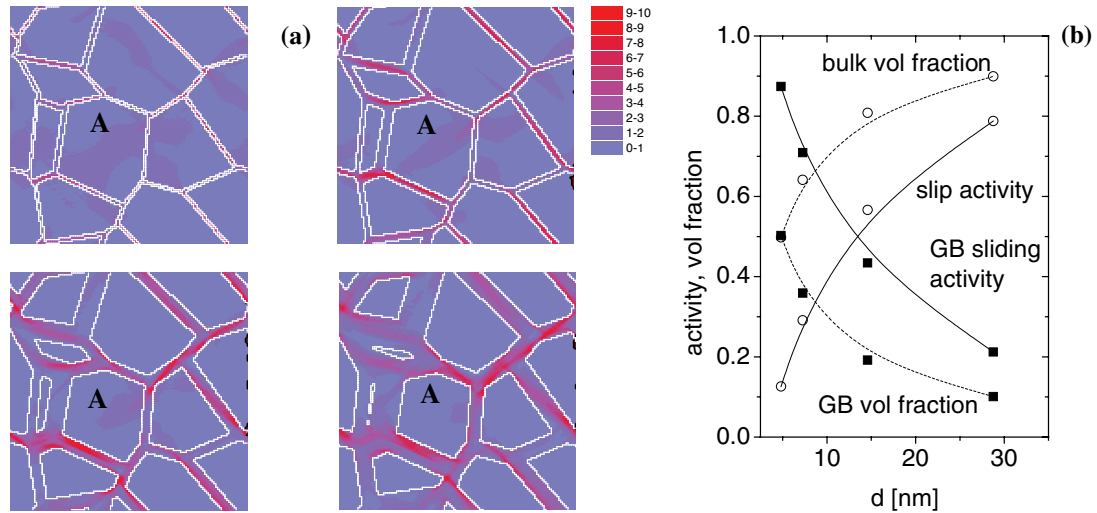

Figure 2. (a) Predicted 2-D section maps (YZ plane at X = 64) of local von Mises equivalent strains, relative to the applied macroscopic von Mises equivalent strain, for axisymmetric compression along the z-axis. (b) Strain patition and volume fraction as a function of grain size.

relative activity at GB exceeds the corresponding GB volume fraction. This trend is more marked as the grain size decreases. At 5 nm, the grain interiors contribute only with 10% to the overall strain, while they still represent almost half of the volume. This result is consistent with most of the reported MD simulations at these grain sizes [2, 5].

3.2 Pressure dependence of yield strength under shock-loading

Figure 3a shows the pressure and grain size dependence of the yield strength of nanostructured samples, as predicted by the present model, using the constitutive equations and parameters for Cu given by Eqs (1-3) and in the text of section 2. The main characteristics of these curves are: a) as expected from the type of pressure dependence adopted for the local constitutive behaviors (Eqs. 3), the yield stress increases with shock pressure, for all grain sizes, b) the Hall-Petch inversion is observed at around 10 nm for all pressures, c) the line joining the yield strength maxima exhibits a negative slope. This result is the outcome of the present model that considers a detailed partitioning of strain between GBs and grain interiors and is therefore to be compared with Fig. 1 of Ref. [5] in which a qualitative minimum criterion for the flow stress was used. From this comparison the differences in the location and the slope of the Hall-Petch inversion curves are apparent. The inversion curve predicted by the present model is located at around 10 nm with very little variations as pressure increases, while from Fig. 1 of Ref. [5] this curve can be located at around 30 nm, with a 5 nm decrease from 0 to 22 GPa. Thus, the significant decrease of the Hall-Petch inversion point with increasing pressure reported in [5] that would maximize the yield strength increase effect of these materials under shock loading for smaller grain sizes as pressure increases, seems to very marginal in the context of the present, more refined approach.

Figure 3b shows the effect of pressure on the strain partition between slip and GB accommodation (for sake of clarity, only the relative slip activity curves are shown, the GB relative activity can be obtained subtracting the slip activities from 1). As shock pressure increases, slip activity increases. The inserted figure shows in more detail the predictions for the smallest grain sizes. While for a grain size of 5 nm the slip activity at 22 GPa shows about 50% increase with respect to the P = 0 case, for larger grain sizes this relative increase is only marginal. Given that the dislocations are the carriers of plastic deformation inside the grains, the significant increase of slip activity at smaller grain sizes and higher pressures should involve a larger number of dislocations gliding through the grains. Such increase in the number of dislocations as grain size decreases and shock pressure increases has also been found in MD simulations [5].
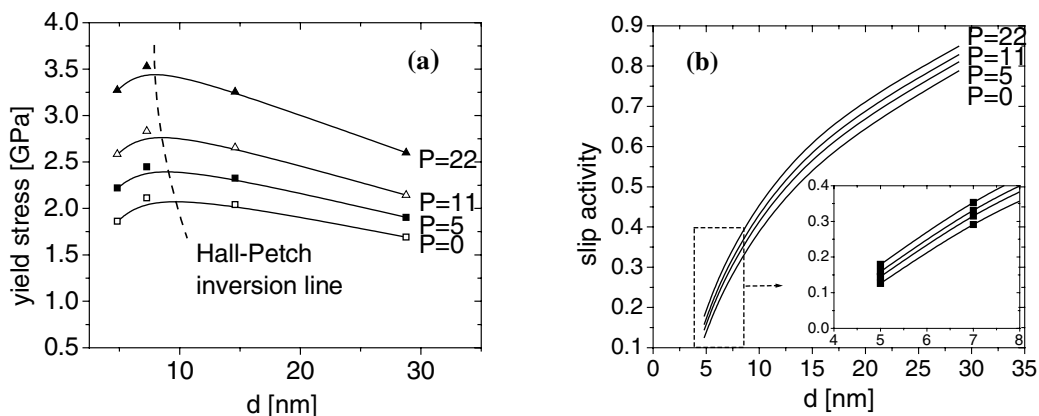

Figure 3. (a) Predicted yield strength, and (b) slip activity, as a function of grain size for different shock pressures (in GPa). Constitutive parameter values for nanocrystalline Cu, after Bringa et al [5].

4. CONCLUSIONS

We have presented results of a micromechanical approach based on Fast Fourier Transforms to study the role played by dislocation glide and GB accommodation in determining the plastic behavior of nanostructured materials. The local strain fields exhibit a strong concentration in GBs. In grain interiors, the strain is higher in grains which can serve as links between GB regions well-oriented with respect to the direction of maximum macroscopic shear, to form transgranular deformation bands. Under shock-loading conditions, the qualitative pressure-sensitive model of Bringa et al. [5] was physically grounded by explicitly considering a material with two different constitutive behaviors, and by solving the strain partition between slip and GB activity. As expected from the type of pressure dependence adopted for the local constitutive behaviors, the predicted yield stress versus grain size curves are shifted upwards as the shock pressure increases, while the predicted Hall-Petch inversion occurs (for the set of parameters adopted here) around 10 nm and shows a weak decrease with pressure. The relevance of these results appears clearly when we note that current constitutive models for shock-loading [4] do not typically include grain size effects. These effects would be significant, for instance, for the design of the nanostructured targets under consideration for the National Ignition Facility [8].

References

[1]   M.A. Meyers, K.K. Chawla, Mechanical Behavior of Materials. Prentice-Hall, Upper Saddle River, NJ (1999).
[2]   Schiøtz J., Jacobsen K.W., Science 301, 1357 (2003).
[3]   Lund A.C., Schuh C.A., Acta Mater. 53, 3193 (2005).
[4]   Steinberg D.J., J. Appl Phys. 51, 1498 (1980).
[5]   Bringa E.M., Caro A., Wang Y.M.,Victoria M., McNaney J.M., Remington B.A., Smith R.F., Torralva B.R., Van Swygenhoven H., Science 309, 1838 (2005).
[6]   Michel J.C., Moulinec H., Suquet P., Comput. Model. Eng. Sci. 1, 79 (1999).
[7]   Lebensohn R.A., Acta mater 49, 2723 (2001).
[8]   Dittrich T.R. et al., Laser Part. Beams 17, 217 (1999).