TECHNOLOGICAL REGULATIONS OF APPLICATION OF ELECTROLYTE-PLASMA TREATMENT FOR SURFACES DECONTAMINATION OF NPP REMOVABLE EQUIPMENT

Nagula Petr

Head of the laboratory of the State Scientific Institution «The Joint Institute for power and nuclear research – Sosny» of National Academy of Sciences of Belarus, Minsk, the Republic of Belarus

Abstract

The paper is devoted to the development of technological regulations for the surfaces decontamination of NPP removable equipment using the EPT method. The technological regulations are a document that completes research on the development of a new technology for the production of products, in our case, the technology of surfaces decontamination of metal products. The technological regulations is the main document establishing the mode, technical means, procedure and norms for carrying out technological operations, safe operating conditions, environmental protection and fire safety requirements. The paper considers the key points of the technological regulations: a decontamination unit, a technological scheme, the disposal of radioactive waste, and the use of purified products.

Keywords: NPP, electrolyte-plasma treatment, technological regulation.

1. INTRODUCTION

The use of physicochemical processes in electrolyte solutions at high voltages of electric current is one of the common and effective methods for treating complex profiled surfaces of critical parts, products made of various metal materials and conductive alloys (stainless and heat-resistant steels, soft non-ferrous alloys, difficult to process titanium alloys, etc.). In this way, electrolyte-plasma treatment (EPT) is based on electric-discharge phenomena in the metal-electrolyte system. [1-4, 10-15].

The paper is devoted to the development of technological regulations for the surfaces decontamination of NPP removable equipment using the EPT method. The technological regulations are a document that completes research on the development of a new technology for the production of products, in our case, the technology of surfaces decontamination of metal products. The technological regulations is the main document establishing the mode, technical means, procedure and norms for carrying out technological operations, safe operating conditions, environmental protection and fire safety requirements. The paper considers the key points of the technological regulations: a decontamination unit, a technological scheme, the disposal of radioactive waste, and the use of purified products.

2. TECHNOLOGICAL SCHEME FOR THE DECONTAMINATION OF REMOVABLE EQUIPMENT OF NPP

According to the results of previous researches [1-7] (theoretical, experimental and practical on the EPT installation), the technological scheme for decontamination by the EPT method has undergone a number of fundamental changes (Figure 1).

Given the peculiarities of working with radioactive substances, pollutants of NPP removable equipment, the entire decontamination zone is divided into two part: «clean» and «dirty».

An electric power unit, control and monitoring panels, and a fresh electrolyte processing system are located in the «clean» zone.

The electric power unit is designed to: convert a three-phase ac voltage into dc voltage with a value of 360 V; to provide energy to electric motors of agitator, a system for moving products to / from a bath, pump drives, valves and other mechanisms.
The control panel is designed for automated process control and monitoring of operating parameters.

![Diagram of the installation for decontamination of NPP removable equipment using EPT](image)

**Fig. 1.** Structural diagram of the installation for decontamination of NPP removable equipment using EPT

Given the peculiarities of working with radioactive substances, pollutants of NPP removable equipment, the entire decontamination zone is divided into two part: «clean» and «dirty».

An electric power unit, control and monitoring panels, and a fresh electrolyte processing system are located in the «clean» zone.

The electric power unit is designed to: convert a three-phase ac voltage into dc voltage with a value of 360 V; to provide energy to electric motors of agitator, a system for moving products to / from a bath, pump drives, valves and other mechanisms.

The control panel is designed for automated process control and monitoring of operating parameters.

The fresh electrolyte processing system is intended for electrolyte processing and is a cylindrical reservoir with a jacket for heating the electrolyte to operating temperature and feeding it into a working bath. The system includes an agitator with an electric drive, a dosing system for water and chemicals, a pump and valves. The working volume of the reservoir for the preparation of electrolyte corresponds to the volume of the working bath.

A system for preparing products before processing, a system for electroplasma processing of products, a system for evaporating and disposing of radioactive undissolved wastes are located in the «dirty» zone.

A list of equipment, that will be processed, is formed for the product preparation system before processing in advance at the NPP design stage. Places of optimal fastening are determined through mathematical modeling, and equipment is being prepared. In the case of complex products, full-profile models are manufactured and tested without the presence of radioactive substances.

The product fastened on a snap is fixed in the upper position of the working bath (it is shown conventionally by dashed lines). The working bath is sealed by the main connector. Then cold water is supplied for cooling and voltage is applied to the anode. The product is lowered into the bath using the...
lowering lifting system until product is completely immersed and the set time is maintained. The processing time does not exceed 4-5 minutes.

After completion of the process, the product is automatically lifted from the solution, the voltage at the anode is turned off, – the product is washed with clean water and sent as directed.

When the radioactivity reaches a critical level in the working bath [8, 9], the use of the electrolyte and processing of the products are terminated, the radioactive electrolyte is pumped into the evaporation, concentration and disposal system.

![Image](image_url)

**Fig. 2.** Vacuum equipment after electrolyte-plasma treatment

Reagents for binding radioactive isotopes into insoluble compounds are fed into the system.

In the vessel radioactive substances are heated to a temperature of 95 ... 98 ºС (without boiling). Water vapor is drained and cooled for reuse. The concentrate (pulp) is sent by a special pump for disposal and cementing to individual metal barrels.

Metal cleaning to a safe level of residual contamination allows us to send units and parts for repair work after processing, and for remelting and reuse at the stage of NPP decommissioning.

Figures 2, 3, 4 show examples of EPT processing of various stainless steel products at the EIP-800 pilot production unit (800 - power with kW). The gained experience in the installation allows us to confidently confirm the prospects of the chosen direction.
3. PURIFICATION AND CONDITIONING OF LIQUID RADIOACTIVE WASTE GENERATED DURING THE DECONTAMINATION OF METAL SURFACES BY EPT METHOD

One of the main contaminants of the equipment of the first circuit of NPP with VVER-type reactors (water-cooled power reactor) are 137Cs, 85Sr и 60Co radionuclides. In addition to radioactive elements, the coolant contains activated corrosion products (mainly radionuclides of iron, chromium, nickel, manganese, plumbum, etc.), as well as components included in decontamination solutions (acids,
alkalis, complex-forming or surface-active substances, etc.). The study of the forms of radionuclides in solutions of various compositions is the basis for the development of new technological solutions for the disposal of radionuclides. In solutions, depending on the concentration of \( H^+ \)-ions (pH) and radionuclide concentrations (metal ion), for example Fe (III), various complexes can be implemented [16–18]:

\[
Fe^{3+} + H_2O \leftrightarrow Fe(OH)_n^{3-n} \rightarrow Fe_{p}(OH)_{q}^{3+} \rightarrow \text{colloids} \rightarrow \text{precipitate}
\]

monospheric hydro complexes
 polyspheric hydro complexes
 pseudo-colloids Fe(OH)\(_3\)

The properties possessed by all these forms (sorption, the ability to penetrate through a membrane designed for ions, the ability to precipitate during centrifugation, and participate as catalysts) depend precisely on the form of the radionuclide (metal ion) in solution.

The most effective is the method of selective coprecipitation of 137Cs, 85Sr and 60Co radionuclides with heavy metal ferrocyanides – Fe (III) and Cu (II). For one cleaning cycle, the concentration of each of the radionuclides 137Cs and 60Co in solution intensified 250 times, 85Sr radionuclides by 25%. Thus, the activity of the purified solution is determined mainly by the content of strontium radionuclide.

The study of the effectiveness of EPT for the decontamination of metal surfaces was carried out according to GOST 50773 on samples-simulators made of 12X18H10T steel. A series of experiments was conducted on the decontamination of samples with fixed pollution (time fixation) of geometric shapes both simple (rectangular) and complex (cylindrical). 137Cs and 60Co radionuclides with activity of 2·104 Bq were used as a contaminating radioactive solution.

To practise cleaning methods the following solutions were used: 1 – liquid radioactive waste of EPT decontamination (LRW EPT); 2 – solutions-simulators of LRW (simulators of LRW), that were prepared by adding radionuclides to the spent non-radioactive electrolyte solution. The element composition of the spent electrolyte was determined on «ACTIVAM» inductively coupled plasma atomic emission spectrometer, method error is 3–5 rel. %. The electrolyte contains ions of Fe, Ni, Ca, Cr, Mg, Mn, Na, Zn, K, Sn, Cu, Co, Zn, A1, Cd metals in a concentration of 145 to 0,02 mg/l, and Si (11 mg/l) and B (0,5 mg/l).

LRW EPT was sedimented for 15 days, filtered through «blue ribbon» paper filter. LRW simulators were prepared as follows: a radioactive label of 137Cs, 85Sr and 60Co carrier-free isotopes was introduced into the spent electrolyte solution, the solution was sedimented for 30 days, filtered through «blue ribbon» paper filter. 85Sr radionuclide was used as analogue of 90Sr radionuclide.

Measurement of activity was carried out on a MKC AT-1315 gamma-beta spectrometer with a scintillation detector. The normalized limit of permissible basic measurement error is ± 20%.

The degree of purification (S) for each radionuclide was calculated by the formula:

\[
S = \frac{A_0 - A_1}{A_0} \times 100\%
\]

where \( A_0 \) – initial activity of the solution, Bq, \( A_1 \) – solution activity after cleaning, Bq/kg.

It should be noted that radionuclide 85Sr, as \( \beta \)-radiant, only slightly affects external radiation dose for personnel. Internal irradiation resulting primarily from radioactive aerosols inhalation will make dominating contribution into the whole dose.

One of the ways to convert strontium ions into the insoluble form is coprecipitation with hardly soluble calcium phosphate residue with solubility product of (washing solution or Ksp) 2·10^{-29}.

The following experiments were made:

method 1 –radionuclides 137Cs, 85Sr and 60Co coprecipitation with ferrum ferrocyanide (II) and calcium phosphate with their simultaneous presence in the solution;
method 2 — consecutive radionuclides coprecipitation with ferrum ferrocyanide (II) and, after ferrum ferrocyanide (II) residue detachment, radionuclides coprecipitation with calcium phosphate.

The results obtained show that during simultaneous radionuclides coprecipitation with ferrum ferrocyanide (II) and calcium phosphate (method 1) after two consecutive steps, degree of purification from strontium radionuclides made up 73 %. At the same time, cobalt and cesium radionuclides content in the solution was lower than the minimal detectable specific activity levels (20 Bq/l).

During consecutive radionuclides coprecipitation first with ferrum ferrocyanide (II) and then with calcium phosphate (method 2), after one cycle of purification (one step of ferrum ferrocyanide (II) coprecipitation and one step of calcium phosphate coprecipitation), degree of purification of the solutions electrolyte-plasma treatment (EPT) from strontium radionuclides made up 65 %. At the same time, cobalt and cesium radionuclides content in the solution was less than 20 Bq/l. Degree of purification from cesium radionuclides made up 99,92 %. However, I37Cs volumetric activity in the purified solution exceeded its 10-time benchmark level content in the drinkable water.

The obtained data thus show that cesium and cobalt radionuclides are effectively detached from the solutions imitating LRW EPT, both by the method of double coprecipitation with ferrum ferrocyanide (II), and by the method of double coprecipitation with ferrum ferrocyanide (II) and calcium phosphate with their simultaneous presence in the solution.

For strontium radionuclides, maximum purification degree made up 73% after simultaneous double radionuclides coprecipitation with ferrum ferrocyanide (II) and calcium phosphate.

The second research set included testing of the method of Sr coprecipitation with barium sulfate (II) residue from the solution, previously purified from 137Cs and 60Co radionuclides. During Sr radionuclides coprecipitation with barium sulfate (II), LRW simulators purification meets the necessary discharge into the environment level.

Work with the samples of various degrees of isotopes adhesion on the surface showed that EPO method might be used for decontamination and cleaning of the surface of the NPP equipment, made of stainless steels, from hard-set residue and scale, with polishing effect.

For LRW purification and decontamination by EPO method, one might effectively use the method of consecutive cesium and cobalt radionuclides coprecipitation with ferrum ferrocyanide (II) and then strontium radionuclides coprecipitation with barium sulfate (II). During this method, level of LRW purification from radionuclides makes up 99,98±0,02%; LRW volume reduction factor makes up ~20. The purified electrolyte might be reused after correction, or, dumped into drain system after purification from heavy metals and anions to the discharge into the environment level.

The results obtained might be used at technologies development for equipment decontamination at NPPs and other nuclear facilities, as well as for ensuing purification of the liquid radioactive waste resulting from decontamination.

4. THEORETICAL PART

Modeling of the processes taking place in the electrolyte at electric discharge treatment of the shaped surfaces is based on the differential equation system [5–7]:

– Navier-Stokes equation in Boussinesq approximation;
– Poisson’s electric field intensity equation;
– Nernst-Plank diffusion equation;
– heat conduction equation;
– equation of exchange.
Physical and chemical model was proposed to enable setting of nonsteady concentrations of electrolyte density, temperature and electric field in electrolyte and on the surface of the detail in process with the account of exchange.

Systems of equations with boundary conditions were solved by numerical methods using the software complex COMSOL Multiphysics.

Mathematical model enables definition of details (anode) power location, details placement in the electrolyte volume, treatment modes (time, temperature, electrolyte concentrations, etc.) at the stage of treatment processes preparation.

5. CONCLUSIONS

Research of many years undertaken at the scientific institution «JIPNR – Sosny» of the National Academy of Sciences of Belarus has shown that the method of electrolyte-plasma treatment of surfaces of metal objects is an effective instrument for decontamination and cleaning of NPP removable equipment, made of stainless and carbon steel, from hard-set radiation contamination. This method is highly effective, quick to decontaminate NPP equipment and assembly, and cost-effective.

The paper introduces the key sections of process procedures, schematic plan for decontamination, including the whole process from contaminated equipment delivery, its decontamination and radioactive waste management. NPP equipment and assembly decontamination to the safe level enables their use for maintenance work, and at the stage of NPP withdrawal – for remelting.

REFERENCES


