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# EFFECT OF ULTRASONIC PROCESSING ON THE PROPERTIES OF ULTRAFINE-GRAINED BRASS

## VPLIV ULTRAZVOČNE OBDELAVE NA LASTNOSTI ULTRA DROBNO ZRNATE MEDENINE

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This article establishes the relationship between the parameters of an ultrasonic action on the microstructure and the complex properties of ultrafine-grained brass. Ultrasonic action on brass after radial-shear rolling at certain amplitudes of mechanical stresses helps to relax the nonequilibrium structure of grain boundaries and relieve internal stresses. In this case, the relaxation effect of the structure depends on the amplitude of the ultrasound. It has been shown for the first time that ultrasonic processing is an effective way to relax the structure of highly deformed materials, which leads to an increase in the proportion of large-angle grain boundaries and triple grain joints, without leading to a significant grain growth.

Keywords: ultrasonic processing, ultrafine-grained, brass

V članku avtorji opisujejo postavitev zveze med parametri delovanja ultarzvočne obdelave na mikrostrukturo in kompleksne lastnosti ultra fino zrnate medenine. Delovanje ultrazvoka na medenino z določeno amplitudo po radialnem strižnem valjanju, ki povzroči notranje mehanske napetosti pomaga sprostiti le-te ter uravnoteži strukturo kristalnih mej. V tem primeru je učinek sprostitve mikrostrukture odvisen od amplitude ultrazvoka. Avtorji so prvič pokazali, da je ultrazvočna obdelava učinkovit način relaksacije mikrostrukture močno deformiranih materialov, ki povzroči povečanje deleža deleža veliko kotnih kristalnih mej in trojnih vezi kristalnih mej, ne da bi pri tem prišlo do pomembne rasti kristalnih zrn.

Ključne besede: ultrazvočna obdelava, ultra drobna kristalna zrna, medenina

## 1 INTRODUCTION

The requirements for the properties of structural materials in modern conditions are becoming higher and more differentiated. First of all, this applies to responsible metal products operating under extreme operating conditions in aerospace, chemical, medical and other industries. One of the methods of increasing the strength of materials is to reduce the size of crystallites from micron to ultrafine-grained (UFG) and nanoscale (NS) states.

The formation of an ultrafine-grained structure of metals and alloys with severe plastic deformation methods (SPD) makes it possible to exert a significant influence on the deformation behavior and mechanical properties of metals and alloys, which makes it possible to consider SPD a very promising method for controlling the structure and properties of metal materials.[1–3] The greatest interest in nanostructured materials is due not only to their unique physical-chemical properties, but also to very high mechanical properties: strength, ductility, wear resistance. Due to the fact that ultrafine-grained materials have appeared relatively recently, their resistance to external influences, in particular, to plastic deformation and heating, has not been sufficiently studied. The processes and methods of processing such materials have not yet been sufficiently studied.[4–6]

Significant advantages of radial-shear rolling is a complete structural processing of a metal along an entire section and its sealing, and an increase in the physical and mechanical properties of rolled products; due to a significant feed angle, an increase in the technological plasticity of workpieces by more than 300 % is achieved. In the deformation zone created with radial-shear rolling, a stress-strain state scheme is implemented, close to comprehensive compression with large shear strains, which is favorable for the formation of an UFG structure. The main feature of radial-shear rolling is the nonmonotonicity and turbulence of deformation, as well as differences in the plastic flow and the structure processing in different zones of a workpiece due to the trajectory-velocity features of the process.[7,8]

What is common to the UFG and NS materials obtained with deformation methods of nanostructuring is that their microstructure is nonequilibrium (metastable). Electron microscopic studies show a diffuse diffraction contrast of grain boundaries in UFG and NS materials, indicating the presence of high internal stresses. The sources of these stresses are nonequilibrium grain

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boundaries formed during plastic deformation. The high density of crystalline defects and significant stored energy lead to low thermal stability and conductivity of nanometals, which makes their application difficult. Materials scientists propose various processing methods such as incomplete recrystallization annealing, gradient deformation, etc., to produce a heterogeneous microstructure of a metal and then optimize the strength/ductility/conductivity ratio.

It has been shown that ultrasonic exposure leads to a noticeable increase in the thermal stability of the structure of UFG nickel, associated with the relaxation of the grain boundary structure, reduction of internal stresses and stabilization of the material structure at the same time.[9–11] The effect of increased plasticity in ultrasonic processing has been established, occurring simultaneously with an increase in the yield strength and tensile strength of UFG nickel obtained with equal-channel angular pressing (ECAP), which may be practically important for increasing the complexity of mechanical properties of UFG materials.

The Institute of Technical Acoustics (ITA) of the National Academy of Sciences of Belarus has developed designs of ultrasonic oscillatory systems and tools for obtaining and processing extended materials with an ultrafine-grained structure using the ECA-stretching method. This process increases the strength properties of materials while maintaining high plastic properties, allowing the production of extended products.[12]

It has been shown that ultrasonic processing reduces the defect density of ultrafine-grained nickel samples,[11] and affects the deformation behavior and the nature of martensitic transformations in materials.[13]

## 2 EXPERIMENTAL PART

It is proposed to use concentrator waveguides of various designs for processing UFG materials, depending on the geometric dimensions of the samples.

Since the maximum amplitude of displacements of ultrasonic transducers in the resonant mode is usually about 10–12 μm, a booster is used to increase the amplitude of vibrations of the working tool and match the transducer with the load. Boosters are transformers of the amplitude of longitudinal ultrasonic vibrations and are cylindrical rods with a variable cross-section, which are usually made of steels, titanium and aluminum alloys.

The most used boosters with respect to the possibility of obtaining significant displacement amplitudes at a low load are step boosters, whose amplitude gain is equal to the ratio of the input and output cross-sections. An ultrasonic oscillatory system using a step booster has a narrow range of operating frequencies and, therefore, a limited possibility of frequency adjustment with load changes. In this case, it is necessary that the ultrasonic generator has the function of automatic frequency tuning since a small deviation of the resonant frequency of the oscillatory system with a step booster in the absence of this function leads to a noticeable decrease in the efficiency of the entire oscillatory system. By selecting the transformation coefficient, it is possible to achieve the optimal load, matching with the converter. A designed titanium-alloy booster with a gain coefficient of 2.25 for ultrasonic processing of UFG samples is shown in **Figure 1**. The booster is designed for an operating frequency of 17.8 kHz of the PMS15A-18 magnetic-friction converter.

The presence of a transition section with an R10 rounding radius reduces the stress concentration in the concentrator material and provides more favorable conditions for the propagation of ultrasonic vibrations.

The calculation and manufacture of the components of an ultrasonic oscillatory system and a tool allowing processing samples of copper and brass in the form of discs with a diameter of up to 20 mm were performed. **Figure 2** shows a photo of the booster, waveguide and clamping screw manufactured according to the drawing presented above.

The PMS15A-18 converter was powered by ultrasonic generators UZG2-4M with an operating frequency of 16.8–19.2 kHz and an output power of 4.5 ± 0.5 kW, and UZG2-1M with an operating frequency of 22 ± 1.65 kHz and an output power of 1 ± 0.2 kW.

We calculated and manufactured the components of an ultrasonic oscillatory system and a tool allowing processing samples of copper and brass in the disc form with a diameter of up to 20 mm, as well as extended cylindrical samples with a diameter of 15 mm of half-wave

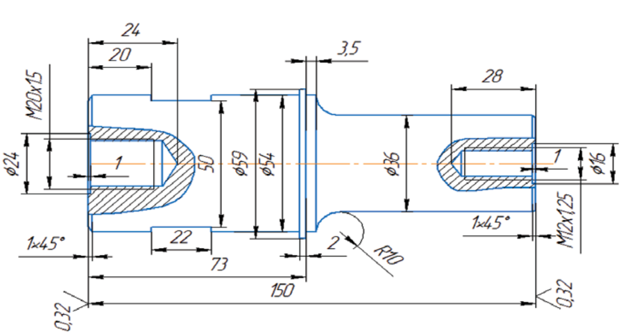


**Figure 1:** Drawing of a booster for UFG samples in the disk form


**Figure 2:** General view of the acoustic-system elements: 1 – booster, 2 – waveguide, 3 – clamping screw

**Figure 3:** General view of the samples in the disc form for ultrasonic processing

length. The studies were carried out on L63 brass samples with a thickness of 0.45–0.6 mm and in the form of discs with a diameter of 15 mm, which were cut from cylindrical samples after radial-shear rolling (**Figure 3**).

A sample was fixed inside the cavity of the waveguide in the antinode of mechanical stresses using a special screw, providing a reliable acoustic contact between the waveguide and the sample.

Ultrasonic processing of the samples was carried out with an amplitude of alternating stresses in the samples of up to 70 MPa using the scheme shown in **Figure 4**. The processing was performed with the same compression force in the voltage range of the waveguide while the tightening torque of the screw was 90 N·m.

The second part of the samples from the materials was made in the form of a cylindrical rod with a diameter of 15 mm and a length of 155 mm. It was the waveguide of the acoustic system.

**Figure 5** shows a diagram of ultrasonic processing of a long-length sample made of copper and brass, while **Figure 6** shows a drawing of a titanium-alloy booster with a gain coefficient of 2.5.

The booster is designed for an operating frequency of 21.8 kHz of a magnetic-friction converter. An M12×1.25 threaded hole is made on the left end surface of the booster for mounting it with a stud to the PMC1-1 converter.

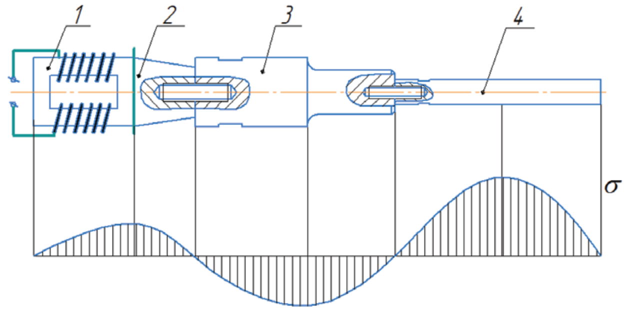


**Figure 5**: Scheme of ultrasonic processing of a long sample: 1 – ultrasonic transducer, 2 – concentrator, 3 – booster, 4 – waveguide

A hole with a thread for an M8 stud was made on the end surface of the samples, with which attachment to the booster was carried out.

During ultrasonic processing of the samples, the temperature of the waveguide was controlled, which did not exceed 45 °C at maximum values of alternating voltages. The amplitude of ultrasonic displacements at the end of the waveguide was measured with a non-contact vibrometer, whose measurement error was 2 % + 0.1 micron.

Some of the cylindrical samples were subjected to ultrasonic processing at low temperatures. After being placed in a Dewar laboratory glass vessel, the studied brass samples were pre-aged for 5 min before USP to equalize the temperature throughout the volume. The temperature in the processing zone of cylindrical samples was controlled by a copper-kopel thermocouple. After USP at low temperatures, the distribution of the microhardness over the sample surface was studied.

X-ray diffraction analysis (XRD) of the structure of copper and brass samples after ultrasonic treatment was carried out on X-ray diffractometers of the DRON-2 type (CuKa radiation). X-ray images were decoded using the Crystallographica Search-Match software. X-ray photography was carried out with Bragg-Brentano focusing in the scanning mode.

The obtained samples were also examined with differential scanning calorimetry (DSC) on a DSC 214 Polyma device. The process of measuring and processing the output information in the calorimeter was controlled by an IBM-compatible personal computer using Proteus,

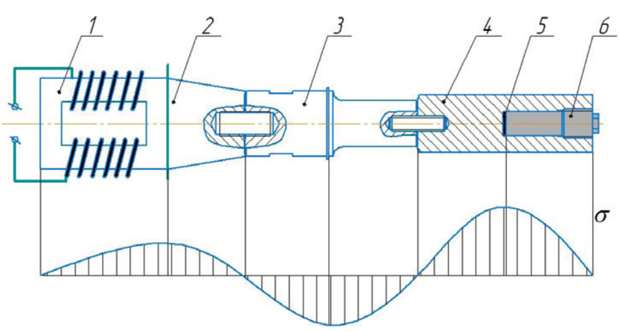


**Figure 4:** Scheme of ultrasonic processing of UFG samples in the disc form: 1 – ultrasonic transducer, 2 – concentrator, 3 – booster, 4 – waveguide, 5 – sample, 6 – preload screw

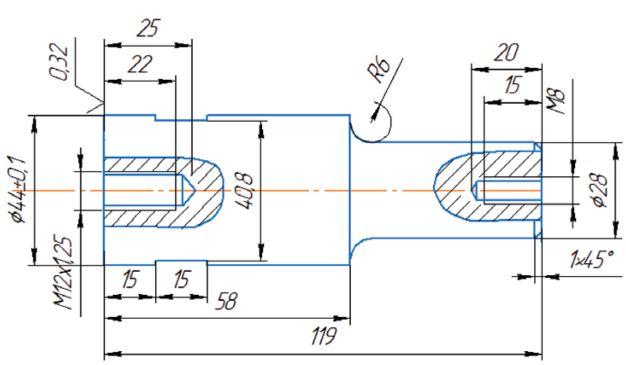


**Figure 6:** Drawing of a booster for ultrasonic processing of long samples

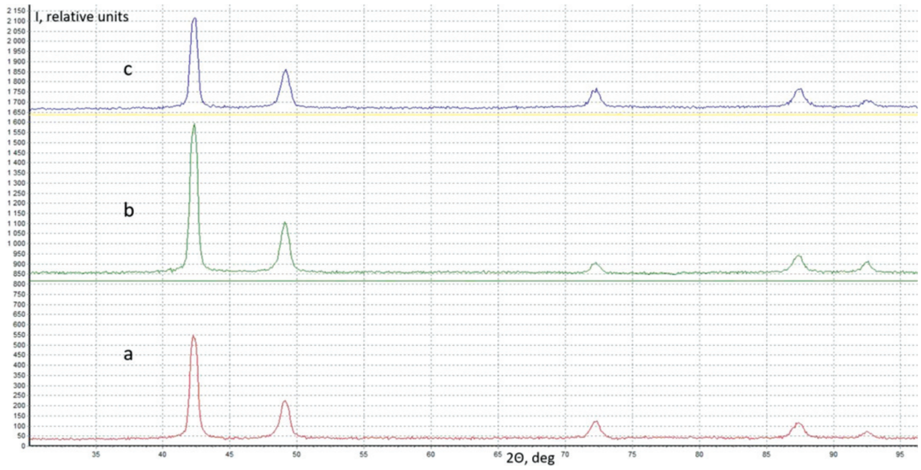


**Figure 7:** Diffractogram (CuKa) of the L63 brass sample after radial-shear rolling: a – without ultrasonic processing; b – ultrasonic processing at 40 MPa; c – ultrasonic processing at 70 MPa

a special software package. The samples were examined in a temperature range of 25–500 °C with a heating rate of 10 °C/min.

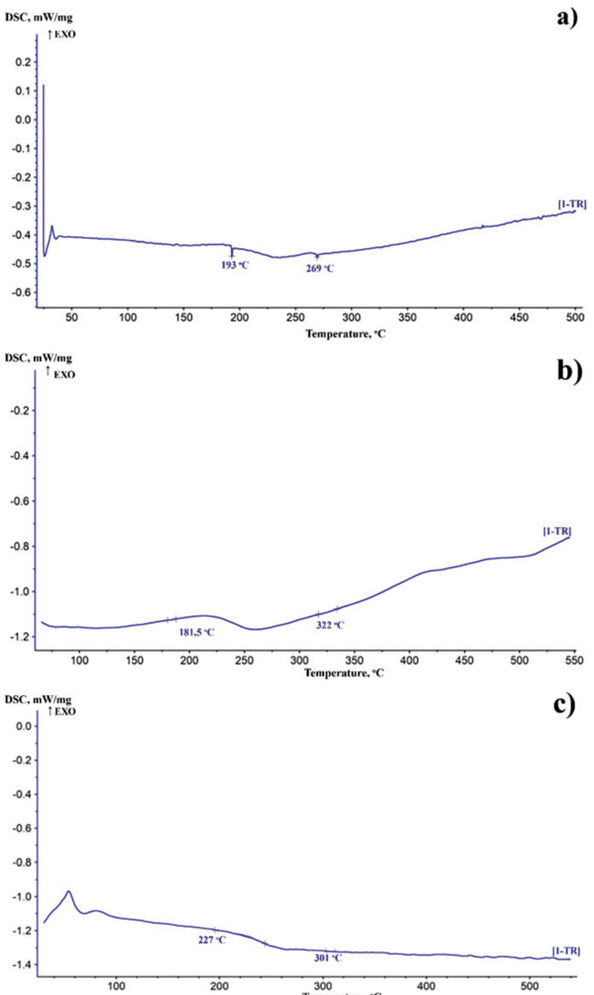


**Figure 8:** DSC curves of L63 brass samples: a – without ultrasonic processing; b – ultrasonic processing at 40 MPa; c – ultrasonic processing at 70 MPa

The study of the structure and determination of the grain size were carried out using a MeF-3 light microscope from Reichert (Austria) at a magnification of 100×.

The mechanical properties of copper and brass samples in the ultrasonic state before and after ultrasonic treatment at different values of the amplitude of alternating stresses were studied by measuring the microhardness on a Micromet-II microhardness meter with a load of 50 g according to GOST 9450-76.

## 3 RESULTS

The diffractograms of the studied samples obtained as a result of X-ray diffraction are shown in **Figure 7**.

The results of the DSC analysis are shown in **Figure 8**.

**Figure 9** shows photographs of the microstructure of L63 brass after radial-shear rolling without and with ultrasonic processing at different capacities.

The results of the microhardness measurements of various copper samples are shown in **Figure 10**.

## 4 DISCUSSION

A decrease in the broadening of X-ray lines indicates a decrease in the microstresses in the sample after the X-ray diffraction analysis, as well as a slight increase in coherent scattering regions (CSRs) (**Figure 7**). The size of the CSRs is usually identified with the average size of the crystallites; when this size is determined, it is possible to assess the nature of the change in the size of the crystallites in the material depending on the processing conditions. Based on the results of the X-ray diffraction analysis, the size of the CSR samples was estimated by broadening the reflexes on the obtained diffractograms according to the Scherrer–Selyakov formula:

$$D = \lambda_{Cu}/I \cdot \cos\Theta,$$

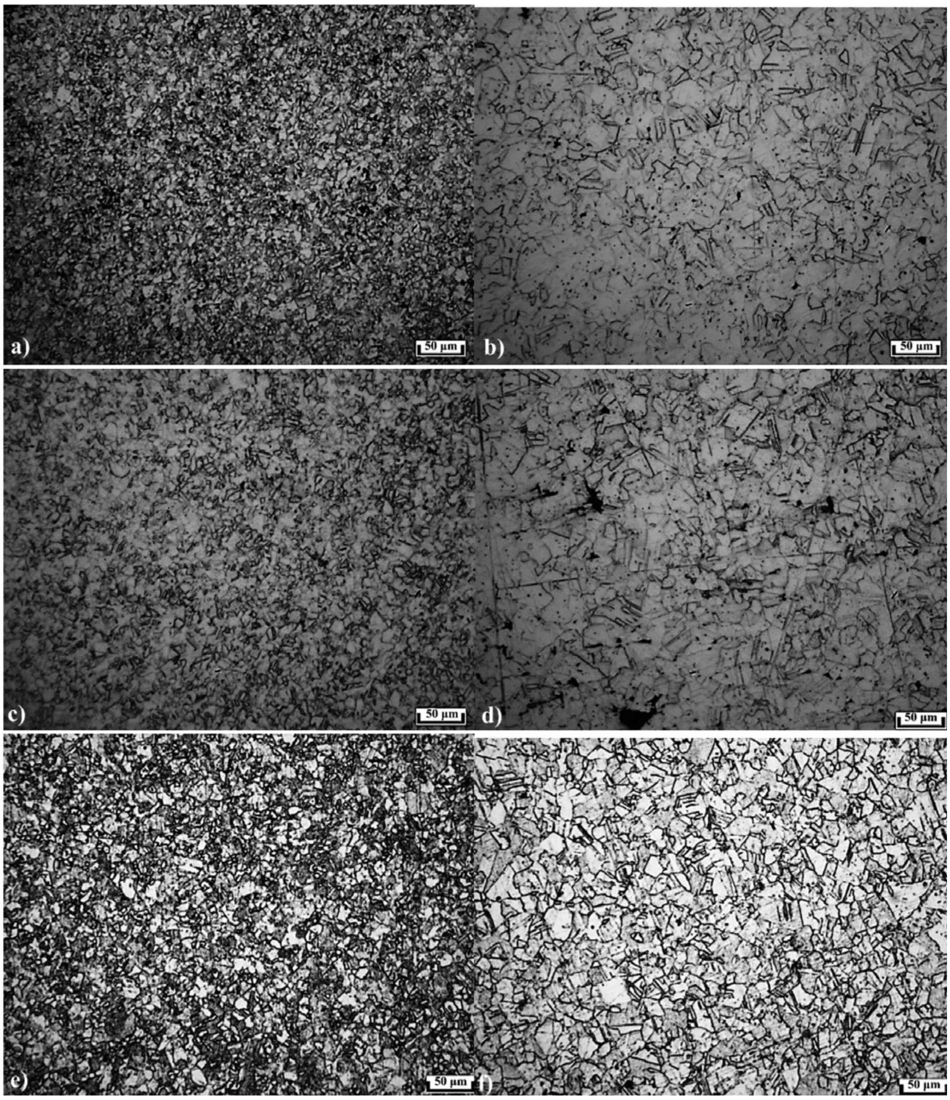


**Figure 9:** Microstructures of L63 samples at the edge of the surface (a, c, e) and in the center (b, d, f); a, b – without ultrasonic processing; c, d – ultrasonic processing at 40 MPa; e, f – ultrasonic processing at 70 MPa

where $\lambda_{Cu}$ – the radiation wavelength of the copper anode (0.1540 nm), $\Theta$ – the scattering angle, B – physical broadening.

The size of a CSR is usually identified with the average size of the crystallites; having determined which, it is possible to assess the nature of the change in the size of the crystallites in the material depending on the processing conditions. Ultrasonic processing led to an increase in the CSR by 8–20 % in all the samples. It was also established with the XRD method that brass of the L63 brand is a mixture of two main phases according to the equilibrium diagram of the Cu–Zn system. The main phases are the CuZn phase ($\beta$-phase) and a solid solution of zinc in copper ($\alpha$-phase). In addition to the basic alpha solution, there are copper and zinc form a number of phases of the electronic types beta, gamma and epsilon. Most often, the structure of brass consists of alpha or alpha + beta' phases: the alpha phase is a solid solution of

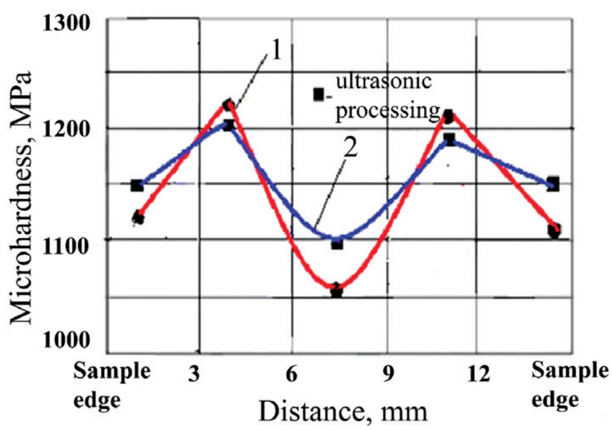


**Figure 10:** Distribution of microhardness: a) – over the diameter of a L63 disc after radial-shear rolling: 1 – without ultrasonic processing; 2 – ultrasonic processing at 40 MPa; b) – along the length of an L63 half-wave cylindrical sample after radial-shear rolling and ultrasonic processing at temperatures of: 1) 40 °C; 2) –100 °C; 3) diagram of alternating stresses at ultrasonic processing along the length of the sample

zinc in copper with a crystalline lattice of copper HCC, and the beta' phase is an ordered solid solution based on the $Cu_5Zn_8$ electron compound with a BCC lattice.

As the DSC analysis showed, there is one exothermic peak found for the L63 samples. The peak is caused by the $\alpha \rightarrow \beta$ phase transition in temperature ranges of 193–269 °C (**Figure 8a**), 182–322 °C (**Figure 8b**) and 227–301 °C (**Figure 8c**). Also, after ultrasonic processing, the peak area increases, which is associated with a change in the level of internal stresses.[14]

As can be seen from **Figure 9**, the microstructure of the surface layer of the L63 brass rod (**Figure 9a**) after radial-shear rolling it to a diameter of 15 mm (from the initial workpiece diameter of 30 mm) differs significantly from the microstructure in the central part of the bar (**Figure 9b**). In the surface zone, the average grain size is in a range of 5–10 μm, in the central zone, the grain size is in a range of 25–40 μm, i.e., there is a gradient distribution of the microstructure over the cross-section of the deformed rod. Subsequent processing of brass rods subjected to radial-shear rolling by ultrasonic processing with amplitudes of alternating stresses of 40 MPa and 70 MPa showed that USP practically does not affect the microstructure evolution in the central zone of a rod. Thus, with USP, with amplitudes of alternating stresses of 40 MPa and 70 MPa in the central part of the rod being formed, the grain size remains at the same level, i.e., in the range of 25–40 μm (**Figures 9d, f**). If neither ultrasonic processing itself nor the change in the amplitude of alternating stresses during USP does not significantly affect the change in the grain size in the central part of a workpiece, then the opposite is observed when studying the surface layers of the rods. In particular, USP leads to both the growth formed in the surface layer during radial-shear rolling of the grain and its alignment, i.e., a more uniform structure is obtained. At the same time, the higher the amplitude of alternating stresses in USP, the more the grain grows. Thus, with USP with an amplitude of alternating stresses of 40 MPa, the average grain size increased to 9–10 μm (**Figure 9c**), while with USP with an amplitude of alternating stresses of 70 MPa, the average grain size was in a range of 14–15 μm (**Figure 9e**). That is, the conducted studies indicate that although USP does not significantly affect the evolution of the microstructure in the central part of a rod, it allows a reduction in the gradient of the structure distribution over the cross-section of a rod deformed with a radial shear rolling mill due to an increase in the size of its surface zone. In addition, the higher the amplitude of alternating stresses during USP, the smaller is this gradient. USP also helps to obtain a more uniform structure (with a smaller grain-size spread) in the surface area of the deformed rod (**Figures 9c, e**).

Graphs of changes in the microhardness along the cross-section of a bar after radial-shear rolling and USP (**Figure 10a**) are generally consistent with the studies of the microstructure evolution during these processes. Thus, in both cases, the gradient of the microhardness distribution over the cross-section of a bar is observed. It is found that the gradient after radial-shear rolling and USP is lower than that after just radial-shear rolling. The highest values of the microhardness in the cross-section of a bar formed on a radial-shear rolling mill are observed in the intermediate zone (on average 4 mm from the surface of the bar). This is because two gfrinding effects are observed. First, due to the implementation of severe plastic deformation, intensive grain grinding occurs in the surface layer, which should lead to an increase in microhardness values. However, at the same time, another type of grinding – more intensive than in the intermediate and central zones – occurs in the surface layer, specifically targeting carbides, leading to a slight decrease in the microhardness of the surface zone (**Figure 10a**). Conducting a subsequent USP with an amplitude of alternating stresses of 40 MPa leads to a slight alignment of this gradient along the cross-section of the rod. At the same time, there are a slight increase in the microhardness (by about 25–30 MPa) in the surface layer, a more significant increase (by 50 MPa) in the central zone, and a slight decrease in the microhardness of the molded brass in the intermediate zone (by 25 MPa). In total, this leads to a decrease in the gradient of the microhardness distribution over the cross-section of a rod made of L63 brass alloy and formed on the radial-shear rolling mill.

A study of the microhardness distribution over the surface of a cylindrical sample made of brass alloy L63 after RSR and USP at temperatures of 40 °C and –100 °C (**Figure 10b**) showed that there is a dependence of the microhardness distribution on the magnitude of the alternating stresses during USP along the length of the sample and on the USP temperature. An increase in the intensity of ultrasonic processing leads to a decrease in the microhardness. At the same time, at a temperature of 40 °C, a systematic dependence of the distribution of microhardness on the surface of a rod, along the length of the sample, is observed in accordance with the diagram of alternating stresses during USP along the length of the sample, as opposed to USP at a temperature of –100 °C. This indirectly indicates that the L63 brass alloy exhibits more stable properties after its deformation on a radial-shear rolling mill and subsequent USP at the temperature of 40 °C than after USP at the temperature of –100 °C.

## 5 CONCLUSIONS

For ultrasonic processing of ultrafine-grained materials, an ultrasonic oscillatory system was developed, consisting of an ultrasonic transducer, booster and waveguide, providing an amplitude of oscillating stresses in the processed samples of up to 70 MPa. Ultrasonic equipment for processing brass samples was tested. The relationship of the ultrasonic exposure parameters with the microstructure and complex properties of the UFG copper and brass samples were established. It was shown that an ultrasonic action carried out on the materials after radial-shear rolling at certain amplitudes of mechanical stresses promotes the relaxation of the nonequilibrium structure of grain boundaries, thus relieving internal stresses. The scientific understanding of the mechanism and patterns of the structure formation in UFG metals and alloys under ultrasonic exposure has been expanded.

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