

# Physics of Equilibrium and Non-equilibrium Deformation Processes in the Nickel Surface Layer

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**Abstract**— *Based on the Hamilton's principle, invariants are proposed to describe the processes of formation and evolution of the structure of the metal interface under friction. These invariants can be used, among other things, in the creation, evolution and destruction of nanomaterials.*

**Keywords**— *nano, submicro, micro, meso and macro levels of deformation, equilibrium and non-equilibrium deformation, invariants.*

## I. INTRODUCTION

The mechanisms of hardening and destruction of the surface layer of metals, as well as the conditions for its self-organization under equilibrium deformation are well studied [1, 2], which cannot be said about the self-organization of processes under non-equilibrium deformation under conditions of phase instability of the crystal lattice [3]. In accordance with nonlinear mechanics and mesomechanics, plastic flow in a loaded solid is a multi-level process and is associated with the loss of shear stability at the nano, micro, meso, and macro-scale levels [4]. The intensity of external influence determines the structure, properties and mechanisms of destruction of surface layers of metals. The physics of processes occurring on the metal surface under multi-cycle, low-amplitude and alternating tribo-loading is described in [5-8]. The mechanisms of formation of elements of the defective structure for each of the structural levels of deformation (nano, micro, meso and macro) of the surface layer of Nickel under multicycle low-amplitude and alternating deformation, where the high density of dislocations and the local orientation gradient of structural elements in the Nickel crystal lattice plays a fundamental role at each of the scale levels of deformation [3]. Using various model representations, namely, extinction contours, local curvature of the Nickel crystal lattice, and dislocation contributions to the hardening mechanisms, a quantitative assessment of the values of internal stresses and parameters of the defective structure of the Nickel surface layer was carried out under conditions of its phase instability, where the value of internal stresses is comparable and exceeds the value of the elastic modulus of Nickel  $\approx 2 \cdot 10^{11}$  Pa. The literature sources contain limited data on such highly dispersed materials and their physical and chemical properties [4, 5]. It is generally accepted that the formation of nanoobjects occurs under high-energy influence or intense plastic deformation and / or equal-channel angular compression in the top-down direction, when the material is fragmented from macro, meso, micro to nanoscale as a result of external influence at contact pressures in the GPa [5]. Data on the formation of nanostructures at contact pressures of  $\approx 0.1 \div 0.2$  kPa in the presence of chemically active substances under conditions of non-equilibrium deformation are extremely limited.

The question of determining the mechanisms and dominant factors that determine the physical, chemical and mechanical properties (amorphous, superplasticity, catalytic activity, etc.) of the metal interface under non-equilibrium deformation remains open. Determination of the basic laws in the field of equilibrium and nonequilibrium deformation is an urgent problem in the creation of nanomaterials with unique properties.

The aim of this work is to study the mechanisms of plastic deformation at various structural-scale levels under conditions of phase instability of the surface layer of nickel under tribo-loading and to determine the basic laws describing the kinetics of hardening and destruction of the surface layer of metals.

This work presents the results of electron microscopical investigation of surface layer microstructures resulted from sliding friction according to dislocation structure study using the method of ferromagnetic resonance (FMR). It analyzes the micro structural destruction mechanism of nickel on the basis of complex data.

## II. EXPERIMENTAL EQUIPMENT AND SAMPLES

The dislocation structure change in the nickel near the surface layer under friction loading was studied by the methods of electron microscopy and ferromagnetic resonance. Polycrystalline nickel of 99.99 % purity was investigated. The samples in

the form of thin disks 5 by 0.1 mm were electrically polished and annealed in a vacuum of 0.133 mPa at 973 K. Friction tests were carried out on an AE-5 type machine with precision positioning of the contact surface. Sliding friction was carried out in the pair Ni-Mo in air, using CIATIM-201 lubricant under a load of 82.3 kPa at a linear speed of 0.5 m·s<sup>-1</sup>. The average volume temperature of the sample did not exceed 40° C. The correct use of load-speed parameters or the correct use of the scale factor, along with the high sensitivity of the ferromagnetic resonance (FMR) method to structural changes in a thin surface layer with a thickness of ≈ 0.1 μm, made it possible to see oscillating kinetics of changes in the dislocation density and wear intensity in the analysis of experimental data, which allowed to correctly interpret the results obtained, which are in good agreement with the results of work [1]. The electron microscopic investigation of nickel was carried out under a EMV-100 AK and Hitachi-H800 microscope by the thin foil method at a "transillumination". The resolution of the Hitachi H-800 is ≈ 0.1 nm. The foil was produced by thinning down the disks electrolytically from the side opposite to the friction surface using a jet blower polishing unit equipped with a sensitive photodiode bridge that allows controlling the segment's transparency at 0.1 μm depth from the friction surface. The technique for preparing nickel samples for transmission electron microscopy is given in [2].

### III. INVESTIGATION RESULTS AND THEIR DISCUSSION

An analysis of the experimental data given in [3, 5–12] allowed the author, on the basis of Hamilton's principle or principle of least action [13], to establish two main fundamental regularities and to propose a third one, formulated in [3, 14].

This is the first pattern. An asymmetric kinetic relationship between the density of dislocations and the intensity of destruction or wear has been determined, namely, an increase in the density of dislocations to a certain critical value causes hardening of the surface layer. The latter determines the decrease in the intensity of destruction. An inversely proportional relationship has been established between the local gradient of the orientation of the boundaries of the structural elements of the crystal lattice or the density of dislocations at each of the structural-scale levels of deformation and the intensity of destruction of the surface layer of nickel. Thus, the dependence of dislocation density on time is unipolar to the kinetics of wear intensity [5], and the expression is fulfilled:

$$\rho \cdot I = const \quad (1)$$

where  $\rho$  is the dislocation density and  $I$  is the wear intensity.

This is the second pattern. The rate of increase ( $K_1$ ) and decrease ( $K_2$ ) in dislocation density at tribo-loading is determined by the formed structure and disorientation of the internal interface boundaries. The rate of increase in the density of dislocations during hardening of the surface layer of nickel determines the rate of decrease in the density of dislocations and, accordingly, the relaxation of deformation stresses for one and each cycle of change in strength properties. The expression is executed [13]:

$$\frac{K_1}{K_2} = const \quad (2)$$

where  $K = \Delta H/t$ .

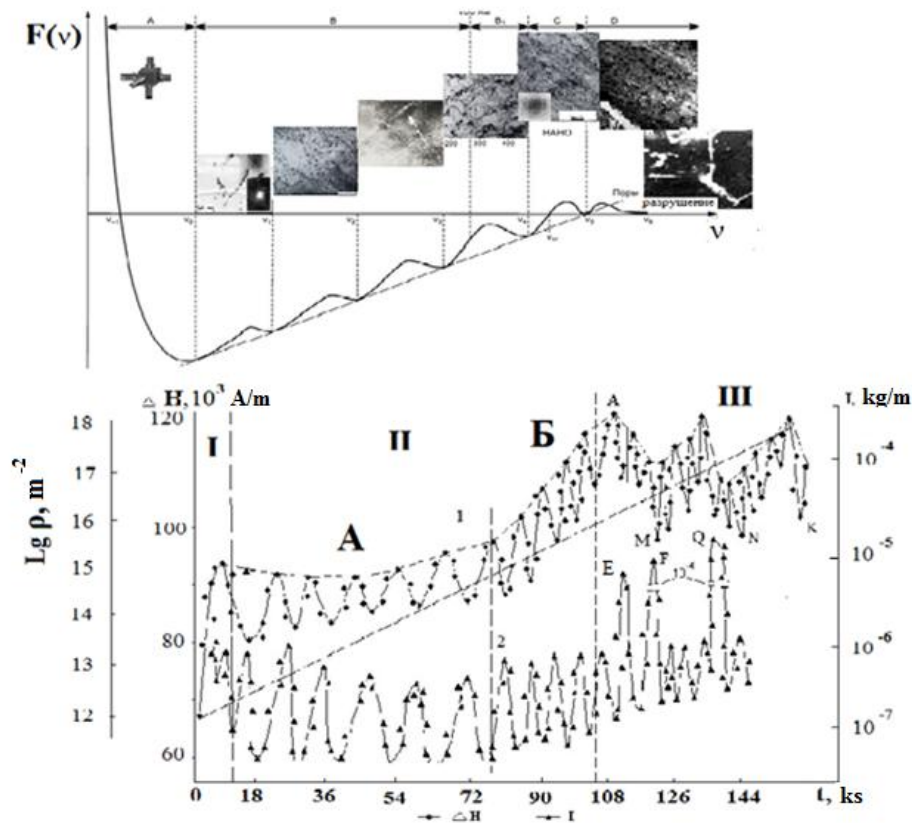
It should be noted that the constant in expression (2) is close to unity. However, with the accumulation of energy by subsurface layers over time, the fragile fracture mechanism can dominate, which leads to an insignificant increase in the relaxation rate of broadening of the ferromagnetic resonance line ( $K_2$ ).

This is the third pattern. The main condition for the compatibility of deformation at the scale-structural levels: nano, micro, meso and macro is the fulfillment of the law of conservation of angular momentum, namely, the sum of the angular momenta is zero, which ensures the fulfillment of the condition for grain boundary sliding of structural elements during plastic deformation. The law of conservation of angular momentum is fulfilled [3, 14]:

$$\sum_{i=1}^N RotJ_i = 0 \quad (3)$$

where  $J_i$  are streams of defects at the  $i$ -th structural-scale level.

We use methods of mathematical physics to describe the processes occurring in the surface layer of Nickel at tribo-loading. The dependence  $\Delta H(t)$  has a nonlinear wave character (fig. 1).



**FIGURE 1: Dependence of the kinetics of structural changes in the surface layer of nickel during friction on the Gibbs potential, broadening of the ferromagnetic resonance line ( $\Delta H$ ) and wear rate (I).**

From the point of view of methods of mathematical physics, if one of the independent variables describing the state of the surface layer of nickel [15] is taken as the broadening of the FMR line ( $\Delta H$ ), then the change in the state of the surface layer of nickel under tribo-loading can be described by partial differential equations. We use the wave equation to describe the physics of the processes of deformation and destruction of the nickel surface:

$$\frac{\partial^2 \Delta H}{\partial x^2} = \frac{1}{v^2} \frac{\partial^2 \Delta H}{\partial t^2} \tag{4}$$

Where  $v$  – the rate of change in the dislocation density,  $\Delta H$  – the broadening of the ferromagnetic resonance line.

It is known that the value of the FMR line broadening ( $\Delta H$ ) is directly proportional to the dislocation density ( $\rho$ ) [5]. We will analyze the wave equation using the experimental data shown in Fig. 1. Consider the region of equilibrium deformation (fig. 1, region II) and the region of non-equilibrium deformation (fig. 1, region III). It should be noted that the ratio of the broadening of the FMR line ( $\Delta H$ ) to time ( $t$ ), defined as the rate of change in the broadening of the FMR line (constant  $K$ ), is the first derivative ( $\frac{\partial \Delta H}{\partial t}$ ). The second derivative of the broadening of the FMR line with respect to time ( $\frac{\partial^2 \Delta H}{\partial t^2}$ ) is the rate of change ( $\frac{\Delta H}{\Delta t}$ ) of the envelope function  $\Delta H(t)$  in Fig. 1, which is shown by the dotted line. Let us analyze this envelope for regions II and III in Fig. 1 using the experimentally established boundary conditions.

Equilibrium strain region II consists of two parts: part A and part B (fig. 1). The envelope of the function  $\Delta H(t)$  for area A can be approximated by a straight line parallel to the axis of time in Fig. 1. Then the expression is executed:

$$\frac{\partial^2 \Delta H}{\partial t^2} = 0. \tag{5}$$

The product of two terms  $(1/v^2)$  and  $(\frac{\partial^2 H}{\partial x^2})$  in the wave equation (4) is equal to zero and, therefore, the expression is fulfilled:

$$\frac{\partial^2 \Delta H}{\partial x^2} = 0. \quad (6)$$

Fick's first law is:

$$J = -\frac{D \cdot dC}{dx} \quad (7)$$

where J is the diffusion flow of the dislocation density passing through a unit area per unit time, D is the diffusion coefficient, dC/dx is the concentration gradient of the dislocation density in the direction of diffusion. It follows from Fick's first law that the dislocation density flux through a unit area per unit time is a constant value. It is natural to assume that this flow is directed inward from the surface. Considering that the local gradient of the orientation of structural elements ( $\chi$ ) is directly proportional to the dislocation density, it follows that:

$$\frac{\partial \chi}{\partial x} = const \quad (8)$$

The local gradient of orientation of structural elements ( $\chi$ ) in the direction (OX) is a constant value, which decreases inversely with increasing distance from the surface. Then const must be defined as in the minus of the first degree, and then expression (9) is defined as:

$$\frac{\partial \chi}{\partial x} = const^{-1} \quad (9)$$

From the analysis of area A of region II in fig. 1, two conclusions follow: the flux of dislocation density through a unit area per unit time is a constant value directed inward from the surface; change in the local gradient of the orientation of structural elements in the direction from the surface is a constant value that decreases in accordance with an inversely proportional relationship with increasing distance from the surface.

Let's go to the analysis of area B of region II in fig. 1. Analysis of the wave equation (4) of area B in fig. 1 shows that the envelope of the rate of change  $\Delta H/\Delta t$  is a straight line directed at an angle of  $45^\circ$  to the time axis, since

$\frac{\partial^2 \Delta H}{\partial t^2} \approx \frac{120\kappa A/m - 95\kappa A/m}{25\kappa s} \approx 1A./m \cdot s$ . Then the expression is executed:

$$\frac{\partial^2 \Delta H}{\partial t^2} = \frac{1}{v^2} \frac{\partial^2 \Delta H}{\partial x^2} \approx 1A./m \cdot s \quad (10)$$

The product of two quantities is equal to one, when each of them is equal to one or minus one. Since the velocity of movement of the dislocation density in the physical sense cannot take a negative value, a system of equations is performed:

$$v = 1 \quad (11)$$

and

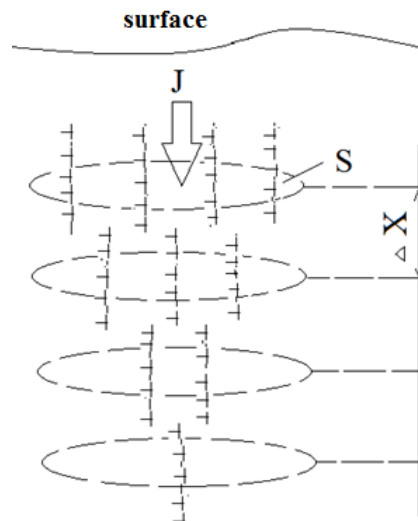
$$\frac{\partial^2 \Delta H}{\partial x^2} = 1 \quad (12)$$

The velocity of movement of the dislocation density deep from the nickel surface is a constant value, which follows from expression 11. From expression 12, taking into account that, where J is the flow of dislocation density through the unit area per unit time, the expression follows:

$$\frac{\partial J}{\partial x} = 1 \quad (13)$$

The gradient of dislocation flow deeper from the nickel surface is a constant value. In other words, the number of linear defects (dislocations) passing through a unit of sites built at some distance from each other deep from the surface, per unit of time is a constant value (fig. 2). Quantitative evaluation of expression (13) using shows that for each cycle of change in strength properties in section B of fig. 1, the flow of dislocation density ( $J$ ) through a unit area per unit time is  $\approx 0.25 \text{ A} / \text{m} \cdot \text{s}$ .

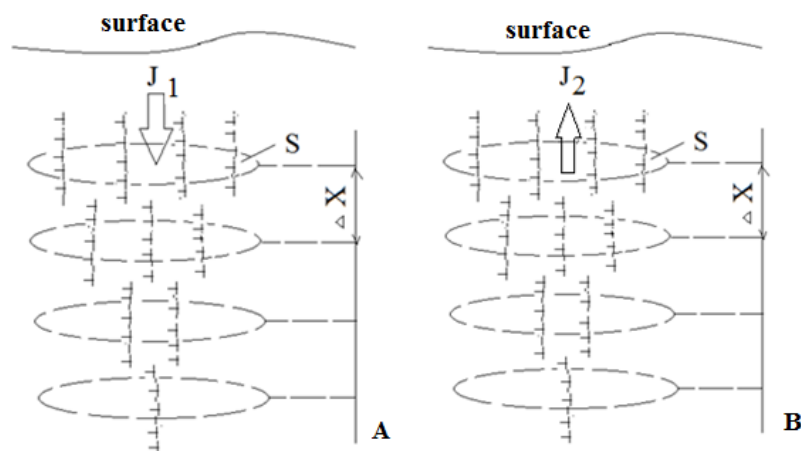
Taking into account the fact that the depth of the surface layer of nickel, in which the energy of friction loading is accumulated, is  $\approx 100 \mu\text{m}$ . Then the depth distribution of the flow (flow gradient) with a step of  $\Delta X = 25 \text{ microns}$  (fig. 2) through the unit area per unit time is  $\approx 0.06 \text{ A} / \text{m} \cdot \text{s}$ .



**FIGURE 2: Scheme showing a constant gradient of dislocation density flow with increasing distance or depth from the surface**

Two conclusions follow from the analysis of area B of region II in Fig. 1: the velocity of movement of the dislocation density deep from the surface is the constant  $\approx 1 ((\text{m} \cdot \text{s})^{-1})$ ; the flow of the dislocation density gradient through a unit of area per unit of time is a constant equal to  $\approx 1 (\text{m}^{-4})$  and is directed deep from the surface.

Analysis of the wave equation (4) in the field of non-equilibrium processes (fig. 1, region III) shows that the envelope of the rate of change  $\Delta H / \Delta t$  is an oscillating (periodically increasing and decreasing) dependence over time, that is, the gradient of the dislocation density flow ( $\frac{\partial J}{\partial X}$ ) periodically increases and decreases (fig. 3) and changes its sign, that is, changes its direction.



**FIGURE 3: Scheme showing a qualitative change in the direction of the flow gradient of the dislocation density with increasing and decreasing changes in the value of stresses inside the nickel crystal lattice with increasing distance or depth from the surface: A-increase in the value of internal stresses; B-decrease in the value of internal stresses.**

It should be noted that for area A and B of region II in Fig. 1, only an increase in the absolute value of the envelope was observed, and there was no low-frequency (LF) component (fig. 1, region III). The period of the LF component of the dislocation density flow gradient is 23÷25 ks. The rate of increase of the LF component

$$\left( \frac{\partial^2 \Delta H}{\partial t^2} \approx \frac{120\kappa A/m - 95\kappa A/m}{10\kappa s} \approx 2,5 A./m \cdot s \right) \text{ is designated as } K_1',$$

and the rate of decrease of the LF component

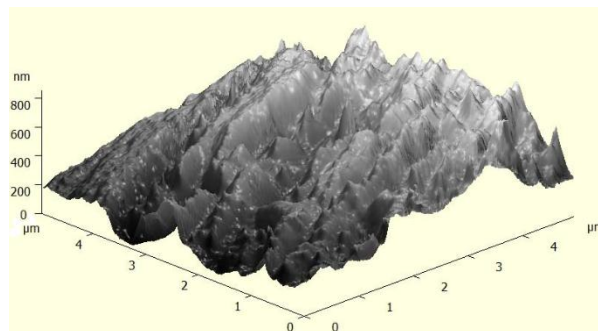
$$\left( \frac{\partial^2 \Delta H}{\partial t^2} \approx \frac{120\kappa A/m - 95\kappa A/m}{6\kappa s} \approx 4,2 A./m \cdot s \right) \text{ is designated as } K_2'. \text{ The ratio of increase } (K_1') \text{ to decrease } (K_2') \text{ of the LF component is } \approx 0.6.$$

The expression is being executed:

$$\frac{K_1'}{K_2'} = \frac{J_1}{J_2} = 0.6 \quad (14)$$

The gradient of the dislocation density flow ( $J_2$ ) with a decrease in the stress value exceeds the gradient of the dislocation density flow ( $J_1$ ) with an increase in the stress value by  $\approx 1.7$  times. Taking into account the fulfillment of the relation (3), where the sum of defect flows is zero, it follows that to fulfill the condition of plasticity and amorphous surface layers of Nickel within a single cycle of the LF component (not including point M, N and K in fig. 1), the dislocation density flow increases by  $\approx 1.7$  times when the stress value decreases.

The descending and ascending flows of the defect structure and the oscillating nature of the stresses determine the formation of a highly developed relief (fig. 4), the folds of which can be interpreted as a corrugated surface layer. A high density of dislocations in local zones, in the form of pronounced white dots, determines dynamic recrystallization with the formation of structures with a high modulus of elasticity.



**FIGURE 4: Three-dimensional image of the AFM image of the nickel surface with nanostructured formations**

At the minimum points (M, N and K) of the dependence of the FMR line broadening ( $\Delta H$ ) on time (t) (fig. 1, curve 1, region III), where an avalanche – like or selective destruction mechanism occurs localized in time with an increase in the wear intensity by two or three orders of magnitude (fig. 1, region III, curve 2, p. F, Q), there is a violation of the law of conservation of momentum, i. e. expression (3).

It implements the principle of least action, where selective destruction mechanism of the porous layer defines the removal of foci of discontinuities of the material (microcracks, pores, twins, defects, etc. ) that maintains the durability of the material from the positions of synergetics and prevents the penetration of cracks into the material.

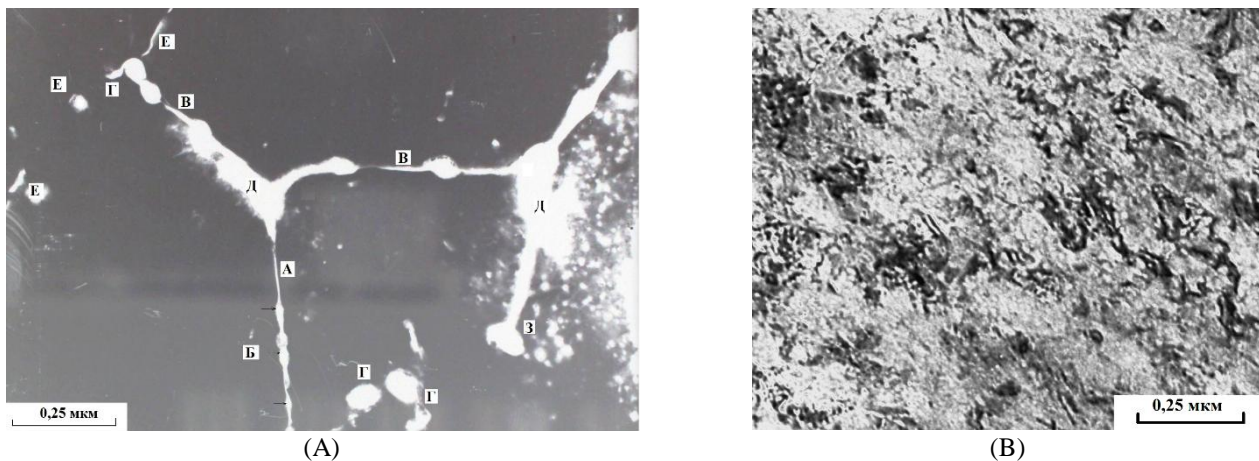
Four conclusions follow from the analysis of area III in Figure 1: the gradient of flow of dislocation density ( $\frac{\partial J}{\partial X}$ )

periodically increases and decreases on time at tribo-loading; the gradient of flow of dislocation density changes its direction; an increase in the intensity of the gradient of ascending and descending flows of dislocation density causes an increase in the lower limit of the change in the wear intensity by at least  $\approx 7$  times, and the upper limit of the change in the wear intensity in the avalanche-like selective mechanism of destruction of the surface layer by three orders of magnitude; the gradient of the

flow of dislocation density ( $J_2$ ) with a decrease in the stress value exceeds the gradient of the dislocation density flow ( $J_1$ ) with an increase in the stress value by  $\approx 1.7$  times.

Thus, the amount of accumulation of defects, their size and interaction determine the process of self-regulation in the area of nonequilibrium deformation. The accumulation of defects by the surface layer and the associated increase in the latent deformation energy lowers the activation energy of relaxation processes by so much that these processes, with further deformation, play the role of a kind of regulator of both the number of defects and the way of their interaction and distribution [5].

Analysis of the results obtained using transmission electron microscopy showed [8] that the main microstructural elements of dispersion of the surface layer are: 1) zones with a high density of dislocations, with the time of loading acquiring the form of thin bundles and oriented along the sliding direction; 2) slip bands and numerous thin twins along their boundaries, which are the sources of the initiation of small cracks; 3) numerous micropores inside the deformed lattice and along the grain boundaries, their coagulation leads to the formation of foci of transcrystalline and intercrystalline destruction (fig. 5, A). Prolonged frictional loading leads to progressive loosening of the surface layer of nickel (fig. 5, B), associated with an increase in the number of fracture centers [10]. Nonequilibrium vacancies at nickel lattice sites under plastic distortion form microporosity by the coalescence mechanism, which is a precursor of plastic shear [16 – 18] meso and macroscales that determine the lobe-layer-by-layer mechanism of destruction [12, 19] and avalanche-like selective destruction of the surface [8] into local moment of time.



**FIGURE 5. A– Pore formation and fracture along grain boundaries and through grain; B – Dispersion and loosening of the surface layer ( $t = 150$  ks)**

In a strongly deformed crystal lattice, high porosity develops reaching  $\approx 25\%$  of the total volume of the material, and a high concentration of microcracks and other discontinuities in the material [5].

It should be noted that it is necessary to conduct further systemic studies to establish the main invariant regularities.

#### IV. CONCLUSION

Analysis of the kinetics of the processes of deformation and destruction of scale levels: nano, submicro, micro, meso and macro using boundary conditions applied to the wave equation shows that:

- In the area of the equilibrium deformation in the absence of a gradient flow structural defects (fig. 1, area A, region II): the flow of the dislocation density is a constant value directed inward from the surface; the change of local gradient of the orientation of structural elements is a constant value that decreases in accordance with an inversely proportional relationship with increasing distance from the surface;
- The intensity of external influence determines the duration of the cycle of changes in strength characteristics, the amount of deformation energy accumulation and the degree of fragmentation of the crystal lattice of metals, and, accordingly, the local gradient of orientation of the boundaries of structural elements, where their geometric size, quantity, density, and interaction determine the dominant role of one or another scale level of plastic deformation at a given time of the kinetics of structure formation, and the mechanism of its destruction in accordance with the

minimum potential energy of interaction of the formed structure in the area of equilibrium deformation in the presence of a flow gradient of structural defects: in the area of equilibrium deformation in the presence of a flow gradient of structural defects: the velocity of the dislocation density movement (fig. 1, area B) deep from the surface is a constant value equal to  $\approx 1 \text{ (m}\cdot\text{s)}^{-1}$ ; the flow of the dislocation density gradient through a unit of area per unit of time is a constant value of  $\approx 1 \text{ m}^{-4}$ ;

- In non-equilibrium deformation ((fig. 1, region III)): the flow gradient of dislocation density changes its direction; the flow gradient of dislocation density ( $\frac{\partial J}{\partial X}$ ) is oscillating in time; an increase in the intensity of the gradient of ascending and descending flows of dislocation density causes an increase in the lower limit of the change in the wear intensity, at least  $\approx 7$  times, and the upper limit of the change in the intensity of wear by three orders of magnitude at the avalanche selective mechanism of destruction of surface layer; the gradient of the flow of dislocation density ( $J_2$ ) at decrease in the stress value exceeds the gradient of the dislocation density flow ( $J_1$ ) at increase in the stress value by  $\approx 1.7$  times.

The principle of least action is implemented, where the selective mechanism of destruction of the porous and amorphous layer determines the removal of foci of discontinuity of the material (microcracks, pores, duplicates, packaging defects, etc.), which determines the preservation of the integrity and continuity of the material.

It has been established that the kinetics of structure formation and evolution of the interface between metals under tribo-loading proceeds in accordance with the following provisions of nonequilibrium thermodynamics:

- Each stable state of the metal interface will have its own structure with a certain value of free energy and, accordingly, with the types of its redistribution between the elements of the boundaries and within the structural formation;
- The system tends to occupy a position or form such a structure of the interface, which corresponds to the minimum thermodynamic Gibbs potential;
- If the action of load-speed parameters or external influence exceeds some critical value of the energy supplied to the system, then it passes into a new structural state characterized by a lower value of free energy;
- The intensity of external influence determines the duration of the cycle of changes in strength characteristics, the amount of deformation energy accumulation and the degree of fragmentation of the crystal lattice of metals, and, accordingly, the local gradient of orientation of the boundaries of structural elements, where their geometric size, quantity, density, and interaction determine the dominant role of one or another scale level of plastic deformation at a given time of the kinetics of structure formation, and the mechanism of its destruction in accordance with the minimum potential energy of interaction of the formed structure.

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