Structure Evolution in Copper Resulting From the Effect of Powerful Current Pulses

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(Submitted 31 March 2002; in revised form 16 September 2002)

Features of structure formation and changes in microhardness of pre-deformed copper 99.9%Cu resulted from the effect of current pulses of high density (~10 kA/mm^2) and short durability (~10^2 μs) at the heating rate of 10^6-10^7 K/s have been studied. The changes occurring are interpreted as a result of a rapid rate process of thermally activated recrystallization. The characteristics of the process are compared with the ones taking place during thermal annealing. The mechanisms responsible for the differences observed are discussed.

Keywords annealing, current, powerful electropulse treatment (PEPT), recrystallization, structure

1. Introduction

Processing of materials by concentrated sources of energy (laser, E-beam, plasma, etc.) is widely used at present in machine building to directionally change the chemical composition and structure-phase state of near-surface layers and, as a result, service properties of machine components. One of the methods for modifying structure and mechanical properties of conductive materials is the exposure to current pulses of high density or powerful electropulse treatment (PEPT). Electropulse processing is an advanced method providing superplastic conditions at high strain rates.[1]

There are known a number of papers devoted to the influence of current pulses of high power on properties of conductive materials.[1-8] According to one known approach, the effect observed is connected with migration of structure defects resulted from electron flow.[2,3] In this case the specified conditions of the experiment are so that the density of Joule heat evolution for one pulse should be insufficient for heating material in the zone of exposure. The other group of experiments is connected with investigation of the thermal effect of current, i.e., with parameters that cause a significant increase of temperature in the zone of processing.[4-8] These works are devoted to investigation of specific features of structure resulting from pulse current. However, the non-uniformity of temperature and current fields as well as the short-term period of the process complicate determination of the quantitative dependence of structure parameters on conditions in the zone of processing.

The present paper deals with systematic studies of structure changes in copper at powerful electropulse treatment in a wide range of changing temperature in the zone of exposure.

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2. Experimental Procedure

Experiments were performed on flat specimens cut out from strips. The strips were produced from annealed copper (99.9%Cu, 0.005%Fe, 0.005%Pb, 0.002%Sn, 0.002%Sb, 0.001%Bi, 0.002%As, 0.005%S, 0.002%Ni, 0.003%Ag, 0.005%Zn, 0.05%O_2) by cold rolling to the strain value $e = 0.3$ and $e = 0.8$ ($e = \ln (h_0/h)$, where h_0 and h are the initial and final thickness of strip, respectively). The current pulse, ~10^2 μs in duration, was generated by discharge of a high-voltage bank of capacitors (Fig. 1a). The amplitude and duration of the discharge were regulated by the voltage of capacitor charge and the value of inductance in discharge loop. To protect the tested specimens from evaporation and in some cases to discontinue rapidly the current in specimen circuit after the first oscillation period an explosive copper tie plate was used. The padding resistor provided overvoltage protection.

The specimens, 0.4 mm thick and 4 mm wide, were gripped by massive copper jaws through which the current was supplied to the processing zone of the specimen, 1 mm long. After pulse heating, the specimen was cooled for 1 μs by means of the heat sink to the gripped specimen edges (air cooling can be neglected). In case of the pulse duration of about 10^2 μs the depth of current penetration into the copper was 1 mm. So, the temperature distribution in the specimen was similar to the one observed during electropulse treatment of near-surface layers of massive articles when cooling of a thin heated layer occurs due to heat sink to the metal bulk.

With allowance for the small thickness of the specimen the non-uniformity of current distribution on cross section is negligibly small due to the skin effect. Testing measurements have shown that distribution of microhardness along the zone treated by current is also almost uniform except boundary (transition) zones between current treated and untreated portions, about 0.1 mm wide.

Current impulse in the specimen was detected by means of the Rogovskyi's loop method.[9] The signal from this loop was given to the input of a memory oscilloscope C8-17 (Mashpriborintorg, Moscow, USSR). The change in the temperature of the specimen was determined by the equation:

$$\frac{j^2}{\sigma_e} = \rho c \frac{\partial T}{\partial t} \quad \text{(Eq 1)}$$

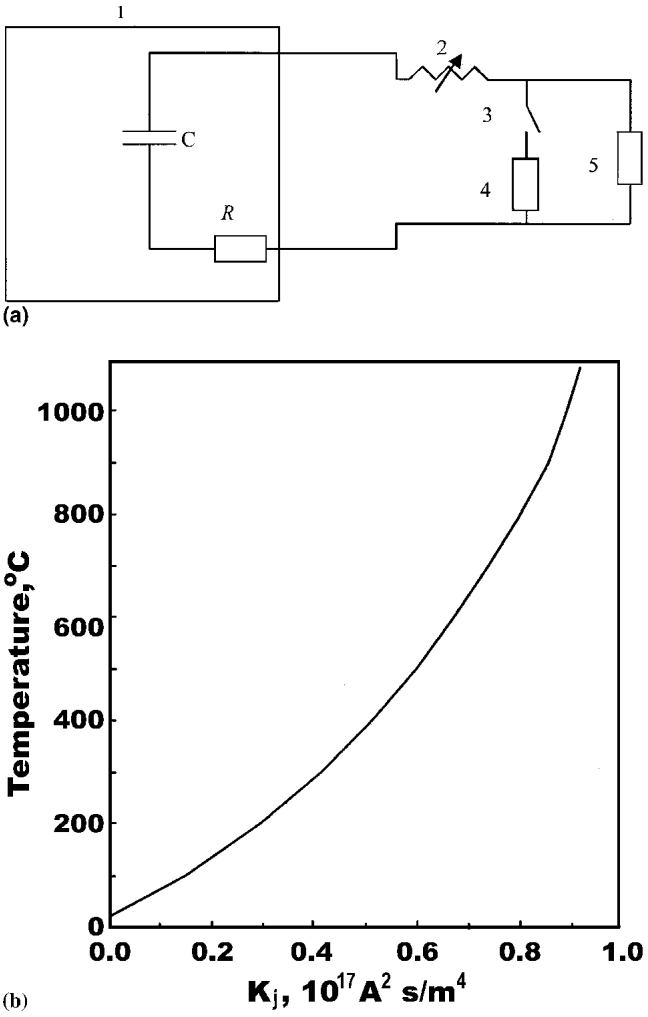


Fig. 1 **(a)** Scheme of device, 1) pulsed oscillator, 2) inductor, 3) connector, 4) specimen, 5) padding resistor; **(b)** dependence of calculated temperature on current integral

where ρ, c, and σ_e are, respectively, the density, heat capacity, and electroconductivity of copper and j is the current density.

Integrating Eq 1, we get $S(T_k) = K_j$, where $K_j = \int_0^{t_k} j^2 dt$, $S(T_k) = \int_0^{Tk} \rho c \sigma dT$, ($t_k$ is the time of PEPT; T_k is the temperature attained in the specimen).

So, knowing the dependence of density, heat capacity and electroconductivity of the material on temperature one can determine the calculated temperature of PEPT by using the experimental current oscillogram.[9] After integrating the first portion of the Eq 1, one can also calculate the density of Joule heat evolution. Figure 1(b) shows the dependence of calculated temperature on a value of K_j, which weakly differs from linear (K_j is the so-called conventional temperature PEPT).

Metallographic analysis of specimens was made on an optical microscope (Neophot-32, Carl Zeiss Jena GmbH, Jena, Germany) and semi-automatic analyzer (Epiquant, Carl Zeiss). Microhardness was measured by a special adapter to the optical microscope (Neophot-2; the error measurement was 8%). Fine structure was studied by an electron microscope (JEM 2000 EX, JEOL Ltd., Tokyo, Japan).

3. Experimental Results and Discussion

Microphotos of structure of specimens at the strain values $e = 0.3$ and $e = 0.8$ are shown in Fig. 2 and 3. Rolling until the strain value $e = 0.3$ leads to occurrence of small metallographic texture. At the strain value $e = 0.8$, the metallographic texture is more distinct (Fig. 2a and 3a). Texture is also observed in the cross section. So, at $e = 0.8$, the mean grain size in the longitudinal section is $d_x = 47.8$, $d_y = 28.5$ μm, and in the cross section, $d_y = 26.3$, $d_z = 15.1$ μm. The value $d = \sqrt{d_x d_y}$ was used as a mean grain size for processing the results of the experiment. In all specimens copper sulphide inclusions were observed arranged in a line along rolling (Fig. 2a and 3a, indicated by arrows).

Thermal or static annealing of copper specimens was conducted over the temperature range 100-500 °C for 1 h. Figure 4 shows changes in microhardness and grain size depending on temperature of annealing. As one can see from Fig. 4, the microhardness sharply decreases at annealing temperatures above approximately 150 °C for the strain $e = 0.3$ and above approximately 125 °C for $e = 0.8$. At annealing temperatures above 200 °C, microhardness is stabilized, having attained the smallest value.

In specimens subjected to rolling at $e = 0.3$, the formation of new grains begins at the temperature of annealing 125 °C. At a temperature 300 °C, the structure of the specimen is recrystallized completely (Fig. 2b). With an increase in the temperature of annealing above 300 °C, growth of recrystallized grains occurs. In case of the strain $e = 0.8$, new grains begin to form already at $T = 100$ °C (Fig. 4b). Complete recrystallization of grains takes place at 200 °C (Fig. 3b). Further increase in temperature leads to a small growth of grains.

Figure 5 shows changes in microhardness and mean grain size in specimens subjected to current pulse treatment depending on conventional temperature of PEPT. In most cases the current diagram corresponded to damped sinusoid with a period of 140-145 μs. In a number of cases whose results are also shown in Fig. 5, the current in the specimen circuit was discontinued after the first oscillation period. The decrease in the time of current exposure did not exert a noticeable effect. This can possibly be attributed to the fact that time of thermal exposure is determined by typical time of cooling, which far exceeds the time of the heating effect of current.

In the determined range of calculated temperature of PEPT ($T = 600$-650 °C for $e = 0.3$ and $T = 500$-550 °C for $e = 0.8$), a decrease occurs in microhardness and refinement of grains. As the conventional temperatures 0.7×10^{17} A^2s/m^4 for $e = 0.3$ and 0.65×10^{17} $A^2s/m^4$for $e = 0.8$ are attained, the material is recrystallized completely (Fig. 2b and 3b). These results allow one to assume that the PEPT, like the usual static annealing, initiates recrystallization processes. Similar to the static annealing, an increase of the preliminary strain results in a decrease of the recrystallization temperature. Note that in submicrocrystalline copper 99.9%Cu at the strain $e = 7$ (cold deformation between Bridgman anvils),[10] the recrystallization initiated by PEPT starts already at 200 °C.[1]

In spite of the qualitative similarity of static recrystallization and recrystallization at PEPT a number of significant differences exist between them. First, if thermal recrystallization

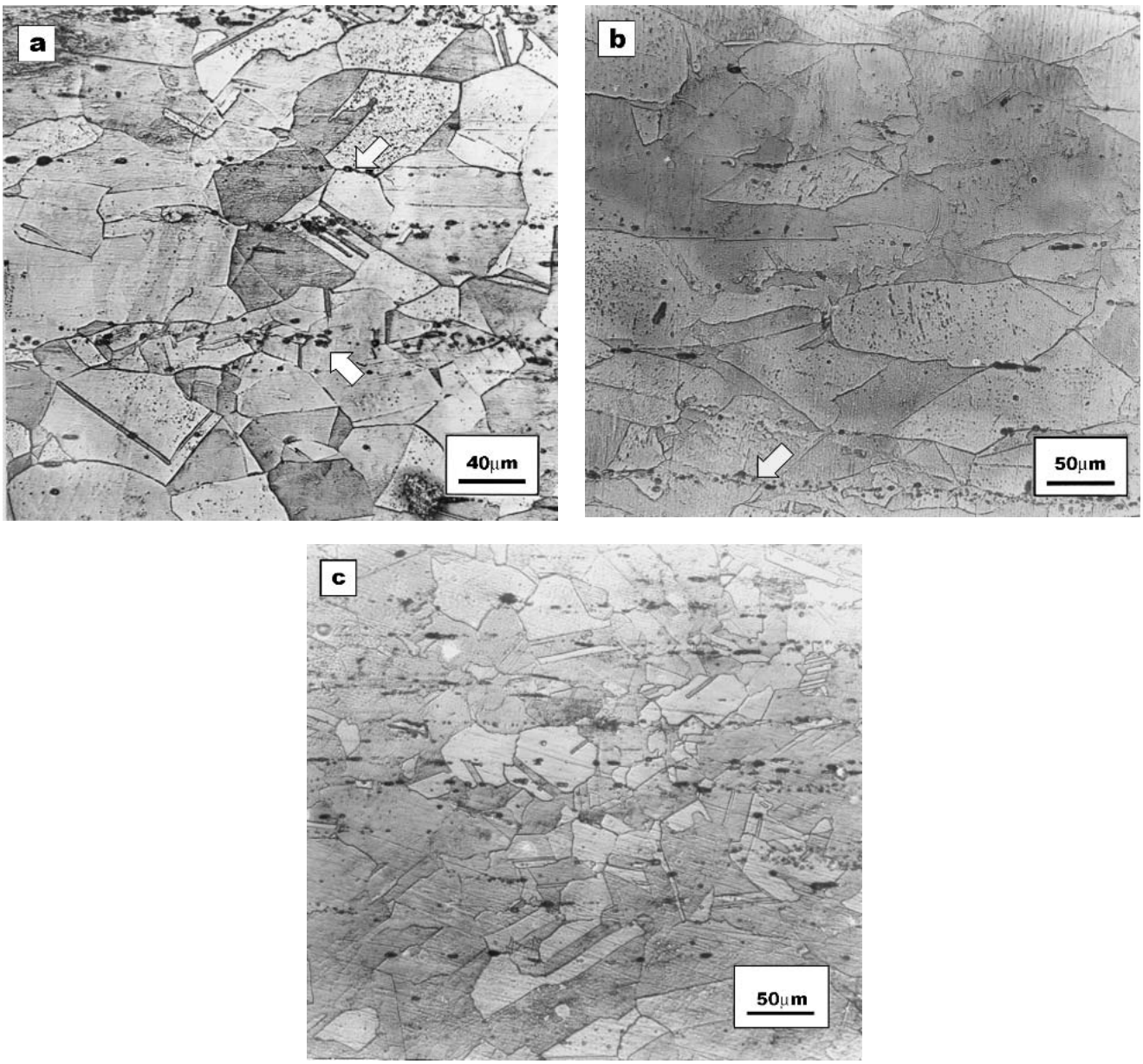


Fig. 2 Microstructure of copper at strain $e = 0.3$: **(a)** initial state, **(b)** after static annealing at $T = 300$ °C, **(c)** after PEPT; $K_j = 0.80 \times 10^{17}$ A^2s/m^4

takes place in the range 100-200 °C, then recrystallization at PEPT occurs at the calculated temperatures 500-600 °C. This is not surprising since it is known that even a comparatively small increase in the velocity of heating increased the temperature of recrystallization significantly. For example, the temperature onset of recrystallization of armco-iron after deformation by 60% is 500 °C at furnace heating and 700 and 900 °C at heating with velocities of 500 and 600 °C/s, respectively.[11] As a rule, in case of high-velocity heating, recrystallization results in formation of a more fine-grained structure that is attributed to formation of a larger number of centers of recrystallization. As follows from comparison of Fig. 4(b) and Fig. 5(b), such a phenomenon does not take place in this case. Moreover, for the strain $e = 0.3$, the average grain size after PEPT (25 μm) is significantly larger than that after primary recrystallization during static annealing (10-15 μm).

One more difference is in the fact that during static annealing, the decrease in microhardness starts at temperatures much lower than the onset of recrystallization that, as known, is connected with redistribution of defects of crystal lattice whereas microhardness of specimens during PEPT remains constant within a wide range of K_j (Fig. 5a). One can assume that retardation of the onset of recrystallization during PEPT is attributed to the lack of time required for defects of crystal lattice to diffuse to grain boundaries. In this connection it should be noted that during PEPT the significant decrease in grain size (portion AB for $e = 0.3$ and portion CD for $e = 0.8$) begins only after the sufficient decrease in microhardness as

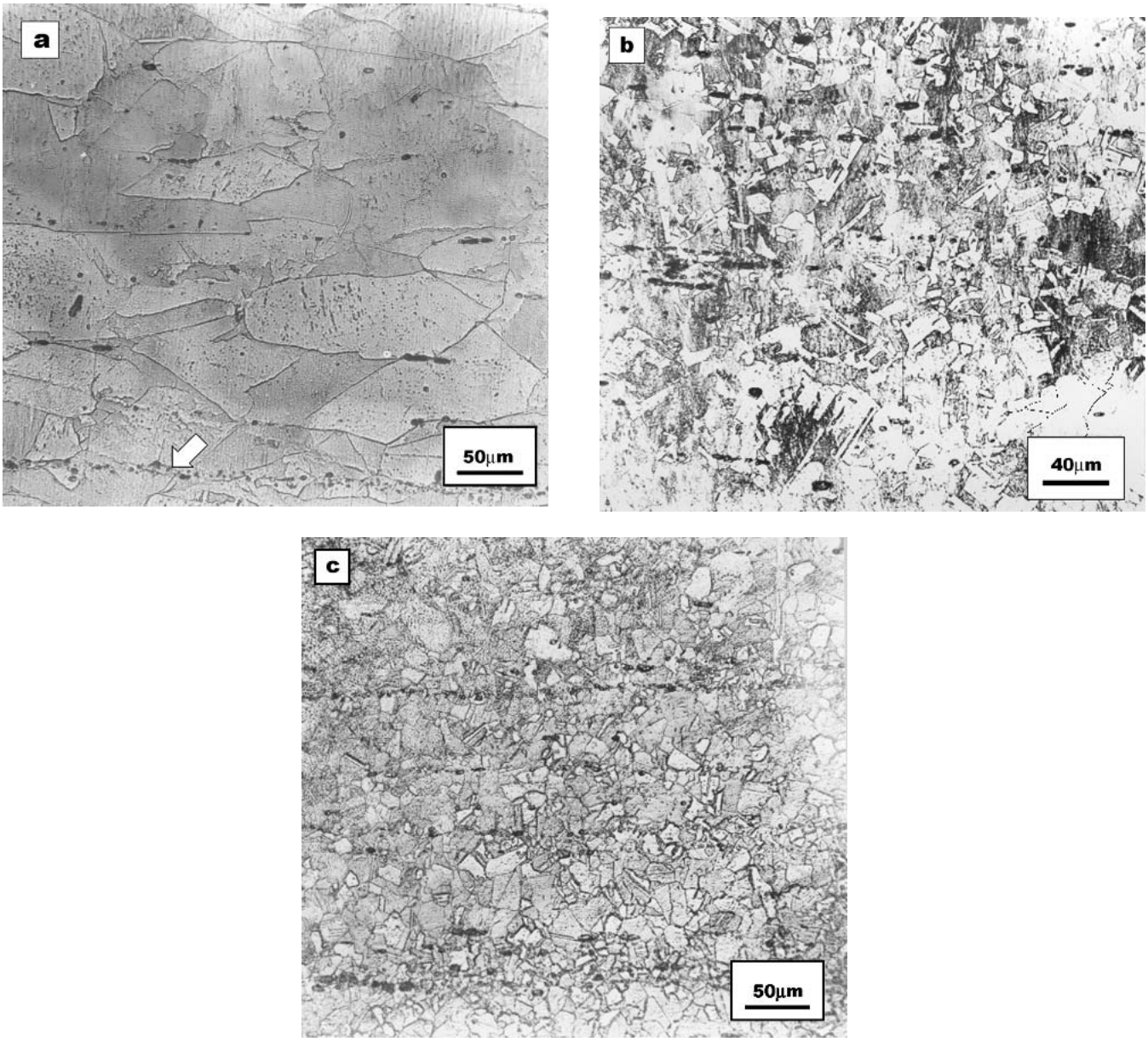


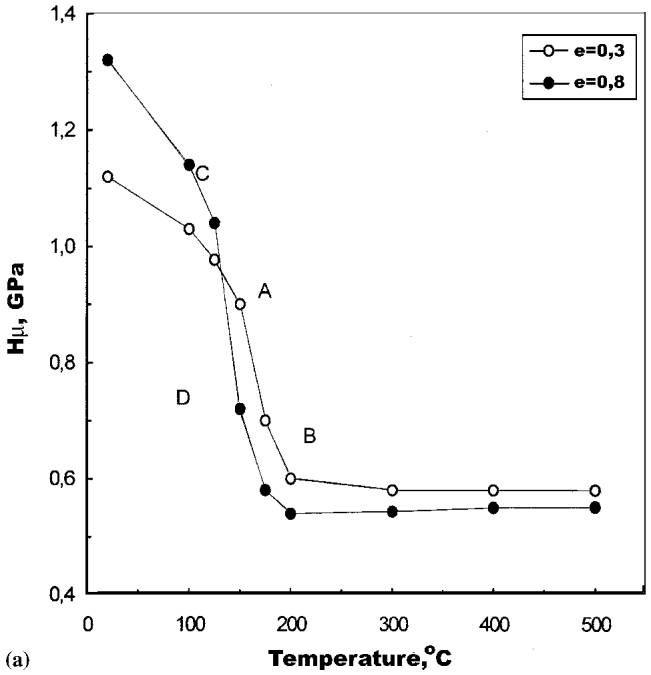
Fig. 3 Microstructure of copper at strain $e = 0.8$: **(a)** initial state, **(b)** after static annealing at $T = 200$ °C, **(c)** after PEPT; $K_j = 0.87 \times 10^{17}$ A^2s/m^4

compared with the initial state. During static annealing the most significant decrease in grain size (areas AB and CD in Fig. 4) occurs synchronously with decreasing microhardness. During PEPT for $e = 0.8$, the significant decrease in microhardness occurs after grain refinement (below point D).

It should be noted that the increase in the calculated temperature of PEPT did not lead to grain coarsening occurring during static annealing. Moreover, microhardness also did not reduce to the values about 0.6 GPa, which are typical for the states preceding the grain coarsening during the static annealing. TEM studies of the fine structure show that as a result of PEPT with a high value of calculated temperature the dislocations form a cell structure (Fig. 6).

Thus, essential differences in the structural evolution and the behavior of microhardness during the static annealing and PEPT have been revealed. These differences suggest that there are also significant dissimilarities in the kinetic of the transformation of defect structure at the high temperatures that is necessary for a recrystallization during short-time thermal treatment. Peculiarities of the structural evolution at high-temperature high-rate recrystallization have also been found by other methods for producing a thermal pulse; for example, during the laser-treatment.[12] The significant influence of the annealing temperature on the kinetics of structural relaxation in submicrocrystalline copper has been established in Ref. 13, too.

Earlier it was noted, that a non-homogeneous distribution of electric conductivity within the metal could lead to a mosaic distribution of pulse current and non-homogeneitics of the temperature.[4] In the present experiments, the inhomogeneous



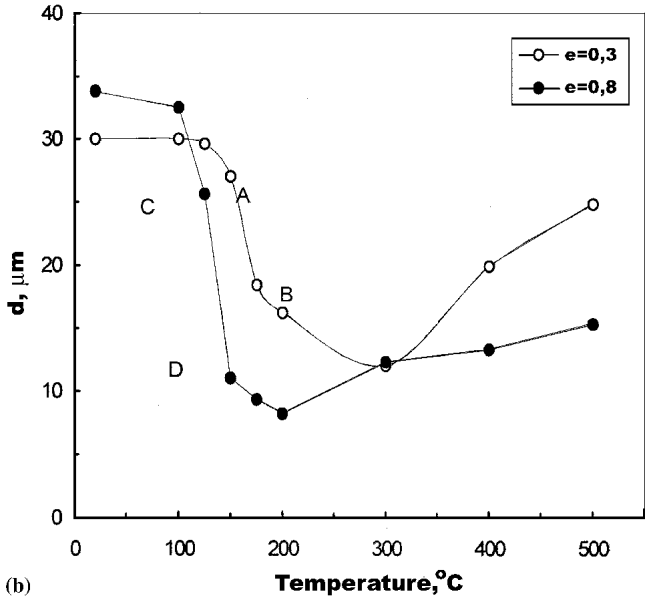
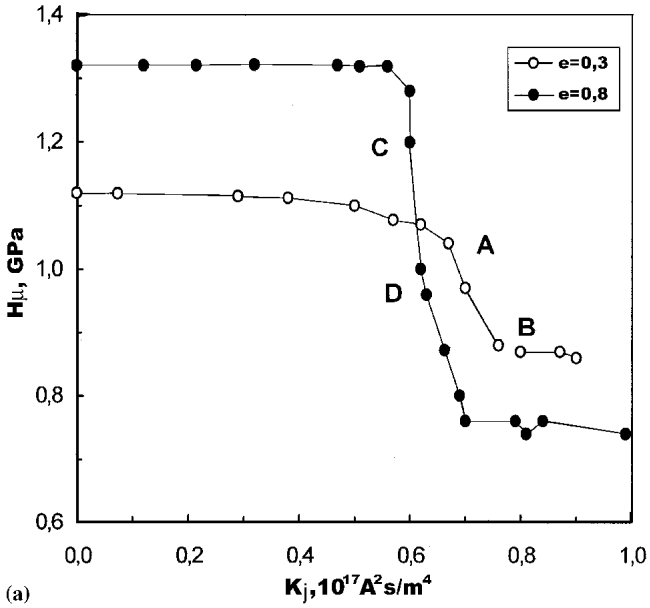


Fig. 4 Change in **(a)** microhardness and **(b)** grain size depending on annealing temperature



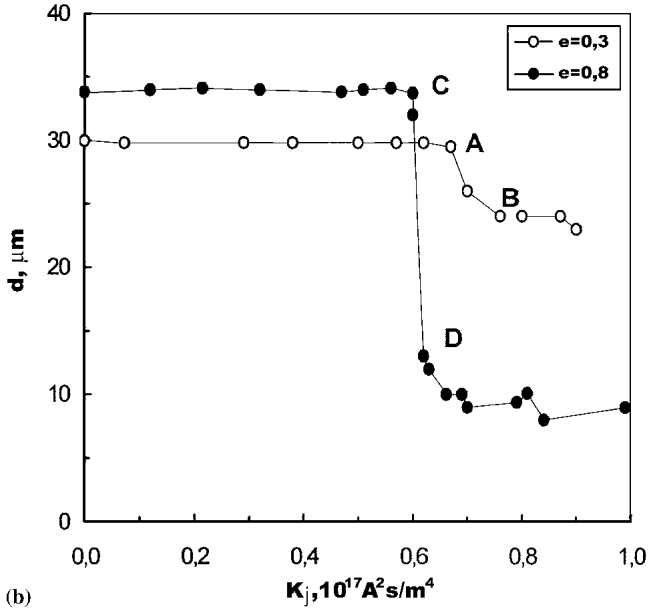


Fig. 5 Change in **(a)** microhardness and **(b)** grain size depending on conditional temperature of PEPT

character of conductivity is attributed to dispersion impurities of copper sulphide, several micrometers in size, whose conductivity is relatively low. At high current densities, corresponding to K_j = (0.85 to 1.0) × 10^{17} A^2s/m^4, zones of recrystallization with a grain size of 1 μm and less—i.e., much less than in the rest volume of the metal—occur in the vicinity of these additions impurities (Fig. 7, arrows). The size of these zones in cross section exceeds the size of copper sulphide impurities by about 2. During static annealing, such features in the structure of copper are not observed in the vicinity of copper sulphide impurities. The probable reason for such phenomena is concentration of current in the vicinity of impurities as pulse current flows around them, which leads to creation of an overheat zone near remaining relatively cool impurities.

4. Conclusions

The action of current pulses of high density leads to changes in microhardness and structure of copper 99.9% Cu. The character of these changes depends on a preliminary strain value, and as in case of static annealing, they are caused by thermal activation of recrystallization processes

However, dependencies of microhardness and grain size on

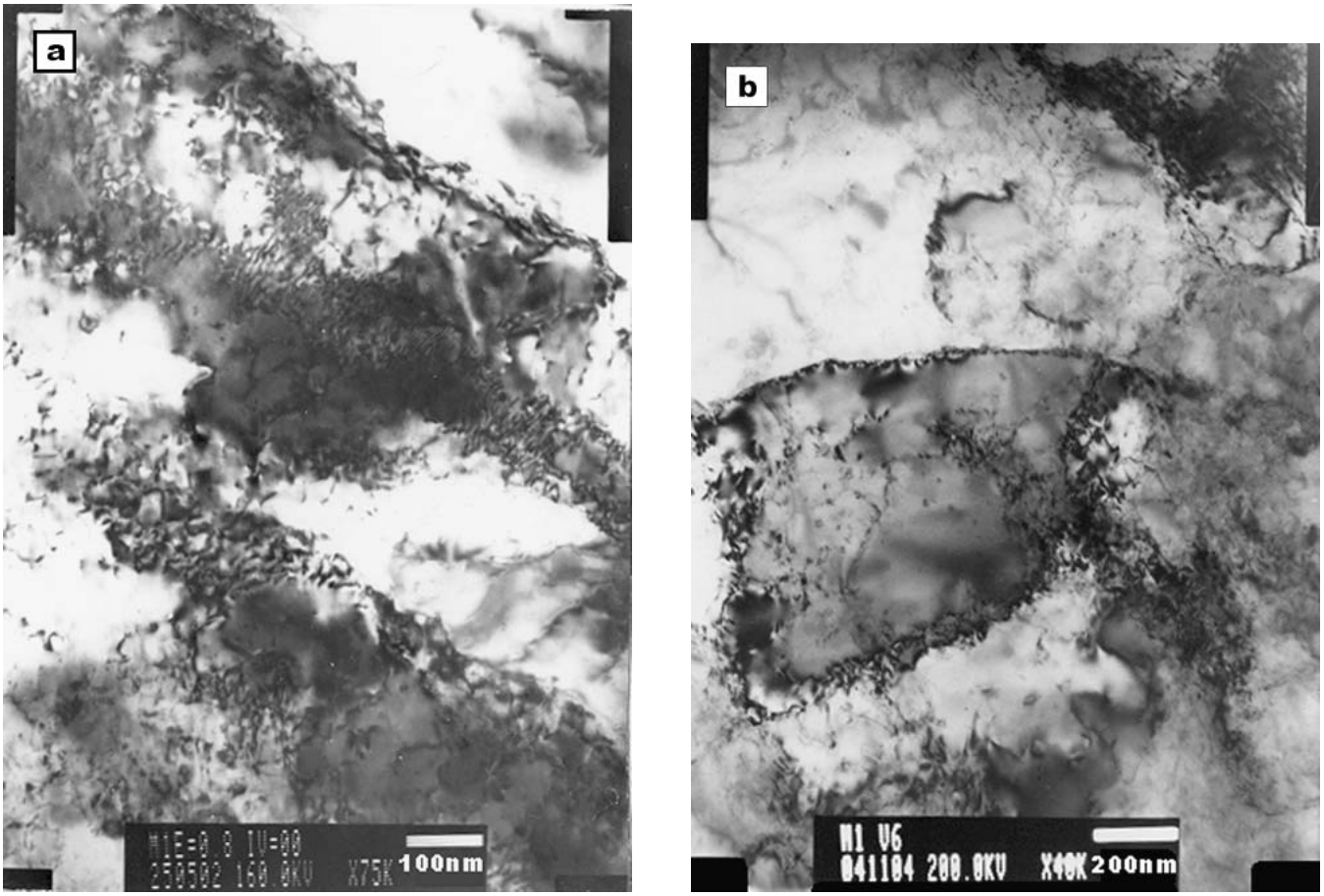


Fig. 6 Fine structure of copper at strain $e = 0.8$: **(a)** before PEPT, **(b)** after PEPT; $K_j = 0.67 \times 10^{17}$ A^2s/m^4

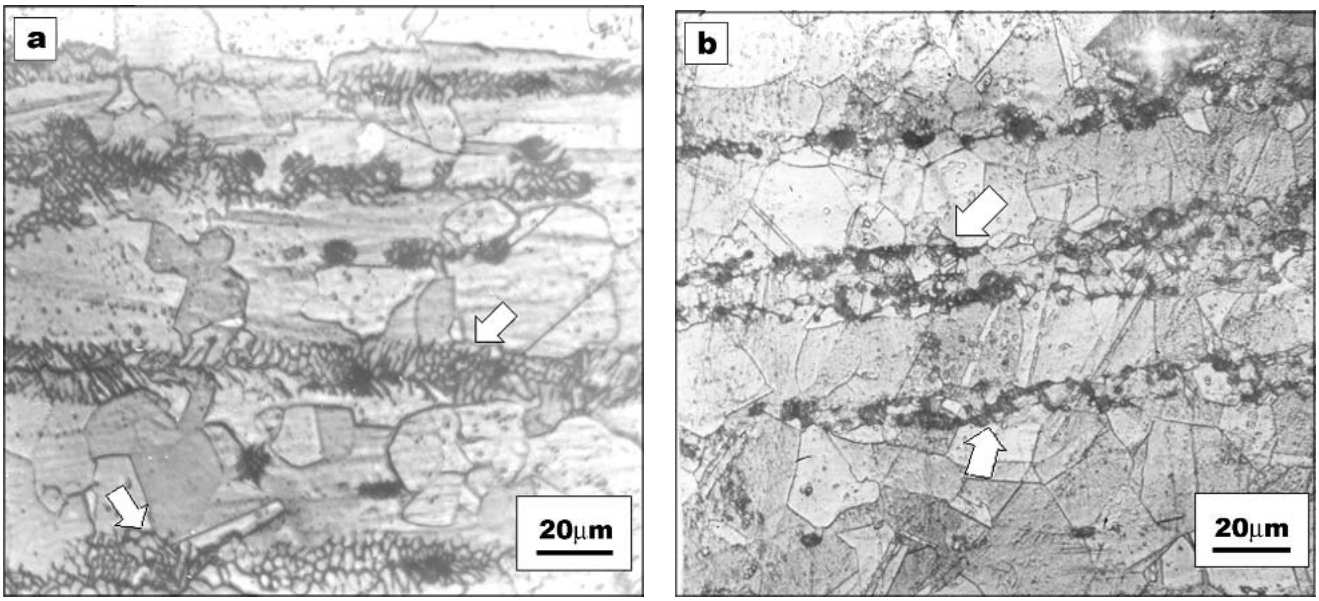


Fig. 7 Microstructure of copper after PEPT in the vicinity of copper sulphide impurities: **(a)** e = 0.3, $K_j = = 0.87 \times 10^{17}$ A^2s/m^4; **(b)** $e = 0.8$, $K_j = = 0.85 \times 10^{17} A^2s/m^4$

temperature are qualitatively and quantitatively different for processes of PEPT and thermal annealing. The temperature of recrystallization increases significantly, and with increasing velocity of annealing no grain refinement is observed, which occurs at smaller velocities of heating. In case of PEPT the significant decrease in microhardness can occur already after grain refinement.

These data testify that kinetics of redistribution of structure defects in the process of recrystallization caused by the short-term exposure of high temperatures differs significantly from that of static annealing.

Acknowledgment

This investigation was supported by the Institute for Metals Superplasticity Problems, Russian Academy of Sciences.

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