

# Formation and stability of the ultrafine-grained structure of commercial-purity copper deformed at 80 K

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The structure and microhardness of commercially pure copper have been studied after cryogenic (80 K) high-pressure torsion performed at an angle of the anvil rotation from 15° to 10 revolutions. The retardation of dynamic softening processes due to impurity dragging allows us to establish the stages of the structural change in commercially pure copper upon deformation, as compared to high-purity copper, in which recrystallization rapidly develops upon heating to room temperature, significantly distorting the deformed structure and decreasing the microhardness of the deformed copper. Two stages of deformation have been observed. The stages change at a true strain of  $\epsilon = 7.3$ . Dislocation slip and mechanical twinning are the main structure-forming mechanisms at the first stage. No mechanical twins are found at the second stage. The second stage is characterized by misoriented microcrystallites which play the role of recrystallization centers upon heating up to room temperature. The average microcrystallite size is 0.1–0.2  $\mu\text{m}$ . Microcrystallites provide a low thermal stability of the structure. Some grains in the structure formed at the second stage of deformation can grow up to several microns in 1–2 days; the fraction of the recrystallized structure is 20%. Holding for 3 years almost completes the recrystallization; the maximum size of recrystallized grains is 100  $\mu\text{m}$ . Recrystallization at room temperature develops slowly in the structure with deformation twins: the early signs of recrystallization are observed 1.5 years later after the end of the deformation; and the fraction of the recrystallized structure does not exceed 10%.

**Keywords:** cryogenic deformation, copper, twinning, recrystallization.

## 1. Introduction

Severe plastic deformation creates nonequilibrium structures in materials and changes the temperature, the kinetics, and the degree of structural and phase transformations [1–4]. In particular, severe deformation decreases the recrystallization temperature [5], which can change structure-forming processes in a number of materials, for example, strain hardening changes to dynamic recrystallization (DR) [6]. Such a change in structure-forming processes is an important factor that is responsible for the final structure and properties of a deformed material. DR was observed in copper during deformation by high-pressure torsion (HPT) at room temperature and unavoidable postdynamic recrystallization (PDR) after the deformation [7,8]. PDR has the most significant effect on the formation of a structure in a high-purity copper (99.99 wt % Cu) and causes the growth of individual grains. As a result, the structure continues to change continuously after the end of the deformation. This makes it difficult to ascertain the regularities of DR. Alloying with a small amount of impurity is known to be an effective way to retard grain growth [1]. PDR in copper of a different purity manifests itself in different ways: in high-purity copper, the growth of individual anomalously coarse grains is observed, whereas PDR in copper of commercial purity (99.9 wt %) can be recognized owing to the geometrically regular shape of the found grains and the formation of annealing twins [8–10]. DR can be suppressed and structure refinement can

be intensified by decreasing the deformation temperature. A high density of microcrystallites can be expected during cryogenic deformation, and a uniform submicrocrystalline structure can be expected as a result of static low-temperature recrystallization during room-temperature heating of a material. Recrystallization in copper upon room-temperature heating was observed after cryogenic deformation by both rolling, extrusion [11–13], and high-pressure torsion [14, 15]. In addition, a decrease in the deformation temperature activates mechanical twinning in copper [12,15,16]. However, the effect of twinning on static recrystallization upon subsequent heating was not analyzed in the above-mentioned works. The issue of various deformation mechanisms causing macroscopic deformation or not, has been considered in recent years in literature. In the first case, deformation twins that form in a coarse-grained material are considered, and the latter mechanism (zero-macrostrain deformation twinning) is typical of nanocrystalline materials [17,18]. It was found in [19] that there were no signs of recrystallization upon heating to room temperature in the regions occupied by deformation twins of the first type in high-purity copper subjected to cryogenic HPT, but the formation of microcrystallites caused a rapid development of recrystallization in the bulk of the material. The aim of this work was to study the formation of an ultrafine-grained structure and its subsequent room-temperature recrystallization in commercial copper, where twinning took place upon cryogenic deformation performed by HPT.

## 2. Experimental

The object of the investigation was commercial copper (99.9 wt % Cu) with a coarse-grained isotropic structure, having an average grain size of 60  $\mu\text{m}$ . Samples of 5 mm in diameter and 0.3 mm in thickness were deformed by HPT at a pressure of 5 GPa and an angle of the anvil rotation from 15° to 10 revolutions. During cryogenic deformation, the anvils together with a sample were immersed in an installed bath which was filled with liquid nitrogen (Fig. S1 of Supplementary Material). Before deformation, the sample was cooled for 30 min. The deformation was carried out at a rate of 0.3 rpm. The anvils together with the sample were kept in liquid nitrogen during the entire period of deformation. The temperature was measured with a thermocouple attached to a stationary anvil. The time from deformation to microhardness measurements and structure examination did not exceed 48 h. The structure was again examined after holding at room temperature from several days to 3 years. The true strain, which depended on the angle of the anvil rotation and the distance to the sample center, was calculated taking into account upsetting [6]. The values of  $e$  are listed here for  $r=1.5$  mm. The thickness of samples was measured along two mutually perpendicular diameters at a step of 0.5 mm. The sample thickness at distance  $r$  was averaged over four measurements. The minimum thickness at  $r=2$  mm after 10 revolutions of the anvil was 80  $\mu\text{m}$ . The Vickers microhardness was measured using a PMT-3 hardness tester at a load of 0.25 N. The load was taken so that the sample thickness exceeded the diagonal of an indentation print by a factor of 3. The measurements were conducted along two mutually perpendicular diameters at a step of 0.25 mm to calculate the average value for each radius (Fig. S2 of Supplementary Material). To plot  $H=f(e)$ , hardness values taken from different samples were averaged over the true strain ranges  $\Delta e=0.4$ .

The structure was examined using a JEM 200CX transmission electron microscope. The distance from the center of a sample to an examined region was determined with an accuracy of  $\pm 0.2$  mm. The size of structural elements was determined from bright- and dark-field (in a  $(111)_\gamma$  reflection) electron microscopy images by the direct measurement method with an error of less than 10%. The fraction of the recrystallized structure was determined using

the fraction of the area occupied by recrystallized grains. All studies were performed in the plane perpendicular to the anvil rotation axis.

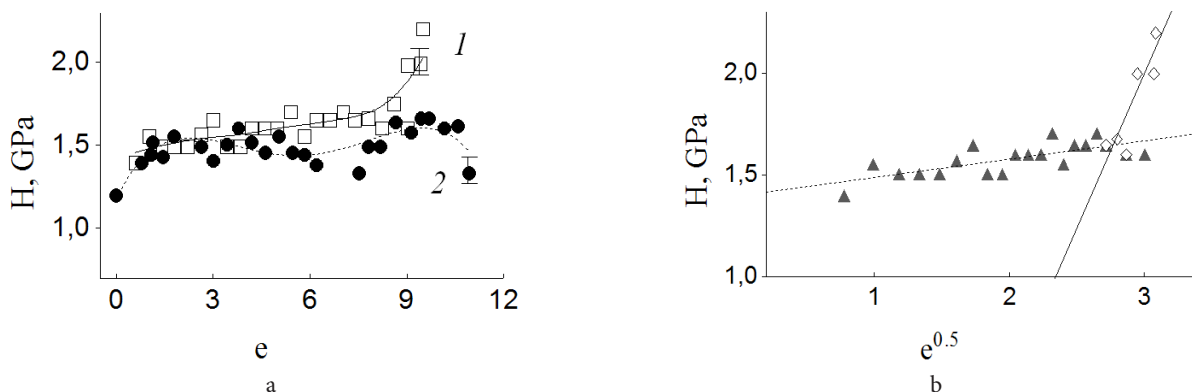
## 3. Results and discussion

Fig. 1a shows the microhardness of the commercial copper deformed at 300 K [9] and 80 K as a function of the true strain. The nonmonotonic dependence obtained after deformation at 300 K can be explained by DR and PDR. These processes result in the formation of a structure with nonuniformly distributed defects and some grown recrystallized grains [9]. After deformation at 80 K, the microhardness increased monotonically over the entire true strain range. In this case, up to  $e=5$  the microhardness is practically independent of the deformation temperature. Because of the monotonic microhardness behavior, we use dependence  $H=f(e^{0.5})$  to identify deformation stages, as it was done in [6]. In these coordinates, data can be extrapolated by two straight lines with different slope angles. The point of intersection of these lines corresponds to a true strain of  $e=7.3$  (Fig. 1b). It can be seen that the first stage is characterized by slow hardening, and in the second stage, the rate of hardness growth significantly increases.

Usually, different stages of deformation correspond to different structures.

Electron microscopic examination showed groups of parallel twins in addition to deformation cells in the structure (Figs. 2a–2d) after cryogenic deformation performed in the anvil rotation angle range from 15° ( $e=2.3$ ) to 360° ( $e=6.1$ ). No signs of recrystallization are observed. After deformation for 2 revolutions ( $e=7.5$ ) and more, there are no mechanical twins in the structure, but microcrystallites (average size 0.1  $\mu\text{m}$ ) and recrystallized grains formed during heating to room temperature are mainly observed (Fig. 3). Individual grains can grow up to several microns in 1–2 days (Figs. 3,4). Annealing twins, indicating static recrystallization, appear in both coarse and fine recrystallized grains.

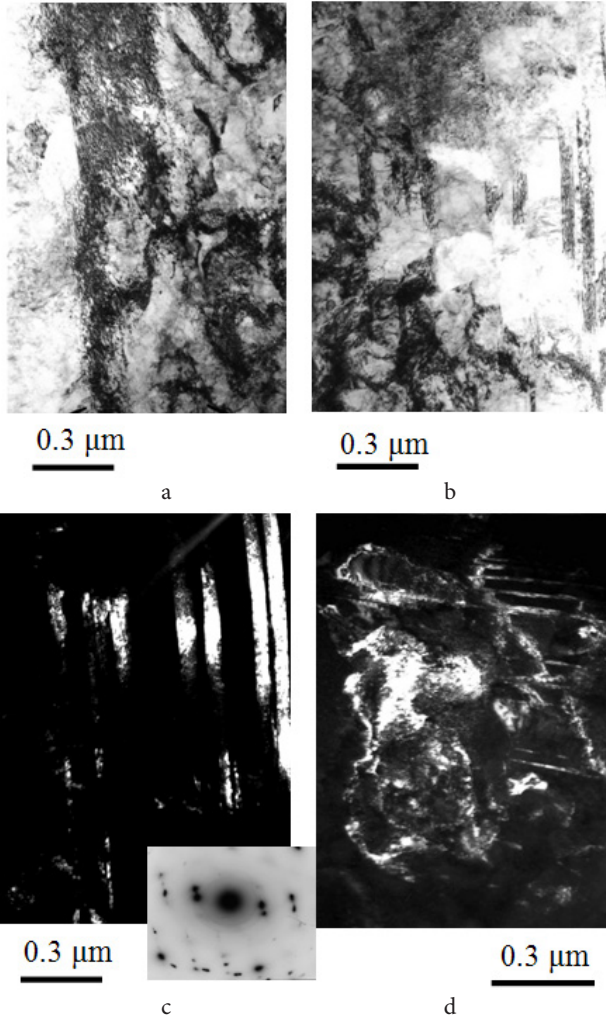
Thus, the structure changes fundamentally at  $e>7.3$  (inflection in Fig. 1b). At the first stage of deformation, revealed in this work, defects are accumulated and mechanical twinning occurs. At the second stage, structure refinement and the formation of misoriented microcrystallites occur.



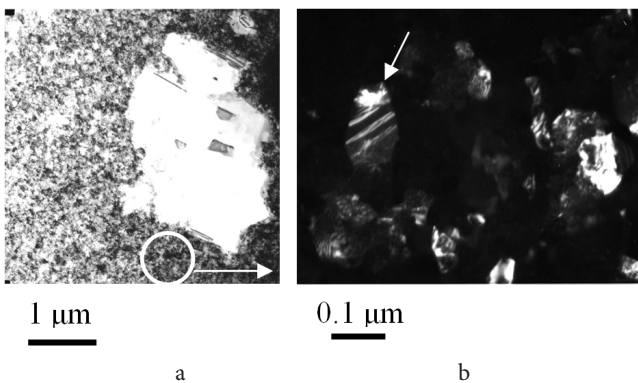
**Fig. 1.** Microhardness of the commercial copper as a function of true strain: (1) 80 K and (2) 300 K (a); stages of the deformation at 80 K:  $\blacktriangle$  — I stage,  $\diamond$  — II stage (b). Curve 2 was built using the data taken from [9].

Since the size of microcrystallites is about 100 nm, then, according to the data of [18,20], mechanical twinning causing macroscopic deformation has no chance to take place in them. The deformation of commercial copper at room temperature [9] was performed under other conditions. It was accompanied by dynamic recovery and

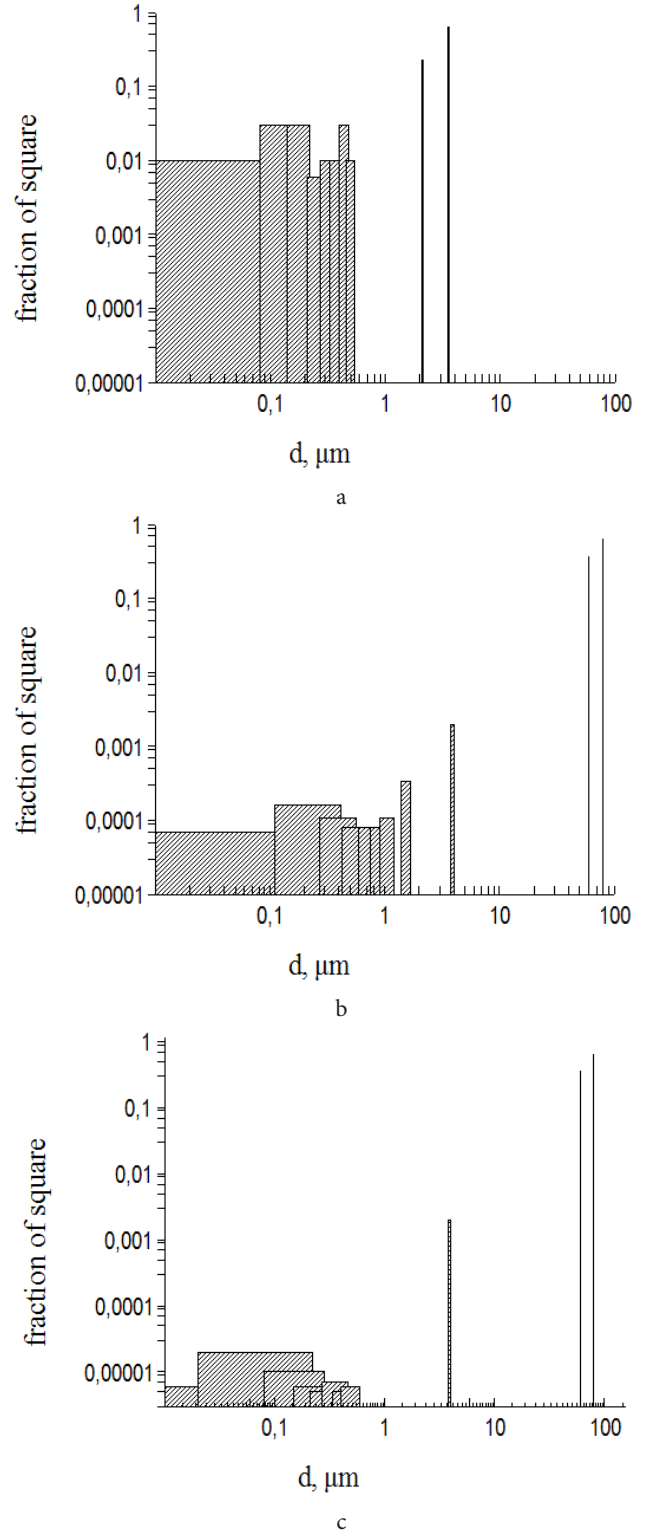
DR without deformation twinning. As a result, misoriented structural elements formed at a smaller true strain: the first microcrystallites were observed after 45° ( $e = 3.5$ ) (Fig. S3 of Supplementary Material). The hardness reached the level of saturation and PDR led to steady grain growth and a decrease in the hardness under these conditions (Fig. 1a).



**Fig. 2.** Microstructure of the commercial copper deformed at 80 K and an angle of the anvil rotation of 15° (a,b,c), and 360° (d). A dark-field image in a twin reflection (c), and a dark-field image (d) taken in matching reflections from the matrix and a twin.



**Fig. 3.** Microstructure of the commercial copper after deformation at 80 K for 5 revolutions: a bright-field image (a) and a dark-field image in the (111)<sub>y</sub> reflection (b).



**Fig. 4.** Area fraction distribution of structural elements after 5 revolutions of HPT performed at 80 K. 48 h later after deformation (a), two weeks later (b), and three years later (c). Arrows show the peaks that correspond to the maximum grain size.

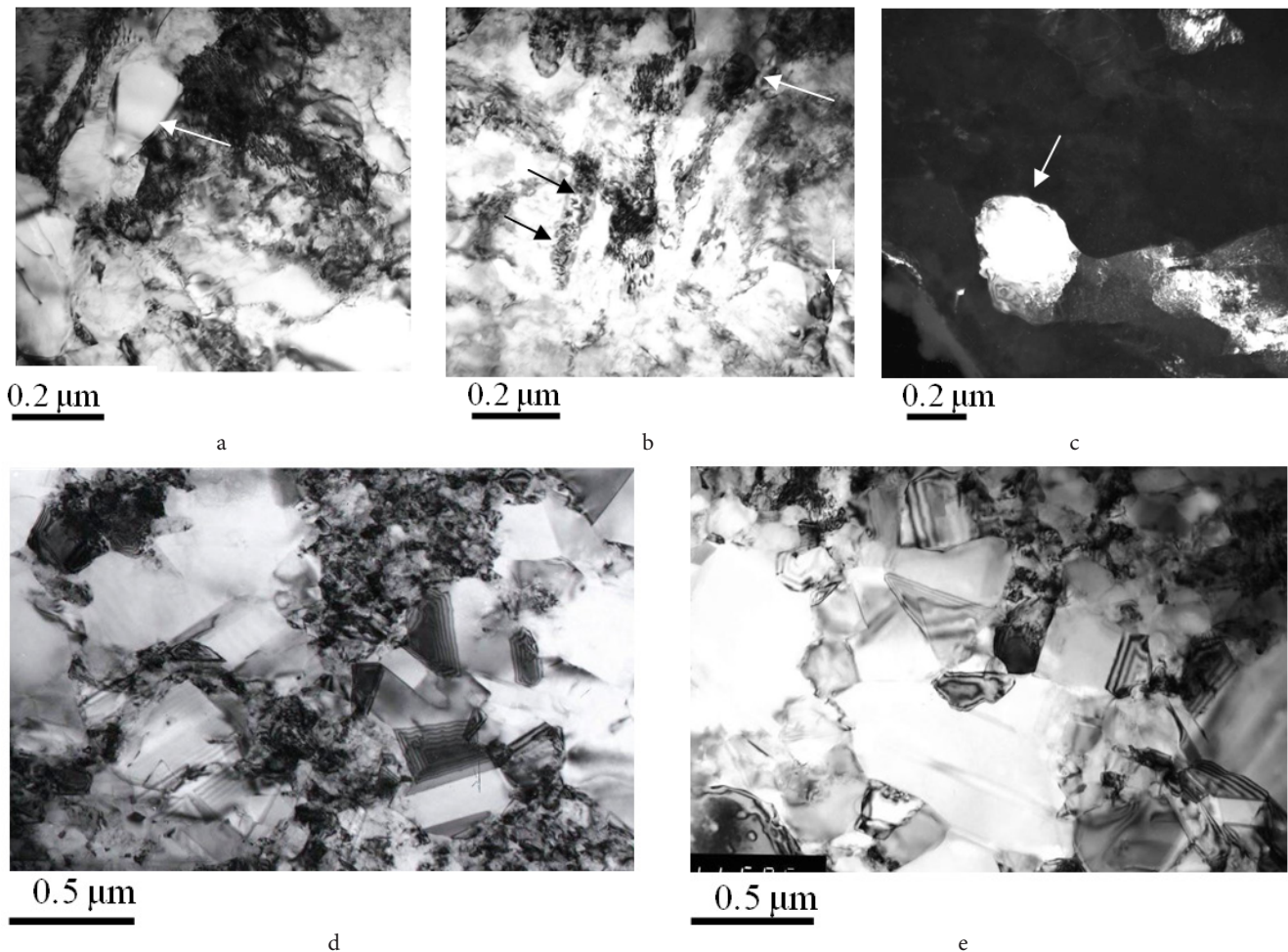
As previous studies of high-purity copper showed, there are dislocation cells, deformation twins, microcrystallites, and recrystallized grains in the structure after cryogenic HPT at an angle of the anvil rotation from 15° to 10 revolutions (Fig. S4 of Supplementary Information). The grains were formed during the heating of the material up to room temperature [19]. The simultaneous presence of all these structural components after the deformation in the investigated range, as well as rapid grain growth at 300 K, did not allow us to reveal the stages of deformation.

The first microcrystallites among dislocation cells are observed even after upsetting without torsion. That is, despite a significant decrease in the deformation temperature, dislocation mobility in high-purity copper is sufficient for the formation of a misoriented dislocation structure and its transformation into microcrystallites, which are nuclei of static recrystallization [5]. Since the average size of structural elements in pure copper after cryogenic deformation is at least 0.3 μm, there is a probability of mechanical twinning [18].

The study of commercial copper showed that impurities during cryogenic deformation substantially retarded the formation of a misoriented structure. First of all, this has an effect on the formation of microcrystallites and the structure

stability. In the case when there are no deformation-induced recrystallization nuclei (microcrystallites), a long incubation period is required for the formation of nuclei during holding at room temperature: submicron grains after deformation by both 15° ( $e=2.3$ ) and 1 revolution ( $e=6.1$ ) are observed only after holding for 1.5 years (they are indicated by white arrows in Figs. 5a–5c); the volume fraction of the recrystallized structure is less than 10%. Fig. 5 shows that there are still traces of deformation twins in the structure (indicated by black arrows in Fig. 5b). The formation of microcrystallites at the second stage of deformation fundamentally changes the recrystallization kinetics at 300 K. 48 h later after the end of the deformation, about 20% of the structure recrystallizes (Fig. 3). Holding for 2 weeks increases the fraction of the recrystallized structure to 50% and holding for 3 years completes the recrystallization (Figs. 5c, 5d).

The histograms shown in Fig. 4 demonstrate the dynamics of grain size variation during the holding of copper having a structure formed at the second stage of cryogenic deformation. It can be seen that after 2 weeks, individual grains reach 100 μm (corresponding peak is indicated by the arrow in Fig. 4b). 3-years holding increases the fractions of such grains, but the size of the coarsest grains remains the same (Fig. 4c).



**Fig. 5.** Microstructure of the commercial copper after deformation at 80 K to 15° (a), 360° (b,c), and 5 revolutions and after holding at room temperature (d,e): 1.5 years (a,b,c), 2 weeks (d), and 3 years (e). A dark-field image in the (111)<sub>y</sub> reflection (c).

#### 4. Conclusions

0.1% impurities (0.09% more than in the previously studied pure copper) in the commercial copper decreased the mobility of defects upon cryogenic deformation and subsequent room-temperature heating. This allowed us to establish in the commercial copper the presence of two stages of structure formation.

The first of these stages involves the accumulation of defects, the formation of a cellular structure, deformation twinning, and the fragmentation of previously formed twins. The second stage begins at  $e=7.3$ . The structure here consists of microcrystallites with high-angle deformation boundaries and recrystallized grains formed upon heating to room temperature. Recrystallization at room temperature develops slowly in the structure with deformation twins: the early signs of recrystallization are observed 1.5 years later after the end of the deformation. The presence of microcrystallites in the structure fundamentally changes the recrystallization kinetics: 48 h later after the end of the deformation, the fraction of the recrystallized structure is 20%. Holding for 3 years almost completes the recrystallization.

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**Supplementary Material.** The on-line version of this paper contains supplementary material available free of charge at the journal's Web site ([www.lettersonmaterials.com](http://www.lettersonmaterials.com)).

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