Int. Journal of Refractory Metals and Hard Materials 43 (2014) 59–63

Contents lists available at ScienceDirect

Int. Journal of Refractory Metals and Hard Materials

journal homepage: www.elsevier.com/locate/IJRMHM

Structure and hardness of molybdenum upon deformation under pressure at room and cryogenic temperatures

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ARTICLE INFO

Article history:
Received 25 June 2013
Accepted 29 October 2013

Keywords:
High pressure torsion
Microstructure
Twinning
Deformation temperature

ABSTRACT

This article considers the effect of the stacking fault energy and deformation temperature on the structure formation of a molybdenum single crystal with an initial orientation (110). The deformation is performed via high-pressure torsion at room and cryogenic temperatures, 290 and 80 K, respectively. The structural analysis and hardness behavior of the studied material indicate that lowering the temperature does not change the deformation mechanism from dislocation slip to twinning. We observe the localization of the deformation, band structure formation, and the formation of misoriented microcrystallites. The initial single crystal converts into a polycrystal immediately after the early stages of deformation ($e > 2$). Misoriented submicron-scale elements are formed at 290 K at a smaller true strain than those formed at 80 K. This deformation results in the structural refinement at both temperatures but does not allow for the formation of a homogeneous misoriented structure with high-angle boundaries. A decrease in the deformation temperature leads to a fall in the plasticity of molybdenum, that is, the application of high pressure fails to prevent its brittle fracture.

1. Introduction

For several decades, researchers have been interested in the deformation-induced structural changes, which lead to the formation of a nanocrystalline structure in metallic materials. As a rule, fcc metals are used for such studies [1–7]. The use of new technologies can result in unexpected discoveries, even though the structural evolutions of many fcc metals have been extensively studied up to the maximum achieved refined states. For example, recent studies suggest that the transformation of fcc metals in the nanocrystalline state activates twinning via the emission of partial dislocations [8,9]. According to molecular dynamic simulations [10], such a mechanism is also possible in bcc nanocrystalline molybdenum. Interestingly, a similar twinning mechanism is experimentally observed in fcc metals, but there are no published data on this mechanism in bcc metals. Regardless of the refinement degree of a structure, twinning is a prevalent deformation mechanism in metals and alloys that have a low stacking fault energy (SFE) and a limited number of slip systems. In materials with medium and high SFE, twinning is activated by decreasing the deformation temperature and increasing the deformation rates [5,11–13].

Currently, there are few studies devoted to bcc metals, especially to refractory metals, because these metals are difficult to deform, their yield strength is highly temperature-dependent, they have high Peierls-Nabarro barriers, and they are highly sensitive to impurities. Therefore, refractory bcc metals can be deformed only at elevated temperatures [14–17]. A well-known method of plasticizing and reducing the ductile–brittle transition temperature of refractory metals is refining the grain structure via severe plastic deformation. The application of a high pressure allows for the deformation of materials without fracturing them at room temperature [18,19]. Nevertheless, their deformation at cryogenic temperatures is a technically challenging problem.

The fact that the SFE of bcc metals lies in a fairly wide range (from medium to high) allows one to study the effect of SFE on their structure evolution. Data on the twinning activation in Fe (SFE $\approx$ 150 mJ/m^2 [20]) deformed by high pressure torsion (HPT) were obtained at the temperature of liquid nitrogen upon deformation [13]. The SFE of molybdenum (>300 mJ/m^2) is twice that of Fe. It is interesting to examine the structure evolution of molybdenum upon HPT because this unique method allows for ultra-high strains without fracturing. In addition, we assume that a low deformation temperature of 80 K may both intensify the structure refinement and change the deformation mechanism from dislocation slip to twinning.

2. Materials and methods

The single-crystal molybdenum was subjected to deformation at 80 K (cryo-deformation) and 290 K (room-temperature deformation) under a quasihydrostatic pressure of 12 GPa in Bridgman anvils [21]. Prior to cryogenic deformation, the unit of anvils together with a sample was drowned into a bath filled with liquid nitrogen, which was a part of the Bridgman installation, and kept for 25–30 min until full cooling. The temperature was controlled with a thermocouple attached to a stationary anvil. The minimum temperature achieved was 80 K and it remained unchangeable with increasing the cooling time up to several hours. As soon as the desired temperature had been achieved, we

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http://dx.doi.org/10.1016/j.ijrmhm.2013.10.022

started the deformation. When evaporated, nitrogen was poured into the bath to constantly keep the anvils with a sample in the medium of liquid nitrogen in the process of the deformation. The pressure was released after the slow natural heating of the anvils with the sample to the room temperature. Before the deformation, disk-like samples have a diameter of 5 mm and a thickness of 0.3 mm. The sample plane was parallel to the (110) one of the single crystal. The rotation angle of a movable anvil was varied from 0 (upsetting) to 10 revolutions at a rotation velocity of $\omega = 0.3$ rev/min. The true strain was calculated taking into account upsetting, the rotation angle of the anvil (φ), and a distance to the center of a sample (R_i):

$$e = \ln\left(1 + \left[\frac{\phi R_i}{h_{iR}}\right]^2\right)^{1/2} + \ln\frac{h_0}{h_{iR}}$$

where h_0 and h_1 are the thickness of the sample before and after deformation at a given R_i; this method of calculation was described in detail in [22].

In the case of room-temperature deformation, the true strain was varied in the range from 0.2 to 9.6; in the case of cryo-deformation, from 0.3 to 5.5. The hardness was measured by the Vickers method along the radii of the samples under a load of 0.5 N. In constructing the dependence of hardness, its values taken from different samples were averaged over the intervals of the true strain $\Delta e = 0.4$. The structure was examined by electron microscopy at a distance of 1.5 mm from the center of the sample. The distance from the sample center to the area of structure under study was estimated with an accuracy of ± 0.2 mm. The sizes of such structural elements as dislocation cells and microcrystallites were determined using bright- and dark-field images by at least four hundred measurements, the error being no more than 10%.

3. Results

3.1. Hardness

Fig. 1 shows the hardness dependences of Mo on the true strain for both temperatures under study. Since the plasticity of Mo sharply decreases with decreasing the deformation temperature to 80 K, we managed to deform the samples at this temperature to a maximum of three revolutions of the anvil without fracturing them. As a result, the maximum achievable true strain at 80 K turned out to be significantly lower than that at 290 K ($e = 7.6$, and 9.6, respectively). As can be seen in Fig. 1a the hardness of the material deformed at 80 K is lower than that of the material deformed at 290 K until $e = 4$; but it becomes higher after a greater true strain. The hardness curves as a function of the square root of the true strain were used to select deformation stages characterized by different hardening rates and the types of the structure formed at them. The method, according to which the stages were selected, was described in detail in [22]. As Fig. 1b illustrates, the curves are approximated by two straight lines with various slope angles, so $\mathrm{tg}\alpha_1 = 1.6$ and $\mathrm{tg}\alpha_2 = 17$ for 290 K and $\mathrm{tg}\beta_1 = 1$ and $\mathrm{tg}\beta_2 = 3$ for 80 K, which corresponds to hardening coefficients (Fig. 1a): $k_1 = 0.7$, $k_2 = 2.9$ at 290 K and $k_1 = 0.4$, $k_2 = 1.1$ at 80 K. At a deformation temperature of 290 K, the intensity of hardening rises sharply after $e = 8.5$ and, at 80 K, this occurs after a much lesser strain $e = 2.8$. Thus, we selected two stages of structural states with edges corresponding to the inflection points for both deformation temperatures.

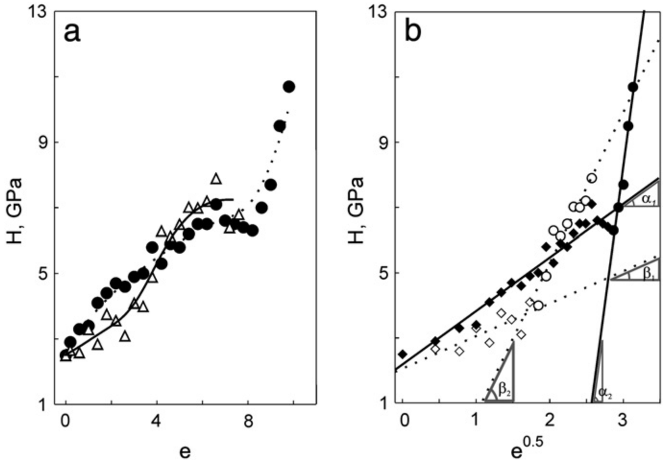

Fig. 1. (a) Hardness and (b) staged character of deformation in molybdenum at a temperature of 290 K (filled symbols) and 80 K (open symbols).

3.2. Structure

Electron microscopic study of the structure was performed on samples deformed at the same angle of rotation of the anvil at both temperatures. It should be mentioned that true strains may differ noticeably in this case. When loading the molybdenum sample by a pressure of 12 GPa at room temperature, the true strain of upsetting was $e = 0.7$. The electron microscopy study indicates the beginning of fragmentation of the single crystal. Dislocation cellular structure and individual boundaries that separate structure regions with significant misorientations up to several degrees can be seen in Fig. 2a and b. Such boundaries were previously observed in molybdenum after upsetting with $e \sim 1$ [23].

These boundaries (indicated by arrows in Fig. 2b), named "knife" ones in [23,24], form as a result of the movement of partial wedge disclinations [25]; and their formation is accompanied by localized shear deformation. In fcc materials, such deformation-induced boundaries are called geometrically necessary boundaries [26]. Together with knife boundaries, broken low-angle boundaries with misorientations >1° are observed (see a callout in Fig. 2b), which were previously found in [23,24]. Thus, regions with various crystallographic orientations, which are large-scale as compared with dislocation cell sizes, appear in the cellular structure.

The yield strength of molybdenum increases with decreasing the deformation temperature to 80 K; therefore, we managed to reach true strain only $e = 0.3$–0.5 after upsetting at a pressure of 12 GPa, which is lower than that at room temperature. In this case, a weakly misoriented cellular structure (Fig. 2c) and deformation-induced shear bands (Fig. 2d–e) form as a result of upsetting. However, no deformation twins similar to those observed in iron and nickel after deformation at 80 K were found [5,13]. The interpretation of the electron diffraction pattern (Fig. 2f), taken from the structure region that includes matrix and a band, disclosed three lattices with the following zone axes: [1 3–2], [1 3 5], [1 1 1]. The analysis showed that these lattices are not in the twinning relationship with each other. In addition, the contrast of the matrix in the dark-field image (Fig. 2d–e) remains unchanged outside the band. These both facts indicate that this band is a kink band [20].

An increase in the true strain up to $e = 2$ leads to the loss of the original single-crystal state at both temperatures. At 290 K, after such a relatively small strain, microcrystallites (Fig. 3a) and misoriented fragments with sizes larger than one micron and almost free of dislocations (Fig. 3b) arise due to the high dislocation mobility in molybdenum. After the cryo-deformation, low-misoriented cellular structure regions are still retained, and the fragmentation occurs within the band structures. In keeping the deformation, no new band structures form.

A further increase in the true strain is accompanied by the gradual refinement of structural elements and by a growth in the number of microcrystallites at both temperatures. However, even after the maximum true strain is reached, the structure remains heterogeneous: the elements with close misorientations are retained in addition to misoriented microcrystallites (Fig. 3c–d).

Fig. 4 presents the sizes of the misoriented structural elements excluding low-misoriented element sizes. The misorientation was