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Article

Application of processes stimulated by nonequilibrium plasma for large-tonnage decontamination of soils

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APPLICATION OF PROCESSES STIMULATED BY NONEQUILIBRIUM PLASMA FOR LARGE-TONNAGE DECONTAMINATION OF SOILS

The object of research is a new, potentially effective and practical process for the decontamination of radioactive soil, based on combination of plasma hydroseparation and plasma activation. The cleaning effect is ensured by the destruction of the bonds of radionuclides with soil particles due to a series of electrophysical discharges at which active particles and shock waves appear. In a designed setup, the process of plasma-chemical treatment is implemented in a plasma cell with a self-sustaining pulsating mode of burning an electric discharge, which occurs in an aqueous solution. The setup realizes a resonant increase in the intensity of shock waves, turbulence and multiple expansion of the core, such that the expansion of the plasma-liquid interface becomes a real basis for scaling up the setup. Regardless of the material of the electrodes and in a wide range of electrical conductivity (measured from 100 to $5,000 \,\mu$ S/cm), the restructuring of the combustion regime is accompanied by an increase in the size and stabilization of the luminous zone, fragmentation of bubbles, and an increase in the rate of their evacuation from the discharge zone. The main factors of such a restructuring are the channel dimensions and temperature of the solution. Various materials of the walls of plasma-chemical reactor have been tested: plexiglass, ceramics and stainless steel with the thickness of 2 mm. The maximum increase in the amplitude of resonance oscillations depends on the cell radius. A dynamic pressure, which in an individual discharge is about 5-15 mm of the water column at the mouth of the discharge, increases to 150-200 mm of the water column at the bottom of the plasma cell at resonance. An increase in efficiency is achieved by an optimal choice of the duration of the current phase and the distance between the electrodes, which is 15-30 mm. The voltage drop is 70-80 % across the spark discharge, the rest falls across the solution. The transition of the discharge to a periodic pulsating current mode with an increase in the temperature of the solution has been found. Tests on a mobile plasma-chemical facility for the process of plasma co-precipitation of radionuclides ¹³⁷Cs, ¹³⁴Cs and ⁹⁰Sr with ferrocyanide sorbents under real conditions of hydroseparation of contaminated soil from fields around the Fukushima Daiichi have shown a decrease in organic substances in water by 40 times, and of radioactivity by 75 times.

Keywords: electric discharge, water solution, electrohydraulic resonance soil decontamination, radionuclides, plasma cell, active particles.

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1. Introduction

The problem of radioactive waste (RW) is an extremely complex issue of nuclear activity in general and nuclear energy in particular [1]. The experience of eliminating the consequences of accidents at the Chornobyl and Fukushima nuclear power plants [2-7] indicates that the existing system of radioactive waste management in real conditions has shortcomings, and the technologies necessary for the urgent effective solution are not available [8–11]. There is no information about new breakthrough scientific approaches and practical measures to solve this problem in the available literature.

Huge amounts of radioactive cesium ¹³⁴Cs and ¹³⁷Cs were released during the Fukushima Daiichi accident, causing great concern due to its concentration, long half-life, and source of gamma radiation. Cesium-137 is produced in high yield by the fission of ²³⁸U and is a common pollutant from nuclear weapons testing and nuclear fuel reprocessing. The concentration and subsurface migration of ¹³⁷Cs are determined by its strong sorption and desorption from soils and sediments. The sorption and desorption of cesium is affected by the properties of the sorbent, such as mineralogical composition and cation exchange capacity, competing cations such as Na and K, and the age of the sorption complex. Understanding the Cs sorption/desorption process is important for environmental impact assessment and recovery.

Following the Fukushima Daiichi accident in March 2011, the Japanese authorities decided to carry out largescale decontamination works in the affected territory, which covered an area of more than 9,000 km². On December 12, 2019, when most of this work was completed, the researchers presented an overview of the decontamination strategies used and their effectiveness [3]. The analyses carried out focus on the future of radioactive cesium $^{134}\mathrm{Cs}$ and $^{137}\mathrm{Cs}$ and tritium in the environment because these radioactive isotopes have been released in large quantities during the accident, contaminating an area of more than $9,000 \text{ km}^2$ and water. Over the past decade, many studies have been aimed at solving the problems of soil decontamination and rehabilitation of territories affected by the nuclear accident at the Fukushima Daiichi [9, 12-16]. However, the main efforts were directed towards the desire to apply traditional approaches to the new problem, which turned out to be expensive and ineffective.

The decontamination work carried out by the Japan Atomic Energy Agency (JAEA) was the first of its kind. Nowhere in the world has decontamination work been undertaken on such a scale, and therefore there has been no real experience [17]. The main method used by the Japanese authorities to clean arable land was to remove the surface layer of soil to a thickness of 5 cm, which reduced the concentration of cesium by about 80 %. Removing the topsoil, which proved effective in cultivating the land, cost the Japanese state about 24 billion euros. However, this method generates a significant amount of waste that is difficult to handle, transport and store for decades in the immediate vicinity of the power plant, which is necessary before the waste is taken out to the final disposal sites outside Fukushima Prefecture by 2050. By the beginning of 2019, about 20 million cubic meters of waste had been generated as a result of the decontamination of Fukushima. An extensive research was performed and a program was developed by JAEA to increase the efficiency of decontamination and minimize waste generation. Attempts have been made to use various technologies, such as soil washing, phytoremediation, heat treatment and electroremediation, to rehabilitate radioactively contaminated soil.

Studies conducted in the 1960s showed that mica clay minerals very selectively sorb Cs. A good sorbent is illite, which is located in the interlayer structures of clay closer to the worn edges – «marginal wear areas» [18, 19]. The authors showed that the interaction of cesium with precipitation and soil particles can be described mechanistically in terms of ion exchange. Many chemical methods have also been tried to remove cesium from soil and it has been demonstrated that, due to the strong bonding of cesium to the lattice of clay minerals present in soil, its uptake is virtually irreversible within current technologies. This has shown a preference for physical methods such as size fractionation or wet separation (more cost effective) than chemical processing. Although considerable progress has been made in the development of methods for extracting some radionuclides from soil, such as U and Co, however, the removal of radioactive cesium from soil has been relatively inefficient or requires high energy consumption. As a result of the research, several methods have been proposed. Many of them have proven to be ineffective and some are still under development.

It is believed that there are no reliable technologies suitable for the industrial extraction of radioactive cesium particles from soil and tritium from water. Soil contamination with cesium is a worldwide problem; therefore, effective approaches to its solution are needed. Currently, there are a number of methods.

Given the recent contamination of large areas of soil with radioactive substances, such as the Fukushima accident, it is necessary to be able to treat large amounts of contaminated soil quickly. In addition, it is necessary to be able to carry out decontamination work over a large area. In this regard, it is necessary to apply treatment, which can reduce secondary waste. The study [20] proposed a new approach to recultivation of soil contaminated with Cs+ using magnetic composites to improve both the efficiency of separation of clay particles and desorption of Cs+. The process of ultrasonic dispersion and magnetic separation was first used to selectively remove clay particles from soil (Fig. 1). Fe₃O₄ nanoparticles were selectively bound to clay particles due to electrostatic attraction, which made it possible to easily separate these particles from the solution under the action of an external magnetic field. The addition of a magnetic adsorbent (Fe₃O₄@SiO₂@KTiFC, where KTiFC is titanium (IV) potassium hexacyanoferrate) significantly improved Cs+ desorption (95.2 %), preventing Cs+ re-adsorption on the sand. This idea seems to be the most tempting, but for its practical implementation it is necessary to conduct a large amount of research using new technical means.

The area around the Fukushima nuclear power plant (NPP) remains polluted because it takes an astronomical amount of money to restore the land, and the accumulated tritium water will apparently be drained into the ocean. The emerging JAEA decontamination pilot projects are the basis for the iterative development of a set of technologies and communications that will facilitate problem solving. Given this situation, let's believe that the solution lies in the development and application of new non-traditional approaches.

Based on the above analysis, the purpose of the authors' research is:

 obtaining the main regularities in the new process of excitation of resonant electro-hydraulic oscillations in the zone of an incomplete spark discharge in order to intensify the plasma-chemical effect on aqueous solutions;

- development of a universal plasma cell with a power supply system that can be easily integrated into an aqueous solution processing line.



Fig. 1. A new two-stage process for the restoration of soil contaminated with cesium using magnetic composites [20]

2. Materials and Methods

2.1. Object and hypothesis of the study. *The object of research* is a new, potentially effective and practical process for the decontamination of radioactive soil, based on combination of plasma hydroseparation and plasma activation.

The problem of decontamination of radioactive soils under real conditions can be solved by a combination of two methods: plasma-stimulated hydroseparation to intensify the disintegration process and treatment of the solution with highly active selective sorbents with their plasma activation [21-24].

2.2. Soil fractionation method. The first stage of the technology is the fractionation of the soil by washing and the separation of the finely dispersed fraction containing the largest amount of radioactive impurities. Soil leaching is one of the most common industrial technologies for the treatment of non-radioactively contaminated soils. A flushing process for tillage contaminated with U, Ra, Th, Sr and Cs radionuclides was also demonstrated. Soil washing typically separates wet particles by size because radionuclides have the highest affinity for particles that have the highest surface area to volume ratio, such as silts and clays. Up to 80 % of pollutants are concentrated in the finely dispersed fraction with a particle size of 5–150 $\mu m,$ the amount of which does not exceed 20 wt. % [21]. These particles form dispersed systems in the aquatic environment and are easily separated from larger ones. However, due to the low processing speed, this method can only be used to decontaminate soils with a low content of radionuclides. The degree of extraction of radioactive cesium using hydroseparation alone is about 16 %. Soil flushing is only effective if the radionuclides can be easily desorbed from coarse soil fractions and the contaminated soils optimally contain less than 25 % silt and clay.

This limits the use of the hydroseparation method for the decontamination of soils contaminated with radionuclides of the specified origin. Physical-chemical and reactive methods have been developed for deeper soil restoration. Any washing of the soil eventually raises the problem of decontaminating radioactive water. In addition, only soluble radioactive substances can be easily removed from the soil, while the cesium radionuclides found in the soils affected by the Chornobyl and Fukushima accidents are predominantly in the ionic form.

Soil is a complex system in which soil minerals, organic matter and microorganisms interact with each other in many ways. These soil components vary greatly in quantity, quality and properties depending on the location. Therefore, it is highly likely that the interactions of 137 Cs

with these soil components, and hence the predominant ¹³⁷Cs retention mechanisms caused by these interactions in surface soils, also differ between geographic locations.

Soil samples were taken from fields, gardens and forests around Fukushima in July 2011; the samples were then separated into three soil fractions with different mineralorganic matter interaction characteristics using the density fractionation method, and then analyzed for ¹³⁷Cs content, mineral content, and organic matter content. The results showed that 20-71 % of ¹³⁷Cs was retained in combination with relatively mineral-free fractions, predominant in the form of solid particles of organic matter, in the surface soil layers of gardens and forests [25]. However, there are also studies suggesting that the behavior of ¹³⁷Cs in mineral soil cannot be simply assessed based on the interaction of ¹³⁷Cs with clay minerals, and there are many factors that can affect the mobility and bioavailability of ¹³⁷Cs in soils. These factors, in addition to the mineralogy of clays, include the physicochemical properties of the soil. Given that Japanese soils are rich in organic matter, and soil organic matter is a dynamic component that is constantly changing, interactions between ¹³⁷Cs, minerals, and organic matter are likely to be critical in predicting the future of Fukushima-derived ¹³⁷Cs in Japan's terrestrial ecosystems.

2.3. Plasma-stimulated methods for removing contaminants. Tests of plasma coprecipitation of ¹³⁷Cs, ¹³⁴Cs and ⁹⁰Sr radionuclides with ferrocyanide sorbents in real conditions of hydroseparation of contaminated soil from fields near Fukushima [22, 23] using a mobile plasma-chemical setup showed a decrease in the content of organic matter in water by 40 times, and of radioactivity by 75 times.

Plasma treatment is accompanied by clarification of the washing water and a weakening of the effect of organic pollutants on the sorption-selective characteristics (Fig. 2). The proposed method is a potentially efficient and practical approach to the decontamination of radioactive soil.

Currently, various electric discharge reactors with nonthermal plasma for the purification of wastewater from pollutants, as well as many other applications, are being intensively studied. They are developing rapidly and based on the interaction of key reactive particles in both plasma and liquid phases [26–28]. For many specific processes, some degree of chemical and physical methods has been identified through reactive species, including hydroxyl radicals, ozone and hydrogen peroxide, as well as ultraviolet (UV) light and shock waves. Further progress in this area is determined by the possibility of more precise control of the density and diffusion into the liquid of chemically active particles formed in the plasma.



Fig. 2. Plasma treatment of contaminated soil in Fukushima Prefecture: a - mobile plasma-chemical setup; b - water before and after plasma treatment

To bring the technology of soil recovery and decontamination using plasma treatment to industrial application, several critical obstacles have to be overcome. The key to process scaling is the expansion of the plasma-liquid interface, which determines the throughput to be processed. Therefore, a reactor configuration with high productivity should significantly increase the volume of plasma formation in the liquid from a single discharge. The problem is that plasma water treatment has shown efficiency that is superior to conventional advanced oxidation methods for small treatment volumes, typically less than a few liters. For practical applications, a flow rate of 20 l/min or more is considered a usefulness threshold [29]. The throughput problem is related to the plasma-liquid interface. To treat contaminants, reactive particles must diffuse into the main solution. And their diffusion rates are slow, which means that the water flow rate for adequate contact time must be correspondingly slow. Therefore, when developing an industrial plasma reactor, measures should be taken that increase the diffusion rate and the production of chemically active substances, which can significantly increase productivity.

The fact of ¹³⁷Cs accumulation in the upper soil layer of 0-5 cm is undeniable, regardless of its composition. The sorption capacity of clays and organic matter is high enough to retain ¹³⁷Cs in the upper layers. In general, both clay and clay fractions contribute almost equally to cesium sorption. As it is mentioned above, the research results show that up to 71 % of ¹³⁷Cs can be retained in combination with relatively mineral-free fractions, predominantly in the form of solid particles of organic matter.

3. Results and Discussion

3.1. The concept. Our concept is based on solving the problem of soil contaminated with radionuclides using a hydroseparation technology that generates soil particle size fractionation and wet separation to break ¹³⁷Cs binding with soil particles. Since most of the radioactivity accumulates in the smallest particles of soil, which are clay and organic matter, then by dividing the soil into fractions, it is possible to isolate and concentrate the largest part of the contamination in one of them, namely, finely dispersed. The key element of purification is electric discharge plasma. This innovative technology for the treatment of aqueous solutions stimulated by nonequilibrium plasma belongs to the group of reagent-free treatment methods with the possibility of implementing large-scale soil decontamination processes. It is aimed at the industrial extraction of radioactive cesium particles from soil

and water with decontamination work over a large area without the formation of secondary waste.

3.2. The method essence. The cleaning effect based on the destruction of the bonds of radionuclides with soil particles is achieved by a complex of electrophysical effects of electric discharges, which generate active particles and shock waves. The main factor for process scaling is the expansion of the plasma-liquid interference which determines the throughput being processed. The configuration of the high-capacity reactor should significantly increase the amount of plasma liquid formation even at a single discharge. Therefore, when developing an industrial plasma reactor, measures should be taken to increase the diffusion rate and the production of chemically active particles. If these obstacles are overcome, the facilities should demonstrate economic expediency of the process. Such a process is implemented in a plasma cell with a self-sustaining pulsating mode of burning an electric discharge in the aqueous solution studied. Here, a resonant increase in the intensity of shock waves, turbulence and multiple expansion of the active zone are provided, which is the basis for scaling the soil decontamination facility. The plasma cells are built into a standard spiral classifier between discs below the water level.

In such a case, the active particles generated in the discharge channel are carried by the shock wave far beyond its limits, increasing the diffusion rate and, correspondingly, increasing the actual reaction volume.

3.3. Experimental setup and the procedure. To study the described effect, a setup was assembled (Fig. 3).

The setup contains a flowing electric discharge plasma cell with a Piezotronics 113B21 pressure sensor placed in it, a probe sensor for measuring the floating potential of the near-electrode plasma, a dynamic pressure sensor, a vibration sensor, an electrode system: stainless steel electrodes with a diameter of 2.5 mm in a ceramic insulator. Power is supplied from an isolating transformer of the network 220/2600 V and 50 Hz with increased dissipation. The discharge ignition phase is controlled by a triac power regulator.

All signals are recorded by the DSO3202A electronic oscilloscope. The temperature of the water at the inlet and outlet is controlled by resistance sensors DTSxx4, electrical conductivity of the solution COND 5021, pH meter D51 HORIBA. The distance between the electrodes is set in the range of 15–30 mm based on the condition that the discharge does not overlap the interelectrode gap. The electrical conductivity of the solution is 100–5,000 µSim/cm, the solution pH varies in a range of 3–12 and the value of discharge current is 0.5–1.5 A.



Fig. 3. Scheme of the setup for studying the discharge parameters with resonant amplification of a standing pressure wave

Fig. 4 shows a typical picture of the usual mode of combustion of a spark discharge in a liquid, with resonant amplification of the shock wave.

Regardless of the material of the electrodes and in a wide range of electrical conductivity (measured from 100 to 5,000 $\mu S/cm$), the restructuring of the combustion mode is accompanied by an increase in the size and stabilization of the luminous zone, fragmentation of bubbles, and an increase in the rate of their evacuation from the discharge zone. The main factors of such a restructuring are the channel dimensions and the temperature of the solution.



Fig. 4. Image of the discharge zone:



Fig. 5 shows typical oscillograms of the current and voltage across the discharge gap of the plasma cell of Fig. 3.



Fig. 5. Oscillograms of voltage (green curve) and current (yellow) in the discharge gap of the plasma cell shown in Fig. 2: a – operation without a power regulator: load current is 1.14 A; b – operation with a power regulator: load current is 0.69 A (a, b – electrical conductivity of the solution is 300 µS/cm, solution temperature is 30 °C); c – electrical conductivity of the solution is 1,500 µS/cm; d – effective current value is 0.4 A, solution temperature is 80 °C

The nature of the discharge can be judged from the instantaneous dependences of the floating potential in the near-electrode zone versus the discharge current (Fig. 6) in accordance with the scheme of Fig. 3. The fact and parameters of shock wave generation are recorded by the pressure sensor synchronously with the impact of the shock wave on the discharge current (Fig. 7).

According to estimates [30], in the resonant mode, the wall of the plasma cell receives energy equal to 14.0-17.5 % of the energy in the discharge channel E_c and 42-53 % of the work A_c expended on expanding the discharge channel.

The minimum energy transferred to the wall is 5 % of the E_c value and 15 % of the A_c value. Various wall materials were tested: plexiglass, ceramics and stainless steel with the same thickness, 2 mm. In all cases, the dependence of the maximum increase in the amplitude of resonant oscillations on the cell radius has been observed. So with a dynamic pressure, which in an individual discharge is about 5–15 mm of the water column at the mouth of the discharge, increases to 150–200 mm water column at the bottom of the plasma cell at resonance (Fig. 8).

Fig. 9 shows a typical record of vibration acceleration on the plasma cell wall at resonance.



Fig. 6. Dependence of the floating potential (green curve) between the probe and the electrode versus the discharge current (yellow) in the discharge gap of the plasma cell shown in Fig. 2. Temperature: a - 30 °C, b - 82 °C. The electrical conductivity of the solution is 1,250 µS/cm



Fig. 7. Readings of the pressure sensor with the discharge current in the operating mode (Fig. 4, a)



Fig. 8. Dependence of the pressure at the bottom of the plasma cell at resonance as a function of the cell size



Fig. 9. Typical recording of the plasma cell wall vibration at resonance: a - the vibration acceleration; b - the spectrum of vibration velocity

The result of excitation of resonant modes of electrohydraulic oscillations in the discharge chamber of the plasma cell is a powerful turbulent removal of matter from the discharge zone (Fig. 10).



Fig. 10. Turbulent removal of matter from the plasma cell discharge zone: *a* – pure water; *b* – solution of water with soil with a ratio 2:1

3.4. The optimal mode of operation. The formation of a special form of a spark electric discharge in an aqueous solution is described. An electric discharge between electrodes installed in parallel and burning on alternating current of industrial frequency (50 Hz) contains two zones: cathode and anode (Fig. 4, a). With a change in the direction of the applied voltage, they change places. Outside the discharge gaps, the current comes through the solution. Two near-electrode zones are useful from the point of view of generating active particles and shock waves. At a voltage of 1,500–2,500 V (depending on the electrical conductivity of the solution), after each phase of the current passing through zero there is a breakdown of both discharge gaps with the development of a spark discharge.

An increase in efficiency is achieved by an optimal choice of the duration of the current phase and the distance between the electrodes. It is found that an optimal distance varies from 15 to 30 mm. In this case, 70-80 % of the voltage drop across the spark discharge is provided, the rest falls across the solution.

The transition of the discharge to a periodic pulsating current mode with an increase in the temperature of the solution has been detected (Fig. 6). Apparently, this is due to the explosive nature of the release of oxygen from water in the near-channel zone of the discharge. In the near-electrode zone of the spark discharge, a directed flow is established with the removal of bubbles and the injection at its mouth. The dynamic pressure in a single discharge at a distance of several millimeters from the electrode surface is 5–15 mm of water column.

In this case, the discharge in the current phase generates periodic shock waves (Fig. 7). The picture fundamentally changes if the electrodes are placed in a cylindrical body. On condition, the amplitude of oscillations increases with an increase in pressure up to 150–200 mm of water column. The vibration of the walls, the movement and mixing of the solution grow. Besides, the external injection and the velocity (about 2 m/s) of the solution flowing out of the discharge zone also increase. The phenomenon is resonant at the fundamental frequency of 100 Hz, which modulates high-frequency oscillations (Fig. 9). Resonant pressure pumping mainly depends on the dimensions of the plasma cell channel (Fig. 8). The discharge acquires a diffuse form (Fig. 4, *a*, *b*). In the discharge zone, gas bubbles with a diameter of about 1-1.5 mm are generated, which are clearly visible, and at resonance, the magnitude of the amplitude and the rate of growth of the deformation oscillations ensure the fragmentation of the bubbles into smaller ones to a level at which it is impossible to determine their size by the optical method.

A typical power consumption of the setup is about 20 kW per processing of 1 m^3 .

Therefore, the possibility of multiple expansion of the treatment zone of the aqueous solution studied from the local energy source, i. e. the spark electric discharge burning in the plasma cell is proved. Further logical steps of practical implementation are aimed at the optimal spatial arrangement of plasma cells in the flow of aqueous solution. The ultimate goal is to effectively purify the entire volume of running water.

3.5. The optimal mode of operation. This paper shows the results of developing a new process of resonant excitation of electrohydraulic oscillations in the zone of spark discharge as applied to decontamination of radioactive soil, based on a combination of plasma hydroseparation and plasma activation.

However, the effective practical implementation of this process will require a deep understanding of a number of limitations and features. Namely:

hydraulic vibrations in a water solution cause vibrations of structural elements in the operating mode.
Measures are needed to ensure the long-term performance of the structure as a whole;

electrical discharge causes erosion of the electrodes.
It should be slowed down as much as possible by the optimal choice of operating parameters, material and cooling of the electrodes.

The processes associated with shock waves in a liquid and plasma treatment using generated active particles are well known and applied separately in practice. In our case, there is a combined effect on impurities in the volume of an aqueous solution from one source. Synergistic effects are to be expected. The mechanism of such effects is a topic for further research.

4. Conclusions

A new process of resonant excitation of electrohydraulic oscillations in the zone of spark discharge has been studied. This provides a combined improved pulse-mechanical and plasma-chemical effect on the material being processed. In the mode of resonant excitation of hydraulic oscillations in the plasma cell, the dynamic head increases from 5-15 to 150-200 mm of water column. This provides powerful turbulent mixing in the treated volume, which removes diffusion restrictions for plasma-chemical reactions.

A universal plasma cell has been developed based on this effect with an energy supply system that can be easily integrated into the system for processing aqueous solutions. The cleaning effect based on the destruction of the bonds of radionuclides with soil particles is achieved with a complex of electrophysical effects of electric discharges: generated active particles and shock waves.

The proposed method is a potentially efficient and practical approach to the decontamination of radioactive soil and wastewater in large quantities. A typical power consumption of the setup is about 20 kW per processing of 1 m^3 .

Conflict 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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