

Decrease in the activity of irradiated graphite and liquid radioactive waste

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Abstract.- Nowadays, the problem of nuclear waste deactivation is very urgent. The method proposed consists of exposing the aqueous solution of radionuclide to powerful nanosecond electromagnetic pulses by placing the treated solution between two electrodes connected to the outlets of the generator. Single-pole current pulses with a length of 1 ns, an amplitude of more than 5 kV, and a repetition frequency of 1 kHz are used. Such pulses can be obtained from the generator units with an amplitude of 5 kV to 15 kV. They have a pulse capacity of 1 to 4 MW, and the mains capacity is below 50 W. The pulse action leads to radiolysis of water and accelerated decay of radionuclides. As a result of radiolysis, hydrated electrons, hydrogen atoms, and various radicals are formed that causes precipitation of radionuclides, such as ^{137}Cs and ^{90}Sr . The description of several experiments and results of experimental investigations of the influence of powerful nanosecond electromagnetic pulses on the properties of water solutions with radioactive nuclides ^{137}Cs and ^{90}Sr and on items of irradiated graphite is presented. Under the influence of pulses on real liquid waste and aqueous solutions, radioactive nuclides in water are reduced by 5 to 50 times during 15 minutes. The effect described is preserved for 20 days. The reduction of beta activity twice during 25 min is observed in big blocks of the irradiated graphite. Flow-through unit for processing of liquid waste is developed, tested, and patented (RU 2726145. MPK G21F 9/28, July 7, 2020). This technology can be used to treat emergency solutions with tritium at Fukushima NPP. of the irradiated graphite.

Keywords: nanosecond electromagnetic pulses; graphite; radiation; radioactive liquid.

Disminución de la actividad del grafito irradiado y los residuos radioactivos líquidos

Resumen.- Actualmente, el problema de la desactivación de residuos radioactivos es muy urgente. El método propuesto consiste en exponer la solución acuosa de radionúclido a potentes pulsos electromagnéticos de nanosegundos colocando la solución tratada entre dos electrodos conectados a las terminales del generador. Se aplican pulsos de corriente de polo único con una longitud de 1 ns, amplitud de más de 5 kV y frecuencia de repetición de 1 kHz. Dichos pulsos se pueden obtener de las unidades generadoras con una amplitud de 5 a 15 kV. Tienen una capacidad de pulso de 1 a 4 MW y la capacidad de la red debajo de 50 W. Los pulsos causan la radiólisis del agua y la descomposición rápida de radionúclidos. La radiólisis, causa la generación de electrones hidratados, átomos de hidrógeno y varios radicales que provocan la precipitación de radionúclidos, como ^{137}Cs y ^{90}Sr . Se describen las pruebas y se presentan los resultados experimentales de la influencia de fuertes pulsos electromagnéticos de nanosegundos en las propiedades de las soluciones acuosas con los núclidos radioactivos de ^{137}Cs y ^{90}Sr y en piezas de grafito irradiado. La influencia por 15 minutos de los pulsos en los residuos líquidos reales y soluciones acuosas reduce el tamaño de los núclidos en el agua en 5 a 50 veces. El efecto descrito permanece por veinte días. Se observa el doble de la disminución de actividad beta por 25 minutos en trozos grandes de grafito irradiado. Se desarrolla, ensaya y patenta la unidad de flujo (RU 2726145. MPK G21F 9/28, 2020). Se puede usar esta tecnología para tratar soluciones de emergencia con tritio en la central nuclear de Fukushima.

Palabras clave: pulsos electromagnéticos de nanosegundos; grafito; radiación; líquido radioactivo.

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1. Introduction

Nowadays, a promising trend in nuclear waste deactivation is the use of powerful energy depositions. Urutskoev *et al.* [1] is worth highlighting as significant work on the subject. The

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influence of high pulse current upon the electric explosion of titanium foil in distilled water is described: a capacitor bank with charging voltage $U = 4,8$ kV and energy $W = 50$ kJ is discharged on a sheet of titanium foil. The foil is placed in a polyethylene explosion chamber shaped like a torus with openings evenly bored circumferentially, where 30 to 50 mL of liquid was poured. An increase followed the explosion in the content of Mn, Fe, Ni, Cu, Zn in the samples; plus, such foreign elements were found in the samples as Na, Al, K, Si, Ca, Cl, Mo, Ag, In, Sn, Sb.

The absolute number of atoms of the elements generated in the samples after the electric explosions ranged from 10^{15} to 10^{18} .

The check tests conducted at the Joint Institute for Nuclear Research (Dubna) Kuznetsov *et al.*, [2] confirmed the main test findings.

The electric explosion of titanium foil in uranyl sulfate solution distorts the initial isotopic ratio of U. The maximal distortion of isotopic composition is observed in a vapor gas release in favor of enriching ^{235}U .

Nigmatulin *et al.* [3] describes the joint action of ultrasound oscillations and a powerful flow of neutrons on the acetone from deuterium. The test results can be rendered as the conversion of deuterium to tritium under the action of cavitation.

A. F. Kladoy [4] proposes a method of making radioactive nuclides less active by mechanical activation. The required oscillation intensity must exceed 1 MW/m^2 . The laboratory setup consisted of a rotary hydrodynamic generator of sound and was powered by a 15 kW motor. The setup's oscillation intensity is $5,9 \times 10^3$ Hz, its operational pressure is 1 MPa, and the active treatment zone size is 250 cm^3 . A detailed description of the activator's design is provided in [4].

This setup was used to test the chloride solution of ^{137}Cs . In the initial test phase, the setup's sorption characteristic was checked. The contribution of the sorption component did not exceed 1 %. The decrease in the solution's activity for the 20 hours of the setup's operation exceeded 60 %.

Jung *et al.* [5] presents the test results of the beta decay under the influence of the external fields of

absolutely stable atoms ^{163}Dy and ^{193}Ir , the half-life of which is also determined.

Andreev and Shafeev [6] presents a detailed description of the effect of laser beams on solutions of radioactive nuclides. The method uses a laser beam to irradiate an absorbing target placed in a transparent water solution with a radioactive nuclide. The gold nanoparticles pushed out of the metal target migrate to the water and speed up the decay of radioactive nuclides. The experiment involved studying the ^{137}Cs and ^{238}U radioactive nuclides, and a golden plate was used as the target. The laser's energy was 2 mJ per pulse, the pulse duration was 10 ns, the repeating frequency was 10 kHz, and the exposure time was 10 hours. The laser beam was used to scan across the tray with the solution of ^{137}Cs .

In the time of exposure, the concentration of ^{137}Cs goes down by 5 % and remains unchanged for 15 hours. The reduction in the concentration for exposure time is formally equal to the 1 200-fold acceleration of decay. This method can be used only with small quantities of solution.

The provided examples show that the isotopic properties of elements can change under an external energy influence.

Nowadays, NPP units with uranium-graphite channel-type reactors are being removed from operation all around the world. The issue of discarding irradiated graphite complicates this process. The amount of irradiated graphite in one Russian uranium-graphite channel-type reactor reaches 1.798 tones. The total amount of irradiated graphite in Russia and worldwide is approximately 60.000 and 250.000 tons, respectively [7]. Irradiated graphite is referred to as class 2 and 3 radioactive waste (RW). The induced activity of irradiated graphite is determined mainly by ^{14}C , ^{36}Cl , ^{60}Co , and ^3H . The share of activity of carbon ^{14}C in the overall activity is 95 %. The intensity of its activity in irradiated graphite reaches 108 - 109 Bq/kg. This radioactive nuclide is a source of beta-radiation, and its half-life is 5.700 years.

In addition to the foregoing radioactive nuclides, irradiated graphite also contains small quantities of ^{235}U (fuel), such elements of its transmutation

as ^{238}Pu , ^{241}Am , and such products its fission as ^{134}Cs , ^{137}Cs . An element especially hazardous in the burial of waste is ^{94}Nb . It follows from the foregoing that the weakening in irradiated graphite activity is a relevant task at the global level.

Many patents describe the existing processing methods spent on graphite materials from nuclear reactors ([8, 9, 10, 11]. The main weaknesses of these methods are complex equipment, high energy costs, processing duration, absence of concrete results.

There are other non-electric methods [12, 13, 14, 15, 16, 17, 18, 19, 20, 21, 22, 23] for reducing the content of radioactive cesium and strontium in aqueous solutions.

All these methods involve the use of additional chemicals. For example, the effectiveness of potable water treatment processes that consist of the stages of coagulation-flocculation-decantation using iron-based coagulants was studied [12]. It was found that the resulting decontamination depends on the chemical behavior of each of the radionuclides considered, on the pH at which the process of coagulation is carried out, and on the concentration of the other stable cations present.

It has been determined that radioactive ions in water can be readily immobilized onto the getter surface by forming precipitates due to the high local concentrations of precipitants on the getters [13]. The secured ionic pollutants can be subsequently removed from water by filtration or sedimentation for safe disposal.

Granular and powdered activated carbon was used to remove radioactive iodine and cesium [22]. The efficiency was approximately 30 %–40 % in some cases.

The methods described in [24, 25] allow for efficient external influence on radioactive nuclides but are characterized by high energy costs and processing periods.

The work examines the influence of nanosecond electromagnetic pulses (NSEMPs) on the activity of water solutions of radionuclides and irradiated reactor graphite without the use of additional reagents.

2. Materials and methods

2.1. Equipment used

Method of NSEMPs exposing on solutions consisted in placing processed solution between two electrodes, which were connected to the outlets of the generator. The construction of the unit is shown in Figure 1.

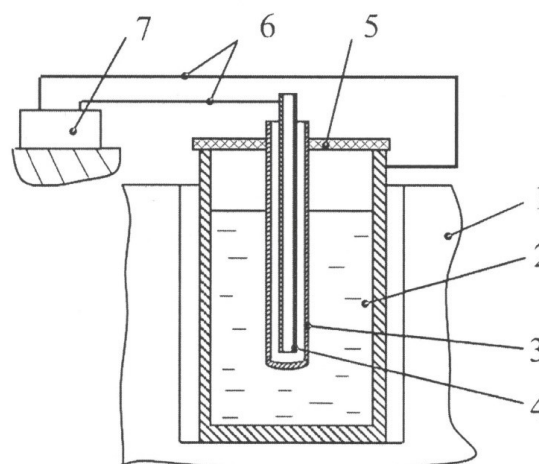


Figure 1: Unit construction: 1 - housing for glass; 2 - current-conducting glass with solution; 3 - insulating tube; 4 - rod; 5 - cover, 6 - supply cable; 7 - pulse generator

The emitters used to test the liquids are described below. The rod-in-the-glass emitter had a round copper rod or a rod with a silver coating. This rod was submerged right in the jar filled with liquid or in an isolated test tube. The rod was connected to the outlet of the center core of the generator's cable. In the case of a metal jar, the cable braiding outlet was connected right to the jar; in case of a glass jar, it was placed into a copper foil shield. The shield was connected to the cable braiding outlet. To ensure more efficient performance, the rod's and jar's diameters had to be correlated in a particular proportion depending on the properties of the irradiated liquid. This emitter was similar to a coaxial line.

Large quantities of liquid were exposed to radiation on a flow-through unit with nozzles. Similarly to the previous one, this unit was shaped as a coaxial line and had the following dimensions:

the inside rod diameter was 150 mm; the outside jar diameter was 400 mm; the unit height was 2000 mm. The generator's outlets were connected to the rod and the jar. The four nozzles were located right between them. The processed solution with a radioactive nuclide was pumped bottom-up to the nozzles under pressure.

When the generator was switched on, an electromagnetic pulse field was generated between the jar and the rod and affected the pulverized liquid. The unit required thoroughly purifying the water solution of solids for ensuring the sustainable performance of the nozzles.

The flow-through unit for processing water with electric pulses is described in RF patent 2531814 and consists of two coaxial copper cylinders. The nonconductive tube (hose), along which the radioactive solution flows, is coiled around the inner cylinder. Each of the cylinders is connected to one of the generator's outlets. The solution feed rate is adjusted using an external pump or flow control valve. This unit complies with the maintenance of staff safety requirements. The construction of the unit is shown in Figure 2.

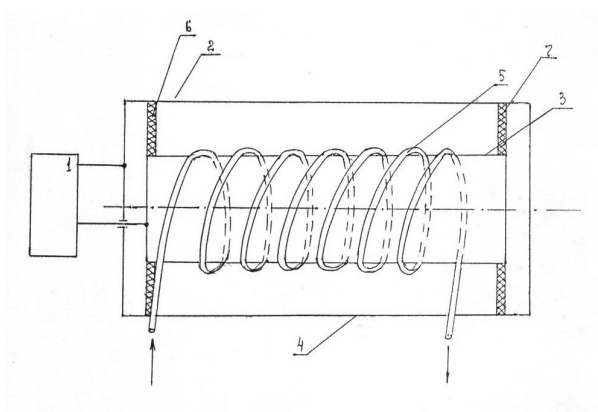


Figure 2: Construction of the flow-through unit: 1 - the pulse generator; 2 - solution exposure element in assembly ; 3 and 4 - coaxial copper cylinders; 5 - dielectric tube (hose); 6 and 7 - dielectric rings

The unit contains shut-off and control valves. The structure of the installation is shown in Figures 3 and 4. An important feature of research was the use of single-pole positive or negative current pulses. A current pulse was shaped like a triangle with a short forefront.

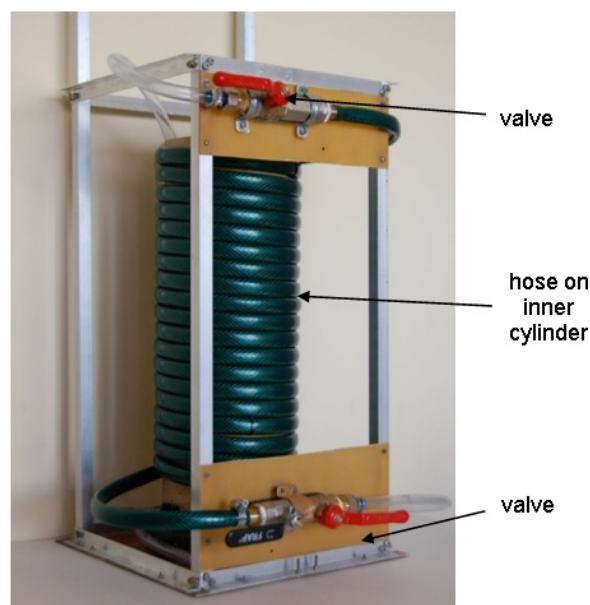


Figure 3: Unit without external cylinder

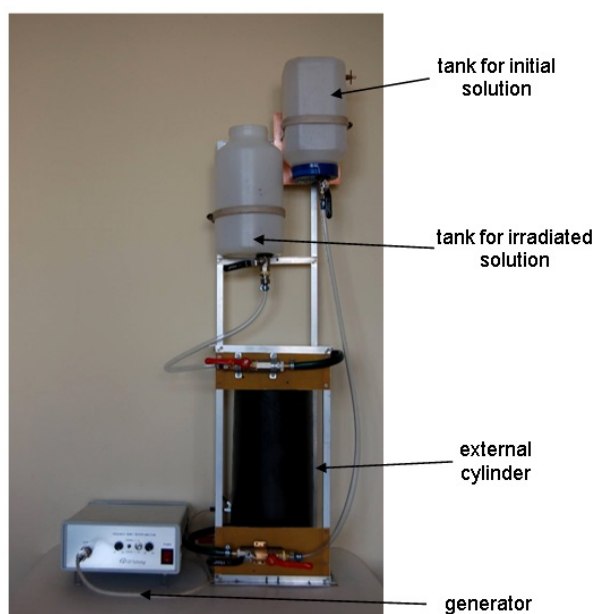


Figure 4: Experimental unit assembly

The principle of operation of the generators used is described in the patent [26]. The generators used in these tests had single-pulse energy of up to 10^{-2} J. That was why the exposure to NSEMPs was not followed by such energy effects as a significant rise in temperature. However, the pulse capacities were even higher than 10 MW.

The main generators used in the tests were the two generator units at the Budker Institute

of Nuclear Physics: GNP with an amplitude of 5 kV and GNI (LEGN) with an amplitude of 6 kV. Other generators used were the ones made by Feed Tekhnologiya company from Saint-Petersburg; they produced pulses with an amplitude of 10 or 15 kV, length of 1 ns, the forefront of 0,1 ns, and repeating frequency of 1 kHz. A distinct feature of all these generator units was their low mains power of up to 50 W.

The main condition for the repeatability of experiments was the use of a similar generator.

2.2. Materials used

The model solutions were prepared using standard elements ^{90}Sr and ^{137}Cs , which were dissolved in various acids. The amount (concentration) of the elements to be dissolved was selected based on the ability to measure their activity most accurately. Real radioactive waste was simply poured into a glass.

In the reactor graphite processing, the test object was a $120 \times 195 \times 325$ mm block of graphite taken from an IVV-2M reactor, graphite of VVG grade. The block's butt surfaces were connected to two copper foil electrodes. The electrodes were connected to the outlet of the generator. When the generator was turned on, a very large pulse current passed through the unit since graphite has a small resistance.

The materials used, the initial and the experimental conditions are given below for each specific test.

2.3. Conditions of experiments

The tests for exposing the radioactive nuclides to NSEMPs were conducted in the radiation control lab of the Chelyabinsk branch of the Ural Territorial District division of the RosRAO FSUE (RosAtom). The lab is certified in the Rosakkreditatsiya system. The solution activity was measured using a GAMMA-1P spectrometrical unit with a DGDK-150V germanium diffusion drift detector, a SEA-13P alpha spectrometrical system with a D30A silicon detector, and a UMF-2000 low-background radiometrical unit. The sampling was made according to the standard documents in effect [27].

The characteristic features of taking the samples and the procedures of measuring their activity will be indicated for each of the conducted tests. The general scheme of the experiments included the following phases: preparation of initial solutions, measurement of their activity, pulse processing, repeated measurement of solution activity.

3. Results and discussion

3.1. Processing solutions with ^{90}Sr

The results of processing the model solutions with ^{90}Sr are presented below. The ^{90}Sr element is a radioactive nuclide with the main beta decay and half-life of 28 years. It is always detected together with ^{90}Y ; to record it in a simpler way; however, we shall more frequently denote it as only ^{90}Sr .

Test 1. The model solutions of ^{90}Sr were bombarded with NSEMPs at various pH in a plastic vessel. The pulses were emitted using the rod-in-the-jar unit. The jar diameter and height were 105 and 130 mm, respectively, the irradiated solution volume was 0,5 L. The rod was a CuZn tube of 6 mm in diameter and 140 mm in length. The generator unit used was GNP, the pulse amplitude was 5 kV, and the pulse repeating frequency was 1 kHz. The exposure continued for 20 minutes. The pH value was set at the beginning of the test. The pH measurement was performed using instrument PH150MI. The pH value was reduced and increased by adding HNO_3 and, respectively, NaOH .

The solution activity was measured by the standard technique. The sample activity measurement results are shown in Table 1. It follows from Table 1 that the specific activity of the solution of ^{90}Sr is much weaker at high pH.

Test 2. The solutions were irradiated in a flow-through unit. Three and a half liters of the strontium nitrate solution with $\text{pH} = 6$ were prepared. The solution was poured into a consumption tank, and then the generator was switched on. The generator used was the one made by Feed Tekhnologiya company and had an amplitude of 15 kV, a length of 0,5 ns; and a repeating frequency of 1 kHz.

The shutoff valve was opened on the consumption tank, and the solution was supplied

Table 1: The specific activity of ^{90}Sr , $\text{N-N}_{\text{bckgrn}}$, cps

Solution	Background	Initial	Exposure pH=1	Exposure pH=4	Exposure pH=8	Exposure pH=12
β	0,0655	$10,053 \pm 0,279$	$10,223 \pm 0,281$	$8,042 \pm 0,279$	$8,393 \pm 0,255$	$3,855 \pm 0,174$
Change in level of β %	-	-	+1,7	-20	-20	-62

to the irradiation unit. After all the solution passed through, the generator switched off, and the shutoff valve closed. Solution treatment rate was 2,1 L/min.

The solution's activity was measured by the standard procedure. A sample of 1,5 mL in volume was taken from the analyzed solution utilizing a measuring graduated cylinder. The sample was placed on a substrate and dried. The substrate was then placed in the measuring chamber of the instrument UMF-2000. A quadruple measurement of the count rate of β -activity was carried out within 1000 seconds. At the same time, the background activity was measured. The initial sample was taken from the prepared solution. The irradiated sample was taken from the receptacle.

The repeated measurements of the activity of irradiated solutions after 1 and 6 days were performed by taking 50 mL samples and placing them in receptacles with a strained plug. Samples of 1,5 mL were taken from these vessels to measure the activity. The results of measuring the activity of the samples of the initial and irradiated solutions are shown in Table 2. It follows from this table that the solution's activity was weakened by 20 % after one run within 6 days after exposure.

Table 2: Specific activity of strontium, $\text{N-N}_{\text{bckgrn}}$, cps

Sample, time	Source	1 run	Change, %
0	$7,45 \pm 0,12$	$8,70 \pm 0,13$	+16
1 days		$6,702 \pm 0,113$	-10
6 days		$5,98 \pm 0,11$	-20

The comparison of all of the experiments with the radionuclide ^{90}Sr allows making the following conclusions: the shape of the vessels and the type of irradiator affected the weakening in the activity of the solution; in all of the experiments, the sorption component was significantly less than the

change in the activity of the solution; the influence of pulse amplitude was insignificant, whereas the influence of the pH value was significant. After treating the solutions with the average pH value by NSEMPs, a reduction in the activity of the solution by 15-20 % can be obtained without taking into account the average sorption component ≈ 5 %. The conclusions on the sorption component do not apply to flow-through units since no such studies were carried out there. At high pH, the activity of ^{90}Sr can be reduced by 60-80 %.

3.2. Irradiation of solutions with ^{137}Cs

The ^{137}Cs element is a radioactive nuclide with the main beta decay (95 %), a low level of gamma radiation, and a half-life of 30 years.

Test 3. One hundred milliliters of the mixed solution of ^{137}Cs were poured into a 64 mm vessel diameter and 94 mm in height. An irradiation source shaped like a copper rod of 6 mm in diameter and 100 mm in height was inserted into the vessel. The end of the rod was 14 mm away from the vessel bottom. The generator unit used had a pulse amplitude of 15 kV and a repetition frequency of 1 kHz. The initial pH value was 5,72.

The activity of the solutions was measured on a UMF-2000 low-background unit: two parallel samples of 0,1 mL each were taken, dried under the bulb, and the count pulses were measured thrice for 100 s. The activity was measured right after and within one day of the exposure. The solution activity measurement results are presented below:

- The initial solution activity was 61 kBq/L;
- After two minutes of exposure, the activity went down by 36 % to reach $39,0 \pm 1,8$ kBq/L;
- After five minutes of exposure, the activity went down by 31 % to reach $42,0 \pm 0,5$ kBq/L;

- After ten minutes of exposure, the activity went down by 31 % to reach $42,0 \pm 1,1$ kBq/L;
- After one day, the solution irradiated for ten minutes and poured into a plastic vessel showed an activity of $39,0 \pm 2,6$ kBq/L, a decrease of 36 %.

Test 4. The solutions were irradiated in a flow-through unit with coaxial cylinders. Three and a half liters of the cesium chloride solution with $\text{pH} = 6$ were made. The solution was poured into a consumption tank, and then the generator was switched on. The generator unit used a pulse amplitude of 15 kV and a repetition frequency of 1 kHz. The shutoff valve was opened on the consumption tank, and the solution was supplied to the irradiation unit. After all the solution passed through, the generator switched off, and the shutoff valve closed.

The solution activity was measured by the standard technique on a GAMMA-1P gamma spectrometer. The vessel with 50 mL of the source solution of ^{137}Cs was placed right to the spectrometer's measuring chamber. Then the solution was irradiated, and the 50 mL of it were poured from the receiving tank into a vessel of the same kind that was placed in the measuring chamber. The source and the irradiated solution samples were left in those same vessels tightly closed with a cap after being taken. The activity measurements were repeated within 1, 3, and 6 days. The activity levels of the source and the irradiated solution are shown in Table 3. The repeated runs were performed similarly to the maiden run after taking a sample for activity measurements.

It follows from Table 3 that, after the maiden run, the solution became less active by 40 %. There was no weakening in the solution's activity after the second and third run. The activity level decreased after the maiden run remains unchanged after 1, 3, and 6 days.

In sum, the parameters tracked during the irradiation of the solutions with radioactive nuclides were the influences of the vessel shape, the type of irradiation units, the pulse amplitude and repetition frequency, the solution's pH, and the

effect of various added salts. All in all, more than 50 tests were conducted. The sorption component was not examined because it was measured most frequently in the same vessel before and after irradiating the solution.

The most interesting results were obtained on the flow-through electropulse machining unit. The passage of the solution through the unit was attended by a decrease in the solution's activity by 40 %. A decrease in activity by 50 % (half-life period) occurs for thirty years in natural conditions. It is seen that this period shortened to minutes. The essentially new result obtained on that unit was that the change in the solution's activity had its limit, irrespectively of the number of runs through the unit. The solution's activity remains decreased for a long time.

3.3. Irradiating actual radioactive waste

This subsection describes the tests aimed at exposing actual liquid radioactive solutions to NSEMPs. The research subject was the water solution of various radioactive nuclides of complex chemical composition.

Test 5. The pulses were emitted using the rod-in-the-jar unit (description was given in section 2).

This test was conducted using the jar made as a glass bottle of 75 mm in diameter, 140 mm in height, and 600 mL in volume. The bottle was wrapped on the bottom and sides in the foil of 100 μm in thickness. The rod was an M0 copper bar of 6 mm in diameter, 195 mm in length, and the rod's tip was 40 mm away from the bottle bottom.

The generator produced pulses of 0,6 ns in length and 15 kV in amplitude with a repetition frequency of 1 kHz. The pulse power was 4,5 MW. The energy of one pulse was $\approx 10^{-2}$ J. The generator's dimensions were $320 \times 210 \times 80$ mm, its main power was below 50 W, and it was lighter than 5 kg. The processing time was 15 minutes.

At the time of the test, the solution of complex chemical composition had $T = 18^\circ\text{C}$ and $\text{pH} = 5$. The bombardment of the solution with NSEMPs left large flakes on its surface, the test results are shown in Table 4.

The specific activity reduction coefficient was determined as the ratio of the specific radioactive

Table 3: Activity of caesium solutions, kBq/L

Sample, time	Source	1 run	2 runs	Decrease, %
0	$37,0 \pm 3,7$	$22,3 \pm 3,1$	$23,0 \pm 2,3$	-
1 days	$37,0 \pm 3,3$	$23,8 \pm 2,4$	$25,1 \pm 2,7$	35
3 days	$38,0 \pm 3,7$	$23,3 \pm 2,6$	$25,3 \pm 2,6$	35
6 days	$39,0 \pm 3,2$	$22,8 \pm 2,3$	$24,8 \pm 2,7$	33

Table 4: Specific activity reduction coefficient after test 5

Radioactive nuclide	Reduction coefficient
^{54}Mn	8,75
^{60}Co	10
^{94}Nb	9,5
^{125}Sb	3,38
^{133}Ba	4,02
^{134}Cs	4,6
^{137}Cs	4,46

Table 5: Specific activity reduction coefficient in the solution after test 6

Radioactive nuclide	Reduction coefficient
^{54}Mn	58
^{60}Co	20
^{94}Nb	46
^{125}Sb	3,2
^{133}Ba	-
^{134}Cs	7,7
^{137}Cs	8,6

activities of a radioactive nuclide before and after processing the solution with NSEMPs.

It follows from Table 4 that the specific activity of all the radioactive nuclides became much weaker. The higher was the radioactive nuclide's atomic number, and the lower was the reduction coefficient.

Test 6. This test differed from test 5 by adding NaOH alkali to the solution with a pH of 10. A glass bottle was replaced with a steel vessel of 100 mm in diameter, 120 in height, and 800 mL in volume. The rod was an M0 copper bar of 6 mm in diameter, 195 mm in length, and the rod's tip was 40 mm away from the bottle bottom.

The 15-minute bombardment with NSEMPs left dark flakes in the solution and on the rod that became black. The processed solution also had many deposits.

The deposit activity was analyzed separately. The sorption effect was measured by soaking a gauze piece in water and using it to pick up the flakes and particles from the vessel's surface. Table 5 provides the values of the specific activity reduction coefficient for the water solution after test 6 compared to the initial solution.

It follows from Table 5 that the specific activity of some of the radioactive nuclides became much weaker. The reduction coefficient of the specific activity decreased with an increase in the sequence

number of the element.

Table 6 provides the values of the specific radioactive nuclide activity rise coefficient for the deposit after test 6 compared to the initial solution. It follows from Table 6 that the specific activity of all the radioactive nuclides in the deposit became much higher. Table 7 provides the values of

Table 6: Specific activity rise coefficient in the deposit after test 6

Radioactive nuclide	Reduction coefficient
^{54}Mn	6,7
^{60}Co	5,3
^{94}Nb	5
^{125}Sb	6,8
^{133}Ba	7,9
^{134}Cs	5,6
^{137}Cs	7,2

the coefficient of changes in the activity of the radioactive nuclides on the vessel walls after test 6 in comparison with the initial solution. It follows from Table 7 that most of the radioactive nuclides were by two to three orders of magnitude as active on the vessel walls as in the initial solution. As shown by the tests, the bombardment of the water solutions of ^{90}Sr , ^{137}Cs , and of the actual waste with powerful nanosecond electromagnetic pulses allowed making the solutions much less active

Table 7: Coefficient of changes in the specific activity on the walls after test 6

Radioactive nuclide	Reduction coefficient
^{54}Mn	$0,5 \times 10^{-3}$
^{60}Co	$0,9 \times 10^{-3}$
^{94}Nb	$7,1 \times 10^{-2}$
^{125}Sb	$0,8 \times 10^{-3}$
^{133}Ba	$1,5 \times 10^{-2}$
^{134}Cs	$2,5 \times 10^{-3}$
^{137}Cs	2×10^{-2}

for a short time. The proposed unit designs make possible a simple increase in the amount of processed solution. For the technical reasons for waiting in the queue, the activity measurements were carried out 20 days after processing. This period can be considered as the time of minor changes in the measured activity level.

3.4. Processing irradiated graphite

The test object was a $120 \times 195 \times 325$ mm block of graphite taken from an IVV- 2M reactor. The block's 120×195 mm bottom butt surface had an indentation of 65 mm in diameter and 7 mm in depth. The upper butt surface had a projection of 65 mm in diameter and 7 mm in depth. The block's butt surfaces were connected to two copper foil electrodes. The electrodes were connected to the generator of pulses of 1 ns in length, 15 kV in amplitude, and a repetition frequency of 1 kHz. The generator's pulse power was 4,5 MW; and the consumed power was 35 W. The periods it took to process the block after three consecutive tests were 5, 15 (5+10), and 25 (5+10+10) minutes, respectively.

The graphite samples were taken from the block's 195×325 mm side surface before the test session and after each test. The sampling from the block for analysis was made in three points by drilling the bottom part of the block (30 mm from the bottom edge).

The drilling was carried out using bore bits of two diameters. First of all, the graphite was drilled with a bore bit of 12 mm in diameter to a depth of 10 mm. After the bored graphite was removed, an aperture of 8 mm in diameter was drilled to a depth of 60 mm. The extracted

graphite powder was collected after each drilling into numbered canisters. The powder was then stirred, and samples of 0,1 g were collected from the canisters for activity measurements.

The measurements were made on a UMF-2000 alpha-beta radiometer. Each counted sample was exposed to five measurements of 100 s each. The measurement results are shown in Table 8. The maximal power of a gamma radiation dose from the block's surface before the processing was 2 mSvph. Its small changes were revealed by the gamma spectrometric analysis conducted before and after the processing. Patent [28] has been obtained for a processing method of irradiated graphite blocks and a unit for applying it.

The processing of a graphite block of such dimensions without its destruction was carried out for the first time in the world. There is no information in the literature about similar experiments.

3.5. Comparison of results

The effect on ^{137}Cs can make the best comparison of results. In Kladov [4](2019), a hydrodynamic generator of mechanical vibrations of the rotary type was powered by a 15 kW engine. The volume of the processing zone was 250 cm^3 . The decrease in the activity of the solution was more than 60 % during the 20 hours of unit operation.

In [6], a cuvette of 5 mL in volume with the solution of ^{137}Cs was scanned for 10 hours with a laser with an energy of 2 mJ per pulse. During the irradiation, the concentration of ^{137}Cs went down by 5 %.

In Filippov [29], the theoretical justification is provided for an increase in the probability of the $^{137}\text{Cs} \rightarrow ^{137}\text{Ba}$ disintegration by 2,9 times under the influence of the high-power field of a sinusoidal electromagnetic wave.

In the flow-through unit (test 4), the solution of ^{137}Cs became less active by 40 % during the passage of the solution through the unit for about 2 min. The consumed power of the pulse generator was 35 W. Unfortunately, and there is no such data for other radionuclides in the available literature.

Table 8: Activity of aggregate beta-emitting radioactive nuclides in graphite

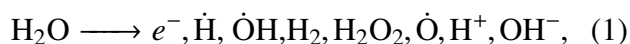
Sampling area	Sampling depth (mm)	Processing time (min)	Specific activity Bq/kg $\times 10^7$	Fold change	Specific activity after 6 days Bq/kg $\times 10^7$	Fold change
Block bottom	10	initial	3,5	-	3,8	-
		5	3,1	-1,1	2,8	-1,4
		15	4,3	+1,2	4,6	+1,2
		25	3,4	-1,02	2,5	-1,52
	60	initial	2	-	1,8	-
		5	9,3	+4,15	10	+5,5
		15	2	1	1,9	+1,05
		25	0,74	-2,85	0,82	-2,2

Thus, the suggested flow-through technology has advantages in operation, processing speed, and energy consumption.

According to existing terminology, it can be attributed to the acceleration of the decay of radionuclides under the influence of high-power energy fields.

3.6. Radiolysis of water

Deposit in test 6 was caused by a change in the chemical composition of the solution. Comparing the liquid irradiation tests with the literary data from [30, 31] allows assuming that the observed effects were similar to the action of radioactive emissions. In this case, the close analogs are X- and γ - radiation. These are defined as sine wave oscillations with a frequency from 3×10^{16} to 3×10^{22} Hz. The frequency spectrum of these pulses is 10^9 Hz. The radiolysis of water followed the bombardment with NSEMPs. The radiolysis of water can be recorded as equation (1)



where e^- is the hydrated electron, $\dot{\text{O}}\text{H}$ is the radical, $\dot{\text{H}}$ is the atom of hydrogen, H_2 is the molecular product, H_2O_2 is hydrogen peroxide H^+ , OH^- are ions. The radiation chemical output of $\dot{\text{O}}$ is very low and can be neglected.

The average radiation chemical output values of the components of radiolysis found by experimentation at various pH are enumerated below: $g(e^-)$ was 2,6 to 3, $g(\dot{\text{H}})$ was 0,55 to 0,6; $g(\dot{\text{O}}\text{H})$ was 2,7 to 2,95; $g(\text{H}_2\text{O}_2)$ was 0,68 to 0,8; $g(\text{H}_2)$ was 0,4 to 0,45.

A hydrated electron is one of the main products of radiolysis of water and water solutions. The

physical properties of this electron are taken from [30]:

- the oxidation and reduction potential is 2,7 V;
- the average charge distribution radius is 0,28 nm;
- the theoretical origination time is 10^{-11} s; as found by test, the hydrated electron is formed in less than 0,02 ns; the equivalent conductivity is $185 \text{ Cm} \cdot \text{cm}^2$.

The hydrated electron is the simplest and a very powerful reducer rapidly reacting with many substances, including most water radiolysis products. The groups in which the reactions with e^- are divided are

1. The addition of e^- to ions.
2. Joining neutral molecules.
3. Dissociative addition.

The H atom is important to the radiolysis of acid water solutions. The H atom is a weaker reducer than e^- and reduces the ions of metals with oxidation-reduction potential that are less negative than -2,3V (Fe_3^+ , Cu_2^+ , Ce_4^+ , and others).

The molecular products of the radiolysis of water are H_2 and H_2O_2 . As a rule, molecular hydrogen does not interact with dissolved particles.

Peroxide can be either a reducer or an oxidizer, which depends on the dissolved substance's essence and conditions; the oxidizing and reducing properties of hydrogen peroxide are more pronounced in acid and alkaline media, respectively.

The bombardment of double-distilled water with NSEMPs made its electrical resistance much weaker, which meant that new particles had

been generated called hydrated electrons. i.e., the bombardment with NSEMPs produced the radiolysis of water. Thus the radiolysis of water significantly modified its composition and physical properties and resulted in precipitation of dissolved substances.

4. Conclusion

The tests carried out have shown that the exposure of powerful NSEMPs on the water solutions of radionuclides leads to a decrease in their activity. In a flow-through unit, the solution of ^{90}Sr is reduced by 20 %, the activity of the solution of ^{137}Cs is reduced by 35 % for 2 minutes of processing time. The measured activity value was maintained for 6 days.

The processing of real radioactive waste showed a decrease in the activity of various radionuclides by 5-50 times. The measured decrease in activity was maintained for 20 days.

The results of the pulsed processing of the irradiated graphite block indicate that the proposed processing method is promising.

The decrease in the solution activity is due to two processes: acceleration of the disintegration during pulse action and radionuclides precipitation due to reactions with water radiolysis products.

There is the repeatability of the results, which is determined and due to the characteristics of the generators used.

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