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PLASMACHEMICAL PLANT FOR NPP DRAIN WATER TREATMENT

Introduction. *As a result of the operation of nuclear power plants, liquid radioactive waste (LRW) is formed, which consist mainly of drain water. Technologies and technical means of drain water treatment have a number of disadvantages that make the treatment long and expensive.*

Problem Statement. *One of the most promising advanced methods of LRW treatment is the plasma chemical method that has not been used in domestic practice. An experimental study of plasma chemical method for the treatment of radioactive waste from nuclear power plants has been performed for ensuring safe and efficient treatment of liquid radioactive waste in order to make it suitable for temporary storage.*

Purpose. *Development of modern domestic equipment for concentrating and removing the radioactive elements from solutions of complex salt composition.*

Materials and Methods. *When creating a plasma chemical plant, methods for mathematical and computer modeling, full-scale prototyping, and machine design have been used. Special solutions containing Cs^+, Sr^{2+}, Co^{2+}, Cu^{2+}, and Mn^{2+}ions, as well as solutions with organic components, have been prepared to study the technical characteristics of the developed plasma chemical plant and its features.*

Results. *An experimental electric discharge plant for using the plasma chemical technologies for the treatment of liquid radioactive waste, in particular, drain water, has been created and studied; its characteristics have been determined.*

Conclusions. *The developed prototype plant enables to carry out integrated treatment of LRW and to significantly reduce its volume. Due to one-stage process, the plasma-chemical method is effective in solving the problems of LRW utilization.*

Keywords: nuclear power, radioactive waste, drain water, environmental safety, and plasma chemical method.

Radioactive waste (RAW) is a product of normal operation of nuclear power plants (NPPs). They are constantly accumulated, which is one of the serious problems both in the field of NPP safety and in the industrial ecology as a whole. In the structure of RAW, a special place is occupied by liquid radioactive waste (LRW).

In view of the fact that RAW poses a serious danger to the environment and humans, the removal and processing of LRW, in particular drain waters, is an important task today.

Now, there are the following methods of LRW utilization: evaporation, ion exchange, ultrafiltration, cementation, bitumen embedding (bituminization), vitrification, deposition, sorption and membrane methods. The choice of one or an-other method depends on LRW chemical and radionuclide composition and is determined by the state of radionuclides in the solution.

One of the main methods for reducing the LRW amount is evaporation that enables to downsize it 20—100 times. The formed concentrates have complex radionuclide and chemical composition, they contain a significant amount of suspensions, petroleum products, colloids, surfactants, and complexing agents. However, this method has significant disadvantages which results in the need to develop new cost-effective and environmentally sound technologies for concentrating RAW.

Radionuclides can also be removed from solutions by physical and chemical methods based on sorption, deposition, and membrane processes.

The use of sorption methods leads to an increase in secondary RAW. In addition, they require selective inorganic sorbents that bind specific radionuclides from complex salt solutions.

The deposition method is used as universal means for removing radionuclides both in ionic and in colloid states. The main disadvantage of the method is a poor degree of solution purification and formation of large amount of secondary waste, which is explained by the lack of methods for selective radionuclide deposition.

The membrane methods are widely used for water treatment, desalination of seawater, in food industry. Their use for RAW treatment has not gone beyond pilot trials.

In general, the disadvantages of existing technologies for converting LRW into nonradioactive waste by concentrating radioactive isotopes in lesser volume and in condition suitable for further long-term storage are as follows:

- ignoring the dispersed state of radionuclides in LRW;
- presence of organic and complexing agents in the solution;
- the need to concentrate the withdrawn components with the associated formation of an additional amount of secondary radioactive waste from sorbents, membranes, ion-exchange resins, etc., which are used in the technological process;
- the formation of a significant amount of secondary RAW;
- the need for using systems for pretreatment of radionuclide solutions;
- the formation of highly soluble and highly active still bottoms requiring special utilization methods.

Therefore, in order to increase the efficiency of purification of LRWs of nuclear fuel cycle facilities (NFCs) and environmental safety as a whole, it is necessary to use state-of-the-art equipment that eliminates the abovementioned disadvantages. Such equipment should provide a significant reduction in electricity consumption for the LRW treatment process, be compact, raise the degree of purification of solutions, and downsize the amount of waste that needs to be buried.

To fulfill the assigned task, the Department of Nuclear and Physical Technologies of the Institute of Environmental Geochemistry of the NAS of Ukraine (IEG NAS of Ukraine) has developed and tested a modern electrodischarge laboratory plant for LRW purification at NFC facilities.

THE CHARACTERISTICS OF DRAIN WATERS

Fig. 1 shows sources of drain water formation [1].

Drain water contains a significant number of background metals (sodium, potassium, calcium, aluminum); the majority of solutions are nitrates (with a concentration of nitric acid ranging from 2 to 6 mol/l), but there are also alkaline solutions. Typical concentrations of macrocomponents of solutions are quite high and varies from 10 to 150 mg/l.

Depending on the origin of drain waters, they may contain the following components (g/l):

- dissolved salts: 0.1—40.0;
- dissolved organic matter: 0.1—5.0;
- micro-suspensions and colloids: 0.01—0.1;
- petroleum products: 0.0—1.0.

The concentration of organic compounds (spent lubricants, solvents, etc.) in drain water can range within 0.4—0.6 g/dm^3, and chemical oxy-

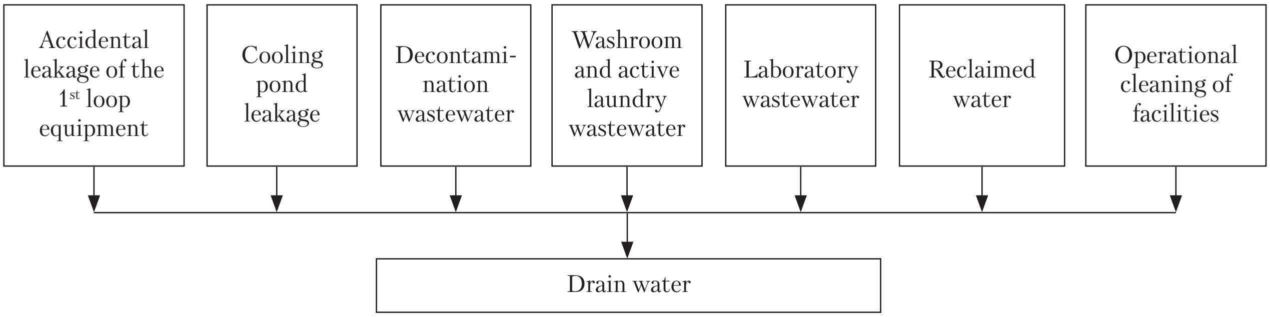

Fig. 1. Sources of drain waters

gen demand (COD) is 2000—3500 mg O_2/dm^3. For the further LRW treatment it is necessary to reduce the content of organic substances to COD $\leq$ 200 mg O_2/dm^3 [2].

The specific feature of drain water is that, in addition to radionuclides, it contains a significant number of stable isotope ions, the amount of which substantially exceeds the content of radionuclides.

In addition to radionuclides in the ionic form, in drain water, there is a significant amount of radionuclides in colloidal or pseudo-colloidal form. Phase composition of drain water is shown in Table 1.

The pollutants to be removed contain both alkaline, earth, and multivalent metals (Co, Mn, Cu, etc.), as well as surfactants, phosphates, and complexing agents.

In order to achieve the maximum concentration of radionuclides while treating LRW, it is desirable to minimize or to completely exclude the introduction of additional chemical reagents into LRW, as well as to remove the maximum amount of non-radioactive ballast components.

Table 1

Phase Composition of Drain Water

Isotope	Form of radioelement		
	Ionic	Colloidal	Pseudo-colloidal
^{90}Sr	Almost at all pH: Sr^{2+}	—	At pH > 8.5, forms negatively charged pseudo-colloids
^{90}Y	Can exist at pH of up to 9.0	At pH = 4.5—9.0; At pH = 7.0, colloid particles of yttrium hydroxide and oxide $Y(OH)_3$, Y_2O_3	At pH > 8.0, pseudo-colloids start to be formed
^{137}Cs	At all pH: Cs^+	—	At pH > 7.2, pseudo-colloids are formed on impurities in the solution
^{140}Ba	At pH = 4.0—5.0 can exist as Ba^{2+}	Colloid aggregates $BaSO_4$ are formed	—
^{140}La	Up to pH = 6.0–7.0, in ionic dispersive La^{3+}	At pH > 9.0, real colloids of lanthanum hydroxide $La(OH)_3$, La_2O_3 started to be formed	—
^{144}Ce	At pH<2.5 can exist as Ce^{3+}	At pH > 2.5 Ce (IV) as real colloids At pH > 6.0 main forms are hydrolysis products	—
^{106}Ru	—	At pH = 4.5—7.0, Ru (IV) in the colloid form At pH = 5.5—8.0, Ru (III) in the colloid form	—
^{95}Zr	At pH = 0–1.6, Zr^{4+}; At pH > 11.0–12.0, zirconates started to be formed	pH 4,0—12,0— $ZrO(OH)^{3-}$	—

To destroy organic waste contained in drain waters, the technology for plasma-chemical reactions in liquid has been considered [3].

In the course of research, the method and laboratory complex for modified plasma-chemical treatment of such wastes have been substantiated and developed.

PLASMA-CHEMICAL METHOD OF DRAIN WATER PURIFICATION

The low-temperature plasma electrolysis method is unique due to the fact that it affects the liquid by means of electrical and photochemical effects, as well as electrochemical reactions occurring on the electrode immersed in liquid. In the solution there simultaneously occur photolysis, electrolysis, and chemical reactions of active radicals with the components of the solution. The products of these reactions can interact with each other.

Under the influence of electrical discharge in the solution, water molecules fall into OH radicals (oxidizing agents) and H atoms (reducing agents): $H_2O \rightarrow OH + H$. In accordance with the law of conservation of charge, the number of oxidizing elements is equal to the number of reducing ones. Electric discharge in the gas phase (air) creates a significant concentration of ozone. Hydrogen atoms interact with ozone with the formation of OH radicals ($H + O_3 \rightarrow OH + O_2$). Therefore, in the presence of air, in the discharge, only oxidizing elements are formed.

The technology for plasma-chemical treatment of water and liquid industrial wastewater belongs to the so-called destructive methods based on chemical changes in the structure and composition of molecules of impurities. Interaction of active plasma particles with chemical compounds or microorganisms leads to their destruction. Moreover, the most effective transformation is the oxidation of substances.

High-voltage discharge (10—70 kV) in water generates a shock wave and causes an electrohydraulic effect (104—105 kg/cm^2) and elastic fluctuations of the medium (≈ 100 Hz). The discharge has a powerful integrated effect on the fluid and intensifies the processes in it.

In general, the plasma discharge in the drain water is characterized by the following components, which together contribute to water purification: the thermal component — temperature of plasma reaches 5000 °C (narrow zone of the gas-liquid section); temperature of the treated liquid with organic matter reaches 44 °C; acoustic component — shock wave; electromagnetic component; ultraviolet radiation; ozonization; and chlorination.

The complex of factors influencing the drain water treatment by plasma discharges leads to the destruction of surfactants and activation of sorbents, which contributes to the effective removal of radionuclides.

Basic components and features of the plant

The pilot plasma-chemical plant consists of the following elements: liquid purification reactor; electronic unit; high-voltage unit; liquid supply and filtration system; system for liquid purification from coarse impurities; hydrodynamic cavitator; and control panel.

The plant is based on a flow reactor that is a vertical cylinder with a multipin cathode in the form of a rotating brush and an anode located at the bottom of this cylinder. The liquid is fed to the reactor through the system of supply and purification from coarse impurities. Schematically, the reactor structure is shown in Fig. 2.

The developed plasma reactor can be classified as a bubble discharge reactor, where the electrical discharge is generated directly in the aqueous medium by adding air bubbles from outside [4]. This technology for obtaining plasma is based on fluid bubbling, i.e. pumping gas from the bottom upward through the anode nozzle, which is located under the grounded electrode, as shown in Fig. 2. The nozzle electrode is placed inside the dielectric tube to avoid any release of energy into the water.

By generating plasma in the form of bubbles, the total surface area of plasma-liquid interface

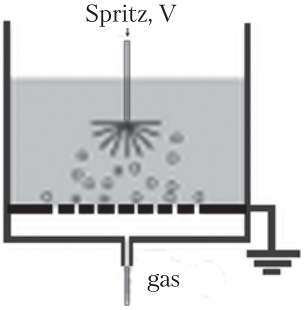

Fig. 2. Scheme of the plant reactor: bubbles ascend towards the high-voltage multipin electrode

increases. It is expected that an increase in the surface area leads to raising effectiveness of the destruction of organic matter.

Bubbling has also one more advantage — it creates conditions for stirring the solution. In addition, initiation of discharge in the gaseous phase minimizes the electrode erosion, which results in extended life of the plant. Gas bubbles passing through the area of discharge greatly increase the density of radicals in plasma. The air in the plant is bubbled from a grounded electrode in the direction of high-voltage multipin electrodes.

The plasma generator uses a specific type of discharge, which is most effective for reducing high concentrations of pollutants. Its main property is to prevent excessive heating of plasma gas and fluid that is purified.

The reactor is based on a rotating electrode design with a rotation speed of up to 1000 rpm. The air is pumped through a stationary electrode and sprayed with the formation of bubble fog between the electrodes. The rotating electrode distributes erosion evenly on a stationary electrode thereby preventing point corrosion and undesirable variation of the distance between the electrodes. In addition, it partially removes the mass transfer limitation that is typical for the pin-to-plate type reactors, as well as reduces voltage loss between the contact electrodes and the stationary electrode. The energy efficiency of decomposition increases as speed of rotation increases. This system is more energy-efficient than the crown-based technologies, but still needs further optimization.

The main technical characteristics of the installation are: discharge current is 10 A; energy consumption is 3—5 kW per 1 m^3 liquid; air consumption per 1 m^3 of purified liquid is 3 m^3; time of continuous operation is 8 hours; operating modes (frequency of electric discharge impulses) range within 1—100 Hz; consumables (sorbents) reach, at most, 1% of the liquid volume; weight is at most, 60 kg.

Pilot plant and the results of its operation

A solution containing Cs^+, Sr^{2+}, Co^{2+}, Cu^{2+}, and Mn^{2+}ions was prepared to simulate drain water. For this purpose, nitric acid salts of the corresponding metals were used. The solution was prepared by adding calculated amount of each individual salt to a small amount of water until the salt is completely dissolved. The approximate concentration of the initial solution was about 100 mg/l of the corresponding ion.

For the experiment, the working solution was made by diluting the initial solution in ratio 1:10. The ion concentration was measured by the atomic absorption method, using a spectrometer. The following results have been obtained: Cs (6.2 mg/l); Sr (6.6 mg/l); Co (7.3 mg/l); Mn (10 mg/l); and Cu 10 (mg/l).

As the working solution is treated with plasma, a suspension of particles that eventually flocculate is formed. The precipitate is not uniform, which indicates the presence of particles of different sizes in the suspension. The most intensively the precipitate is formed during the first minutes after the treatment. At the same time, as the number of impulses increases, so does the precipitate volume. According to the results of visual observations, the precipitation of particles in standing water is completed in 1—2 days after the treatment.

Complex organic compounds decompose with the formation of organic acids, which results in a decrease in pH of the solution. In addition, there is formed carbon dioxide that having dissolved in water leads to a decrease in pH as well. As pH of the solution decreases, the rate of many reactions slows down, and oxidation is inhibited.

Using steel electrodes enables the following processes when electric discharge passes through liquid:

$$2Fe + O_2 \xrightarrow{discharge} 2FeO\downarrow$$

$$2FeO(OH) \xrightarrow{discharge} Fe_2O_3 + H_2O.$$

This means, a small amount of Fe appears in the solution. After the treatment with discharges having the following parameters: U = 20 kV, number of pulses = 100, iron concentration reaches 0.55 mg/l, while before the treatment it was 0.48 mg/l.

Changes in the solution properties in the plant has been observed when graphite electrodes are used. It can be assumed that in the case of the treatment of solution with plasma the reaction occurs according to the following scheme:

$$2H_2O + 2\bar{e} \rightarrow H_2\uparrow + 2OH^-.$$

To check the effect of electric pulse discharge on complex solutions with organic components, a series of experiments on their purification has been carried out. Fig. 3 shows a general view of prototype plant; Fig. 4 features the experiment results. Table 2 presents the summarized results of the experiment.

Table 3 shows changes in the solution parameters after purification.

In order to increase the degree of purification of complex isotope composition solutions, all-purpose sorbents (montmorillonites, zeolites, and nanocomposite compounds SiO_2, Fe_2O_3) were used. During the solution treatment with plasma, the sorbent is destroyed, and nanoparticles (nanosorbents) characterized by a high surface area (ratio of surface area to volume) and activity (the specific sorption surface is 2700 m^2 per 1 g substance) are formed.

Table 2

Effectiveness of TDA* and DNT* Degradation After Their Treatment in the Prototype Plant

Solution	COD* of the initial solution, mg/l	COD of the solution treated with plasma, unfiltered, mg/l	COD of the solution treated with plasma, filtered, mg/l
with TDA	7875	150	90
with DNT	5480	350	240

*TDA — toluylenediamine, DNT — dinitrotoluene, COD — chemical oxygen demand

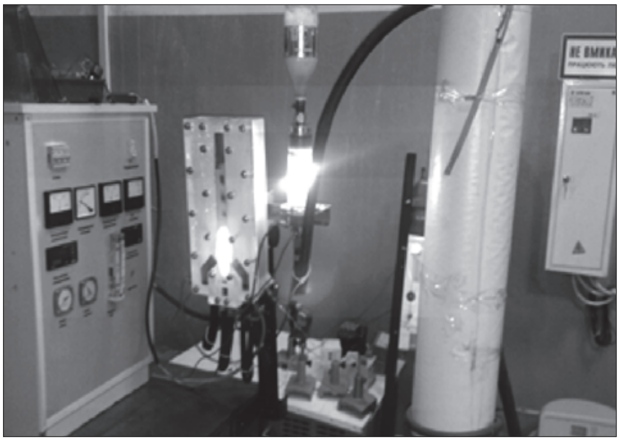

Fig. 3. General view of the prototype plant

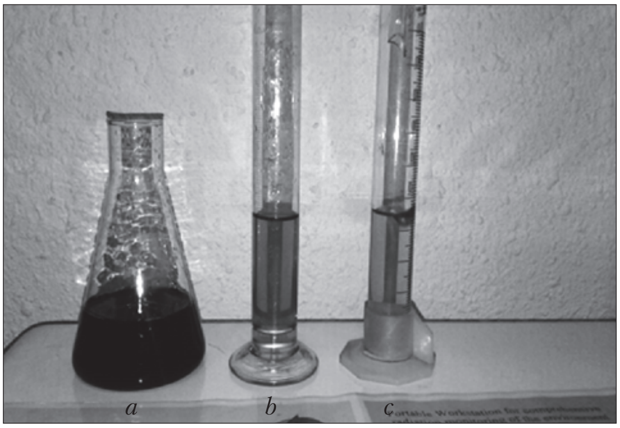

Fig. 4. Plasma effect on the experiment solutions with organic components: *a* — initial solution; *b* — solution treated with plasma, unfiltered; *c* — solution treated with plasma, filtered

Despite the fact that the sorbents reduce the content of metal ions and organic compounds, the use of plasma is justified as it is an all-purpose destructor of any organic compound and initiates a chemical reaction that results in more effective removal (sorption) of organic compounds from contaminated liquids.

Table 3

Experimental Solution Properties after Purification

Element	pH		Salt content, mg/l		COD, mg/l		Organic decomposition, mg/l		Purification coefficient based on salt content, %
	before	after	before	after	before	after	before	after	
Cs	12	6	16.0	0.2	7656	70	8500	170	98.8
Sr	9	6	4.9	0.1	5673	80	7100	210	98.0
Co	14	8	11.3	0.2	7640	90	4300	150	99.9
Cu	11	7	29.2	0.5	8578	80	5900	190	98.3
Fe	12	7	44.8	0.8	8230	70	6300	70	99.98
Mn	11	8	32.4	0.6	8690	90	8100	140	98.2

Having carried out experimental studies of the effect of the plant on the destruction of surfactants in aqueous solutions having a complex chemical composition, the authors can suggest that under the action of a pulsed electric discharge the decomposition of surfactants reaches, at least, 98 %. As a decomposition product, CO_2 has been detected. Its yield amounts to, at least, 60 % of the total carbon content. In the case of surfactant destruction, due to the formation of less toxic products, as compared with the initial products, there is no secondary environment pollution.

CONCLUSIONS

11. Radioactive waste management is the key problem of nuclear energy in terms of environment safety. Therefore, treatment, concentration, LRW storage, and final disposal of radioactive waste are currently a matter of urgency.

2. Based on the innovative approach, a prototype plant has been developed and tested, which enables deep treatment of liquid radioactive wastes and significant reduction in the existing LRW.

3. Electric pulse method is a complex one-stage process for solving problems of industrial liquid waste utilization. The method combines several physical phenomena that arise as a result of electrical pulse discharges and enables purifying drain waters with various compositions and concentrations of radionuclides in the presence of surfactants and organic pollutants.

4. Features and benefits of the designed plant are as follows:

- Oxidation groups are generated directly in the liquid treatment zone. The combination of aeration, generation of oxidants and liquid treatment in one reactor increases the effectiveness of drain water treatment and reduces the cost of both the plant and the purification process.
- Under the action of electric discharge, nanoparticles with high sorption properties are formed as a result of destruction of sorbents.
- The modular structure of the equipment enables to assemble plants having a required productivity, and, if necessary, to place them on the existing sites.
- The possibility of using standard components both domestic and foreign made.

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ПЛАЗМОХІМІЧНА УСТАНОВКА ОЧИЩЕННЯ ТРАПНИХ ВОД АЕС

Вступ. В результаті роботи атомних електростанцій утворюються рідкі радіоактивні відходи (РРВ), в структурі яких значне місце займають трапні води. Технології та технічні засоби очищення трапних вод мають низку недоліків, які роблять процес очистки довготривалим і затратним.

Проблематика. Одним з найбільш перспективних сучасних способів очищення РРВ є плазмохімічний метод, який у вітчизняній практиці поводження з РРВ не використовується. Виконано експериментальне дослідження застосування плазмохімічних методів в процесі поводження з радіоактивними відходами АЕС для безпечної та ефективної обробки РРВ до стану, придатного для тимчасового зберігання.

Мета. Розробка сучасного вітчизняного обладнання для концентрування і вилучення радіоактивних елементів із розчинів комплексного сольового складу.

Матеріали й методи. При створені плазмохімічної установки використано методи математичного і комп'ютерного моделювання, натурного макетування, машинного проектування. Для дослідження технічних характеристик розробленої плазмохімічної установки, її особливостей виготовлено спеціальні розчини, що містили іони Cs^+, Sr^{2+}, Co^{2+}, Cu^{2+}, Mn^{2+}, а також розчини з органічними компонентами.

Результати.Створено та досліджено експериментальну електророзрядну установку з використанням плазмохімічних технологій для очищення РРВ, зокрема, трапних вод, та визначено її характеристики.

Висновки. Розроблена експериментальна установка дозволяє проводити глибоку переробку РРВ та істотно зменшувати їх об'єми. Завдяки одно стадійності процесу плазмохімічний метод є ефективним при вирішення проблем утилізації РРВ.

Ключові слова: атомна енергетика, радіоактивні відходи, трапні води, екологічна безпека, плазмохімічний метод.

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ПЛАЗМОХИМИЧЕСКАЯ УСТАНОВКА ОЧИСТКИ ТРАПНЫХ ВОД АЭС

Введение. В результате работы атомных электростанций образуются жидкие радиоактивные отходы (ЖРО), в структуре которых значительное место занимают трапные воды. Технологии и технические средства очистки трапных вод имеют ряд недостатков, которые делают процесс очистки длительным и затратным.

Проблематика. Одним из наиболее перспективных современных способов очистки ЖРО является плазмохимический метод, который в отечественной практике обращения с ЖРО не используется. Выполнено экспериментальное исследование применения плазмохимических методов в процессе обращения с радиоактивными отходами АЭС для обеспечения безопасной и эффективной обработки ЖРО до состояния, пригодного для временного хранения.

Цель. Разработка современного отечественного оборудования для концентрирования и извлечения радиоактивных элементов и растворов комплексного солевого состава.

Материалы и методы. При создании плазмохимической установки использовались методы математического и компьютерного моделирования, натурного макетирования, машинного проектирования. Для исследования технических характеристик разработанной плазмохимической установки, ее особенностей были изготовлены специальные растворы, содержащие ионы Cs^+, Sr^{2+}, Co^{2+}, Cu^{2+}, Mn^{2+}, а также растворы с органическими компонентами.

Результаты. Создана и исследована экспериментальная электроразрядная установка по использованию плазмохимических технологий для очистки ЖРО, в частности, трапных вод, и определены ее характеристики.

Выводы. Разработанная экспериментальная установка позволяет проводить глубокую переработку ЖРО и существенно уменьшать их объемы. Благодаря одностадийности процесса плазмохимический метод эффективен при решении проблем утилизации ЖРО.

Ключевые слова: атомная энергетика, радиоактивные отходы, трапные воды, экологическая безопасность, плазмохимический метод.