Instability of a molybdenum layer under deformation of a CuMoCu laminate by high-pressure torsion

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A B S T R A C T

High-pressure torsion of a laminate consisting of a layer of monocrystalline molybdenum sandwiched between two layers of copper was investigated. Computed tomography showed that at sufficiently large angles of rotation of the anvils the molybdenum layer loses planarity. It develops periodic folds and vortices, leading to ruptures. EBSD analysis revealed the formation of a blocky substructure in the molybdenum layer and a pronounced fragmentation of copper sheets, with the formation of a significant proportion of high-angle grain boundaries. The proposed mathematical model of the process accounts for the observed phenomena qualitatively. It is based on the gradient plasticity theory and predicts the loss of stability of the harder molybdenum layer when the shear strain in the laminate exceeds a critical value.

Synthesis of nanocomposites by High-Pressure Torsion (HPT) of metal laminates has been attracting a growing attention of researchers (see [1]). A crucial condition for the synthesis is mechanical mixing of the components [2], which is caused by the loss of stability of the layers during HPT, followed by multiple breakage of these layers [3,4].

There is no generally accepted understanding of the physical mechanisms of the instability. Here we report new data that may shed light on its origin and the ensuing consequences for HPT of a copper/molybdenum laminate.

A sample for the HPT deformation was prepared as a stack consisting of two layers of polycrystalline Cu and a monocrystalline Mo sheet sandwiched between them. This sheet was obtained by orientation rolling of a Mo single crystal and had a surface parallel to the (001) plane of the body-centered cubic (bcc) structure.

Computed tomography (CT) measurements on the deformed sample were performed using a ZEISS Xradia 520 Versa X-ray microscope operated at 140 V. A rod-shaped specimen for the CT investigations had the dimensions of 1.5 mm × 1.5 mm × 0.5 mm. It was cut from a penny-shaped HPT sample at a mid-radius location. The CT scans were reconstructed using the ZEISS proprietary software followed by the component segmentation using the Dragonfly software (ORS Inc.).

Structural characterization of the samples after HPT, including electron backscatter diffraction (EBSD), was performed on a ZEISS Auriga 60 scanning electron microscope (SEM) operated at 20 kV. Specimens for SEM and EBSD investigation were prepared by mechanical grinding to 1 μm roughness followed by Ga ion beam polishing.

Fig. 1 shows the geometry of the molybdenum layer after four turns of the anvils.

The 3D reconstruction in Fig. 1b shows that the surface of the Mo layer is wavy, like the sea in a storm. At some places, the layer is ruptured, and Cu phase fills the volume that is void of Mo. For all the stochastic nature of the mixing process, the pictures shown in Fig. 1 look quite regular. This suggests that a certain physical mechanism governs the shape of the structural components inside the sample at the stage of transition from a planar laminar flow to chaotic mixing of the components.

According to the hypothesis suggested in [4,5], the transition to turbulent plastic shear flow is caused by local flow deceleration, which

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causes a torque stress and rotation in the mass transport. A mathematical tool for studying this phenomenon is offered by the gradient theory of plasticity [6]. Based on some insights obtained in this work, we propose a model that provides a qualitative description of the initial stage of distortion of the laminate layers during HPT.

According to [6], the curvature of a material's fibers is given by

$$\chi \sim \frac{d\gamma}{dx} \tag{1}$$

where $\chi = \sqrt{\frac{2}{3}\chi_{ni}\chi_{ni}}$, $\chi_{ni} = \frac{\partial\theta_n}{\partial x_i}$ is the plastic component of the curvature tensor, $\theta_n = \frac{1}{2}\epsilon_{njk}u_{k,j}$ is the plastic rotation vector, $u_{k,j}$ is the antisymmetric part of the plastic displacement gradient, and ϵ_{njk} is the Levi-Civita symbol.

Our approach builds on the work [6,7] relating the lattice curvature of a single crystal to spatial inhomogeneity of strain, which in turn can be described in terms of the distribution of geometrically necessary dislocations (GNDs). According to [6], the GND density ρ_G depends linearly on χ, which is expressed by

$$\chi \sim \rho_G b, \tag{2}$$

where b is the magnitude of the Burgers vector.

During the HPT of laminates, the hard layers inhibit the shear in the soft layers. This leads to a strain gradient, which can be estimated as follows. When the anvils are rotated through an angle φ, the average shear strain γ at any point of the sample undergoing laminar flow is given by

$$\gamma = \frac{r\varphi}{h}, \tag{3}$$

where r is the distance from a point within the sample to the rotation axis and h is the sample thickness. The shear strain γ can be expressed as follows:

$$\gamma = (1 - \vartheta)\gamma_s + \vartheta\gamma_h, \tag{4}$$

where γ_s and γ_h are the shear strains of the hard and soft layers, respectively, and ϑ is the fraction of the hard material in the sample. According to the classical theory of plasticity [7], the relations

$$\gamma_s \sim \frac{\tau_s}{\sigma_s}, \gamma_h \sim \frac{\tau_h}{\sigma_h}, \tag{5}$$

hold, where τ_s and τ_h denote the shear stresses in the soft and the hard layers, while σ_s and σ_h are the flow stresses in the respective layers. From the continuity condition for shear stress at the interface between the layers, $\tau_s = \tau_h$, [7] and Eq. (5) one obtains

$$\frac{\gamma_s}{\gamma_h} = \frac{\sigma_h}{\sigma_s} \tag{6}$$

Eqs. (4) and (6) then yield

$$\gamma_h = \frac{\gamma}{\vartheta + (1 - \vartheta)k} \tag{7}$$

$$\gamma_s = \frac{k\gamma}{\vartheta + (1 - \vartheta)k} \tag{8}$$

where $k = \frac{\sigma_h}{\sigma_s}$.

As the inequality $k > 1$ holds, it follows from Eqs. (7) and (8) that the strain in the soft layer is greater than that in the hard one. This is corroborated by experiments that show that grain refinement with the formation of large-angle grain boundaries is more pronounced in copper than in molybdenum. The distribution of molybdenum in the sample after two anvil revolutions is visualized in Fig. 2. The crystallographic orientation of copper after deformation is shown in Fig. 2 (c). The grains are seen to be highly deformed and their average size is reduced to about

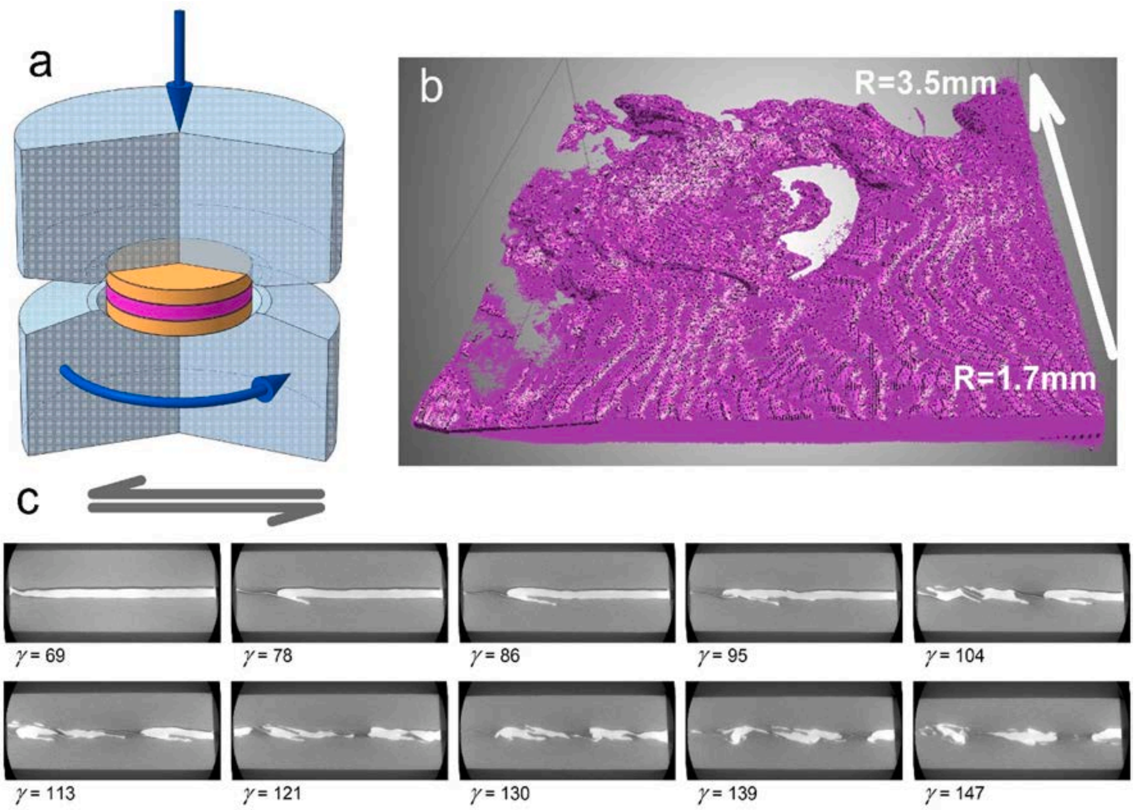

Fig. 1. Rendering of the molybdenum layer after four turns of the anvils: (a) schematics of HPT; (b) 3D reconstruction; the arrow shows the direction from the center to the periphery of the sample; (c) a cross-section normal to the radius; the bright phase is Mo; the arrows show the shear directions.

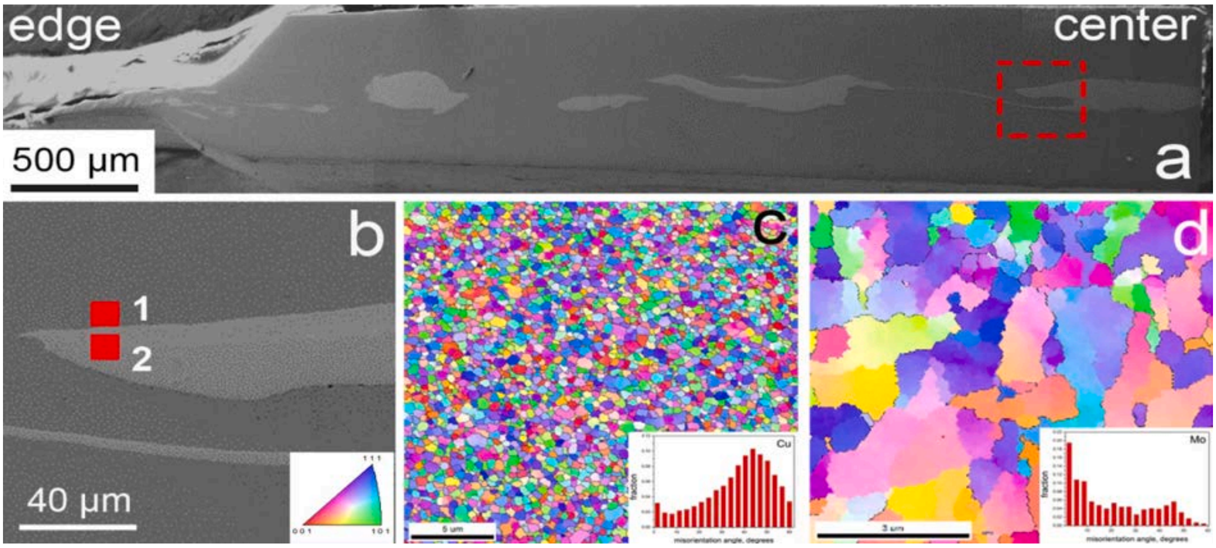

Fig. 2. (a) SEM image illustrating mutual arrangement of Mo and Cu layers after HPT; (b) zoomed-in part of (a) from the dashed rectangle shows two areas for EBSD mapping; (c) EBSD map of Cu (area 1 in b) and corresponding GB misorientation distribution (insert); (d) EBSD map of Mo (area 2 in b) and the corresponding GB misorientation distribution (insert). The insert in (b) is the color code for the EBSD maps.

450 nm. The grain structure exhibits a large proportion of high angle grain boundaries (>15° misorientation). Fragmentation of initially monocrystalline Mo can be seen in Fig. 2 (d).

As a measure of the strain gradient, we take the following expression:

$$\frac{d\gamma}{dx} \quad \frac{\gamma_s \quad \gamma_h}{\Delta}, \tag{9}$$

where Δ is a characteristic layer thickness.

Substituting Eqs. (3), (7), and (8) into Eq. (9), we obtain:

$$\frac{d\gamma}{dx} \quad \frac{(k \quad 1)}{\vartheta + (1 \quad \vartheta)k} \frac{r\varphi}{h\Delta} \tag{10}$$

From Eqs. (1) and (10) the lattice curvature is estimated as

$$\chi \quad C \frac{(k \quad 1)}{\vartheta + (1 \quad \vartheta)k} \frac{r\varphi}{h\Delta} \tag{11}$$

where C is a dimensionless factor. Equation (11) shows that under HPT of laminates of metals with different flow stress ($k > 1$), the layers become more and more curved in the shear plane (in sections normal to the sample radius). This is due to the accumulation of GNDs whose density is given by the equation

$$\rho_G \quad C \frac{(k \quad 1)}{\vartheta + (1 \quad \vartheta)k} \frac{r\varphi}{h\Delta b} \tag{12}$$

Equation (12) determines the *average* value of the GND density at a distance r from the rotation axis of the anvils. The value of ρ_G at a given point should depend on its tangential coordinate, and this can be substantiated by the following reasoning. At the interface, the hard layer constrains the deformation of the soft one. This means that the soft layer is compressed and the hard one is under tension. Accordingly, buckling of their interface towards the hard layer occurs. If ρ_G did not depend on the tangential coordinate, buckling would not be possible. Instead, a uniform decrease in the thickness of the hard layer would take place. Most likely, upon reaching a certain critical value, ρ_G^*, the uniform angle-independent GND distribution loses its stability, and a periodic distribution emerges. A re-arrangement of ρ_G occurs at the interface, so that the layer thickness varies with the tangential coordinate.

These considerations lead us to the conclusion that periodic constrictions should appear in the hard layer, dividing it into fragments. According to [4], these fragments will rotate in the shear flow of the soft material. This scenario is consistent with our experimental data, which demonstrate both periodic fragmentation of the hard layer (Fig. 1b) and the 'frozen' rotation of its fragments (Fig. 1c). The proposed model of laminate deformation contains only one parameter, Δ, which has the dimension of length. We can interpret this parameter as the material length scale of the gradient theory of plasticity [6]. From the dimensional considerations, we can assume that the characteristic size of the fragments of the hard layer should be of the order of Δ. Fig. 1 confirms this conjecture.

Using Eq. (12), the condition for the onset of the described structural changes can be expressed in terms of a critical anvil rotation angle

$$\varphi^* \quad \frac{\rho_G^*}{C} \frac{\vartheta + (1 \quad \vartheta)k}{(k \quad 1)} \frac{h\Delta b}{r} \tag{13}$$

It is seen that φ^* increases with decreasing distance from the anvil rotation axis. This means that the region with layers distorted by HPT expands from the periphery to the center of the sample. As

$$\frac{d\varphi^*}{dk} \quad \frac{1}{(k \quad 1)^2} < 0 \tag{14}$$

holds, it follows from Eq. (13) that an increase in the ratio of the flow stress values of the components promotes the fragmentation of the hard layer. The same effect can be obtained by reducing the layer's thickness.

We conclude that the stability of the laminate under HPT is determined by the quantity ρ_G, which plays the role of an order parameter [8], in that it governs the stability of the shear deformation and can be used to predict the onset of instability. At this stage, the critical GND density, ρ_G^*, can be considered as a model parameter.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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