PHYSIC-MATHEMATICAL MODELING OF TRANSPORT PHENOMENA IN ELECTROLYTES DURING METAL SURFACE DECONTAMINATION BY ELECTROLYTE-PLASMA TREATMENT

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Abstract

This work is devoted to the study of technology and physical-mathematical modeling of transport phenomena in electrolytes during the decontamination of metal surfaces by the method of electrolyte-plasma treatment.

Keywords: *electrolyte-plasma processing, decontamination, nuclear power plant, mathematical modeling*

The most important factor in sustainable economic development is the introduction of new effective technologies that save resources and have higher economic and environmental indicators. These technologies include electrolyte-plasma processing (polishing) (EPP) [1]. This method allows processing stainless steel, carbon steel and non-ferrous metal products in aqueous solutions of salts. The technology uses safe aqueous solutions of salts of moderate concentration. Depending on the purpose of a particular technological process, EPT can be carried out for the purpose of electrolytic heating or quenching of products [2, 3], chemical-thermal treatment (carburization, nitriding, cementation), oxidation, surface cleaning from contamination, removal of coatings, electrolyte-plasma polishing at the stage of finishing processing of surfaces of metals and alloys [1, 4–7].

Studies conducted at the scientific institution "JINR – Sosny" [8, 9] showed that EPP is a promising way to deactivate surfaces of removable equipment of NPP with any complexity of configurations. This method meets all the requirements for methods of decontamination of nuclear power plant equipment, has high efficiency and speed of surface treatment.

1. ELECTROLYTIC-PLASMA POLISHING OF METAL SURFACES

Electrolytic-plasma polishing of surfaces of products from electrically conductive materials is based on the use of the phenomenon of spontaneous pulsed electrical discharges occurring near the surface of a workpiece immersed in a bath with a weak concentration of salt electrolyte. The working bath itself usually serves as the cathode of the electric circuit; the workpiece connected to the voltage source is the anode (Fig. 1). A thin (≈ 50 ÷ 100 μm) vapor-gas shell (VGS) is formed around the anode through which microdischarges flow. The stability of VGS is a necessary condition for the implementation of the process of smoothing microroughnesses.

The main technological parameters of EPP:

- current density 0.2 ÷ 0.6 A/cm²
- constant voltage ~ 350 V
- working temperature of the electrolyte 70 ÷ 90 °C
- average surface polishing time 3 ÷ 5 min.
- non-toxic solutions of salts as an electrolyte (for the processing of stainless steel products according to GOST 5632-72 a 4 ÷ 10% aqueous solution of ammonium sulfate is used).

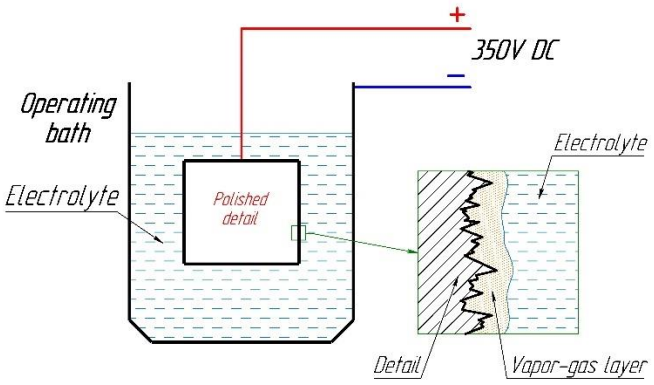


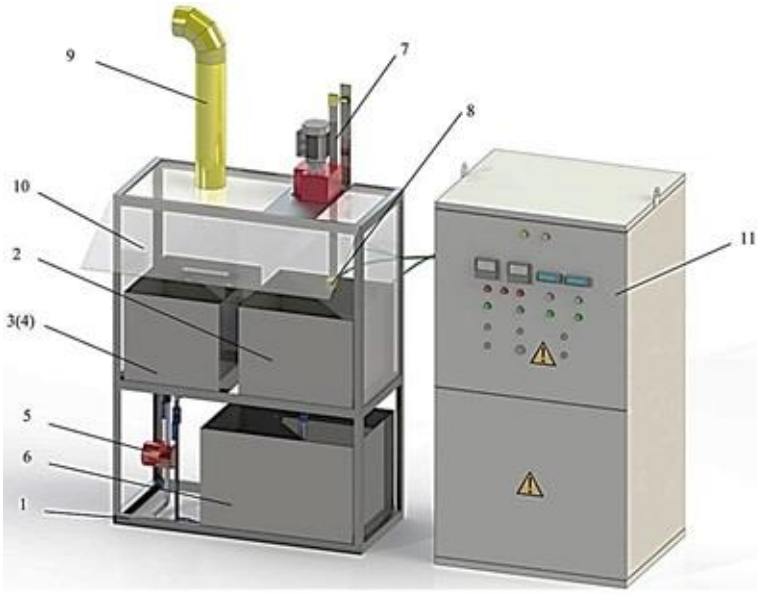
Fig. 1. The principle of electro-plasma polishing

In the State Scientific Institution "Joint Institute for Energy and Nuclear Research – Sosny" of the National Academy of Sciences of Belarus (hereinafter referred to as the scientific institute "JINR – Sosny"), two EPP units are in operation: a laboratory unit EPP-15 with an electric power of 15 kW (Fig. 2a) and industrial installation EPP-800 with an electric power of 800 kW (Fig. 2b). Installations for EPP consist of: a working bath 2, in which the product is processed; electrolyte cooling baths 3 with a cooler 4 installed therein; vortex pump for circulating electrolyte 5; electrolyte sedimentation baths after polishing 6; a lifting mechanism 7 with a mount 8 of the polished product; exhaust systems for gases formed during polishing 9; the control panel 11 and the power control cabinet 12 (in the EPP-15 installation, the power unit and the control panel are mounted in one cabinet 11).

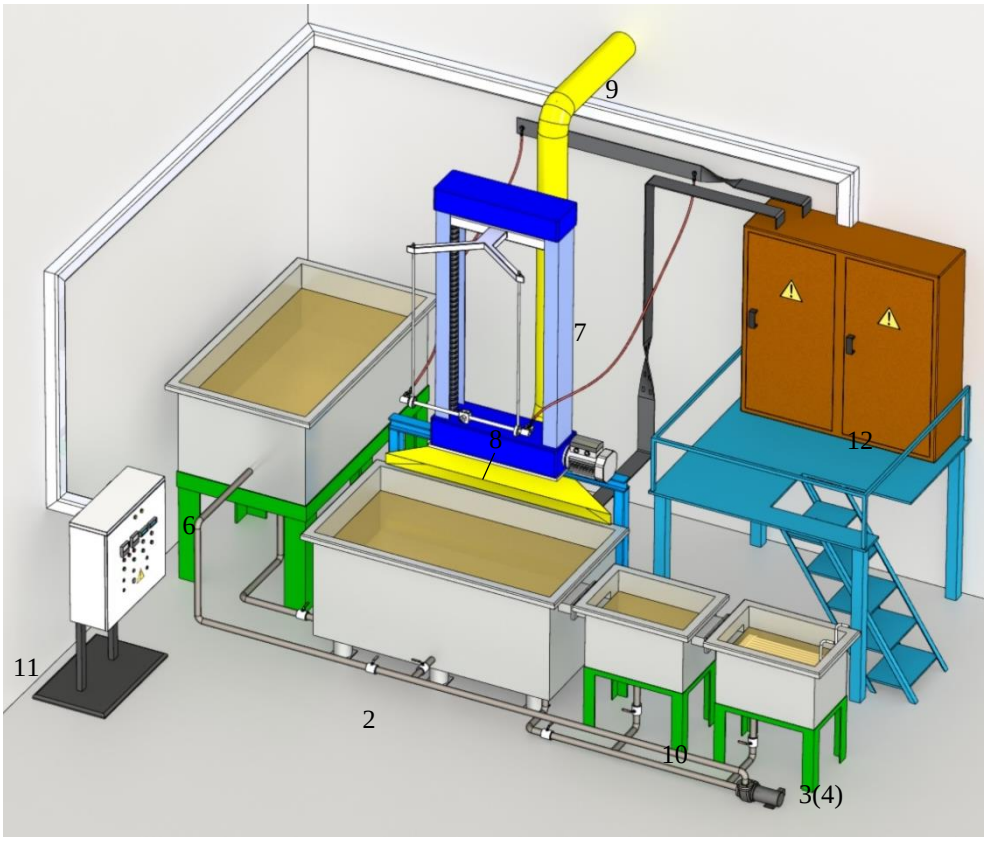
The EPP-800 unit is additionally equipped with an intermediate capacity 10 between the working bath and the cooling bath to precipitate an insoluble electrolyte precipitate during polishing of a large batch of products. The main technical characteristics of the plants are given in table 1.

Parameter, dimension	**EPP-15**	**EPP-800**
Electric power, kW	15	800
Polished surface area (per cycle), cm2	400	10 000
Rated rectified current, A	50	2 000
Volume of a working bath, l	70	1 800
Volume of cooling bath, l	30	600
Overall dimensions of the installation, m (L × H × D)	2,5×2×2	10×4×4

Table 1. The main technical characteristics of EPP installations



a) unit EPP-15: 1 - frame construction; 2 - working bath; 3 - electrolyte cooling bath; 4 - electrolyte cooler; 5 - electrolyte circulation pump; 6 - bath of sedimentation of the electrolyte after work; 7 - lifting mechanism; 8 - mounting unit of the workpiece; 9 - gas exhaust system; 10 - a protective guard; 11 - control panel and power cabinet



b) installation EPP-800: 2 - working bath; 3 - electrolyte cooling bath; 4 - electrolyte cooler; 5 - electrolyte circulation pump; 6 - bath of sedimentation of the electrolyte after work; 7 - lifting mechanism; 8 - mounting unit of the workpiece; 9 - gas exhaust system; 10 - bath of sedimentation of the electrolyte during polishing; 11 - control panel; 12 - power cabinet installation

Fig. 2. General view of the installations for electrolyte-plasma polishing EPP-15 and EPP-800

As mentioned above, the presence and stability of VGS formed around the workpiece during EPP is a prerequisite for the process of polishing the surface – smoothing microroughnesses. The most important parameters that determine the stability of VGSs are the electrical, temperature and hydrodynamic conditions in the near-electrode zone and the electrolyte, which are influenced by the geometric shape of the treated surfaces and the way the polished parts are placed in the working bath. For example, when an article in the shape of a cone was immersed in the working bath of the EPP-800 installation, when the axis of the cone is perpendicular to the free surface of the electrolyte, unstable hydrodynamic flows of electrolyte appeared in the internal volume of the product, emissions of steam and drops of electrolyte through the upper base of the polished part were observed. This led to the appearance of large current surges and, as a result, to the triggering of the installation safety system. A change in the orientation of the immersion of the part (the axis of the cone is parallel to the free surface of the electrolyte) made it possible to get rid of the formation of unstable flows of electrolyte, it was possible to completely immerse the product in the working bath and conduct high-quality polishing of its internal and external surfaces. The experimental and analytical substantiation of this phenomenon is considered in detail in [10].

The complexity of the geometric shape of polished products is also an important factor in the stability of the process of electrolyte-plasma treatment. If, when processing simple flat objects, difficulties usually do not arise, then when processing complex volumetric products, unpredictable circulation of the electrolyte is observed in the working bath with steam emissions through holes and nozzles. The consumed energy increases several times in comparison with the calculated relative to the cultivated area. Often this leads to a breakdown of the VGS and to a violation of the polishing process.

2. STUDY OF THE INFLUENCE OF THE COMPLEXITY OF THE SHAPE OF THE PROCESSED PRODUCTS ON THE EPP PROCESS

To conduct an experiment on the influence of the shape of the workpieces on the thermal and electrical parameters of the electrolyte during EPP, parts of various geometric shapes were made of stainless steel 12X18H10T in accordance with GOST 5632-72. The dimensions of the investigated parts are designed so that their polished surface areas are the same and equal to 0.23 m2. In this case, the peculiarity of the EPP process was taken into account that the inner surfaces of long products in the form of pipes are not processed [4]. The controlled parameters and conditions of the experiment are shown in table 2. The type of parts and their dimensions are presented in table 3.

Parameter	Value
The area of the polished surface of the parts, m2	0,23
Material of polished parts	steel 12X18H10T in accordance with GOST 5632-72
Controlled parameters	Power consumption (A), electrolyte temperature at two points (T1 и T2)
Experimental unit	ЭПП-800
Polishing electrolyte	6% (mass.) aqueous solution of ammonium sulfate $(NH_4)_2SO_4$ according to GOST 9097-82
The volume of electrolyte, l	1 800
The initial temperature of the electrolyte, ℃	80±1
The processing time of one part, min	7,5

Table 2. Controlled parameters and conditions of the experiment on the influence of the shape of the processed products on the EPP process

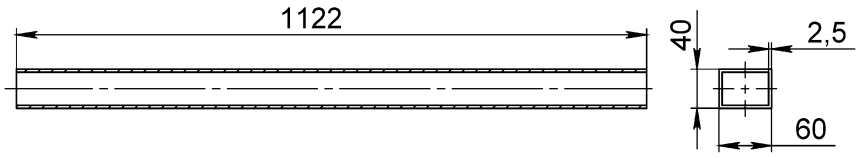
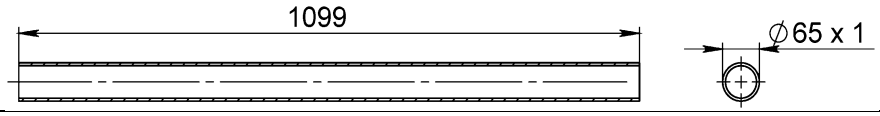
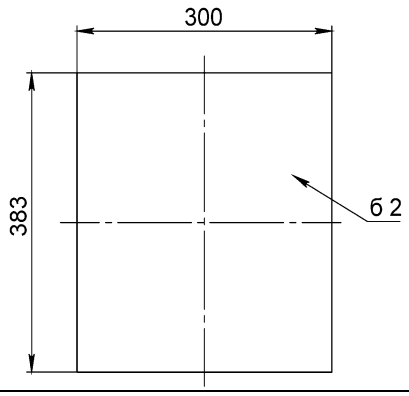
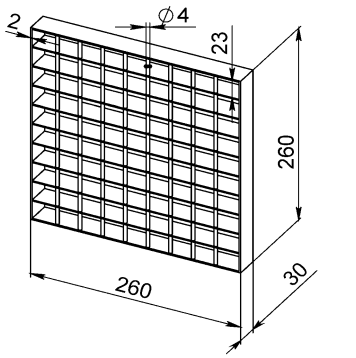
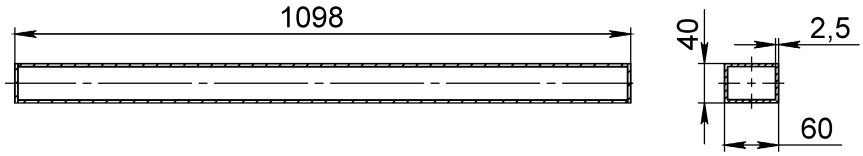
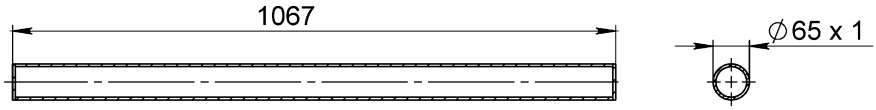
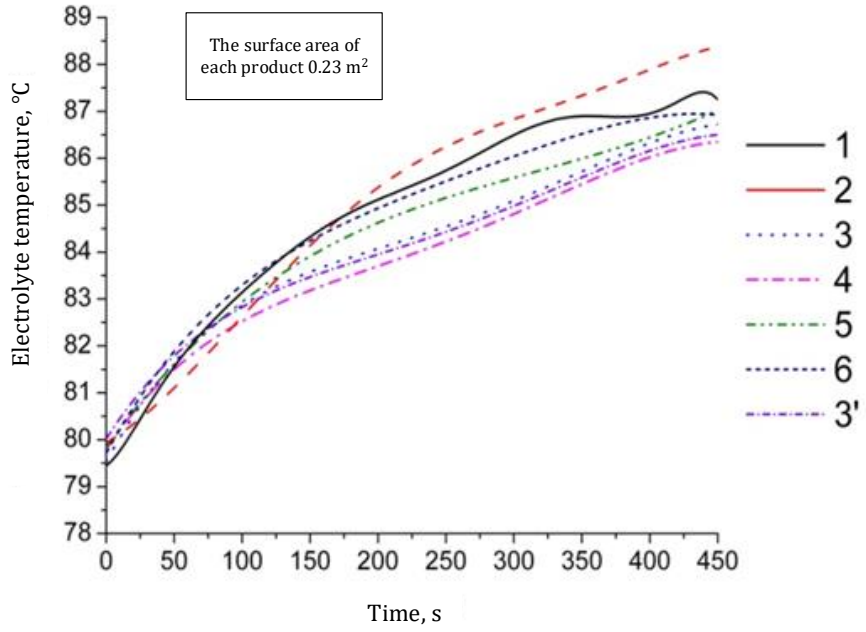
№	Part Type	Part Dimensions
1	Profile pipe (without stubs)	
2	Pipe (without stubs)	
3	Sheet	
4	Greed	
5	Profile pipe (with stubs)	
6	Pipe (with stubs)	

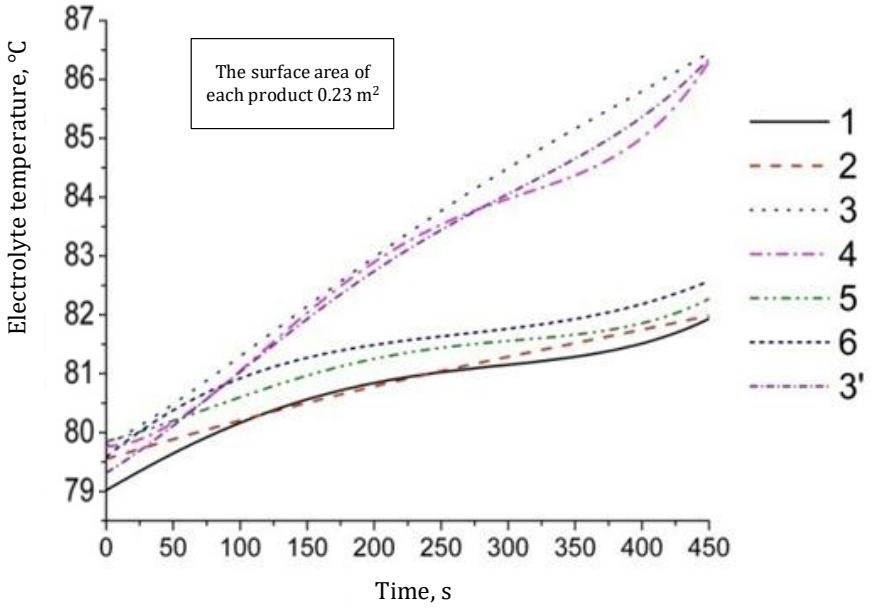
Table 3. Type of processed products and their main dimensions

The experiment was conducted on the installation of the EPP-800. To measure the temperature of the electrolyte during the experiment, two temperature sensors were used installed in the volume of the working bath of the installation: T1 – resistance thermoconverter TSP-1199/011 (standard sensor of the installation EPP-800), T2 – resistance thermoconverter TSP-1199/12. The T1 sensor measures temperature at a depth of 450 mm from the free surface of the electrolyte, T2 – at a depth of 750 mm.

The control of the parameters of the polishing process was carried out using the multichannel meter-controller IR SOSNA-002, mounted in the control panel of the installation. The device is intended for measurement, control and signaling as part of automated process control systems. The device measures technological parameters with the help of primary converters, generates control and information signals for other elements of control systems. The values of current consumption and electrolyte temperatures were transferred from the SOSNA-002 IR to a personal computer using the SysView v.2.1.8 [13] program for collecting and displaying data from SOSNA devices with a recording frequency every 3 seconds.



a) temperature T1



b) temperature T2

Fig. 3. Graphs of changes in electrolyte temperature during polishing of parts No. 1-6. a – temperature sensor T1; b – temperature sensor T2

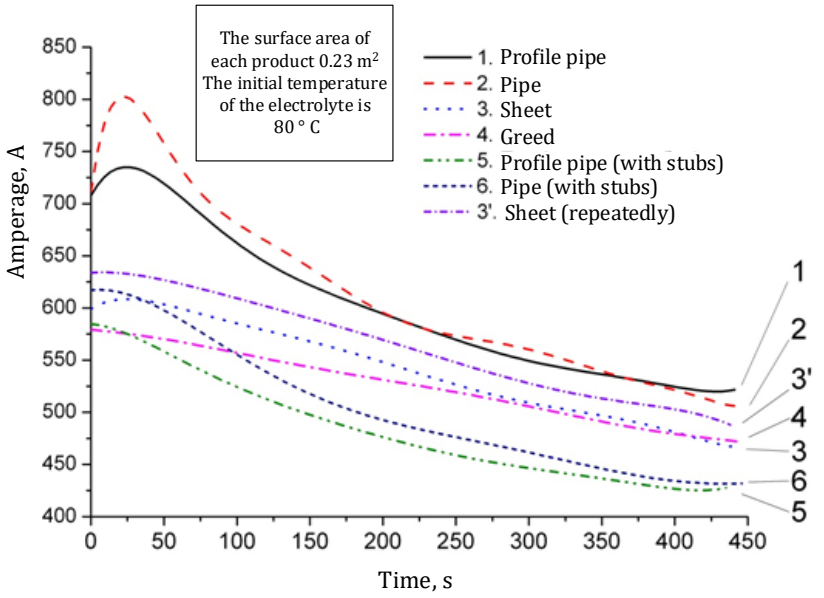


Fig. 4. Graphs of changes in current when polishing parts No. 1-6

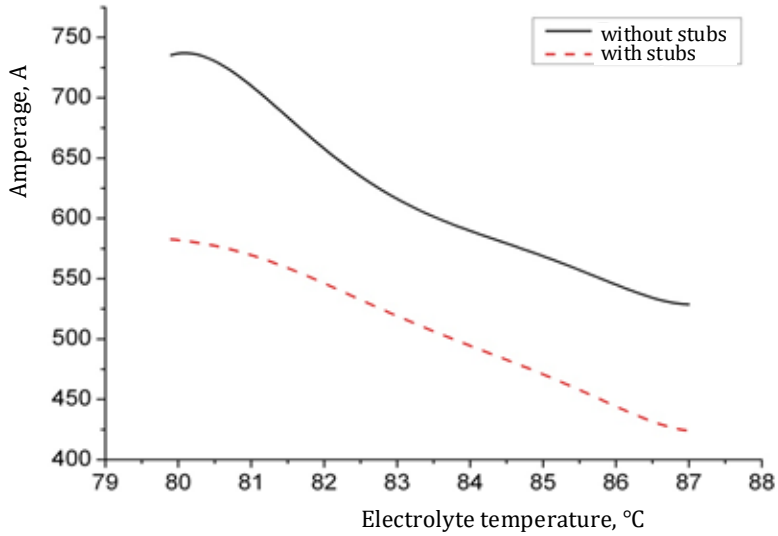


Fig. 5. The dependence of the current strength on the temperature of the electrolyte during polishing of the profile pipe

Fig. 3 shows graphs of changes in the temperature of the electrolyte during processing of parts (smoothed using a polynomial of degree 6). Parts 1, 2, 5 and 6 have a shape elongated along the horizontal axis and small height, during processing their lower edge is lowered into the electrolyte to a depth of no more than 15 cm from the surface, so the temperature of the lower layers of the electrolyte during polishing of these parts increases very slightly (figure 3b). Unlike parts 3 and 4, the lower edge of which falls to a depth of 30-40 cm from the surface, so that lower electrolyte layers are also heated.

Fig. 4 shows graphs of changes in current during the polishing of parts. The greatest current strength is observed when polishing pipes without plugs (parts 1 and 2). When processing these parts, powerful hydrodynamic flows are formed. In addition, the first few seconds after the part is lowered into the electrolyte, current surges occur (degree 6 polynomial is used in the construction of the graphs, so the

current surges are not displayed in the figure, but are smoothed out in the growth of the curves for the first ≈30 seconds). When polishing the grating (detail 4), jumps in the current strength are also noticeable at the beginning, however, they are much weaker and the process stabilizes much faster than with pipes. Also in the lattice cells there are weak hydrodynamic flows. It is best processed flat metal sheet (part 3). The process in this case occurs very stably, no current surges are observed even when the part is quickly lowered into the electrolyte.

After the pipe and the profile pipe were closed with plugs (parts 5 and 6), when polished, the current strength decreased by 17 – 20%, in addition, in this case there were no current surges at the beginning of polishing. Thus, powerful hydrodynamic flows arising in narrow open cavities significantly increase the consumption of electric power during polishing.

During the processing time (7.5 minutes), the current strength for all parts gradually decreased. At the end of the treatment, the current value decreased by 22 – 25% compared with the initial values. The voltage in the electric circuit is constant (~ 380 V) and does not change during the processing process (controlled by a standard voltmeter), which means that a decrease in the current value is associated with an increase in resistance. Figure 5 shows the dependence of the current strength on the temperature of the electrolyte for the profile pipe with plugs and without them. The nature of the curves corresponds to an inversely proportional dependence of the current strength on temperature. Thus, one can judge the increase in resistance in the cathode – electrolyte – VGS – anode system.

The main contribution to the decrease in power consumption is made by the increase in the resistance of the vapor-gas shell associated with its thickening with increasing temperature of the electrolyte, since it is in it that the main resistance of the system is concentrated, significantly exceeding the contribution of the resistances of the electrodes and electrolyte.

3. STUDY OF THE POSSIBILITY OF USING EPP FOR DECONTAMINATION OF REMOVABLE EQUIPMENT OF NUCLEAR POWER PLANTS

The operation of a nuclear power plant is accompanied by radioactive contamination of the internal and external surfaces of the equipment of the circuit, as well as the surfaces of the premises where it is located. The following types of pollution are distinguished: unfixed, weakly fixed and firmly fixed. The first is caused by an adhesive process and is characterized by the presence of an interface between the radioactive substance and the surface. The second is mainly caused by nuclide adsorption and ion exchange and is characterized by contamination of the surface layer of the equipment. The third is associated with corrosion processes, oxide film formation and diffusion and is characterized by contamination of the deeper layers of equipment construction materials. In practice, a combination of various types of radioactive contamination is possible.

The main methods for the decontamination of equipment and facilities at nuclear power plants are: chemical, chemical-mechanical, electrochemical, steam emulsion and hydrodynamic. The choice of this or that method is determined by the nature of the contaminants, the dimensions and configuration of the decontamination facility, the applied structural materials and other factors [14].

The electrochemical decontamination method is used to remove firmly fixed radioactive contaminants when the use of the chemical method is ineffective or technically difficult. Using this method, the decontaminated surface is subjected to electrochemical etching under the influence of direct current with a density of 10 – 20 A/dm2, after which water washing is carried out. The deactivated surface serves as the anode and the surface layer is removed from it along with radioactive contaminants. As an electrolyte, aqueous solutions of oxalic (H2C2O4) or phosphoric (H3PO4) acids with a concentration of 1.5 – 2.0% are used. The method under consideration is used to decontaminate parts and assemblies of pumps, drives, external and internal surfaces of pipelines, etc.

From 2015 to the present, the scientific institution “JINR – Sosny” has been conducting research on the possibility of using EPP to deactivate removable equipment of nuclear power plants as an alternative method of electrochemical processing [15, 16]. EPP is a more environmentally friendly way, since weak solutions of neutral salts are used as the electrolyte.

Experiments on the decontamination of metal surfaces by the EPP method (cleaning from fixed radioactive contamination, fixation by time) were carried out on a modified EPP-15 installation (Fig. 6). For research, a 3-liter working bath was made of steel 12X18H10T according to GOST 5632-72 in order to minimize liquid radioactive waste (LRW) generated during the EPP process. The control cabinet with the power unit of unit 1 was located in a clean room, and the working bath with a cooling jacket 3 and the lifting mechanism 4 were in a fume hood 2 of the room certified for work with open radiation sources of class II according to [17]. In the working bath 3 filled with electrolyte, the samples were processed. Raising and lowering the test samples was carried out in the automatic operation mode of the installation.

Samples for research: plates of size 40×30×5 made of steel 12X18H10T according to GOST 5632-72. Preparation of the bases for contamination and contamination of the samples was carried out according to GOST 50773. 0.15 cm3 of contaminating radioactive solution was applied to the center of each sample — a mixture of 137Cs and 60Co radionuclides with an activity of 20 kBq each. Contaminated samples were dried in air at a temperature of 20 ± 2 ℃ from 18 to 20 hours. Radioactive contamination was on the surfaces of the samples for 6 months.

Measurement of the radioactive contamination of the samples before and after the treatment was carried out using the γ- and β-analyzer MKS-AT-1315, the minimum detectable activity (MDA) of which is 10 Bq.

Experimental electrolyte: 6% (mass.) Aqueous solution of ammonium sulfate $(NH_4)_2SO_4$ according to GOST 9097-82 (analytical grade).

The cleaning process took place in 2 stages: stage 1 – decontamination of samples due to removal of the surface layer of metal (4 minutes); stage 2 (if necessary) – post-treatment of residual radioactive contamination to background activity values.

The results of the experiment are shown in table 4.

After the first stage of processing (4 min), half of the samples were completely cleaned of radioactive contamination: the level of contamination decreased by about 2585 times for 137Cs and 2445 times for 60Co. The second stage of processing was carried out to further purify the remaining samples to a level of contamination below the established standards [18]. The decontamination factor of the second stage ranged from 1 to 5.

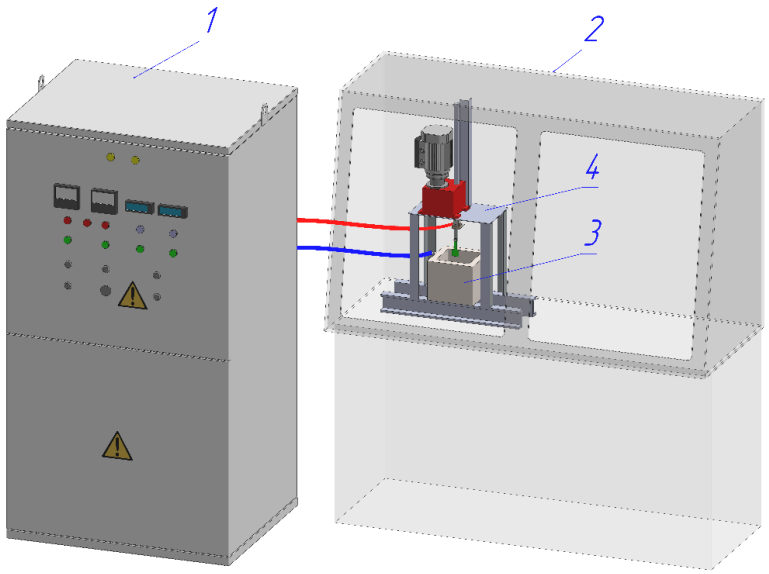


Fig. 6. General view of the decontamination unit (modified by EPP-15) by the EPP method:

1 – control cabinet with the power unit; 2 – fume hood; 3 – working bath with a cooling jacket; 4 – lifting gear

№	Initial activity, Bq		Activity after stage 1, Bq		Decont. factor of 1 stage		Activity after stage 2, Bq		Decont. factor of 2 stage		General decont. factor	
	^{137}Cs	^{60}Co	^{137}Cs	^{60}Co	^{137}Cs	^{60}Co	^{137}Cs	^{60}Co	^{137}Cs	^{60}Co	^{137}Cs	^{60}Co
1	37606± 1031	25586± 1038	<MDA	<MDA	3760	2558	–	–	–	–	3760	2558
2	35426± 1062	24979± 1073	25±10	<MDA	1417	2498	<MDA	–	2,5	–	3542	2498
3	36306± 875	25705± 964	53±11	56±39	685	459	<MDA	< MDA	5,3	5,6	3630	2570
4	36677± 818	25560± 1042	15±4	<MDA	2445	2556	<MDA	–	1,5	–	3667	2626
5	36381± 549	26866± 671	<MDA	<MDA	3638	2686	–	–	–	–	3638	2686
6	35631± 903	26110± 1030	<MDA	<MDA	3563	2611	–	–	–	–	3563	2611

Table 4. The results of the experiment on the decontamination of samples by EPP

Studies have shown that EPP is a promising way to decontaminate removable equipment of nuclear power plants. This method has high efficiency and speed of surface treatment, meets all the requirements for methods of decontamination of nuclear power plant equipment [14]:

1) provides effective removal of radioactive contamination;

2) does not cause significant corrosion and destruction of the deactivated material of the products;

3) the amount of radioactive waste is minimal;

4) the method of decontamination is economical, safe, does not lead to the spread of radioactive contamination, allows the possibility of complete automation of the process;

5) the processing time of the product does not exceed 5 minutes in one step.

As mentioned earlier, the geometry of the workpiece and its orientation in the working bath play a significant role in ensuring the polishing function [10]. If, when processing simple flat objects, difficulties usually do not arise, then when processing complex volumetric products, unpredictable circulation of the electrolyte is observed in the working bath with steam emissions through holes and nozzles. The consumed energy increases several times in comparison with the calculated relative to the cultivated area. Often this leads to a breakdown of the vapor-gas shell formed around the workpiece during polishing, and to a violation of the polishing process. Therefore, in order to develop the technological regulations of the EPP method for equipment decontamination and introduce the method into the technological process at nuclear power plants, it is necessary to create a tool that will predict the behavior of the electrolyte during processing, depending on the complexity of the configuration of the part and the method of its placement in the working bath. It is proposed to use physical and mathematical modeling of the EPP process as such a tool [10–12].

4. PHYSICS-MATHEMATICS MODELING OF THE EPI PROCESS

The system of electrolyte-plasma processing of materials is a complex physical object in which the phenomena of transfer of matter, electric charges and thermal energy are observed. The most important feature of transport processes in EPP is also the presence of singular surfaces (metal – liquid, liquid – gas, gas – solid), which serve as fracture surfaces for one or more quantities (density, velocity, etc.) and are generally moving.

Physically, the EPP system is a spatial system of electrical conductors of the first kind (anode and cathode), separated from each other by electrical conductors of the second kind (electrolyte). It is known that the charge carriers in the first kind conductors are electrons, while the second kind conductors have ionic conductivity [19, 20]. The contact of electrical conductors with different types of conductivities leads to the fact that a double electric layer arises at the interface between them, generating a jump in the electric potential on itself. It is these potential jumps that are the driving force of the electrode processes occurring in EPP systems.

With an open external circuit, the EPP system is in a state of thermodynamic equilibrium; when the external circuit is closed, all physical fields (electric, thermal, field of electrolyte concentrations) become dynamic and depend on the electric mode of the external circuit and time. The stationary structure of the double electric layer is disrupted, gradients of the electric and concentration fields appear in the electrodes and electrolyte, under the influence of which ions of different sorts come into motion. The movement of ions, causing Joule heat [21], directed in such a way as to eliminate the gradients that have arisen.

Thus, in the EPP system, there are three main physical fields: electric, thermal, and the field of electrolyte concentrations (concentration) —functionally interconnected in three ways:

- through the influence of one field on the physical parameters of another;

- through the influence of one field on the boundary conditions of another;

- through the creation by one field of sources for another.

It is convenient to trace the specific form of these relationships using the conceptual model shown in Fig. 7. The following fields are included in the model:

- electric field of electrolyte I;

- concentration field of electrolyte II;

- thermal field of electrolyte III;

- electric field of electrodes IV;

is the thermal field of the electrodes V.

A mathematical description of these fields is given by the well-known equations of mathematical physics, which in each specific problem must be supplemented by their initial and boundary conditions.

Initial conditions set the initial state of the object, and recording them is usually straightforward. When recording boundary conditions, it should be borne in mind that only boundary conditions on the external surface of the EPP system (external boundary conditions) can be explicitly set. These boundary conditions include particulars that are specified in the calculations of the indicators of the electric and thermal modes of the EPP system in the external circuit, the voltage at its terminals, the ambient temperature, etc. The boundary conditions at the interphase boundaries of the EPP system cannot be explicitly specified in the general case due to the mutual influence of the fields on each other.

The boundary condition 1 (see Fig. 7) for the electric field at the phase boundary is specified in the form of potential jumps at the electrode-electrolyte interfaces determined by the Nernst equations. Bilateral relations I – 1 and IV – 1 indicate that this boundary condition is implicitly specified. According to the Nernst equation, potential jumps depend on temperature and electrolyte concentration on the electrode surface. This dependence is reflected in the model by bonds II – 1 and III – 1. The boundary condition 3 for the concentration field is determined by the concentration gradient at the electrode – electrolyte interface. Through this boundary condition, the electric field affects the concentration field (bonds I – 3 and 3 – II). In addition, the concentration gradient of the electrolyte at the interface depends on the diffusion coefficient, which in turn is related to the concentration and temperature.

The most difficult boundary condition in EPP is the boundary condition for the thermal field at the electrode-electrolyte interface. This boundary condition 2 is determined by the thermal effect of the electrochemical reaction, which depends on the distribution of the electric field at the interface between

the solid and liquid phases. The implicit recording form of boundary condition 2 is reflected in the model by the double-sided bonds I – 2, III – 2, V – 2, and IV – 2.

The EPP system is a nonlinear object, since the physical parameters of the fields existing in it depend on the mode of its operation. So, the characteristic B of the concentration field – the diffusion coefficient – depends on the electrolyte concentration and temperature. In the model, this dependence is shown by II – B and III – B bonds. Since the diffusion coefficient itself affects the distribution of electrolyte concentration, the II – B bond is two-way.

Physical characteristics of the thermal field of electrolyte B - heat capacity, density, coefficient of thermal conductivity - also depend on temperature and electrolyte concentration. The II – B to III – B bonds reflect this dependence.

Physical fields in EPP electrodes are also nonlinear in nature. The electrical parameter of the electrodes - the electrical conductivity - significantly depends on the temperature and the operating mode of the EPP system. These dependences are shown in the model by IV – D and V – D bonds. Non-linearity of the thermal field of the electrodes associated with the dependence of their thermal characteristics on temperatures is shown by a two-way V – D bond.

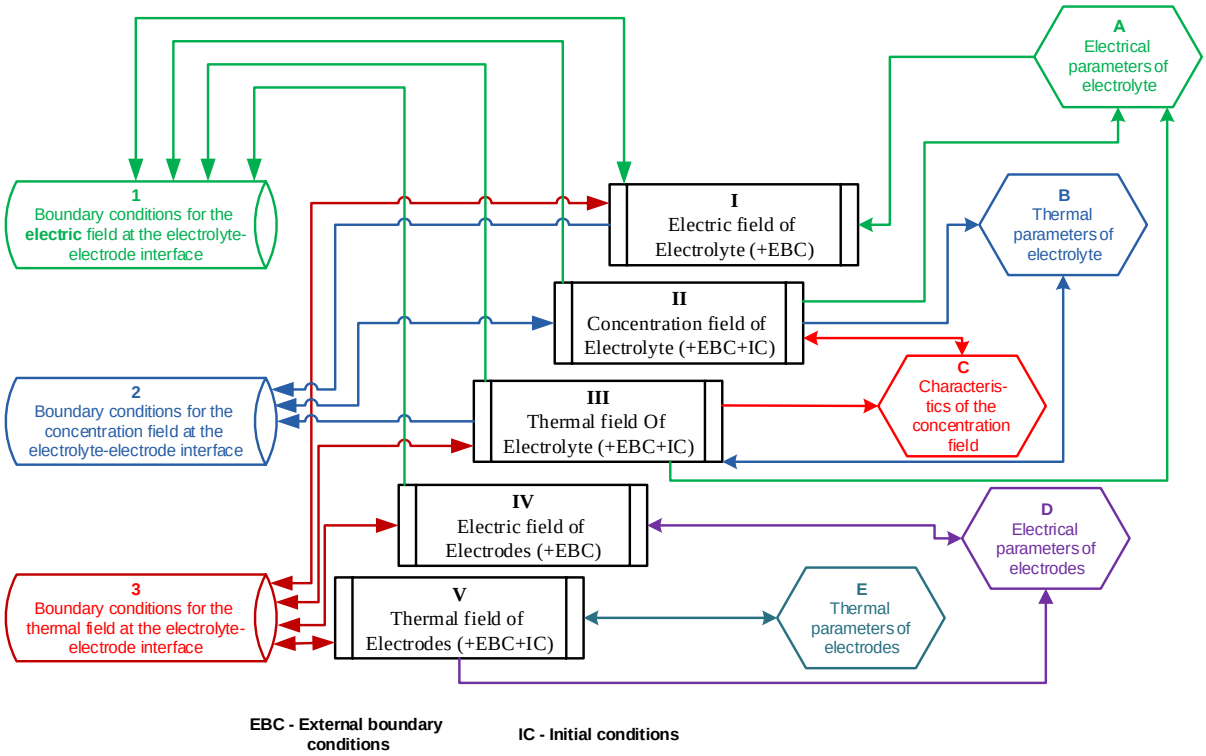


Fig. 7. EPP system model

Dependence of electrolyte conductivity on concentration and temperature (II – A, III – A, A – 1 bonds).

It is necessary to note the direct influence of one physical field on another. So the positive sources for the thermal field in the entire volume of the EPP system is the Joule heat determined by the electric field (the bonds I – III and IV – V are not shown in Scheme 7).

4.1 The system of equations of the conceptual model of EPP

1) The electric field of the electrolyte is described by the Gauss equation, when the dependence of the current density j on the field strength E is considered linear [22]:

$\varepsilon_0 div\ \bar{E} = -\rho_e,$
$\bar{j} = \mu\rho_e\bar{E},$
$div\ \bar{j} = 0,$ (1)

where μ – ion mobility, m2/(V·s); ρ_e – bulk charge density, C / m3; ε_0 – electric constant.

The threshold value of the field strength E_0 is determined from the condition of aperiodic Tonks – Frenkel instability [23]:

$E_0 = \sqrt[4]{64\pi^2 g\rho\gamma}$, (2)

where $g = 9.8$ m/s2 – acceleration of gravity; $\rho = 1.035 \cdot 10^3$ kg/m3 – density of a 6% solution of ammonium sulfate; $\gamma = 58.85$ mN / m - the coefficient of surface tension of the solution at 100 °C. In this case $E_0 = 2.39 \cdot 10^6$ В/м [22].

2) To describe the motion of an electrolyte due to a change in its density, we use the well-known Navier – Stokes equation in the Boussinesq approximation:

$\rho_0\left(\frac{\partial\vec{v}}{\partial t} + (\vec{v}\Delta)\vec{v}\right) = -\nabla p + \mu\nabla^2\vec{v} + \rho_0[\alpha(\mathrm{c} - \mathrm{c}_0) - \theta(T - T_0)]\vec{g}$, (3)

where p is the pressure; α and θ are the expansion coefficients due to a change in concentration and temperature, respectively, g свободного is the acceleration of gravity. c and c_0 – concentration and initial concentration of electrolyte.

$\nabla\ \bar{v} = 0$ – condition of incompressibility of a liquid electrolyte. (4)

3) The heat equation is as follows:

$c_p\rho\frac{\partial\ T}{\partial t} = -\nabla\vec{J}_\tau + \vec{J}_q\vec{E}$, (5)

where $\vec{J}_q = \sigma\left(\vec{E} - D^*\nabla\mathrm{c}\right)$, $\vec{J}_\tau = -k\nabla T$, c_p – heat capacity of electrolyte, ρ – electrolyte density, σ – electrolyte conductivity, D^* – specific electric ambipolar diffusion coefficient, c – electrolyte concentration.

4) The electrolyte concentration field is defined by the following equation:

$\frac{\partial \mathrm{c}_i}{\partial t} = D_i\nabla^2 c_i + U_i\frac{z_i}{|z_i|}grad\phi grad c_i$. (6)

where D_i – effective diffusion coefficient, ϕ – electrolyte potential, z_i – ion charge.

To simulate the EPP system, equations (1) – (6) are supplemented by initial and boundary conditions, and, if necessary, a model of the presence of a vapor – gas shell at the metal – electrolyte interface.

4.2 Boundary conditions of the conceptual model of EPP

Equations (1) – (6) are supplemented by the boundary conditions

- on the walls of the working bath (cathode),

- on the free surface of the electrolyte,

- on the surface of the workpiece (anode).

The boundary conditions are schematically shown in Fig. 9.

1) On the surface of the workpiece (anode) are set:

- for the equation of the electric field: electric potential (e.g. $\phi = \phi_A = 380(V)$);

- for the equation of motion of the electrolyte: the conditions of "sticking" for speeds on the entire surface of the part ($v = 0$);

- for the heat equation: on the entire surface of the anode, a distributed electrochemical heat flux Q_b (W/m^2), is defined, the distribution diagram of which is the result of solving the equation for the electric field (1) (thus, the relationship between equations (1) and (3) is realized);

- For the equation of the field of electrolyte concentration, the boundary condition for the jump in the concentration flux at the metal – electrolyte boundary is set:

$$-n\left[\left(J_{\text{конв},i}+vc_i\right)_{\text{э}}-\left(J_{\text{конв},i}+vc_i\right)_{\text{м}}\right]=N_{0,i},$$

$$J_{\text{конв},i}=-D_i\nabla^2c_i-z_iU_iFc_i\nabla\phi,$$

where $N_{0,i}$ (mol/(m2·s)) – concentration jump.

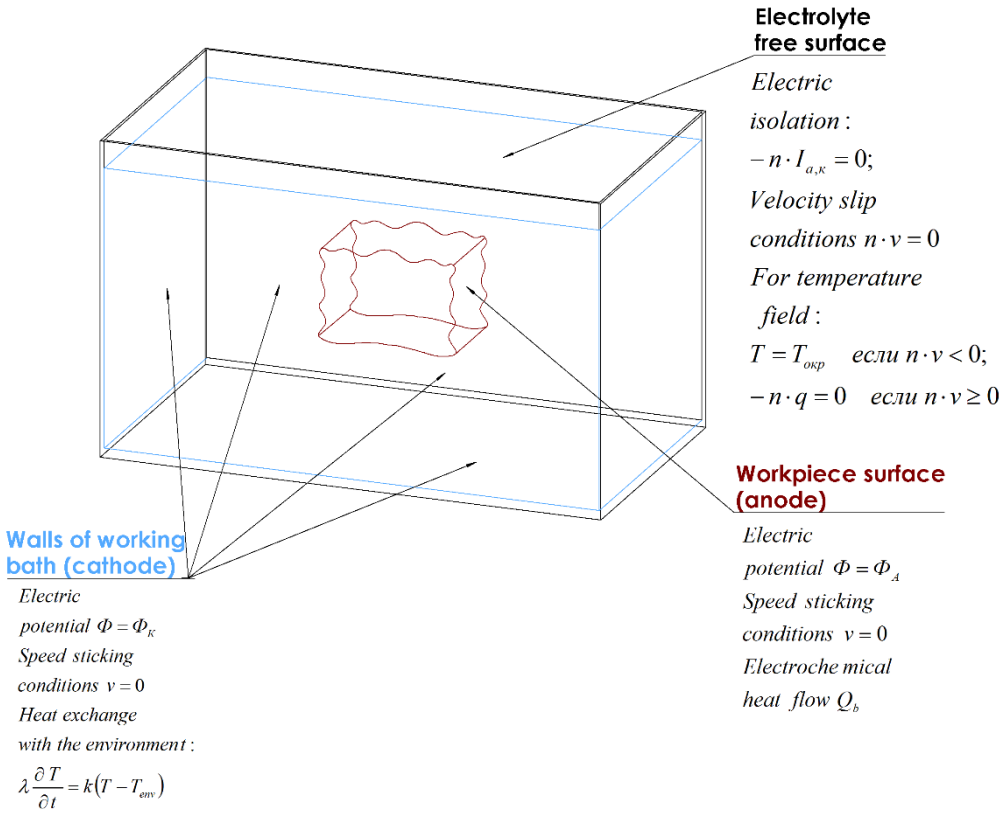


Fig. 8. The boundary conditions of the mathematical model

2) On the walls of the working bath (anode) are set:

- for the equation of the electric field: electric potential (e.g., $\phi=0\ (V)$);

- for the equation of motion of an electrolyte: “sticking” conditions for speeds on the entire surface of a part ($v=0$);

- for the heat equation: heat transfer with the environment

$$\lambda\frac{\partial T}{\partial t}=k(T-T_0),$$

where k – heat transfer coefficient to the room and T_0 – ambient temperature;

- for the field of electrolyte concentration, the conditions for the absence of flow: $-n(-D\nabla c)=0$.

3) On the free surface of the electrolyte:

- for the equation of the electric field: conditions of electrical "isolation" of the surface

$-n \cdot I_a = 0;$
$-n \cdot I_k = 0.$

- for the equation of motion of an electrolyte: conditions of "slipping" of speeds ($n \cdot v = 0$);

- for the heat equation:

$T = T_0$ if $n \cdot v < 0;$
$-n \cdot q = 0$ if $n \cdot v \geq 0.$

- for the field of electrolyte concentration:

$-n \cdot J_{\text{конв}} = 0$ if $n \cdot v \geq 0;$
$\mathrm{c} = \mathrm{c}_0$ if $n \cdot v < 0.$

4.3 Computer implementation of the conceptual model of EPP

The proposed model was implemented using the COMSOL Multiphysics 5.5 software package. COMSOL Multiphysics® is an integrated modeling platform that includes all its stages: from creating geometry, determining the properties of materials and describing physical phenomena, to customizing the solution and the post-processing process [24].

COMSOL Multiphysics has a modular structure. In relation to our formulation of the problem, the following modules are used:

1) To model the electric field in the electrolyte, the Electrochemistry module (Secondary current distribution model) is used. This module allows you to implement the system of Gaussian equations (1), (2) for modeling the electric field in an electrolyte, taking into account the presence of models of a double electric layer at the metal-electrolyte interface.

2) For thermal and hydrodynamic fields – the module "Fluid Flow" ("Fluid Flow", submodule "Nonisothermal Flow"), which includes the models "Laminar Flow" and "Heat Transfer in Liquids" in Fluids "). In the indicated modules, the systems of equations (3) – (5) are implemented with direct connection with the electric field of the electrolyte.

3) The field of electrolyte concentration is modeled using the module "Flows in diluted solutions" ("Transport of Diluted Species"). This module allows you to implement the electrolyte concentration equation (6) taking into account the electric field of the electrolyte and the presence of a double electric layer: the module has the ability to choose various options for the jump in the concentration flux at the metal-electrolyte interface.

Using the proposed physical and mathematical model, we will calculate the distribution of electric, thermal and hydrodynamic fields in the electrolyte for various forms of processed products. To compare the results with experimental data as polished products, we consider the details described in paragraph 2 of this article (see table 3): a profile pipe without plugs, with plugs and a sheet. The calculation area for the product "Profile pipe (without plugs)" is shown in Fig. 9 (for other products, the calculation areas are similar).

The calculation results are shown in fig. 10 – 12. A comparison of the obtained calculation results with experimental data is shown in table 5 and on Fig. 13 – 15. The calculation results are in good agreement with experiments performed on EPP installations: the difference in absolute values of current density and electrolyte temperature is no more than 3%.

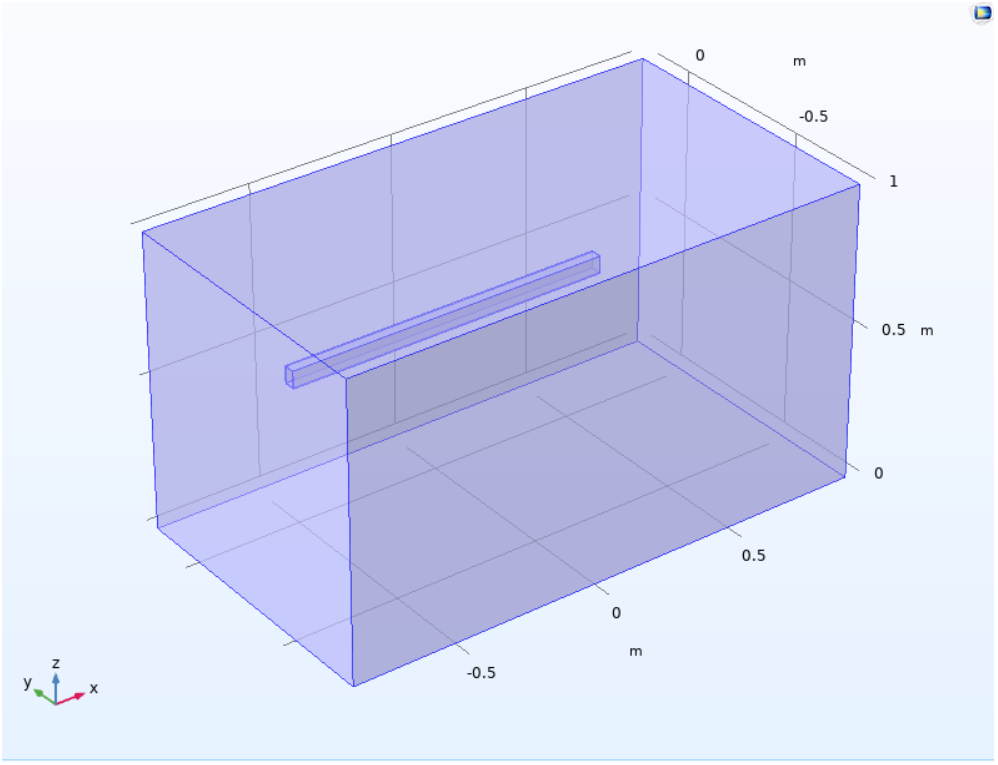


Fig. 9. Calculation area

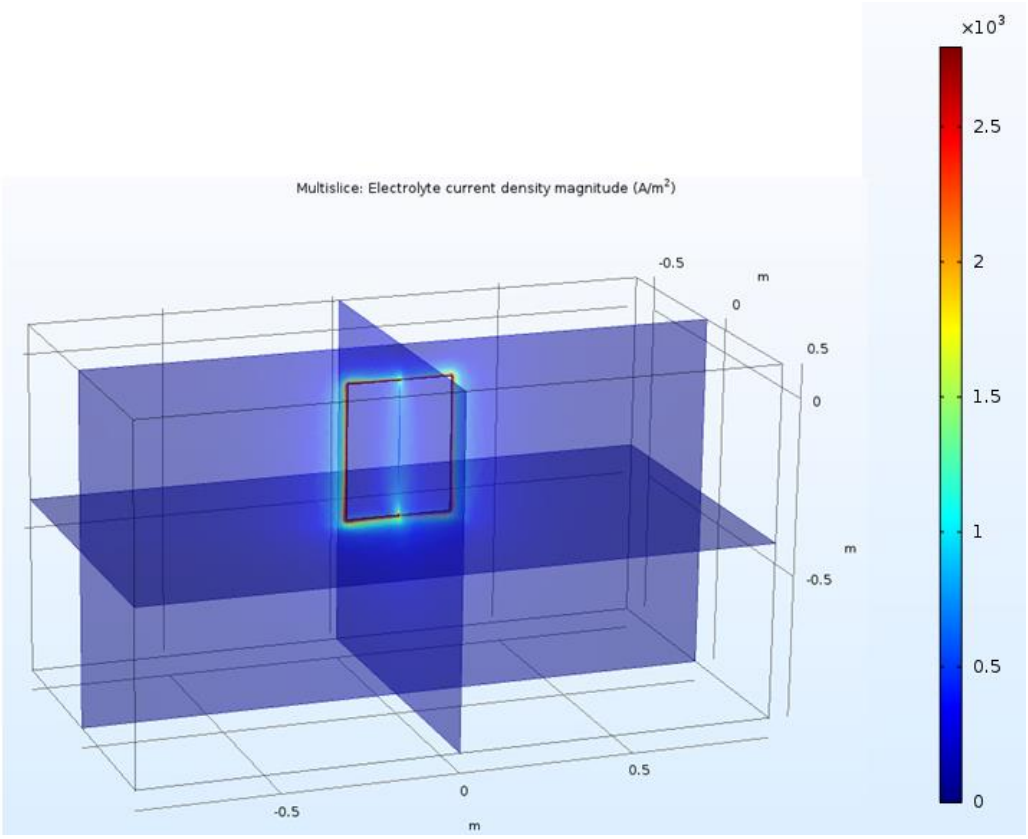


Fig. 10. Distribution of current density during processing of the product "Sheet"

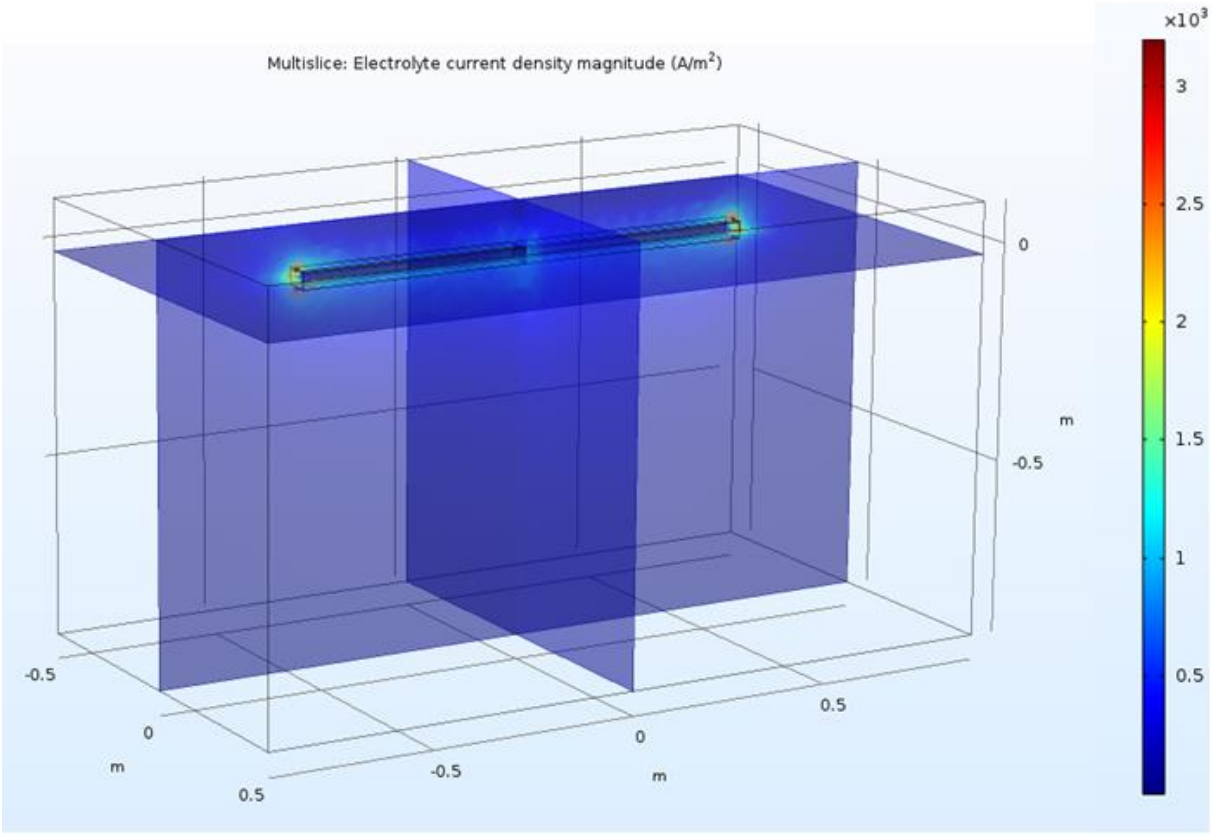


Fig. 11. Distribution of current density during processing of the product «Profile tube»

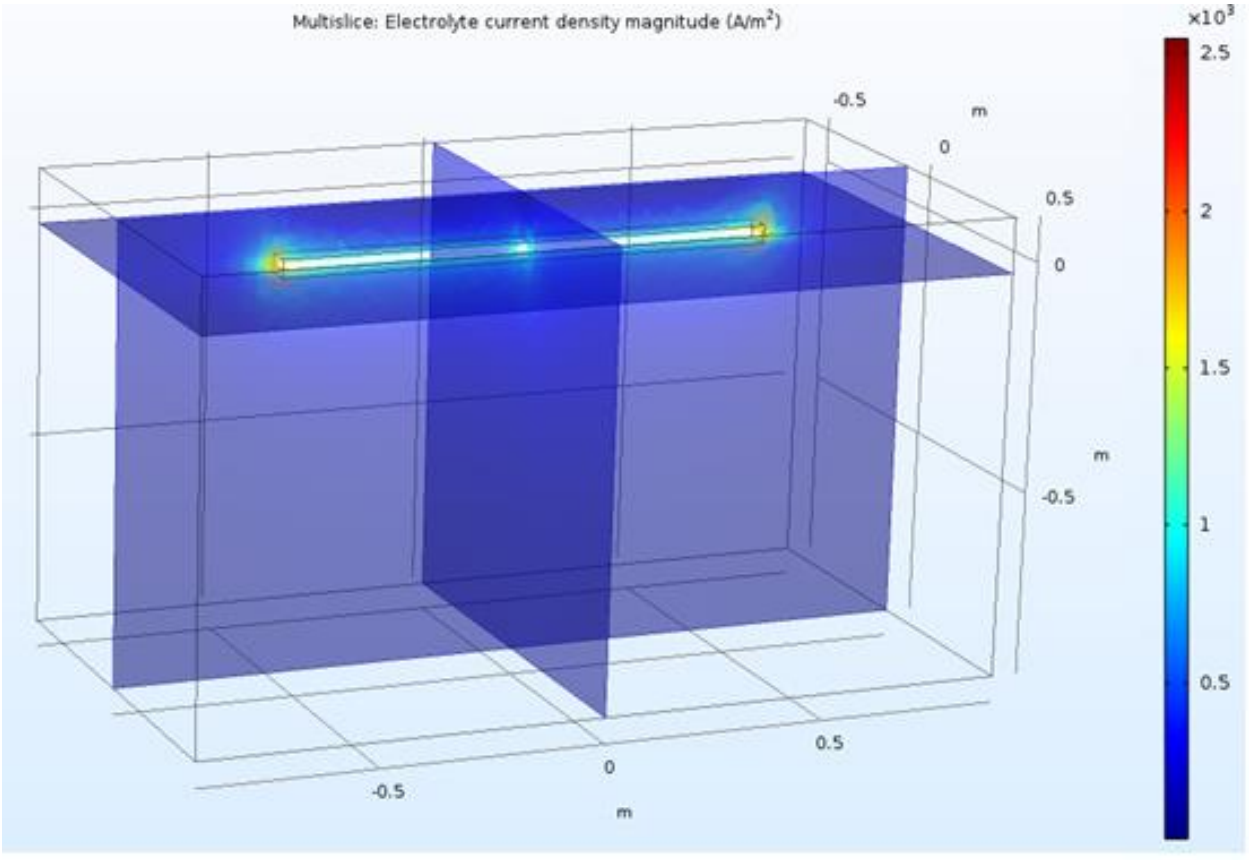


Fig. 12. Distribution of current density during processing of the product “Profile pipe (with plugs)”

Polished product	Maximum current density, A/m^2		Difference
	Experiment	Calculation	
Sheet	2727	2694	1,2%
Profile tube	3174	3221	1,4%
Profile tube (with plugs)	2522	2594	2,9%

Table 5. Comparison of calculated and experimental values of current density during EPP

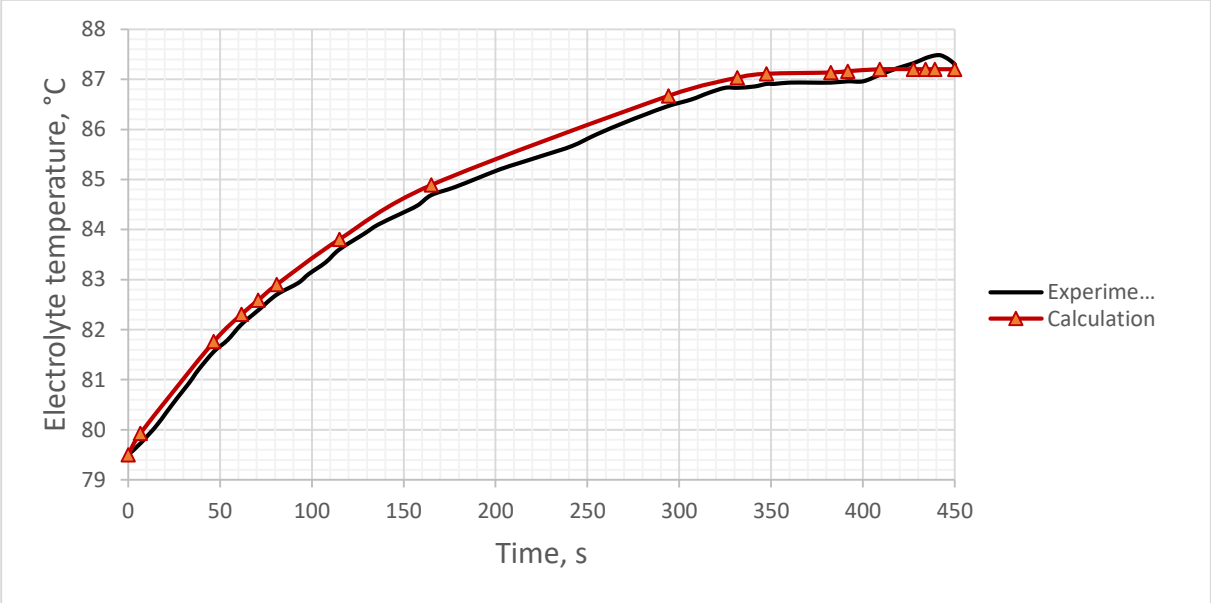


Fig. 13. Comparison of calculation results and experimental data on the temperature of the electrolyte during processing of the product "Profile pipe"

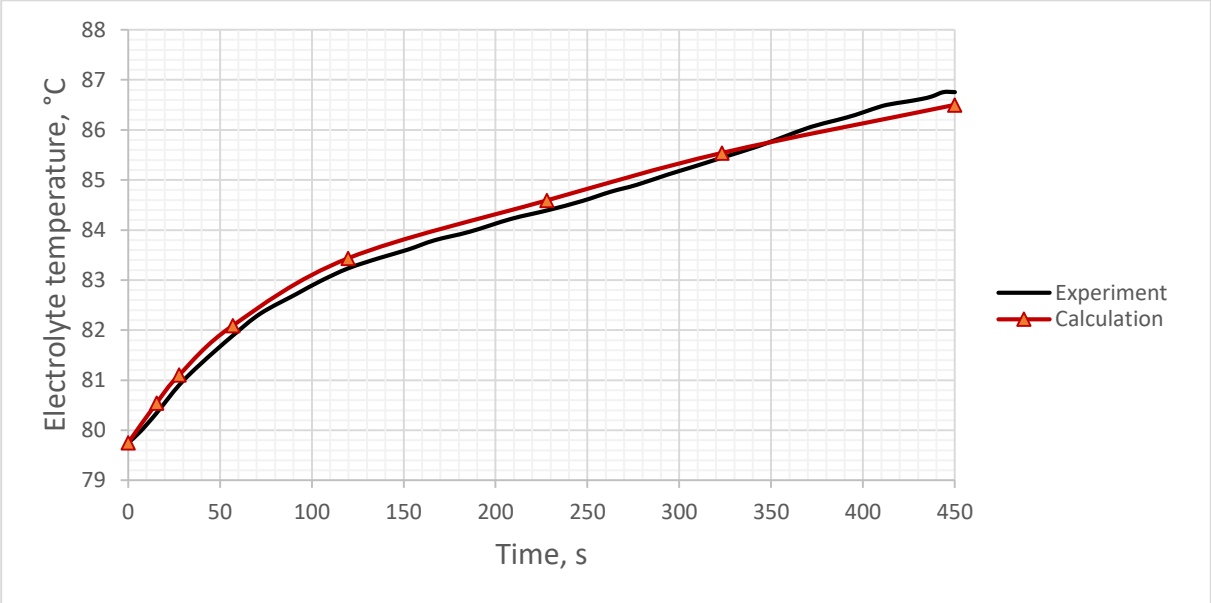


Fig. 14. Comparison of calculation results and experimental data on the temperature of the electrolyte during processing of the product “Sheet”

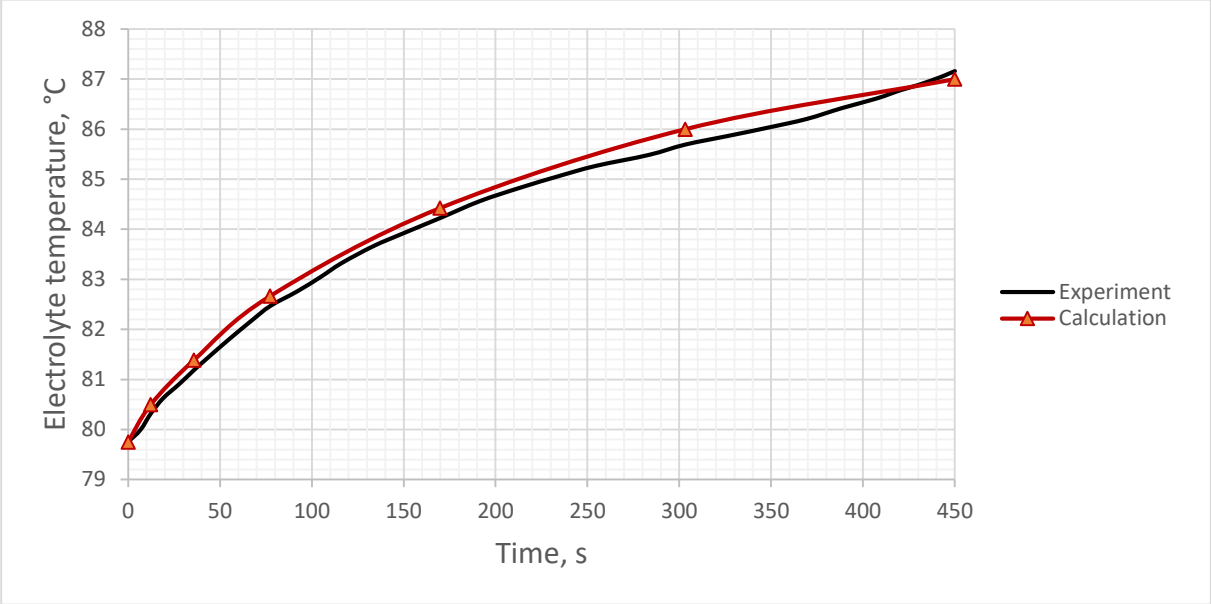


Fig. 15. Comparison of calculation results and experimental data on the temperature of the electrolyte during processing of the product "Profile pipe (with plugs)"

CONCLUSIONS

This work is devoted to the study of technology and physical and mathematical modeling of transport phenomena in electrolytes during the decontamination of metal surfaces by the method of electrolyte-plasma treatment.

The main results of the work:

1. Studies have been conducted on the influence of the complexity of the shape of the processed products on the EPP process. The results of the experiments show that the hydrodynamic flows arising in the narrow open cavities of the processed products significantly (by 17 – 20%) increase the consumption of electrical energy during polishing.
2. Studies have been carried out on the possibility of using EPP to deactivate removable equipment of nuclear power plants. It is experimentally proved that EPP is a promising way to decontaminate removable equipment of nuclear power plants. This method has high efficiency and speed of surface treatment, meets all the requirements for methods of decontamination of nuclear power plant equipment.
3. A conceptual physical and mathematical model is proposed, which allows one to calculate the electric, thermal, hydrodynamic, and concentration parameters of an electrolyte during EPP of metal products of complex geometric shapes. The basic system of equations of the physical and mathematical model is presented, the boundary conditions of the model on the surfaces of the workpiece, the walls of the working bath and the free surface of the electrolyte are proposed. A method for computer implementation of a physical and mathematical model using the COMSOL Multiphysics software package is proposed. The proposed physical and mathematical model and the method of its computer implementation make it possible to obtain realistic output data that are in good agreement with practical experiments conducted on EPP installations.

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