Effect of electric pulse treatment on the structure and hardness of nickel deformed at room and liquid nitrogen temperatures

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Abstract. The influence of the energy of electro-pulse processing (EPP) on the structure and hardness of pure nickel deformed at room and liquid nitrogen temperatures is studied. The metal was subjected to isothermal rolling with a strain of 90% and subsequent EPP with integral current densities $K_j$ in the range from $0.06 \times 10^5$ A$^2$/mm$^4$ corresponding to the calculated temperature range of 130-925°C. It was found that decreasing the rolling temperature of Ni resulted in a more uniform and less misoriented nanocellular structure and a 15-20 HV higher hardness. Subsequent EPP decreased the hardness due to recovery, discontinuous recrystallization with the formation of annealed twins and growth of new grains. It is revealed that a more uniform fine-grained structure with a smaller grain size is formed in the cryorolled metal in a narrower interval and at less EPP energies.

1. Introduction

One of the promising but poorly studied ways of processing the ultrafine-grained (UFG) structure in metals and alloys is the so-called cryogenic deformation, which is carried out at temperatures below 120 K (-153°C) [1-8]. It is considered that low deformation temperatures, suppressing dynamic recovery, promote the accumulation of internal stresses due to the extremely high densities of crystal lattice defects and thus intensify grain refinement under straining and subsequent annealing. However, there are a number of investigations that have shown that deformation even to high strains (with $e > 1$) at cryogenic temperatures does not guarantee the UFG structure processing. For example, severe plastic deformation (SPD) of pure aluminum via cryorolling did not result in the formation of the expected nanocrystalline structure and even did not ensure significant grain refinement [1,7]. The studies of copper have also shown that a decrease in the deformation temperature, suppressing double-cross slip of lattice dislocations, complicates the formation of strain-induced boundaries and thus restricts grain fragmentation (grain subdivision) and the formation of new fine grains via dynamic recrystallization. Nevertheless, in [9-11] it was found that a complex thermomechanical treatment involving cryorolling and subsequent electro-pulse processing (EPP) ensured the formation of the UFG structure. Therewith, the EPP played a dominating role causing processes of static recovery and recrystallization under thermal activation in a highly work-hardened material, allowing extreme grain refinement due to the superf ast heating rate and very short time of annealing.

The purpose of the present investigation was to compare the effect of EPP with different energies on the structuring and softening of pure nickel, initially deformed to equal strains at a temperature of liquid nitrogen and ambient temperature.
2. Material and procedure

Pure nickel (designated as NP2 in accordance to the Russian state standard GOST 492-73) was used as the material for the study. The ingots were preliminary subjected to setting at 850°C followed by annealing at 600°C for 2 h. Subsequent multi-pass rolling of mechanically cut out sheet billets was carried out to a total strain of about 90% at 20 and −196°C. Rolling was performed in isothermal conditions using a laboratory six-roll mill with preliminary cooling of removable working rolls and billets in a bathtub with cold water/liquid nitrogen within 60 minutes. The rolling speed did not exceed 100 mm/min with straining per pass of about 5%.

EPP was carried out on flat samples having the dog-bone shape (as samples for tensile tests) with a thickness of 0.4 mm and sizes of the gauge part of 3×4 mm². The samples were cut out along the rolling direction and fixed in massive clamping jaws with current connectors. The temperature of heating of the gauge part was calculated using the relationship between the temperature and current density:

\[
S(T_C) = \int_{T_1}^{T_e} \rho \sigma_{e} dT, \quad K_j = \int_{0}^{\tau} j^2 dt S(T_C) = K_j,
\]

where \(T_C\) is the calculated temperature, \(T_1\) is the reference temperature of the sample, \(\tau\) is the duration of the electric impulse. For the chosen \(K_j\) current parameters (from 0.06×10⁵ to 0.19×10⁵ A²s/mm⁴), the temperature interval of heating the samples was within 130 and 925 °C. With an impulse time of about 10⁻⁴ s and a current frequency of 10⁴ Hz, the depth of its penetration to the sample of nickel was about 0.2 mm [3]. This means that the skin-effect in the samples used could be neglected suggesting a uniform distribution of current on their section.

The microstructure was analyzed in the rolling plane with the use of a "TESCAN MIRA 3 LMH" scanning electron microscope equipped with "HKL Channel 5" software. EBSD maps were obtained using the data from 6 Kikuchi-line scanning with a step of 0.03 and 0.5 µm. The intercrystalline boundary misorientation of 15° was taken as a criterion for dividing into low- and high-angle boundaries (LABs and HABs). The mean size of subgrains and grains (d and D, consequently) were determined by the method of equivalent diameter on the base of at least 300 measurements of crystallites. The average angle of intercrystalline boundary misorientations (θ) and fractions of HABs and twin boundaries (\(F_{HAB}\) and \(F_{\Sigma3+\Sigma9}\), consequently) were derived from grain boundary spectra, in which boundaries of less than 1° were not taken into consideration. The size and fraction of recrystallized grains (\(D_R\) and \(F_R\), consequently) were also measured using EBSD data and the mentioned software.

Microhardness was determined by the Vickers method with an MVDM 8 "AFFRY" tester at 0.5 N and a loading time of 10 s. To obtain an error not exceeding 5%, more than ten measurements at the point were performed.

3. Results and discussions

The obtained EBSD maps and grain boundary spectra (figure 1) allow one to conclude that the structuring of nickel under room temperature- and cryorolling conditions has a quite similar nature in many respects since it resulted in the formation of the same type of highly work-hardened, partially recrystallized structure. Indeed, in both cases the fragmentation of the initial grains and dynamic recrystallization with the formation of new HABs and fine grains occurred quite inerly and non-homogeneously, especially under cryostraining. Moreover, it could be concluded that the main mechanism of structure transformations under both rolling temperatures was the formation of so called low-energy dislocation structures, which led to the evolution of a predominant and well-developed substructure with nanoscale crystallites of various shapes and sizes divided mostly by LABs. Thus, both deformation structures could be defined, in general, as cellular-type ones, meanwhile in cryorolled condition it was more disperse and less misoriented. As a result, the fractions of HABs and average boundary misorientations were recorded to be a bit higher in the cold-rolled metal (table 1). It should be also noted that the deformed structure is characterized by the absence of deformation twins, which are frequently observed in conventionally cold processed nickel [12].
Judging by the data in figure 2, the higher suppression of dynamic recovery and recrystallization led to a more defected and developed deformation structure, being the main reason for the increased hardness of cryorolled nickel.

From figure 2 also follows that in the low EPP energy range (up to $K_j = 0.130 \times 10^5 \text{ A}^2/\text{mm}^4$), the cold-rolled metal continuously softened to some extent. EBSD analysis showed that such behavior was caused by the activation of static recovery and the local occurrence of recrystallization in the work-hardened structure (figure 3, table 1). With a further increase in the EPP energy, the hardness of nickel decreased more extensively and at $K_j = 0.170 \times 10^5 \text{ A}^2/\text{mm}^4$, the effect of deformation hardening was completely lost (figure 2). Judging by the maps in figure 3, at such EPP energy conditions the processes of discontinuous static recrystallization were accompanied by abnormal growth of separate new fine grains. As a result, at $K_j = 0.152 \times 10^5 \text{ A}^2/\text{mm}^4$, a partially recrystallized structure with an average size of new grains of about 3 μm was initially formed inside the coarse-grained and highly strained structure.

![Figure 1. SEM-EBSD grain boundary maps for Ni after rolling at 20 (a,c) and -196°C (b,d) to a total strain of 90% and corresponding LABs spectrums.](image)

(Hereafter, HABs and LABs are indicated by black and red lines, respectively)
Table 1. Parameters of Ni microstructure after rolling and EPP with different energies

| Rolling temperature, °C | \( K_j \times 10^5 \) A²s/mm⁴ | d, μm | \( D_R \), μm | \( F_{R*} \), % | \( \Theta \), degree | \( F_{HAB*} \), % | \( F_{\Sigma 3+\Sigma 9} \), % |
|------------------------|---------------------------------|-------|---------------|----------------|----------------|----------------|-----------------|
| 20                     | -                               | 0.08  | 2             | 1              | 6              | 9              | 0               |
|                        | 0.152                           | 0.82  | 3             | 26             | 9              | 14             | 4               |
|                        | 0.170                           | 2.10  | 8             | 79             | 39             | 77             | 27              |
| -196                   | -                               | 0.07  | 2             | 1              | 5              | 4              | 0               |
|                        | 0.106                           | 0.84  | 2             | 1              | 5              | 4              | 0               |
|                        | 0.112                           | 1.95  | 3             | 90             | 44             | 84             | 39              |

**Figure 2.** Evolution of microhardness vs the EPP energy for Ni rolled at 20 (○) and -196 °C (●).

**Figure 3.** SEM-EBSD grain boundary maps for Ni rolled at 20 (a, b) and -196 °C (c) and further subjected to EPP with \( K_j = 0.152 \) (a), 0.170 (b) and 0.112×10⁵ A²s/mm⁴ (c).

(For better observation LABs of less 2° misorientations are not plotted)

Therewith, these new grains were mainly settled down in the form of chains (bands) with the interspacing close to the initial grain size, demonstrating that sites of their primary nucleation were located in the regions adjacent to the initial boundaries. With increasing the energy of EPP to 0.170×10⁵ A²s/mm⁴, recrystallization occupied practically the whole specimen volume and the HABs fraction, the fraction of recrystallized grains and the mean misorientation angle of intercrystallite boundaries reached their maximum values (table 1). However, the intense growth of new grains leading to their noticeable coarsening (figure 3b) was simultaneously observed. It should be noted that the presence of annealed twins, the fraction of which exceeded a quarter (table 1), was a character for
the structure processed, confirming the active role of twinning in the transformation of the deformed structure of nickel at EPP.

Unlike cold-rolled nickel, EPP of cryorolled one, performed with small energies (up to $0.106 \times 10^5 \text{ A}^2/\text{mm}^4$ only), did not cause any significant changes in its hardness (figure 2). It revealed a higher stability of the deformation structure, processed by rolling at a lower temperature. Virtually the structure underwent recovery in the form of insignificant transformation (perfection) of dislocation configurations only and even remained highly work-hardened. However, when the current density exceeds $K_j = 0.112 \times 10^5 \text{ A}^2/\text{mm}^4$, the stored energy in the cryodeformed nickel rapidly relaxed in a narrow interval of energies (temperatures) of EPP, which was expressed in an intense loss of its strength owing to the formation of an almost completely recrystallized fine-grained structure with a mean grain size about 3 µm (figure 3c). A feature of this recrystallized structure, as well as in the EPP-ed room-temperature rolled Ni, was a quite high fraction of LABs (table 1). Besides, the fraction of twin boundaries increased to a value of about 40%, testifying the even more significant role of twinning and grain growth at EPP of the cryorolled metal. With further increasing the EPP energy, the microhardness of cryorolled nickel decreased to the level of the starting material due to the loss of the Hall-Petch hardening effect because of the coarsening of recrystallized grains.

4. Conclusions
It is established that a decrease in the temperature of rolling of nickel from room temperature to cryogenic leads to the formation of a more uniform and less misoriented nanocellular structure and to an additional increase in its hardness by 15-20 HV. Subsequent impact on the work-hardened metal by powerful impulses of current at EPP resulted in a decrease in its hardness owing to recovery, discontinuous recrystallization with the formation of annealed twins and the growth of new grains. It is revealed that EPP of nickel deformed at a temperature of liquid nitrogen leads to the formation of a completely recrystallized structure at lower energies than that of nickel deformed at room temperature. At the same time, this process proceeds in a narrower interval of current densities of EPP and forms a more uniform fine-grained structure with a smaller grain size.

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