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Modeling hydrodynamic flows in plasma fluxes when depositing metal layer on the surface of catalyst converters

D A Chinakhov^{1, a}, V D Sarychev², A Yu Granovsky², S A Solodsky¹, S A Nevsky², S V Konovalov²

¹ Yurga Institute of Technology of Tomsk polytechnic university, Russia, Yurga, Leningradskaya str., 26

² Siberian State Industrial University. Russia, Novokuznetsk, Kirova str., 42

E-mail: ^achinakhov@tpu.ru

Abstract. Air pollution with harmful substances resulting from combustion of liquid hydrocarbons and emitted into atmosphere became one of the global environmental problems in the late 20th century. The systems of neutralization capable to reduce toxicity of exhaust gases several times are very important for making environmentally safer combustion products discharged into the atmosphere. As revealed in the literature review, one of the most promising purification procedures is neutralization of burnt gases by catalyst converter systems. The principal working element in the converter is a catalytic layer of metals deposited on ceramics, with thickness 20-60 micron and a well-developed micro-relief. The paper presents a thoroughly substantiated new procedure of depositing a nano-scale surface layer of metal-catalyst particles, furthering the utilization of catalysts on a new level. The paper provides description of mathematical models and computational researches into plasma fluxes under high-frequency impulse input delivered to electrode material, explorations of developing Kelvin-Helmholtz, Marangoni and magnetic hydrodynamic instabilities on the surface of liquid electrode metal droplet in the nano-scale range of wavelengths to obtain a flow of nano-meter particles of cathode material. The authors have outlined a physical and mathematical model of magnetic and hydrodynamic instability for the case of melt flowing on the boundary with the molten metal with the purpose to predict the interphase shape and mutual effect of formed plasma jet and liquid metal droplet on the electrode in the nano-scale range of wavelengths at high-frequency impact on the boundary “electrode-liquid layer”.

1. Introduction

Emission cut of harmful combustion products is a vital problem relevant to the ecological state of our planet, population health, gene pool and cultural treasure preservation, environment safety of foodstuffs. The ecological problem is as important as preservation of peace, and providing world population with foodstuffs. Therefore, each particular measure aimed at cutting harmful emissions of internal combustion engines into the atmosphere is urgent and significant. When manufacturing catalyst converters a number of difficulties are to be overcome; among them there is coating the



working surface with a thin layer of metal-catalysts. As a consequence, attempts are made all over the world to design new efficient procedures of depositing such layers.

The regularities describing generation and evolvement of structure and phase states in materials exposed to concentrated energy flows have been in the focus of researches for a long time [1]. The surface layers of materials modified with the help of these flows are distinguished by high hardness and wear resistance [2, 3]. Heterogeneous plasma fluxes are one of the most efficient modification procedures [4]. This is a quite complex process including formation of droplets on the electrode tip; movement of droplets in the jet; interaction between plasma and droplet jet with the product; processes occurring in the molten layer of the product enriched with droplet component of the jet [5, 6].

The technology of plasma arc finds wide application in metallurgy, powder spraying, surfacing etc. Thermal conducting plasma is formed when consumable electrode surfacing. Its properties and operating conditions are of great importance for processes of weld pool and weld formation, influencing this way on properties of the modified surface. Therefore, computational modeling of hydrodynamic processes and thermal fields are relevant to the development of surface modification procedures [7]. Some researchers [8] explored the effect of cathode casing on the thermal field. It's revealed that different potential boundary conditions on the back surface of the anodic plate can hardly effect on the heat flow in cathode or within the arc, affecting, however, current density and temperature field near the anodic plate. For the case the same computational domain includes cathode and anode arcs, additional conditions on the boundary "plasma - electrode" are required to take into consideration the energy flowing through it. The calculation of isotherms for electric current 200 detects a satisfactory fit with the experiment. The authors [9] investigated formation of self-organizing structures in electrodes. As a result, a three-dimensional double-temperature imitation model was developed and revealed that a dotted structure arises gradually due to the increasing level of anode cool-down: from one fuzzy spot at low cooling levels to small spots covering anode zone at intense cool-down. The authors [10] compared models of turbulent flows for free burning of high-intense argon arc. They modified Navier-Stocks equations via considering the transfer of emission, power consumption and electromagnetic forces. To model the turbulence, which is relevant to the arc boundary because of sharp temperature gradient between the arc column and gas around it they referred to zero and two equation of viscosity. It was found out that the proposed model can provide better description of temperature profiles in the arc. The model of plasma flux in anode zone of plasma gun at constant current is presented in [11]. Maxwell and heat transfer equations are solved within this model both for liquid and solid parts of the electrode. The mean temperature of tungsten and copper parts of anode can be assessed with the help of this model. The second important feature of the model is that it takes into account injection of gas into the plasmatron. The researchers [12] fulfilled computational modeling of high pressure in arc discharge relying on self-congruent simulation of the most components, e.g. electrodes and their interaction. In particular, the arc column and cathode part of the discharge are modeled on the base of the double-temperature hydrodynamic model and the model of nonlinear heating surface, respectively. The results of modeling are presented for free burning arc in the atmosphere pressure of argon, covering the arc currents from 10 to 200 A. It was revealed that electric power delivered to the near cathode layer is given to cathode and arc column, as well. This effect can't be described in terms of local thermodynamic equilibrium (LTE). The enthalpy of electron transfer can significantly increase the thermal input caused by the conductivity of electrons and heavy particles, dominating, therefore, the process of power transfer from the near cathode layer into the arc column. Hence, temperatures calculated in terms of LTE-model are much higher than those taking into account the input of electrons and heavy particles [12].

The processes occurring in the molten pool under the action of plasma are investigated in works [13-17]. The authors [13] addressed to the issue of heat exchange and liquid flows in conditions of pulse arc welding the stainless steel and solved this problem relying on the finite elements method. The proposed model made it possible to explore the time evolvement of the weld pool at constant and pulsed current. Having compared the results, it's concluded that pulsed current furthers formation of a better joint weld. Computational simulation was supplemented with experiments in [14], including

high-speed recording the infra-red range of the weld pool surface. While processing and analyzing the images a satisfactory fit with the computational model was revealed. The effect of various parameters on the weld pool geometry was analyzed in [15]. According to the results, the gradient of surface tension is the most relevant factor influencing the weld pool geometry, whereas magnetic conductivity, coefficient of thermal expansion of material are less important. The paper [16] is dedicated to modeling processes in electric arc and weld pool with the dividing electrode. This electrode influences significantly on temperature and flow of arc plasma but is irrelevant at peak temperature and arc voltage. Having increased the dimensions of this electrode, a conclusion is made that one-peak profile of current density distribution, heat flow and temperature on anode become double-peak. The overall heat release in anode changes a little in spite of some oxygen. A standardized model of the system “welding arc – weld pool” is presented in [17]. It's ascertained that temperatures and velocities of distributions in the arc are not rotationally-symmetric.

Gas (argon) streamline and heating in the diaphragmatic channel are calculated according to the position and dimensions of the diaphragm, consumption and intensity of gas rotation in terms of equilibrium magnetic and gas-dynamical model [18]. The specifics of arc, changing in channels with alternating cross-section was determined for rotational and non-rotational flows. It is indicated that diaphragming of the channel together with intense gas rotation provides localization of plasma flux before the diaphragm in the narrow paraxial zone with the distinct thermal interface and the outer gas flow (effect of vortex thermal insulation); the length of stabilization zone depends on the size of a diaphragm opening and its position in the channel. The authors [19] dealt with the computational investigation of a micro-wave discharge in argon jet, injected axially into the coaxial with the shortened inner electrode. It's ascertained that the inner electrode exit section is of great importance for formation of a micro-wave discharge, the flow of cold gas, moving in its channel, cools its walls down, stabilizing, therefore, the burn of a micro-wave discharge and furthering formation of the sharp front heat edge.

The present day research into modeling the plasma fluxes is focused on the development of a single mathematical model describing processes both in the zone adjacent to cathode and in the weld pool. Therefore, this work aims at modeling hydrodynamic flows in the diaphragmatic electric arc.

2. Problem formulation

It's quite reasonable to begin exploration of magnetic and hydro-dynamic flows in plasma with calculation of electromagnetic force. We write Maxwell equation for its calculation using scalar potential V and vector potential \vec{A} :

$$\begin{aligned} \nabla \cdot \left(\sigma \nabla V + \sigma \frac{\partial \vec{A}}{\partial t} \right) &= 0, \quad \sigma \frac{\partial \vec{A}}{\partial t} + \frac{1}{\mu_0} \nabla \times (\nabla \times \vec{A}) + \sigma \nabla V \\ \vec{E} &= -\nabla V - \frac{\partial \vec{A}}{\partial t}, \quad \vec{j} = \sigma \vec{E}, \quad \vec{B} = \nabla \times \vec{A} \end{aligned} \quad (1)$$

The model based on Navier-Stocks equation and that of thermal conductivity is used to model the plasma flow and temperature distribution in it:

$$\begin{aligned} \rho \left(\frac{\partial \vec{u}}{\partial t} + \vec{u} \cdot \nabla \vec{u} \right) &= -\nabla p + \mu \Delta \vec{u} + \vec{F}_V, \\ \nabla \cdot \vec{u} &= 0, \\ \rho C_p \left(\frac{\partial T}{\partial t} + \vec{u} \cdot \nabla T \right) &= \nabla \cdot (k \nabla T) + S_V \end{aligned} \quad (2)$$

where \vec{u} - velocity vector, p - pressure, ρ - density, μ - dynamic viscosity, \vec{F}_V - volumetric forces, resulting from the sum of Lorenz and gravitation forces, T - temperature, C_p - specific heat, k - thermal conductivity coefficient, S_V - volumetric heat sources.

The volumetric heat source for anode and cathode is Joule effect $S_V = \vec{j} \cdot \vec{E}$, whereas for plasma there is additional heating from electron enthalpy and losses caused by radioactive emission.

Boundary conditions for formula (1) and (2) are formulated. We consider a cylindrical electrode, one of its tips is semi-spherical and the other one is flat (Figure. 1).

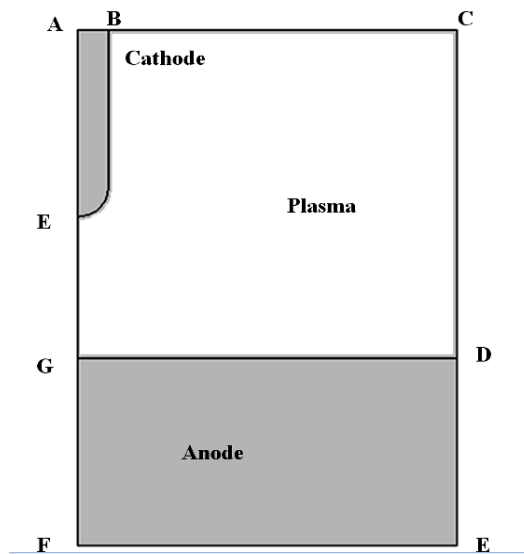


Figure 1. The geometry used in the calculation of the plasma arc

The heat flow on line GD (plasma/anode) is set as a total of plasma heating, and heat arising from electron condensation $|\vec{j} \cdot \vec{n}| \varphi_a$ (where φ_a anode work function of electrons) and radiation losses $\varepsilon \sigma_B T^4$ (where ε - emissivity of anode, σ_B - Stefan-Boltzmann constant). On line EB (plasma/cathode) the heat flow depends on plasma, ion heating $j_i V_i$, radiation losses $\varepsilon \sigma_B T^4$ and thermo-ion cooling $j_e \varphi_c$ (power required for electron emission from cathode, which is a product of electron flow density and cathode work function of material). Boundary conditions for Maxwell and magnetic hydrodynamic equations are provided in Table 1. The properties of materials necessary for calculations are given in Table 2. The kinetic parameters of plasma (heat capacity, thermal conductivity, viscosity, electric conductivity) are dependent on temperature and formulated in the table.

Table 1. Boundary conditions

	AB	BC, CD	DE	EF	AE, FG	EG
T, q	$-\vec{n} \cdot \vec{q} = 0$	T_0	T_0	T_0	$\vec{n} \cdot \vec{q} = 0$	$\vec{n} \cdot \vec{q} = 0$
(u, v, w)	-	$p = p_0$	-	-	-	$\vec{n} \cdot \vec{v} = 0$
(V, \vec{A})	$\vec{j} \cdot \vec{n} = \frac{I_c}{\pi R_c^2}$	$\vec{j} \cdot \vec{n} = 0$ $\vec{A} \times \vec{n} = 0$	$\vec{j} \cdot \vec{n} = 0$ $\vec{A} \times \vec{n} = 0$	$V = 0$ $\vec{A} \times \vec{n} = 0$	$\vec{j} \cdot \vec{n} = 0$ $\vec{B} = 0$	$\vec{j} \cdot \vec{n} = 0$ $\vec{B} = 0$

Table 2. Material properties of cathode, anode and gas

Symbol	Nomenclature	Value
ρ_m	Density anode and cathode metal	7500 kg m ⁻³
C_m	Specific heat of metal	602 J kg ⁻¹ K ⁻¹
k_m	Thermal conductivity of metal	26 W m ⁻¹ K ⁻¹
σ_m	Electrical conductivity of metal	7.7×10^5 $\Omega^{-1} \text{ m}^{-1}$
φ_c	Surface work function of cathode	4.52 V
V_i	Plasma's ionization potential	15.68 V
φ_a	Surface work function of anode	4.65 V
ε	Surface emissivity	0.9

3. Results and discussion

System (1), (2) with boundary conditions was solved with the help of the finite elements method; the diaphragm was ignored and taken into consideration. Cathode had a form of a cylinder, limited by a semi-sphere of the same radius; i.d. 0.8 mm, in the first case anode had a form of a plane, and in the second – a plane with the opening, the radius of which was 2 mm. Cathode-anode distance (EG) was 3.8 mm. The density of current was set on cathode to provide the overall current 100 A. The potential of anode was assumed zero. The initial conditions are in line with gas at rest and material of the electrodes. The temperature at the start time is constant (300K) for the whole zone. The initial density was set constant for each zone: for cathode and anode – the density of steel, for the inter-electrode space – the density of air.

The space-wise step was 0.1 mm, and the time step was set automatically according to Courant condition.

Streamline analyzed for the case without opening in anode.

Streamline is non-stationary in plasma jet at points of time below 1 μs , the heat wave propagates from cathode to anode, and three vortex zones are formed in the area of streamline. One zone caused by the opposite jets is close to cathode and symmetry axis. These jets arise from the action of radial component of electromagnetic force directed towards the axis. The second vortex zone is located near the symmetry axis, approximately in the middle between anode and cathode. The interaction of these zones results further in formation of one stable vortex zone, located at the distance equal to two radii from the axis. The third vortex zone is formed near anode at point of time 0.1 μs and exists for the whole period of calculation.

Therefore, the essentially non-stationary streamline in plasma transforms into the quasi-stationary state, induced by viscous and electro-dynamic forces. The streamline has a complex structure comprising some zones: one close to the electrode, the paraxial flow with the impact transition, and two vortex zones. The distribution of velocity on the axis is presented in Fig. 2, b, which shows that arc plasma velocity increases from zero (E – cathode surface), reaching its peak ~ 500 m/s at the distance 1 mm from cathode surface, reducing afterwards to zero and reversing the sign. Analytical models or numerical computations with a significant spatial step can hardly facilitate identifying the above specifics.

From practical perspective, the analysis of hydro-dynamic situation furthers making two important conclusions. First, the vortex near cathode and its existence for 1 μs facilitates formation of tiny particles approximately 100 nm of the molten material. Second, the vortex zone close to anode causes a counterflow of plasma, preventing small particles from anode surface and complicating

deposition of the layer. Therefore, we suggest using anode with an opening. The calculations for the case with the opening are given in Fig. 3, demonstrating that vortex zones do not arise; this fact is relevant for the development of surfacing procedure.

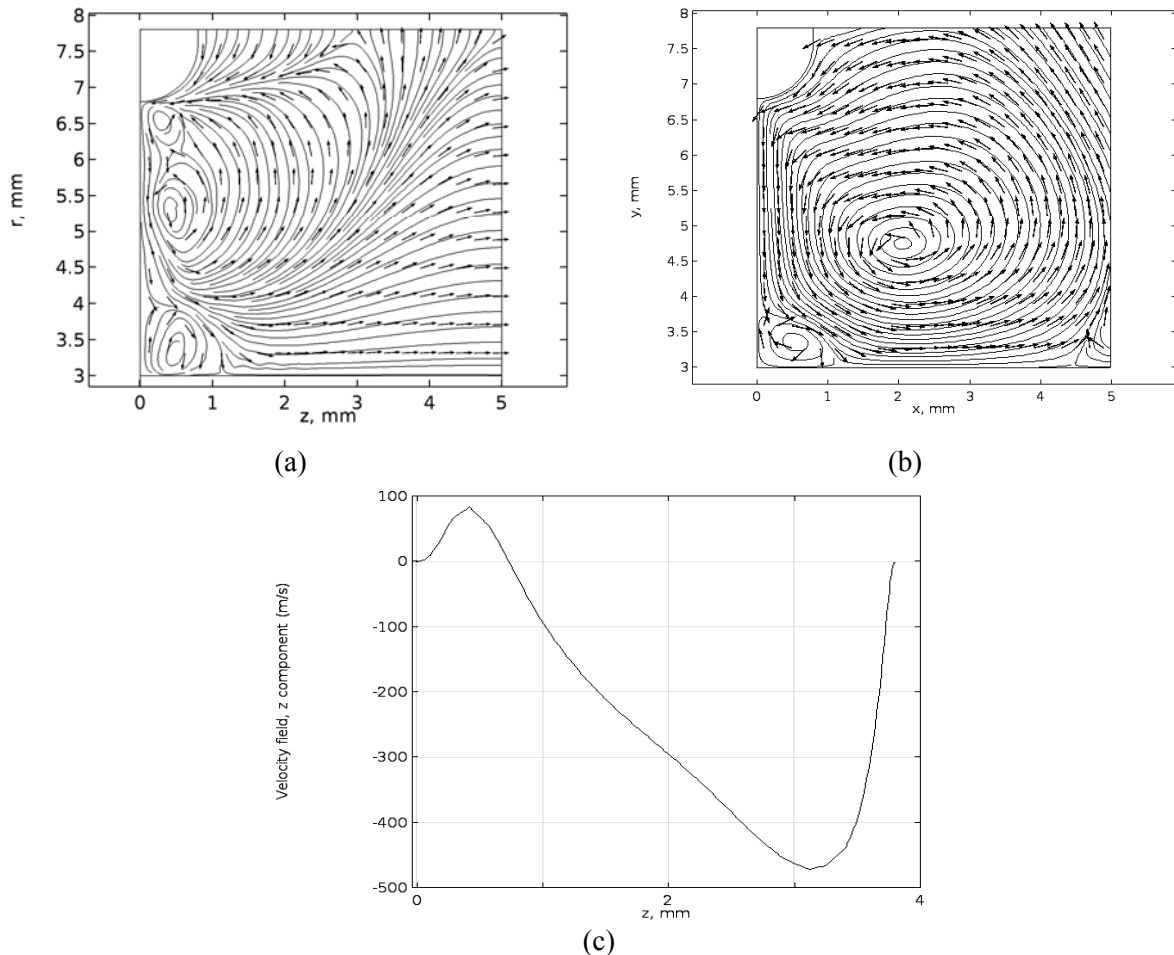


Figure 2. Velocity field of arc: (a) - streamlines at $t=1.1 \mu s$; (b) - streamlines at $t=8$ ms; (c) - v_z on the axis of symmetry GE ($t=8$ ms)

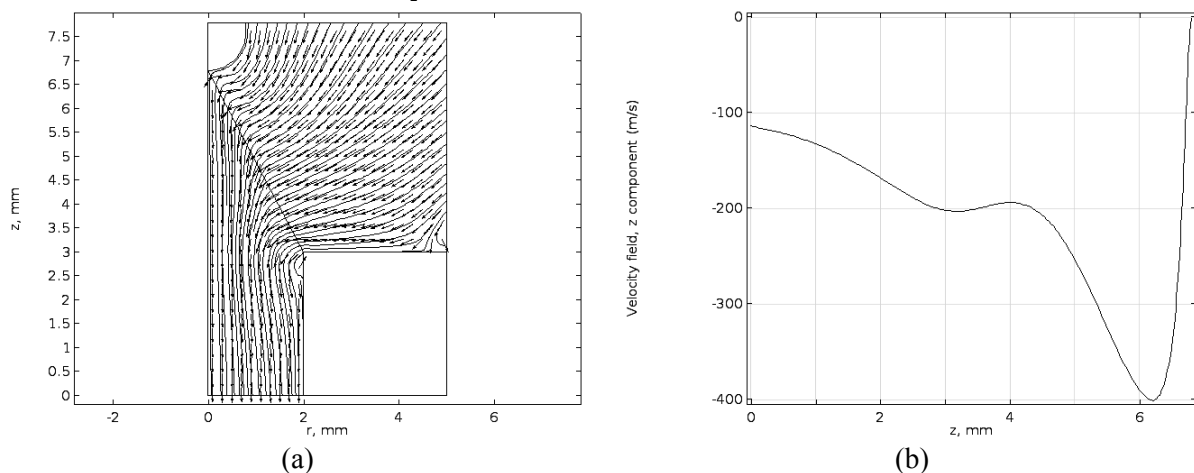


Figure 3. Plasma velocity fields: a) – streamlines of plasma fluxes; b) – velocity of arc plasma on the axis.

As stated before [20], three vortex zones of hydro-dynamic flows in the jet of electric arc plasma arise in the arc plasma and other injection sources of charged particles generated at the consumable electrode tip (between a rod electrode and a flat conductive target located perpendicularly to it). The interaction of these zones results in formation of two stable vortex zones; one of which is at the distance of two electrode radii from the axis and in the middle between the electrodes, the other vortex zone is immediately close to the solid wire electrode. A counterflow of plasma is generated in this zone and emits small particles. The vortex zones do not occur when using a flat electrode with the opening. Therefore, small particles can pass through it and get consolidated. The hydro-dynamic situation analyzed in previous investigations revealed formation of the first vortex close to cathode, which existence for 1 μ s furthers formation of small particles (approximately 100 nm) of molten material.

As reported in [21], small particles of molten metal leave the surface due to high-frequency impact of Rayleigh waves on the boundary “solid-liquid” in conditions of plasma fluxes. The ultrasound impact on the electrode with diameter 0.8 mm and molten tip (material - Fe, Ni, Co) was tested in laboratory conditions and its relative extension was identified $\Delta l/l \approx 10^{-6} \dots 10^{-2}$ m at oscillations 2...50 MHz.

In consideration of elastic anisotropic medium, e.g. metal, it's noteworthy that two types of bulk waves arise in this medium under point energy deposition: longitudinal waves, where particles spread in the same direction as the wave propagates, and transverse waves with particles spreading perpendicularly to the direction of wave propagation. Longitudinal and transverse waves might be accepted bulk free oscillations.

As a magnetic impulse is delivered to the welding wire, consideration of transverse waves can be omitted because a point impulse is applied throughout the diameter; therefore, arising transverse waves are mutually absorbed in each point. The velocity of longitudinal waves is designated (c_n). So the velocity of longitudinal wave propagation through elastic materials is formulated as follows [21]:

$$c_n = \sqrt{\frac{E(1-\nu)}{\rho(1+\nu)(1-2\nu)}}, \quad (3)$$

where ρ – density of material; ν – Poisson ratio; E – Young's modulus.

Thus, surficial drift propagates along axis x in form of an elastic wave and is exponentially damped deep into the wire. This surficial wave is called Rayleigh wave $R(c)$, its velocity is written:

$$R(c) = (2 - m^2)^2 - 4\sqrt{1 - m^2} = 0, \quad (4)$$

where $m=c/c_n$, where c_n – sound speed in the material.

As a consequence, a required frequency of the welding wire can be determined via calculating Rayleigh wave velocity, since we know that the wave becomes concave at velocities approximating but not exceeding $R(c)$, i.d. $c_n < R(c)$, and convex at $c_n > R(c)$.

Provided that a welding wire of ferromagnetic materials is used, a surficial wave is generated by the applied electromagnetic impulses and spreads in both directions from the impact center, i.d. electromagnetic impulse. When mechanized shielded welding such electromagnetic impulses are applied to the wire moving in the welding arc zone. As the velocity of surficial Rayleigh waves exceeds significantly the velocity of wire displacement, the latter can be ignored in calculations.

Therefore, Rayleigh waves approach the liquid layer of anode tip, propagating along the welding wire surface, and on the solid-liquid boundary there are waves, regarded as Love's waves with vertical polarization and quite different structure and velocity as compared with Rayleigh waves. They include a weak inhomogeneous wave in the liquid, the amplitude of which decreases slowly moving from the boundary of two essentially inhomogeneous (longitudinal and transverse) waves in the solid. That is why wave energy and motion of particles are localized mostly in the liquid but not in the solid [21].

Rayleigh wave is generated on anode material due to the applied high-frequency impulses, the amplitude of which decreases slowly moving from the boundary of two essentially inhomogeneous (longitudinal and transverse) waves in the solid. So wave energy and motion of particles are localized mostly in the liquid but not in the solid. When propagating, these waves constantly emit energy into the liquid, generating there an inhomogeneous wave, spreading in both directions from the phase boundary. The phase velocity of this surficial acoustic wave is determined, attenuation constant on the wavelength $\lambda \sim 0.1$. As a consequence, energy of a high-frequency electromagnetic field is delivered through the electrode into the liquid droplet due to Rayleigh and Love's waves. The liquid layer of the electrode surface gets more active because of the obtained energy, as the result, the increase is registered in forces of all-around compression of nano-scale heterogeneity bridges; they get destructed faster, initiating the detachment of droplets from the electrode surface. As identified in previous research [22, 23], heat and mass transfer in the welding zone, frequency and dimensions of formed droplets can be controlled via changing the shielding conditions of molten electrode metal droplet and arc plasma column.

Therefore, conditions required for formation of nano-scale waves on the liquid anode layer are to be provided with specially designed equipment, and detachment of particles is possible due to high-frequency impact on the electrode, making the particles leave the phase interface "electrode – liquid layer". High-frequency (ultrasound) electromagnetic impact on the metallic electrode generating elastic longitudinal oscillations within range 2.5...25 MHz is accepted for the detachment of nano-scale waves from the electrode surface. As demonstrated in experiments, plasma flux treatment results in a structure with cellular crystallization, cells dimensions range 100 to 300 nm (Fig. 4).

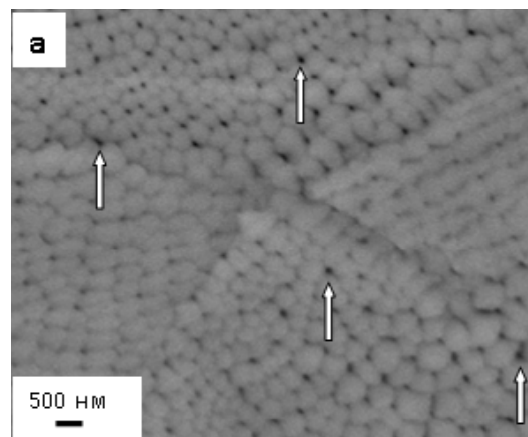


Figure 4. – Microstructure Ni, deposited via high-frequency plasma spraying

Conclusions

1. The conditions are ascertained which are necessary for origination and evolvement of thin layers with surficial-periodic micro-and nano-scale relief on the surface of liquid metal when supplying metallic wires into the zone of heterogeneous plasma of the electric arc in conditions of Kelvin-Helmholtz and thermo-capillary (Marangoni) instabilities. The supplementary magnetic and hydro-dynamic impact of oriented plasma fluxes on thin molten metal layer occurs when using a working element to form required directions of plasma fluxes together with high-frequency (ultrasound) impact, as the result, the flow of micro-and nano-scale droplets is deposited on the substrate (target).

2. Generating elastic Rayleigh waves deliver the longitudinal oscillations to the phase boundary "electrode – liquid layer", creating conditions this way for detachment of particles formed by the vortex flux of the plasma jet. The amplitude of Rayleigh wave decreases slowly (exponentially) moving from the boundary between two significantly inhomogeneous (longitudinal and traverse) waves in the solid. It is the reason for the localization of wave energy and its constant emission into

the liquid, but not into the solid, an inhomogeneous wave is generated in the liquid and propagates from the boundary, increasing the detachment effect of the micro-waves.

3. The proposed deposition procedure of a catalytic layer using the ultra-disperse metal furthers generation of the electricity-conductive foil of good quality with higher catalytic activity of metal due to the nano-scale particles. The above procedure makes it possible to form coatings distinguished by good adhesion on various surfaces, as well as with good continuity, homogeneity and reproducibility of the layer composition.

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