Mini-review
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The use of combined plasma chemistry methods in the design of the main plasma-chemical processing units of the laboratory plant for radioactive waste treatment

Abstract

The article presents the results of the development of a modular laboratory unit for the purification of radioactive water based on the basic principles of plasma chemical technologies at the South Ukraine Nuclear Power Plant. The main technical characteristics of the unit and the algorithm for controlling the radioactive water purification process are given. Preliminary work they been carried out to develop the main algorithms for the purification of radioactive water at the Chernobyl Nuclear Power Plant. We obtained the most significant results using the third algorithm for the purification of radioactive water. The results of the third algorithm show that the sorption of radionuclides by a complex sorbent based on copper ferrocyanide, iron nanoparticles and coagulants with preliminary preparation of radioactive water by plasma electrolysis for 60 minutes and subsequent single- and two-stage treatment by plasma electrolysis in combination with sorbents for 30-45 minutes. Stirring in a magnetic mixer and the use of Zb series sorbents resulted in the removal of 137Cs ions to an activity of 2.48 Bq/dm^3, which is 1.3E7 times lower than the cesium activity (3.2E7 Bq/dm^3) of the initial radioactive water. PK series - removal of 137Cs ions to an activity of 129 Bq/dm^3, which is 2.5E5 times lower than the activity of cesium (3.2E7 Bq/dm^3). Removal of 90Sr ions to an activity of 6.85E2 Bq/dm^3 under the above-mentioned sorption conditions using PK series sorbents, which is 17.1E3 times lower than the activity of strontium (4.9E6 Bq/dm^3) in the initial radioactive water. The use of complex sorbents of the Zb and PK series and the above-mentioned conditions ensured the complete removal of 154Eu and 241Am ions.

The cycle of experiments on the purification of radioactive water shows the effect of plasma electrolysis on the destruction of organic radicals, which contributes to the additional removal of 137Cs and 90Sr ions and their sorption. The result obtained it's probably explained by the fact that the sorption of 137Cs ions and transuranic radionuclides on ferrocyanides and iron nanoparticles occurs in the presence of hydroxyl groups.

Because of cleaning the radioactive liquid waste at the South Ukraine Nuclear Power Plant using sorbents and a plasma discharge unit, it was possible to achieve complete removal of 54Mn, while the activity of 60Co and 134Cs decreased by two orders of magnitude and the activity of 137Cs decreased by one order of magnitude. After cleaning, the initial total activity of liquid radioactive waste decreased by 10% from the initial activity. This indicates the effectiveness of the method and the further prospects for its research and application. As for chemical indicators, permanganate oxidisability decreased by 7.4 times, and the content of ions determined became minimal.

Keywords: nuclear power, radioactive waste, environmental safety, combined plasma-chemical method, modularity, laboratory facility, radioactive waste treatment

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Introduction

Given the global trends in the development of technologies for environmental protection against contamination of territories and water resources by radioactive isotopes, the problem of liquid radioactive waste (LRW) treatment is particularly relevant. Particular attention should be paid to 137Cs, which is a fission product of plutonium and uranium, is one of the main dose-forming radionuclides, has a long half-life (more than thirty years), and is characterised by high volatility, solubility and activity.[1] When released into the environment, even in small amounts, cesium can be deposit and accumulated in the soft tissues of aquatic and terrestrial organisms.[2,3] When 137Cs enters the human body, it causes disorders of the reproductive system, kidneys, liver and other organs.[4–6] Therefore, the task of environmental safety is to solve the problems of cesium-137 extraction from low- and intermediate-level aqueous solutions. To date many technological approaches and methods for the extraction of cesium ions they known, but practice shows that the most effective is a combination of several methods to achieve selective extraction of 137Cs. The most common methods are adsorption, ion exchange, chemical precipitation, chemical reduction, membrane technologies, coagulation, extraction, ion flotation.[2,5,7–10]

The combination of adsorption and plasma-chemical methods is a progressive tool in liquid radioactive waste treatment technologies due to the wide range of adsorbents, process efficiency, simple technology and wide range of applications.

Literary review and problem statement

The known technologies for the treatment of contaminated wastewater and radioactive water from organic substances include cavitation,[11,12] which intensify mass transfer and hydro mechanical processes during the setting of cavitation bubbles and promote the destruction of organic molecules with their subsequent oxidation. A decrease in COD to 384 mgO_2/dm^3 we observed within 15 minutes of treatment. The disadvantage of such technologies is the extremely high-energy intensity of the process.

Based on the composition of surface wastewater contaminants, including radioactive contaminants, a technological scheme we proposed with preliminary removal of suspended solids in sand traps and primary radial settling tanks, followed by biochemical denitrification in corridor aeration tanks. Biochemically treated water should we sent for disinfection by bubbling with oxygen, ozone, chlorine or cavitation.[13] The disadvantage of the proposed solution is the large size of water treatment plants.

It we noted in[14] that traditional water treatment technologies (based on coagulation and sedimentation, as well as on oxidation or ion exchange) are ineffective in the conditions of modern anthropogenic loads and new more stringent requirements to the quality of drinking and technical water. The use of membrane technologies in industry opens wide prospects for the possibility of creating fundamentally new energy-saving, environmentally safe technological schemes.

This paper[15] investigated the processes of physical, chemical, and biological treatment of industrial wastewater. The treatment effects in terms of COD in the case of using iron III sulphate were 76.0% and 72.2% at the initial COD values of untreated wastewater, respectively, 90 and 120 mg/dm^3. The indicators of treated wastewater by the biological method were as follows: COD - 50-80 mg/dm^3; BOD - 15-20 mg/dm^3; suspended solids - up to 15 mg/dm^3; nitrogen and phosphorus compounds - within the normal limits for discharge into natural water bodies. The developed technologies we implemented at industrial enterprises, saving energy costs by up to 40-45%.

This paper[16] considers the theoretical aspects of oxidative-cavitation and electro-hydro-discharge water purification from organic substances. Schematic diagrams of experimental stands for combined treatment methods were developed and presented. The study we carried out on model solutions of sodium lauryl sulphate (SL) ($NaC_{12}H_{25}SO_4$) and Ethylenediaminetetraacetic acid (EDTA) ($C_{10}H_{16}N_2O_8$). LC we chosen because it is the most common surfactant present in various detergent and decontamination mixtures. The use of EDTA is due to its application in technological processes at NPPs and its presence in liquid radioactive waste. The destruction of organic compounds occurs because of oxidation by ozone, which is constantly bubbling through the solution, and they enhanced by the action of electrical impulses or ultrasonic (ultrasonic) cavitation. The paper determines the regularities of the decrease in the concentration of model solutions depending on the treatment method, pH of the medium, and duration of the process. It was been established that the destruction of surfactants and OS is best achieved by the combined use of ozone and ultrasonic cavitation or electric discharge, at high pH values. The highest efficiency indicators of solution purification are as follows: when using discharges and ozone (pH = 6.2), ~ 71 % of surfactants we destroyed, at pH = 10, ~ 61 % of OS they destroyed; when using ultrasonic cavitation and ozone (pH = 10), ~ 83.3 % of surfactants w destroyed.

Paper[17–21] shows that one of the most promising modern methods of radioactive waste treatment is the plasma-chemical method, which is not used in the domestic practice of radioactive waste management. The methods of mathematical and computer modelling, full-scale modelling, and mechanical design were used to create the plasma-chemical unit. To study the technical characteristics of the developed plasma-chemical unit and its features, special solutions containing Cs^+, Sr^{2+}, Co^{2+}, Cu^{2+}, Mn^{2+} ions, as well as solutions with organic components, were made. An experimental electric discharge unit using plasma-chemical technologies for the treatment of radioactive waste, in particular, ladder water, was created and studied, and its characteristics were determined. The developed experimental unit allows for deep processing of LRW and a significant reduction in their volume. Due to the one-stage process, the plasma-chemical method is effective in solving the problems of radioactive waste disposal.

Taking into account the above, the purpose of this work is to create a mobile-modular laboratory unit for short-term low-energy radioactive water treatment based on the combined use of adsorption, membrane, plasma-chemical and cavitation methods, which will allow for deep processing of radioactive water and significantly reduce its volume and solve the problems of radioactive water disposal.

Nuclear power plants in Ukraine are actively implementing the latest approaches in this area, including the PLASMA-SORB technology. The design of the laboratory unit of the proposed technology we based on the use of algorithmic elements for LRW treatment, which consist of phased treatment stages using complex sorbents based on copper and nickel Ferro cyanides, iron nanoparticles, coagulants and plasma chemistry elements (Figure 1).

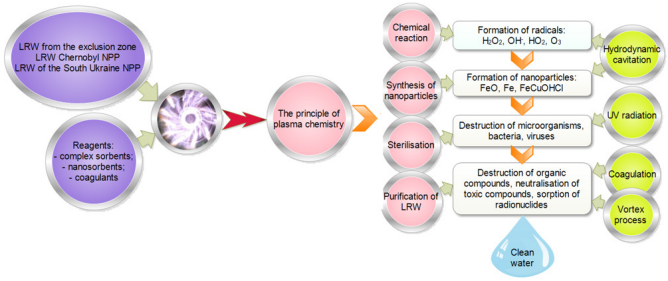

Figure 1 Block diagram of the universal technology for liquid radioactive waste treatment of the PLASMA-SORB technology.

These approaches, at the first stages of the purification process, allow reducing the initial activity of water from 10^7 Bq/dm^3 for 137Cs to 10^2-10^4 Bq/dm^3, for 90Sr from 10^6 Bq/dm^3 to 10^2-10^3 Bq/dm^3, and for transuranics - to complete purification.

However, the development they based on the complete purification of radioactive waste from all radionuclides, so a 2-stage plasma electrolysis method we used with the stepwise involvement of developed sorbents based on Ferro cyanides and coagulants. Previously, the authors conducted a series of experiments under ChNPP conditions to determine in detail the regularities of the plasma-chemical electrolysis process in order to establish the main technological parameters aimed at obtaining industrial water that meets the requirements of current standards. The influence of the plasma-chemical electrolysis time on the sorption process they studied and it we established at what values of this factor complete purification of LRW from 137Cs ions is achieve. The dependence of the process stages on the parameters of the plasma-boiling layer and the criteria for radionuclide ion transfer on the surface of iron and Ferro cyanide nanoparticles they shown. The obtained result is probably because sorption of cesium and transuranic nuclides on Ferro cyanide and nano-iron occurs in the presence of water molecules and hydroxo groups. Strontium were further purified

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to zero activity values by passing water through zeolite filters and an ultra-membrane, which is provided at the third stage of purification.

Structural elements of the plant

To address the issue of liquid radioactive waste treatment, a treatment technology they developed. For its development and testing, a system we designed in the form of a laboratory installation (prototype) (Figure 2), the hydraulic diagram of which we shown in Figure 3.

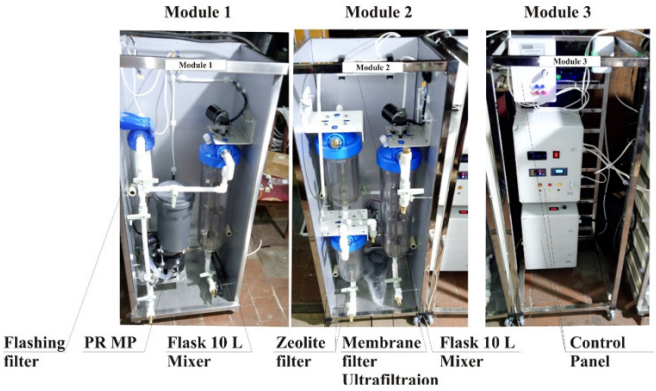

Figure 2 Laboratory installation for radioactive waste processing using PLASMA-SORB technology.

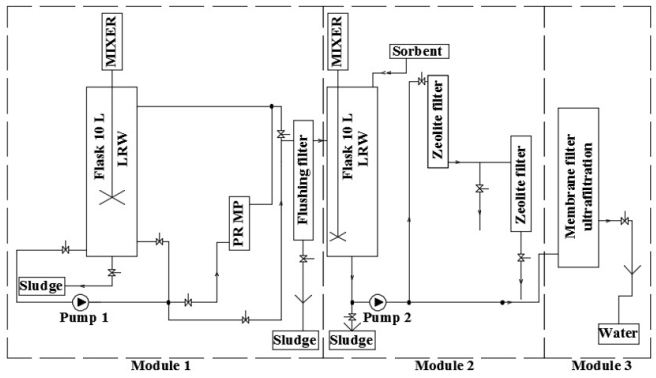

Figure 3 Hydraulic system of a laboratory LRW treatment plant.

The laboratory setup consists of 3 modules:

a) Activation module:

(i) 10 l flask with mechanical stirrer;

(ii) plasma reactor;

(iii) coarse filter, rinsing.

b) Chemical treatment module:

(i) 10 L flask.

(ii) flask reactor with stirrer;

(iii) filter with zeolite filler;

(iv) Filtration module:

(v) Ultrafiltration system.

The laboratory setup works as follows 5 liters of liquid radioactive waste (LRW) we fed into a 10-litre flask with a mechanical stirrer. Using pump 1, the LRW is electrically activate in the activation reactor. The activation time is 10 minutes. The next step is plasma chemical treatment of LRW using a plasma reactor. The treatment time is 10 minutes. After the activation process in the plasma reactor, the sorption process in the flask begins. A mechanical stirrer we switched on. For 40 minutes, the LRW they stirred at low speed. Stop the stirrer and allow the liquid to settle. Pour the sediment into the sediment hopper. Pour the liquid through a flushing filter into another (clean) 10-litre flask. Using pump 2, pump the flask into the reactor with a mechanical stirrer. Turn on the stirrer and add the sorbent.

The processing time is 30-40 minutes. Turn off the stirrer and allow the liquid to settle. Pour the sludge into the sludge hopper. The LRW we transferred to a 10-litre flask. Start the ultrafiltration process. Pour the purified water into a clean container.

Technical characteristics of the laboratory unit:

(i) capacity - 5 l/h;

(ii) power consumption - 800 W;

(iii) power supply network - 220 V.

Description of the control system

The control system of the plant for water purification from radioactive contamination they designed to implement the technological cycle of the plant.

The overall control of the plant operation we provided by a central industrial PC running Windows 10 Pro and installed specialised in-house software. This approach, using a graphical interface, allows the user to set the plant's operating modes, control the operation of the equipment, and record files - the results of the plant's operation, with reference to the date/time, for their further analysis. In addition, the specialised software controls the device that ensures the operation of the plant's technical equipment (dosing units, mixers, pumps, valves, heaters, and reactor) and the radiation measurement equipment (intensity and spectral composition).

The device for ensuring the operation of the plant's technical equipment we based on the AT mega 328 microcontroller, which runs low-level software and controls the end devices - dosing devices, mixers, valves, pumps, heaters, and a reactor - through interface modules. In addition, this controller provides time tracking to ensure the duration of the technological stages of water treatment and the maintenance of the necessary conditions for this. This ensures that the technological stages of the plant's operation are carried out, namely, preparation, the first stage of treatment, interstate procedures with, if necessary, process adjustments, the second stage of treatment and final procedures.

The measuring instruments are original devices based on a cesium-iodine crystal, a HAMAMATSU photodiode, an original preamplifier that generates a signal for the analyser, which they based on the STM 32F411 microcontroller, which runs under the control of low-level software. The measuring instruments we designed to control the degree of contamination of the source liquid, interstage and final control with the determination of intensity and spectral composition, and transmit this information via a wireless communication channel to a central computer. This information makes it possible to determine/adjust the technology of the water treatment process, namely the composition and quantity of reagents required, the conditions of treatment (time, temperature, mixing intensity, application of additional treatment measures).

The control system they constructed of separate functional units with a unified interface. This makes it possible in quickly replace

these units if necessary in the event of a failure during operation and to scale the system as a whole.

The authors of the article conducted a series of experiments under the conditions of the Chornobyl NPP to determine in detail the regularities of the plasma-chemical electrolysis process in order to establish the main technological parameters aimed at obtaining industrial water that meets the requirements of current standards. The influence of the plasma-chemical electrolysis time on the sorption process we studied and it we established at what values of this factor complete purification of LRW from 137Cs ions they achieved. The dependence of the process stages on the parameters of the plasma-boiling layer and the criteria for radionuclide ion transfer on the surface of iron and Ferro cyanide nanoparticles they shown. The obtained result is probably because sorption of cesium and transuranic nuclides on Ferro cyanide and nano-iron occurs in the presence of water molecules and hydroxo groups.

Results

Results of radioactive water purification at the Chernobyl and South Ukraine nuclear power plants

Radioactive measurement methodology

At the Chernobyl Nuclear Power Plant the content of radionuclides 90Sr, 137Cs, 154Eu and 241Am was measure in RPV solutions and RPV solutions after sorption treatment with specific sorbents. The activity of gamma-emitting radionuclides 137Cs, 154Eu, and 241Am was determined by γ-spectrometric measurements on a gamma spectrometric complex consisting of a GL2020R semiconductor detector with ultra-pure germanium CANBERRA and a 16000-channel amplitude pulse analyzer CANBERRA in accordance with the methodology for performing measurements on a gamma spectrometer with a semiconductor detector. The detector has a 500 μm thick beryllium window. The minimum activity determined for the measurement geometry for the gamma line 59 keV (241Am) was 0.1 Bq/sample, for 137 mBa (661.6 keV) – 0.4 Bq/sample. The instrumental spectra were processes in the GENIE-2000 software environment. The measurement error for 137Cs did not exceed 15%, and for 154Eu and 241Am – 25%.

The content of 90Sr in solutions was determine by the precipitation method.[22] Calculated sources of 90Sr were produced on stainless steel substrates in the form of strontium carbonate $Sr(90Sr)CO_3$ precipitate. The chemical yield of 90Sr was determine by the weight method. The activity of 90Sr was determine by β-radiometric measurements after radiochemical separation on a low-background beta radiometer UMF-1500 and on a radiometer RUB-01P, which includes a UI-38P1 measuring device complete with a BDZB-06P1 detection unit in passive lead shielding. The intrinsic background of the β-radiometer did not exceed 0.11 imp/sec. The efficiency of β-particle registration was 25%. The measurement error for 90Sr was 10–15%, depending on the activity of the sample.

Control measurements of purified water samples we performed in the laboratory of the Department of Nuclear Physics Technologies of the State Institution 'IGNS NASU' using the Atoll-3M radiometer with Spectroline software.

The analysis of the radioactive composition of LRW (trap water) at the South Ukrainian Nuclear Power Plant we performed using a SEG50P spectrometer.

Results of radioactive water purification

The research conducted we aimed at the complete removal of radionuclides and obtaining technical water that meets the requirements of current Ukrainian standards. Work on the purification of radioactive water they carried out simultaneously at two nuclear power plants. The laboratory of the Institute for Nuclear Power Plant Safety Problems of the National Academy of Sciences of Ukraine has developed four basic algorithmic approaches to the purification of radioactive water, consisting of sequential purification stages using complex sorbents based on copper and nickel Ferro cyanides, iron nanoparticles, coagulants and plasma chemistry elements (Figure 1).

At the South Ukraine Nuclear Power Plant, similar work we carried out on a pilot plant (Figure 2 & 3) developed at the Institute of Nuclear Power Plant Problems of the National Academy of Sciences of Ukraine.

During the first stage of the work, the following were determined: optimal technological modes of sorption; types and quantities of sorbents to be introduce; duration of plasma-chemical electrolysis, etc. The data obtained we taken into account during the testing of the pilot plant.

According to measurements, the radionuclide composition of LRW (trap water) prior to treatment is presented in Table 1.

Table 1 Radionuclide composition of RPV prior to treatment

Type of radionuclide	South Ukraine NPP	Chernobyl NPP
	Activity, Bq/dm³	
^{54}Mn	4 E 2	-
^{60}Co	4,3 E 2	-
^{134}Cs	2,17 E 4	-
^{137}Cs	6,73 E 2	3,2 E7
^{90}Sr	-	4,9 E6
^{241}Am	-	2,24 E4
^{154}Eu	-	1,8 E3

The most significant results we obtained using the third algorithm for radioactive water purification. The results of the third algorithm show that the sorption of radionuclides by a complex sorbent based on copper ferrocyanide, iron nanoparticles and coagulants with preliminary preparation of radioactive water by plasma electrolysis for 60 minutes and subsequent one- and two-stage treatment by plasma electrolysis in combination with sorbents for 30-45 minutes. Stirring in a magnetic mixer and the use of **Zb** series sorbents resulted in the removal of 137Cs ions to an activity of 2.48 Bq/dm³, which is 1.3E7 times lower than the cesium activity (3.2E7 Bq/dm³) of the initial radioactive water. **PK** series - removal of 137Cs ions to an activity of 129 Bq/dm³, which is 2.5E5 times lower than the activity of cesium (3.2E7 Bq/dm³). Taking into account the contamination of vials in the laboratory of the Institute for NPPSP National Academy of Sciences of Ukraine, control measurements of the activity of samples Zb4E and 1PK-3 in a clean vial on the Atol-3M device confirmed the complete purification of radioactive water from 137Cs ions to a technical state (Figure 4).

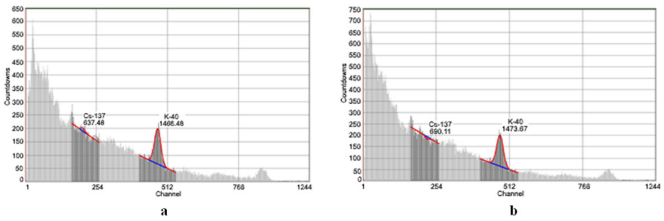

Figure 4 Spectrum of purified water samples treated with complex sorbents ZB4E (a) and 1PK-3 (b).

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Removal of 90Sr ions to an activity of 6.85E2 Bq/dm^3, under the above sorption conditions, using **PK** series sorbents, which is 17.1E3 times, lower than the strontium activity (4.9E6 Bq/dm^3) of the initial radioactive water. The use of complex sorbents of the **Zb** and **PK** series and the above conditions ensured the complete removal of 154Eu and 241Am ions.

The cycle of experiments on the purification of radioactive water shows the effect of plasma electrolysis on the destruction of organic radicals, which contributes to the additional removal of 137Cs and 90Sr ions and their sorption. The result obtained is probably because the sorption of 137Cs ions and transuranic radionuclides on Ferro cyanides and iron nanoparticles occurs in the presence of hydroxyl groups.

Table 2 shows the radionuclide composition of the treated water after purification.

Table 2 Radionuclide composition of RPV prior to treatment

Type of radionuclide	South Ukraine NPP	Chernobyl NPP
	Activity, Bq/dm^3	
^{54}Mn	-	-
^{60}Co	3.37	-
^{134}Cs	7,4	-
^{137}Cs	1,55 E 1	-
^{90}Sr	-	6.85 E2
^{241}Am	-	-
^{154}Eu	-	-

Because of cleaning the radioactive liquid waste at the South Ukraine Nuclear Power Plant using sorbents and a plasma discharge unit, it was possible to achieve complete removal of Mn-54, while the activity of Co-60 and Cs-134 decreased by two orders of magnitude and the activity of Cs-137 decreased by one order of magnitude. After cleaning, the initial total activity of liquid radioactive waste decreased by 10% from the initial activity.

This indicates the effectiveness of the method and the further prospects for its research and application. As for chemical indicators, permanganate oxidisability decreased by 7.4 times, and the content of ions determined became minimal.

Conclusions

Based on theoretical concepts of plasm chemical processes in aqueous medium, the paper presents the main design solutions for the creation of a modular laboratory unit for radioactive water treatment. The unit consists of an activation module, a chemical treatment module, and a filtration module. Data on the operation of the modules, a description of the automatic control system of the working elements and technical data of the plantthey presented. The main technological stages of water treatment we tested in the conditions of the Chornobyl NPP. The developed experimental unit allows for deep processing of radioactive waste and significant reduction of its volume. Due to the one-stage process, the plasma-chemical method is effective in solving the problems of radioactive waste disposal.

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Declaration

Data availability

Manuscript has no associated data.

Use of artificial intelligence

The author confirms that they did not use artificial intelligence technologies when creating the current work.

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Conflicts of interest

The authors declare that they have no conflict of interest in relation to this research, whether financial, personal, authorship or otherwise, that could affect the research and its results presented in this paper.

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