

Combination of cryogenic deformation and electropulse processing as a way to produce ultrafine-grain metals

© 2023

*Mikhail V. Markushev**, Doctor of Sciences (Engineering), senior researcher, Head of laboratory

Elena V. Avtokratova, PhD (Engineering), senior researcher

*Aigul Kh. Valeeva*¹, PhD (Engineering), researcher

*Irshat Sh. Valeev*², PhD (Engineering), researcher

*Rafis R. Ilyasov*³, junior researcher

*Stanislav V. Krymsky*⁴, PhD (Engineering), Head of laboratory

Oleg Sh. Sitdikov, PhD (Physics and Mathematics), senior researcher

Institute for Metals Superplasticity Problems of RAS, Ufa (Russia)

*E-mail: mvmark@imsp.ru

¹ORCID: <https://orcid.org/0000-0003-4305-4538>

²ORCID: <https://orcid.org/0009-0002-5162-7324>

³ORCID: <https://orcid.org/0000-0003-0195-1206>

⁴ORCID: <https://orcid.org/0000-0002-1534-3239>

Received 15.08.2023

Accepted 01.12.2023

Abstract: The data of a comparative analysis of the structure and hardness of pure metals with a face-centered cubic lattice – aluminum, nickel and copper, subjected to complex thermomechanical treatment (TMT), including isothermal cryogenic rolling at liquid nitrogen temperature and subsequent high-density electropulse treatment (EPT) were presented. The main stages, features and advantages of TMT, which first ensure strong work hardening of the processed material due to deformation at low temperatures and then its ultra-fast contact electropulse heating up to a specified temperature, were considered. A multi-level analysis of the metals structure evolution due to TMT was carried out using modern methods of scanning electron microscopy and X-ray diffractometry, recording a wide range of its linear and angular parameters. The kinetics and nature of the processes of the metals structure evolution under cryogenic rolling and EPT, their driving forces and controlling factors, as well as general patterns and temperature intervals of activation of the deformation structure recovery and recrystallization influenced by an electric pulse are identified and discussed. Based on the results of the analysis of the structural and mechanical behaviour of metals, it was concluded that the combination of severe plastic cryogenic deformation and a single-step treatment with ultrashort alternating current pulses is an effective way to obtain semi-finished products with controlled parameters of their structure and properties, including high-strength ultrafine-grain rolled products. At that the phenomenology and nature of the strengthening/softening of metals during cryogenic rolling and subsequent electropulsing are similar to those observed under cold rolling and furnace annealing.

Keywords: FCC metals; cryogenic deformation; cryogenic rolling; electric pulse treatment; ultrafine-grain structure.

Acknowledgments: The work was carried out within the framework of the state assignment of the Federal State Budgetary Institution of Science Institute for Metals Superplasticity Problems of the Russian Academy of Sciences.

Experimental studies were carried out on the base of the Collaborative Access Centre “Structural, Physical and Mechanical Studies of Materials” of the RAS.

For citation: Markushev M.V., Avtokratova E.V., Valeeva A.Kh., Valeev I.Sh., Ilyasov R.R., Krymsky S.V., Sitdikov O.Sh. Combination of cryogenic deformation and electropulse processing as a way to produce ultrafine-grain metals. *Frontier Materials & Technologies*, 2023, no. 4, pp. 53–62. DOI: 10.18323/2782-4039-2023-4-66-5.

INTRODUCTION

The development of effective industrial methods for increasing the service properties of metals and alloys, through the controlled strain and thermal effects on them leading, among other things, to a decrease in the size of crystallites (grains and subgrains) to nanosizes [1–3] is an urgent task of modern materials science. Among these methods are thermomechanical treatment (TMT) based on a combination of cryogenic rolling (CR) and subsequent electric pulse treatment (EPT) [4–6] and ensuring at least the formation of an ultrafine-grained (UFG) structure (with a grain size of less than 10 μm) in the processed material.

Currently, the effect of temperature decrease during the transition from cold (at room temperature) to cryogenic (at temperatures below 120 K) deformation on the structure

and mechanical behaviour of metallic materials has been studied quite well. Generally, it is described as their extra strengthening due to the suppression of dynamic recovery, and an increase in the dislocation density [7–9]. Moreover, in a number of studies, for example in [9–11], strengthening is also associated with crystallite refinement.

At the same time, the effect of electric current on the deformation structure of metals and alloys has been studied to a much lesser extent, especially when using short-time high-density current pulses [12], and in relation to cryodeformed materials. Most investigations in this area connected with direct current, and with its long-term (up to several hours) exposure [13; 14]. Carrying out such experiments was primarily caused by the needs to study the peculiarities of the influence of contact electric heating on the structural and mechanical behaviour of objects during their direct deformation

processing, as well as by the search for a less expensive alternative to their non-contact (furnace, induction) heating. The main result of these studies includes a convincing demonstration of high efficiency when using electric heating of blanks for deformation and post-deformation heat treatment [13; 14]. The papers [15–17] describe the so-called electroplastic effect, which “facilitates” the process of metal plastic flow, when directly exposed to electric current, and propose options for interpreting its nature.

As a result of a few studies, where short-time electric pulse exposure was implemented, it was found that the activation of the recovery, polygonization and recrystallization processes [18] in pre-strained metal materials, improves a number of their technological properties [15; 19; 20]. The reason is obvious – a decrease in strength, and an increase in ductility due to a decrease in the structure defectiveness and an increase in its dispersity [2; 18]. However, there is some literature data with an opposite trends in structure changes, indicating the multidirectionality of the EPT effect on the structure and properties of materials. For example, in [21] it was noted the suppression of recrystallization in steels during EPT.

There is a belief that the softening of work-hardened material during electro-induced heating is accompanied by the so-called “electron wind” effect [22] contributing to an increase in the mobility of dislocations, and the accelerated formation of recrystallization centres. While the latter phenomenon can be explained within the framework of classical ideas on the influence of heating rate on nuclei formation during recrystallization [18], the nature of the former still does not have a clear physical interpretation. Another EPT character feature is the skin-effect [20; 22]. It is caused by the uneven distribution of electric and heat flows inside and on the surface of the blank (conductor) under the electrical impulse action. As a result, the structure formed on the surface may differ significantly from the structure inside the blank. In this case, the degree of grain size nonhomogeneity, and the depth of the subsurface layer directly depend on the nature of the material being processed, the shape and cross-sectional area of the conductor, as well as on the EPT parameters, in particular on the pulse energy [20; 22].

From general point of view, TMT based on a combination of CR and EPT should be the most popular and effective for pure metals and solid solutions. The reason is that in such objects, there are no dispersed particles of second phases, which act as the main factor in controlling the structure and strength of alloys through limiting the rearrangement of dislocations in them and the growth (migration of boundaries) of crystallites [18]. During TMT of metals and solid solutions, their structuring is controlled by dislocation reactions, which are often determined by stacking fault energy (SFE). In our previous works [23–26], we have already analysed and partially published data on the phenomenology and nature of the structural-mechanical behaviour of various pure fcc metals, subjected to rolling up to high strains at liquid nitrogen temperature, and subsequent single-step EPT with different integral current densities. At the same time, in the papers mentioned, the analysis of the data obtained was carried out separately for each metal, and the overall picture of changes in their structure and properties occurring during TMT was not presented.

The purpose of this work is to supplement the published data, identify common patterns, and discuss the nature of

the structural and mechanical behaviour of three fcc metals – Al, Cu and Ni during cryogenic rolling, and subsequent electric pulse treatment, taking into account the differences in their melting temperature and stacking fault energy value.

METHODS

Al (99.99 %), Ni (99.5 %), and Cu (99.99 %) were used as research materials. Plates cut from forged and annealed coarse-grained billets, were subjected to cryogenic deformation to a total strain of 90 % ($\epsilon=2.3$) on a laboratory isothermal six-roll mill with a removable work rolls diameter of 64 mm. Isothermal rolling conditions were ensured by pre-cooling the work rolls and blanks in a bath of liquid nitrogen for 1 h. The deformed blanks were cooled after each pass and the rolls – after 4–5 passes. The absence of nitrogen boiling was the criterion for achieving the required temperature. The deformation temperature of Ni, Cu and Al was about 0.05, 0.06 and 0.08 of their melting point, consequently. The strain per pass did not exceed 7 % at a rolling speed of about 100 mm/s. The rolled strips were stored in a freezer at a temperature of -18 °C. The duration of their room temperature annealing during the subsequent TMT stage, as well as during the objects fabrication and analysis of the structure and properties of metals, was recorded to control the degree of their softening.

Using a MIU-20 device, EPT was carried out on flat samples with a gauge part of 3×4 mm cut on an electric spark machine from cryo-rolled strips 0.4 mm thick along the direction of their rolling. The selected thickness guaranteed the absence of skin effect in the working area of samples, made of all metals and the uniform distribution of thermal and electrical flows over their cross section. The samples were fixed in clamps served as current conductors during the discharge of the capacitor bank. The current pulse was recorded using a Rogowski coil, and an AKTAKOM ASK-3107 storage oscilloscope. As the pulse energy criterion, the integral current density K_j was taken, which was calculated using the following equation [22]:

$$K_j = \int_0^\tau j^2 \partial\tau = \frac{k^2}{S^2} \cdot \frac{A_1^3}{A_2} \cdot \frac{\tau}{4 \ln(A_1 / A_3)},$$

where j is the current density;

τ is the pulse duration;

k is the Rogowski coil coefficient;

S is the cross-sectional area of the sample;

A_1 , A_2 and A_3 are the first, second, and third amplitudes of the damped pulse, respectively.

During EPT, the sample was heated according to the Joule – Lenz law. Due to pulsed thermoelectric action of about $\sim 10^{-4}$ s, reliable measurement of sample heating temperature (T_h) was a difficult technical point. In this regard, as recommended in [22], this temperature was determined by the calculation method, assuming that

$$\frac{j^2}{\sigma_e} = \rho c \frac{\partial T}{\partial t},$$

where ρ , c , σ_e are the density, heat capacity and electrical conductivity of the metal being processed, respectively.

By integrating this equation, the authors obtained the dependences of the heating temperature of metals on the integral pulse current density.

The microstructure of metals was analysed in the rolling plane by the EBSD method using a TESCAN MIRA 3 LMH scanning electron microscope, and the HKL Channel 5 software package. Diffraction patterns were indexed by 6 Kikuchi lines with a scanning step of no more than 0.5 μm . A grain-boundary angle of 15° was used as a criterion for dividing into low- and high-angle boundaries (LABs and HABs). The sizes of grains and subgrains (d_g and d_{sg}) were determined by the equivalent diameter method. The average grain-boundary angle of intercrystalline boundaries (Θ_{av}) and the fraction of high-angle and twin boundaries (F_{hab} and F_{Σ}) were determined from the grain-boundary angle spectra. In this case, boundaries with $\Theta < 2^\circ$ were not taken into account. The fraction of recrystallized grains (F_{rec}) was determined as the ratio of the area they occupy to the area of the map.

X-ray diffraction analysis (XRD) was carried out on a DRON-4-07 diffractometer in Cu-K α radiation at a voltage of 40 kV and a current of 30 mA with a wavelength of $\lambda = 1.54418 \text{ \AA}$. The scanning was carried out with a rotating sample, a step of 0.1° and an exposure time of 4 s, using a graphite monochromator on a diffracted beam. The root-mean-square microstrain of the crystal lattice ($\langle \epsilon^2 \rangle^{1/2}$) and the size of coherent domain size (D) were calculated by full-profile analysis in the MAUD software package. The dislocation density (ρ) was determined as

$$\rho = \frac{2\sqrt{3}\langle \epsilon^2 \rangle^{1/2}}{D \times b},$$

where b is the Burgers vector.

The strength of metals was assessed by the microhardness level determined by the Vickers method using 10 measurements on an MVDM 8 AFFRY hardness tester at loads of 1 and 0.5 N and duration of their application of 10 s in the central part of the rolled strips and samples subjected to EPT.

Structure analysis and assessment of the hardness of metals were carried out in laboratory conditions. In this case, the total duration of cryo-rolled samples at room temperature required for preparation of objects (samples for EPT) and their structural and mechanical analysis did not exceed 24 h.

RESULTS

Tables 1 and 2 show the results of assessing the linear and angular parameters of the structure of Al, Cu, and Ni, as well as their hardness recorded after cryogenic rolling and EPT with different integral current densities. Even a cursory glance at these data allows concluding that the implemented TMT scheme makes it possible to effectively control the processes of deformation structure transformations of all studied metals, to comparatively easy change its type and parameters, and most importantly, to ensure the achievement of its main goal. In particular, to obtain sheets in three main structural states: 1) with a developed dislocation-cellular structure with nanosized cells; 2) with

a homogeneous UFG structure with a grain size of 1–3 μm ; 3) with an “intermediate” composite structure with controlled dislocation density and ratios of the main components, as well as low- and high-angle boundaries.

As should be expected, due to the lowest melting temperature (the highest homologous CR temperature), the least dispersed (with the largest crystallite sizes) structure of all types was recorded in aluminium (Table 1). Such a result was caused by the formation during cryogenic rolling of coarsest and the least developed cellular structure with an integral dislocation density almost an order of magnitude lower than in other metals (Table 2). Another fact that attracted attention was that with an increase in the pulse energy (heating temperature) during EPT, the hardness of cryo-rolled metals noticeably decreased. At the same time, the almost twofold softening observed in the studied range of integral current densities was caused primarily by the activation of recrystallization processes (see changes in F_{rec} and F_{hab} in Table 1 and dislocation densities in Table 2).

Despite the differences in the structure types and parameters and in the level of strength of metals recorded at various TMT stages, there was a similarity in their behaviour during EPT. Thus, the dependences of the hardness of all cryo-rolled metals on the homologous temperature of their heating turned out to be qualitatively similar (Fig. 1). These dependences consist of three characteristic temperature regions indicating a similar nature and kinetics of the development of thermally activated processes under electric pulsing. In the first region, the deformation structures were relatively stable and retained the maximum strengthening effect recorded after rolling. In the second and third regions, the metals became softer. In the second region, intensive softening was occurred in a narrow temperature range. In the third region, on the opposite, it was observed weak softening, leading to an almost complete loss of deformation and structural (due to grain refinement) hardening by cryorolled metals.

It is noteworthy that for all three metals, the boundaries between the marked regions were quite close, although the parameters of their deformation structure and the level of stacking fault energy were different. Thus, the deformation structure of Ni was retained after EPT up to the temperature of heating the sample to 0.33 T_m with its SFE of about 90 mJ/m^2 , Cu – up to 0.38 T_m with the SFE of about 70 mJ/m^2 , and Al – up to 0.42 T_m at SFE of about 200 mJ/m^2 . To define more exactly, the SFE values of metals are given based on the averaged values published in [27–29]. Intense softening of Ni was completed at 0.36 T_m , Cu and Al – at 0.42–0.45 T_m . At first glance, these data indicated the absence of SFE influence on the structural and mechanical behaviour of metals during TMT. However, in reality, this effect took place in the form of structural manifestations of the processes of accumulation of internal stresses during rolling and their relaxation during EPT. Thus, when rolling aluminium, its highest SFE level contributed to the least accumulation of dislocations and lattice distortions among the metals studied due to the easiest dynamic recovery. During EPT, the same factor ensured the development of polygonization in aluminium, which preceded recrystallization. Meanwhile in copper and nickel there were observed an active twinning, resulting in new a smaller size of recrystallized grains due to the lower mobility of twin boundaries.

Table 1. EBSD data on metals structure parameters after cryorolling to 90 % and further single electropulsing with different energies
Таблица 1. Параметры структуры криокатанных до 90 % и подвергнутых однократной ЭИО с различной энергией импульса металлов по данным EBSD-анализа

Metal	State	K_j , $10^4 \text{ A}^2/\text{mm}^4$	T_h , K	T_h/T_m	d_{sg} , μm	d_g , μm	F_{rec} , %	Θ_{av} , deg	F_{hab} , %	F_{Σ} , %
Al	CR	–	293	0.31	2.0±0.3	4.0±0.4	3	7	11	–
	EPT	1.0	398	0.42	2.0±0.4	4.0±0.8	6	16	23	–
		1.2	423	0.45	5.0±0.3	19.0±1.1	66	31	76	–
Ni	CR	–	293	0.17	0.2±0.1	2.5±0.5	3	7	7	<1
	EPT	1.0	573	0.33	1.3±0.2	1.9±0.4	5	5	4	44
		1.1	593	0.34	2.8±0.6	3.2±0.6	92	47	92	42
Cu	CR	–	293	0.20	0.3±0.1	0.7±0.2	15	17	33	2
	EPT	3.5	513	0.38	0.4±0.1	0.9±0.1	37	21	42	5
		3.8	573	0.42	0.8±0.2	1.0±0.2	86	39	79	22
		6.8	923	0.68	1.2±0.2	1.3±0.3	96	43	88	30

Note. K_j is integral current density;
 T_h/T_m is heating/melting temperature;
 d_g and d_{sg} are grain and subgrain size, respectively;
 F_{rec} , F_{hab} and F_{Σ} are fraction of recrystallized grains, high-angle boundaries and twin boundaries, respectively;

Θ_{av} is average angle of grain boundary misorientation.

Примечание. K_j – интегральная плотность тока;

T_h/T_m – температура нагрева/плавления;

d_g и d_{sg} – размер зерна и субзерна соответственно;

F_{rec} , F_{hab} и F_{Σ} – доля рекристаллизованных зерен, высокоугловых границ и двойниковых границ соответственно;

Θ_{av} – средний угол разориентировки межкристаллитных границ.

Data in Table 2 also follow that the maximum strain energy was stored in nickel, the homologous rolling temperature of which was the lowest, and the SFE level was close to copper. Therefore, to activate recrystallization in nickel, processing with the minimum pulse energy for three metals was required. As a result, after EPT, even with an energy of only $K_j=1.1 \times 10^4 \text{ A}^2/\text{mm}^4$, it was possible to fix in nickel a grain structure close in degree of dispersion to copper, and treated with a current pulse with an integral density almost 4 times higher (Table 1). At the same time, it should be noted that EPT of copper with $K_j=3.5 \times 10^4 \text{ A}^2/\text{mm}^4$ did not lead to significant changes in the linear parameters of the structure and hardness with a simultaneous increase in the proportion of HABs and recrystallized grains against the background of a twofold decrease in internal lattice distortions. At the same time, the homologous rolling temperature of copper was slightly higher, and occupied an intermediate position among the metals studied, and its SFE was slightly lower than in nickel. Structural changes during EPT detected in copper were caused by the greater intensity of the processes of static recovery and recrystallization,

leading to a more significant increase in the structure equilibrium, and to a significant compensation of the softening effect by the strengthening one, caused by the formation of submicron-sized grains. In other words, probably the more active formation and improvement of the structure of ultrafine crystallites, and their boundaries were the reason for the less intensive softening of cryorolled copper in this EPT area. When increasing current density to $K_j=3.8 \times 10^4 \text{ A}^2/\text{mm}^4$, the stored deformation energy (and hardness) of copper, similar to nickel and aluminium, sharply decreased due to the transformation of the subgrain structure into a partially recrystallized one with significantly larger subgrains (Table 1). This structure was also characterized by almost complete levelling of lattice microdistortions, due to a decrease in the dislocation density to an equilibrium level (Table 2). A further increase in the pulse energy upon transition to the third EPT temperature range, led to the recrystallization in the full volume of the processed materials accompanied by subsequent normal growth of recrystallized grains (Table 1), which was able to occur even with an extremely short time of exposure of electric current.

Table 2. XRD data on structure parameters and hardness of metals after cryorolling to 90 % and further single electropulsing with different energies

Таблица 2. Параметры структуры по данным РСА и твердость криокатаных до 90 % и подвергнутых однократной ЭИО с различной энергией импульса металлов

Metal	State	K_j , $10^4 \text{ A}^2\text{s/mm}^4$	T_h , К	T_h/T_m	ρ , 10^{14} m^{-2}	$\langle \varepsilon^2 \rangle^{1/2}$, %	D , nm	HV
Al	CR	–	293	0.31	0.5	0.060±0.001	340±4	49±5
	EPT	1.0	398	0.42	<0.01	0.001±0.001	156±2	49±4
		1.2	423	0.45			96±3	31±4
		2.9	623	0.67			123±4	26±5
Ni	CR	–	293	0.17	3.5	0.165±0.002	68±4	246±8
	EPT	1.0	573	0.33	<0.01	0.001±0.001	75±3	242±7
		1.1	593	0.34			129±2	101±8
		1.6	943	0.55			141±2	91±9
Cu	CR	–	293	0.20	4.5	0.186±0.004	57±2	152±7
	EPT	3.5	513	0.38	<0.1	0.001±0.001	48±2	143±8
		3.8	573	0.42			67±4	96±7
		6.8	923	0.68			100±4	81±8

Note. K_j is integral current density; T_h/T_m is heating/melting temperature; ρ is dislocation density; $\langle \varepsilon^2 \rangle^{1/2}$ is root-mean-square microstrain of crystal lattice; D is coherent domain size.

Примечание. K_j – интегральная плотность тока; T_h/T_m – температура нагрева/плавления; ρ – плотность дислокаций; $\langle \varepsilon^2 \rangle^{1/2}$ – среднеквадратичная микродеформация кристаллической решетки; D – размер областей когерентного рассеяния.

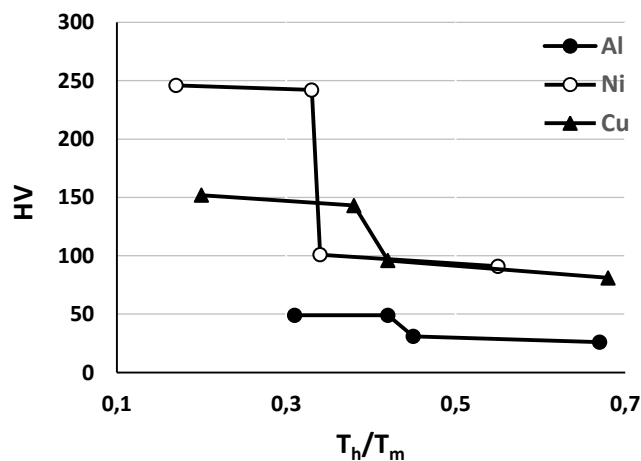


Fig. 1. Changes in hardness of cryorolled metals after room temperature annealing and further heating to a T_h temperature due to electropulsing
Рис. 1. Изменения твердости криокатаных металлов, вызванные отжигом при комнатной температуре и ЭИО с нагревом до температуры T_h

DISCUSSION

The generalized character of behaviour of cryodeformed metals during EPT can be represented as follows. In the first temperature range, the main mechanism for their structure transformation was static recovery, which led to a decrease in the defectiveness of the deformation structure and its improvement. Along with the recovery, static recrystallization developed locally, and prevailed in the second temperature range. As a result, when heating nonequilibrium work-hardened metals within this range, their predominant dislocation-cellular structure was almost completely replaced by an equilibrium grain-type one, which was accompanied by a decrease in their strength almost to the level of undeformed metal. In the third range there was occurred normal grain growth, which led to further softening of metals due to the loss of the Hall–Petch effect caused by the refinement of recrystallized grains [18].

Compared to the continuous static recrystallization, observed during furnace annealing of metals highly deformed at room temperature [2; 18], during EPT in the range of $0.3\text{--}0.4 T_m$, the main process of transforming the deformation structure of cryorolled metals, was a transition process of the structure improvement by the *in situ* type, controlled by a recovery, to normal grain growth. A sharp drop in hardness at the boundary of the first and second temperature ranges in Fig. 1 was caused by two main reasons. On the one hand, the growth of individual nanosized crystallites formed during rolling, which could potentially serve as recrystallization centres, intensified. On the other hand, due to the structural heterogeneity inherent in the deformed state and the different recrystallization kinetics in each individual grain, grains formed *in situ* in the deformed structure acquired the potential for accelerated growth. When the pulse energy/heating temperature, during EPT, reached a certain “threshold” level, some grains were “freed” from defects, which activated their growth, while neighbouring grains still “occupied” with high-densities of dislocations and, could undergo polygonization or twinning. The grain growth was the result of migration of their boundaries, which occurred under the influence of driving forces caused by previous deformation. The migrated boundaries of growing grains absorbed the crystal structure defects (vacancies, dislocations and other boundaries) [18], which led to a decrease in the internal energy of the system and, as a consequence, to a decrease in the metal strength level achieved due to rolling.

It should also be noted, that the boundaries of new grains formed as a result of severe low-temperature deformation had increased specific energy, and accordingly, the ability to migrate more quickly during EPT. In contrast to such boundaries, low-energy low-angle boundaries of both dynamically and statically formed cells had less mobility. As a result, at low heating energies/temperatures, migration of the boundaries of recrystallized grains predominantly took place towards the deformed matrix. At higher energies, in particular, after the deformed structure disappearance (the end of the transition from the polygonization and recrystallization processes to normal grain growth), grain boundaries continued to migrate under the influence of a driving force, caused by a decrease in

the local radius of their curvature. This process was accompanied by an increase in grain sizes due to their normal growth [18].

Based on the above, and considering the results of few investigations [5; 10; 18; 26], one can conclude that the processes occurring during EPT of cryorolled fcc metals are close to the ones occurring at static annealing of cold-strained materials: static recovery, continuous static recrystallization, and grain growth. At that, the short duration (pulse nature) of the thermal effect on the deformed metal during EPT was compensated by the high applied energy. Since recovery and static recrystallization are controlled by self-diffusion [18; 30], then, according to the Arrhenius law, even a slight increase in the EPT temperature/energy should lead to a noticeable increase in the rate of these processes, and consequently, to a decrease in the time of their completion, which is what we observed in the experiments.

Thus, the data obtained cannot testify for either the presence or absence of the “electron wind” effect, which could have a noticeable impact on the structural and mechanical behaviour of cryorolled fcc metals during high-density electropulsing. All experimental results found and discussed can be explained from the well-known, classical positions developed for conventionally work-hardened materials, subjected to annealing without exposure to electric current.

CONCLUSIONS

1. Thermomechanical treatment based on a combination of severe plastic deformation at cryogenic temperatures, and subsequent high-density electric pulsing, is an effective way to produce sheets of pure fcc metals with controlled structure, and strength parameters, including those with homogeneous UFG recrystallized structure with a grain size of $1\text{--}3 \mu\text{m}$, and a developed dislocation-cellular structure with nanosized cells.

2. The processes of softening of cryorolled fcc metals under electric pulse exposure to high-density currents, are similar in nature and kinetics and are characterized by the presence of clearly defined three energy/heating temperature intervals. In the first of them, EPT has virtually no effect on the level of metal hardness after cryogenic rolling to high strains. In the second one, starting from a certain “threshold” value of the integral current density K_j , corresponding to the calculated temperature equal to $0.33 T_m$ for nickel, $0.38 T_m$ for copper and $0.42 T_m$ for aluminium, their hardness is significantly reduced, due to the activation of static recrystallization of the deformation structure. When the heating temperature exceeded the values equal to $0.36 T_m$ for nickel, and $0.42\text{--}0.45 T_m$ for copper and aluminium, the third EPT region is observed, which accompanied by low hardness loss to the values corresponding to the original undeformed states, caused by the recrystallized grain growth.

3. The processes of transformation of a highly work-hardened structures during EPT of cryorolled fcc metals, are close to the processes occurring during static annealing of cold-deformed materials – static recovery, continuous static recrystallization and grain growth.

REFERENCES

- Estrin Y., Vinogradov A. Extreme grain refinement by severe plastic deformation: A wealth of challenging science. *Acta Materialia*, 2013, vol. 61, no. 3, pp. 782–817. DOI: [10.1016/j.actamat.2012.10.038](https://doi.org/10.1016/j.actamat.2012.10.038).
- Zhilyaev A.P., Pshenichnyuk A.I., Utyashev F.Z., Raab G.I. *Superplasticity and Grain Boundaries in Ultrafine-Grained Materials*. Amsterdam, Elsevier Publ., 2020. 416 p.
- Edalati K., Bachmaier A., Beloshenko V.A., Beygelzimer Y., Blank V.D., Botta W.J. Nanomaterials by severe plastic deformation: review of historical developments and recent advances. *Materials Research Letters*, 2022, vol. 10, no. 4, pp. 163–256. DOI: [10.1080/21663831.2022.2029779](https://doi.org/10.1080/21663831.2022.2029779).
- Pan Dong, Zhao Yuguang, Xu Xiaofeng, Wang Yitong, Jiang Wenqiang, Ju Hong. Effect of High-Energy and Instantaneous Electropulsing Treatment on Microstructure and Properties of 42CrMo Steel. *Acta Metall Sin*, 2018, vol. 54, no. 9, pp. 1245–1252. DOI: [10.11900/0412.1961.2017.00562](https://doi.org/10.11900/0412.1961.2017.00562).
- Konkova T., Valeev I., Mironov S., Korznikov A., Myshlyaev M.M., Semiatin S.L. Effect of electric-current pulses on grain-structure evolution in cryogenically rolled copper. *Journal of Materials Research*, 2014, vol. 29, no. 22, pp. 2727–2737. DOI: [10.1557/jmr.2014.299](https://doi.org/10.1557/jmr.2014.299).
- Konkova T., Valeev I., Mironov S., Korznikov A., Korznikova G., Myshlyaev M.M., Semiatin S.L. Microstructure response of cryogenically-rolled Cu–30Zn brass to electric-current pulsing. *Journal of Alloys and Compounds*, 2016, vol. 659, pp. 184–192. DOI: [10.1016/j.jallcom.2015.11.059](https://doi.org/10.1016/j.jallcom.2015.11.059).
- Khaymovich P.A. Cryodeformation of metals under all-around compression (Review Article). *Fizika nizkikh temperatur*, 2018, vol. 44, no. 5, pp. 463–490. EDN: [YTJSLG](https://www.edn.ru/ytjslg/).
- Panigrahi S.K., Jayaganthan R. A Study on the Combined Treatment of Cryorolling, Short-Annealing, and Aging for the Development of Ultrafine-Grained Al 6063 Alloy with Enhanced Strength and Ductility. *Metalurgical and Materials Transactions: A*, 2010, vol. 41, pp. 2675–2690. DOI: [10.1007/s11661-010-0328-x](https://doi.org/10.1007/s11661-010-0328-x).
- Magalhães D.C.C., Kliauga A.M., Ferrante M., Sordi V.L. Plastic deformation of FCC alloys at cryogenic temperature: the effect of stacking-fault energy on microstructure and tensile behavior. *Journal of Materials Science*, 2017, vol. 52, pp. 7466–7478. DOI: [10.1007/s10853-017-0979-8](https://doi.org/10.1007/s10853-017-0979-8).
- Ma E. Eight Routes to Improve the Tensile Ductility of Bulk Nanostructured Metals and Alloys. *JOM*, 2006, vol. 58, no. 4, pp. 49–53. DOI: [10.1007/s11837-006-0215-5](https://doi.org/10.1007/s11837-006-0215-5).
- Krymskiy S., Sitdikov O., Avtokratova E., Markushev M. 2024 aluminum alloy ultrahigh-strength sheet due to two-level nanostructuring under cryorolling and heat treatment. *Transactions of Nonferrous Metals Society of China*, 2020, vol. 30, no. 1, pp. 14–26. DOI: [10.1016/S1003-6326\(19\)65176-9](https://doi.org/10.1016/S1003-6326(19)65176-9).
- Sheng Yinying, Hua Youlu, Wang Xiaojian, Zhao Xueyang, Chen Lianxi, Zhou Hanyu, Wang James, Berndt Ch.C., Li Wei. Application of High-Density Electropulsing to Improve the Performance of Metallic Materials: Mechanisms, Microstructure and Properties. *Materials*, 2018, vol. 11, no. 2, article number 185. DOI: [10.3390/ma11020185](https://doi.org/10.3390/ma11020185).
- Kang Kaijiao, Li Dayong, Wang Ao, Shi Dequan, Gao Guili, Xu Zhenyu. Experimental investigation on aging treatment of 7050 alloy assisted by electric pulse. *Results in Physics*, 2020, vol. 3, article number 103016. DOI: [10.1016/j.rinp.2020.103016](https://doi.org/10.1016/j.rinp.2020.103016).
- Xua Hong, Liu Meng, Wang Yu-peng, Ma Pin-kui, Bai Ming, Jiang Bo, Guo Zhi-peng, Zou Yu-jie. Refined microstructure and dispersed precipitates in a gradient rolled AZ91 alloy under pulsed current. *Materialia*, 2021, vol. 20, article number 101245. DOI: [10.1016/j.mtla.2021.101245](https://doi.org/10.1016/j.mtla.2021.101245).
- Xu Zhutian, Jiang Tianhao, Huang Jihui, Peng Linfa, Lai Xinmin, Fu M.W. Electroplasticity in electrically-assisted forming: Process phenomena, performances and modeling. *International Journal of Machine Tools and Manufacture*, 2022, vol. 175, article number 103871. DOI: [10.1016/j.ijmachtools.2022.103871](https://doi.org/10.1016/j.ijmachtools.2022.103871).
- Kim Moon-Jo, Yoon Sangmoon, Park S. et al. Elucidating the origin of electroplasticity in metallic materials. *Applied Materials Today*, 2020, vol. 21, article number 100874. DOI: [10.1016/j.apmt.2020.100874](https://doi.org/10.1016/j.apmt.2020.100874).
- Ruszkiewicz B.J., Mears L., Roth J.T. Investigation of Heterogeneous Joule Heating as the Explanation for the Transient Electroplastic Stress Drop in Pulsed Tension of 7075-T6 Aluminum. *Journal of Manufacturing Science and Engineering*, 2018, vol. 140, no. 9, article number 091014. DOI: [10.1115/1.4040349](https://doi.org/10.1115/1.4040349).
- Humphreys F.J., Hatherly M. *Recrystallization and Related Annealing Phenomena*. 2nd ed. Amsterdam, Elsevier Publ., 2004. 658 p.

СПИСОК ЛИТЕРАТУРЫ

- Estrin Y., Vinogradov A. Extreme grain refinement by severe plastic deformation: A wealth of challenging science // *Acta Materialia*. 2013. Vol. 61. № 3. P. 782–817. DOI: [10.1016/j.actamat.2012.10.038](https://doi.org/10.1016/j.actamat.2012.10.038).
- Zhilyaev A.P., Pshenichnyuk A.I., Utyashev F.Z., Raab G.I. *Superplasticity and Grain Boundaries in Ultrafine-Grained Materials*. Amsterdam: Elsevier, 2020. 416 p.
- Edalati K., Bachmaier A., Beloshenko V.A., Beygelzimer Y., Blank V.D., Botta W.J. Nanomaterials by severe plastic deformation: review of historical developments and recent advances // *Materials Research Letters*. 2022. Vol. 10. № 4. P. 163–256. DOI: [10.1080/21663831.2022.2029779](https://doi.org/10.1080/21663831.2022.2029779).
- Pan Dong, Zhao Yuguang, Xu Xiaofeng, Wang Yitong, Jiang Wenqiang, Ju Hong. Effect of High-Energy and

- Instantaneous Electropulsing Treatment on Microstructure and Properties of 42CrMo Steel // *Acta Metall Sin.* 2018. Vol. 54. № 9. P. 1245–1252. DOI: [10.11900/0412.1961.2017.00562](https://doi.org/10.11900/0412.1961.2017.00562).
5. Konkova T., Valeev I., Mironov S., Korznikov A., Myshlyaev M.M., Semiatin S.L. Effect of electric-current pulses on grain-structure evolution in cryogenically rolled copper // *Journal of Materials Research.* 2014. Vol. 29. № 22. P. 2727–2737. DOI: [10.1557/jmr.2014.299](https://doi.org/10.1557/jmr.2014.299).
 6. Konkova T., Valeev I., Mironov S., Korznikov A., Korznikova G., Myshlyaev M.M., Semiatin S.L. Microstructure response of cryogenically-rolled Cu–30Zn brass to electric-current pulsing // *Journal of Alloys and Compounds.* 2016. Vol. 659. P. 184–192. DOI: [10.1016/j.jallcom.2015.11.059](https://doi.org/10.1016/j.jallcom.2015.11.059).
 7. Хаймович П.А. Криодеформирование металлов в условиях всестороннего сжатия (обзор) // *Физика низких температур.* 2018. Т. 44. № 5. С. 463–490. EDN: [YTJSLG](https://www.edn.ru/ytjslug/).
 8. Panigrahi S.K., Jayaganthan R. A Study on the Combined Treatment of Cryorolling, Short-Annealing, and Aging for the Development of Ultrafine-Grained Al 6063 Alloy with Enhanced Strength and Ductility // *Metallurgical and Materials Transactions A.* 2010. Vol. 41. P. 2675–2690. DOI: [10.1007/s11661-010-0328-x](https://doi.org/10.1007/s11661-010-0328-x).
 9. Magalhães D.C.C., Kliauga A.M., Ferrante M., Sordi V.L. Plastic deformation of FCC alloys at cryogenic temperature: the effect of stacking-fault energy on microstructure and tensile behaviour // *Journal of Materials Science.* 2017. Vol. 52. P. 7466–7478. DOI: [10.1007/s10853-017-0979-8](https://doi.org/10.1007/s10853-017-0979-8).
 10. Ma E. Eight Routes to Improve the Tensile Ductility of Bulk Nanostructured Metals and Alloys // *JOM.* 2006. Vol. 58. № 4. P. 49–53. DOI: [10.1007/s11837-006-0215-5](https://doi.org/10.1007/s11837-006-0215-5).
 11. Krymskiy S., Sitdikov O., Avtokratova E., Markushev M. 2024 aluminum alloy ultrahigh-strength sheet due to two-level nanostructuring under cryorolling and heat treatment // *Transactions of Nonferrous Metals Society of China.* 2020. Vol. 30. № 1. P. 14–26. DOI: [10.1016/S1003-6326\(19\)65176-9](https://doi.org/10.1016/S1003-6326(19)65176-9).
 12. Sheng Yinying, Hua Youlu, Wang Xiaojian, Zhao Xueyang, Chen Lianxi, Zhou Hanyu, Wang James, Berndt Ch.C., Li Wei. Application of High-Density Electropulsing to Improve the Performance of Metallic Materials: Mechanisms, Microstructure and Properties // *Materials.* 2018. Vol. 11. № 2. Article number 185. DOI: [10.3390/ma11020185](https://doi.org/10.3390/ma11020185).
 13. Kang Kaijiao, Li Dayong, Wang Ao, Shi Dequan, Gao Guili, Xu Zhenyu. Experimental investigation on aging treatment of 7050 alloy assisted by electric pulse // *Results in Physics.* 2020. Vol. 3. Article number 103016. DOI: [10.1016/j.rinp.2020.103016](https://doi.org/10.1016/j.rinp.2020.103016).
 14. Xua Hong, Liu Meng, Wang Yu-peng, Ma Pin-kui, Bai Ming, Jiang Bo, Guo Zhi-peng, Zou Yu-jie. Refined microstructure and dispersed precipitates in a gradient rolled AZ91 alloy under pulsed current // *Materialia.* 2021. Vol. 20. Article number 101245. DOI: [10.1016/j.mtla.2021.101245](https://doi.org/10.1016/j.mtla.2021.101245).
 15. Xu Zhutian, Jiang Tianhao, Huang Jihui, Peng Linfa, Lai Xinmin, Fu M.W. Electroplasticity in electrically-assisted forming: Process phenomena, performances and modelling // *International Journal of Machine Tools and Manufacture.* 2022. Vol. 175. Article number 103871. DOI: [10.1016/j.ijmachtools.2022.103871](https://doi.org/10.1016/j.ijmachtools.2022.103871).
 16. Kim Moon-Jo, Yoon Sangmoon, Park S. et al. Elucidating the origin of electroplasticity in metallic materials // *Applied Materials Today.* 2020. Vol. 21. Article number 100874. DOI: [10.1016/j.apmt.2020.100874](https://doi.org/10.1016/j.apmt.2020.100874).
 17. Ruszkiewicz B.J., Mears L., Roth J.T. Investigation of Heterogeneous Joule Heating as the Explanation for the Transient Electroplastic Stress Drop in Pulsed Tension of 7075-T6 Aluminum // *Journal of Manufacturing Science and Engineering.* 2018. Vol. 140. № 9. Article number 091014. DOI: [10.1115/1.4040349](https://doi.org/10.1115/1.4040349).
 18. Humphreys F.J., Hatherly M. Recrystallization and Related Annealing Phenomena. 2nd ed. Amsterdam: Elsevier, 2004. 658 p.
 19. Conrad H. Electroplasticity in metals and ceramics // *Materials Science and Engineering: A.* 2000. Vol. 287. № 2. P. 276–287. DOI: [10.1016/S0921-5093\(00\)00786-3](https://doi.org/10.1016/S0921-5093(00)00786-3).
 20. Grimm T.J., Mears L.M. Skin effects in electrically assisted manufacturing // *Manufacturing Letters.* 2022. Vol. 34. P. 67–70. DOI: [10.1016/j.mfglet.2022.09.006](https://doi.org/10.1016/j.mfglet.2022.09.006).
 21. He Changshu, Zhang Yudong, Wang Y.N., Zhao Xingyong, Zuo Liang, Esling C. Texture and microstructure development in cold-rolled interstitial free (IF) steel sheet during electric field annealing // *Scripta Materialia.* 2003. Vol. 48. № 6. P. 737–742. DOI: [10.1016/S1359-6462\(02\)00552-3](https://doi.org/10.1016/S1359-6462(02)00552-3).
 22. Shneerson G.A., Dolotenko M.I., Krivosheev S.I. Strong and Superstrong Pulsed Magnetic Fields Generation. Berlin: De Gruyter, 2014. 439 p. DOI: [10.1515/9783110252576](https://doi.org/10.1515/9783110252576).
 23. Валеев И.Ш., Валеева А.Х., Ильясов Р.Р., Автokratova E.B., Крымский С.В., Ситдиков О.Ш., Маркушев М.В. Влияние электроимпульсной обработки на структуру и твердость криопрокатаного алюминия // *Письма о материалах.* 2021. Т. 11. № 3. С. 351–356. DOI: [10.22226/2410-3535-2021-3-351-356](https://doi.org/10.22226/2410-3535-2021-3-351-356).
 24. Маркушев М.В., Ильясов Р.Р., Крымский С.В., Валеев И.Ш., Ситдиков О.Ш. Структура и прочность мелкозернистой меди после криопрокатки и однократной электроимпульсной обработки различной мощности // *Письма о материалах.* 2021. Т. 11. № 4. С. 491–496. DOI: [10.22226/2410-3535-2021-4-491-496](https://doi.org/10.22226/2410-3535-2021-4-491-496).
 25. Markushev M., Valeev I., Valeeva A., Ilyasov R., Avtokratova E., Krymskiy S., Sitdikov O. Effect of electric pulsing on the structure, texture and hardness of cryorolled fine-grain copper // *Facta Universitatis. Series: Mechanical Engineering.* 2022. P. 1–12.

26. Markushev M.V., Valeev I.Sh., Avtokratova E.V., Ilyasov R.R., Valeeva A.K., Krimsky S.V., Sitdikov O.S. Effect of high-dense electropulsing with different energies on the structure and strength of nickel cryorolled to different strains // Letters on Materials. 2023. Vol. 13. № 2. P. 126–131. DOI: [10.22226/2410-3535-2023-2-126-131](https://doi.org/10.22226/2410-3535-2023-2-126-131).
27. Danyuk A., Merson D., Yasnikov I., Agletdinov E., Afanasyev M., Vinogradov A. The effect of stacking fault energy on acoustic emission in pure metals with face-centered crystal lattice // Letters on Materials. 2017. Vol. 7. № 4. P. 437–441. DOI: [10.22226/2410-3535-2017-4-437-441](https://doi.org/10.22226/2410-3535-2017-4-437-441).
28. Sarma V.S., Wang Jun, Jian W.W., Kauffmann A., Conrad H., Freudenberger J., Zhu Yuntian T. Role of stacking fault energy in strengthening due to cryo-deformation of FCC metals // Materials Science and Engineering: A. 2010. Vol. 527. № 29–30. P. 7624–7630. DOI: [10.1016/j.msea.2010.08.015](https://doi.org/10.1016/j.msea.2010.08.015).
29. Zhao Yonghao, Liao X.Z., Zhu Yuntian, Horita Z., Langdon T.G. Influence of stacking fault energy on nanostructure under high pressure torsion // Materials Science and Engineering: A. 2005. Vol. 410–411. P. 188–193. DOI: [10.1016/j.msea.2005.08.074](https://doi.org/10.1016/j.msea.2005.08.074).
30. Belyakov A., Sakai T., Miura H., Kaibyshev R., Tszuzaki K. Continuous recrystallization in austenitic stainless steel after large strain deformation // Acta Materialia. 2002. Vol. 50. № 6. P. 1547–1557. DOI: [10.1016/S1359-6454\(02\)00013-7](https://doi.org/10.1016/S1359-6454(02)00013-7).

Сочетание криогенной деформации и электроимпульсной обработки как способ получения ультрамелкозернистых металлов

© 2023

Маркушев Михаил Вячеславович*, доктор технических наук, старший научный сотрудник, заведующий лабораторией

Автократова Елена Викторовна, кандидат технических наук, старший научный сотрудник

Валеева Айгуль Хамматовна¹, кандидат технических наук, научный сотрудник

Валеев Иршат Шамилович², кандидат технических наук, научный сотрудник

Ильясов Рафис Раисович³, младший научный сотрудник

Крымский Станислав Вацлавович⁴, кандидат технических наук, заведующий лабораторией

Ситдииков Олег Шамилович, кандидат физико-математических наук, старший научный сотрудник

Институт проблем сверхпластичности металлов РАН, Уфа (Россия)

*E-mail: mvmark@imsp.ru

¹ORCID: <https://orcid.org/0000-0003-4305-4538>

²ORCID: <https://orcid.org/0009-0002-5162-7324>

³ORCID: <https://orcid.org/0000-0003-0195-1206>

⁴ORCID: <https://orcid.org/0000-0002-1534-3239>

Поступила в редакцию 15.08.2023

Принята к публикации 01.12.2023

Аннотация: Проведен сравнительный анализ структуры и твердости чистых металлов с гранцентрированной кубической решеткой – алюминия, никеля и меди, подвергнутых комплексной термомеханической обработке (ТМО), включавшей изотермическую криогенную прокатку при температуре жидкого азота и последующую электроимпульсную обработку (ЭИО) токами высокой плотности. Рассмотрены основные этапы, особенности и преимущества ТМО, обеспечивающие сначала сильный наклеп обрабатываемого материала за счет деформации при отрицательных температурах, а затем его сверхбыстрый контактный электроимпульсный нагрев до заданной температуры. С использованием современных методов сканирующей электронной микроскопии и рентгеноструктурного анализа проведено многоуровневое исследование структуры металлов после основных этапов ТМО с фиксацией широкого спектра ее линейных и угловых параметров. Выявлены кинетика и природа процессов трансформации структуры металлов при криопротатке и ЭИО, их движущая сила и контролируемые факторы, а также общие закономерности и температурные интервалы активации возврата и рекристаллизации деформационной структуры под воздействием электроимпульса. На основе результатов анализа структурно-механического поведения металлов сделан вывод о том, что сочетание большой пластической криогенной деформации с последующей однократной обработкой ультракороткими импульсами переменного тока является эффективным способом получения полуфабрикатов с регламентированными параметрами их структуры и свойств, в т. ч. высокопрочного ультрамелкозернистого проката. При этом феноменология и природа упрочнения/разупрочнения металлов при криогенной прокатке и последующей обработке импульсами тока аналогичны наблюдающимся при холодной прокатке и печном отжиге.

Ключевые слова: ГЦК-металлы; криогенная деформация; криогенная прокатка; электроимпульсная обработка; ультрамелкозернистая структура.

Благодарности: Работа выполнена в рамках государственного задания федерального государственного бюджетного учреждения науки «Институт проблем сверхпластичности металлов Российской академии наук».

Экспериментальные исследования были выполнены на базе Центра коллективного пользования ИПСМ РАН «Структурные и физико-механические исследования материалов».

Для цитирования: Маркушев М.В., Автократова Е.В., Валеева А.Х., Валеев И.Ш., Ильясов Р.Р., Крымский С.В., Ситдиков О.Ш. Сочетание криогенной деформации и электроимпульсной обработки как способ получения ультрамелкозернистых металлов // Frontier Materials & Technologies. 2023. № 4. С. 53–62. DOI: 10.18323/2782-4039-2023-4-66-5.