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Characteristics of Bipolar Nanosecond Discharges in Air Formed in the Electrode System “BLADE-SURFACE of Nonmetallic Liquid -BLADE”

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Abstract

The design of the device for producing a high-current, bipolar nanosecond discharge over the surface of a non-metallic liquid (water, electrolytes, alcohols, etc.) in air is given. Air pressure is ranged from 5 to 101 kPa. The distance between the tip of the blade and the surface of water or liquid (5% solution of copper sulfate in distilled water) was 4 mm, and the distance between parallel metal blades was 40 mm. The conditions for uniform plasma overlapping of the electrolyte surface between the metal blades are established. The spatial, electrical, and optical characteristics of the discharge are investigated. It is shown that the discharge under study allows obtaining colloidal solutions of copper nanoparticles in distilled water in a macroscopic amount (1 liter or more). The developed reactor is of interest for use in poisonous chemical solution disinfection systems, solutions based on dangerous bacteria and viruses for which the use of traditional systems with a point spark discharge or a barrier discharge becomes ineffective. The reactor is also promising for the synthesis of colloidal solutions of transition metal oxide nanoparticles from solutions of the corresponding salts. These solutions can be used in micro-nanotechnology and for antibacterial treatment of plants in greenhouses, processing of medical instruments and materials.

Keywords: Nanosecond discharge, Distilled water, Copper sulfate, Electrolytes, Alcohols, Colloidal solutions, Copper nanoparticles.

Introduction

Recently, intensive use of various gas-discharge sources of ultraviolet (UV) radiation in medicine, microbiology, chemistry, sanitation and ecology has been observed [1–9]. Promising for these purposes is the use of “cold” plasma jets, which are also formed mainly in different gas-discharge devices [10]. When using such UV emitters, including open ones, operating in atmospheric air and not having a quartz shell, for the action of biologically or chemically active solutions on the surface, the distance between the lamp and the liquid is several centimeters. The duration of the radiation pulses of such gas-discharge emitters is tens to hundreds of microseconds, and the operating voltage of their power sources does not exceed 10-11 kV. This leads to a decrease in the pulsed power of radiation compared with nanosecond lamps. Simultaneously, the fluxes of active particles (such as hydroxyl radicals - OH, hydrogen atoms - H, and oxygen atoms -O from the plasma are eliminated because they do not have time to

penetrate the surface of the treated fluid during their lifetime (which does not exceed 10 ns). The use of jets of “cold” plasma for surface treatment of biologically and chemically active solutions is often limited to the small working surface of the plasma jet. Therefore, the development of gas-discharge devices to obtain near-surface plasma of a relatively large area, which can be used for treating surfaces of non-metallic liquid media in air to solve microbial and medicine issues that are not able to solve by known sources of UV radiation and plasma, is relevant [11,12].

In previous research [13], the characteristics of the discharge in the air above the water surface are given. Here, a high-voltage tip electrode was used, to which a high-voltage pulse of negative polarity was applied, and a flat, grounded metal electrode was placed in water near the opposite end of an opened discharge cell. The voltage pulse had amplitude of up to 20 kV and duration of 5-1000 μ s. The distance between the electrodes reached 20 cm, and the cathode distance from the water surface was 0.1-1 cm. The discharge channel had a width of 0.3-0.5 cm and its main part was parallel to the water surface and separated from it by a fraction of a millimeter. Since the plasma channel had a small width, it could not cover a large surface area of water. With a current pulse duration of tens of microseconds - 1 ms, heating of the liquid, which is undesirable when working with biosolvents, can be essential.

Therefore, it is important to study the formation of nanosecond discharges over the surface of non-metallic liquids in air, allowing us to obtain large width discharges in the system of electrodes based on metal blades. For these purposes, the technique of forming plasma electrodes developing on the surface of solid dielectrics (fiberglass laminate, leucosapphire, etc.) can be used. Such electrodes have been used in high-pressure lasers pumped with a transverse volume discharge, in particular, on excimer molecules - 308 nm XeCl (X-B) [14]. The main disadvantage of such plasma electrodes based on solid dielectrics is their small service life associated with the destruction of a dielectric by a powerful discharge in an aggressive gaseous medium. In our experiments, it was proposed to obtain a nanosecond discharge over the surface of liquid nonmetallic media, which tend to self-recover in the course of operation, which significantly increases their service life. The aim of the work was to develop a gas-discharge reactor based on a surface discharge with a liquid nonmetallic electrode for applications in medicine, biology, agriculture and nanotechnology.

This article presents the results of a study of the spatial, electrical, and optical characteristics of a high-current nanosecond discharge over the surface of distilled water and a solution of CuSO_4 salt in distilled water, which was formed in air at pressures of 5–101 kPa.

Materials and Methods

Technique and conditions of the experiment

The use of steel blades in the system for obtaining a high-current nanosecond discharge in air at atmospheric pressure when the discharge in the form of 12 plasma sheets was closed to the surface of distilled water or electrolytes was described in [15–16]. But in this case, the ratio of the area of the overlapped plasma to the total area of the electrode based on the system of steel blades did not exceed 0.3-0.4. The characteristics of the discharge and the system of electrodes, consisting of two steel blades installed under an angle varying during the experiment to each other at some short distance from the surface of non-metallic liquids, were described in [17–18]. In this case, it was also not possible to completely overlap the surface of liquid medium by plasma, which was located under the steel blades. As in the first case, and in the second case, colloidal solutions based on nanoparticles of copper, zinc and iron oxides in a volume of about 10^3 cm^3 were synthesized in such gas-discharge plasma-chemical reactors. Therefore, in this study as a basis to create a surface discharge, the design and power supply scheme for the nanosecond surface discharge of plasma electrodes of high-pressure lasers pumped by a transverse volume discharge was chosen [14]. A high-current nanosecond discharge at air pressures in the range of 5–101 kPa was ignited in the “blade – electrolyte surface – blade” electrode system in the discharge module, the circuit of which is shown in **Figure 1**.

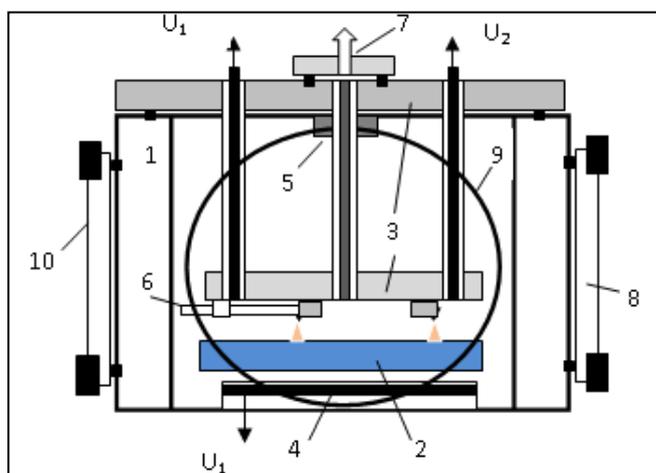


Figure 1. 1 - discharge chamber made of plexiglass, 2 - a cuvette with a non-metallic liquid (diameter - 120 mm, height - 16 mm), 3 - platforms with an electrode system “blade-surface of non-metallic liquid-blade”, 4 - grounded metal plate, 5 - system adjusting the height of the electrodes above the surface of the liquid ($h = 0-15 \text{ mm}$), 6 - the system for regulating the distance between steel blades ($L = 40-80 \text{ mm}$), 7 - the system for pumping out and gases inlet, filling the cuvette with liquid and adjusting the system of electrodes in height, 8 - quartz window for recording spectra and radiation from the plasma discharge gap, 9 - glass

window for visual, photographic, video surveillance, 10 - window for adjustment and access to the system of adjusting the distance between the blades.- U_1 -, U_2 - connection points for the leads from a high-voltage nanosecond source of the discharge ignition.

The electrode system was installed in a sealed discharge chamber made of dielectric. The air pressure was varied in the range of 5-101 kPa for a liquid electrode based on a 5% solution of copper sulfate ($CuSO_4$) salt in distilled water and was 101 kPa for an electrode based on distilled water. The distance between the surface of the non-metallic liquid and the tips of the thin steel blades was 4 mm, and the distance between the parallel steel blades was 40 mm. The level of the working fluid was kept constant with the help of a special drip system and an additional tank with liquid. The discharge cuvette was made of organic glass and could be pumped out with a vacuum pump to a residual pressure of 10 Pa. The overpressure of the gases in the cuvette could reach 200 kPa. The electrodes were made of steel blades, 0.1 mm thick. The discharge cell was installed in the screen of their metal grid, which reduced the influence of the electromagnetic fields of the discharge ignition system on the recording system of its electrical and optical characteristics.

The characteristics of a nanosecond surface discharge in the system of electrodes «blade – electrolyte surface – blade» were studied using an experimental setup described in [18–19]. A high-current nanosecond discharge on the surface of the liquid was ignited using a high-voltage source of bipolar nanosecond voltage pulses with a resonant recharge of the storage capacitor with a capacity of 1.54 nF and a hydrogen thyatron switchboard. The amplitude of the voltage pulses of the same polarity on the voltage input of the high-voltage modulator could vary in the range of 10-25 kV. The amplitude of the main maximum of the current pulse reached 100-170 A. The repetition frequency of nanosecond voltage and current pulses was in the range of 35-1000 Hz.

Radiation of surface discharge plasma was recorded in the spectral range $\lambda = 200-665$ nm using a spectrometer with a diffraction grating of 1200 lines / mm - MDR-2. In the output of the spectrometer, a photomultiplier tube FEU-106 connected to a DC amplifier was used to detect radiation. The signal from the amplifier was fed to an analog-to-digital converter and then fed to a personal computer for processing. To estimate the energy contribution to the discharge plasma, current and voltage oscillograms were recorded using a 6-channel wideband oscilloscope 6LOR-04. The voltage pulses on the electrodes were applied through a capacitive divider to one of the channels of the oscilloscope, and the discharge current pulses were measured using a calibrated Rogowski belt on the other

channel of the oscilloscope. The synchronization of the pulses of the discharge power supply with the launch of the 6LOR-04 oscilloscope was carried out using a G5-15 pulse generator. The disadvantage of the proposed device is the use of static blades as electrodes. During prolonged operation of the reactor, the erosion of steel electrodes occurs and the discharge loses a little in uniformity. In the industrial version of the device, the electrodes must be made of refractory alloys based on tungsten, molybdenum or tantalum, as in high-pressure dischargers in systems for the formation of nanosecond pulses at a current amplitude level of hundreds of amperes.

Results and Discussion

Spatial, electrical and spectral characteristics of surface discharge

In each experiment on the ignition of a surface discharge in the air with a liquid nonmetallic electrode, together with a study of its spectral characteristics, its spatial and electrical characteristics (current and voltage waveforms) were also recorded. Using the oscillograms of the voltage across the discharge gap and the discharge current, we determined the pulsed electric power of the discharge and the energy input to the plasma per pulse (Figure 2).

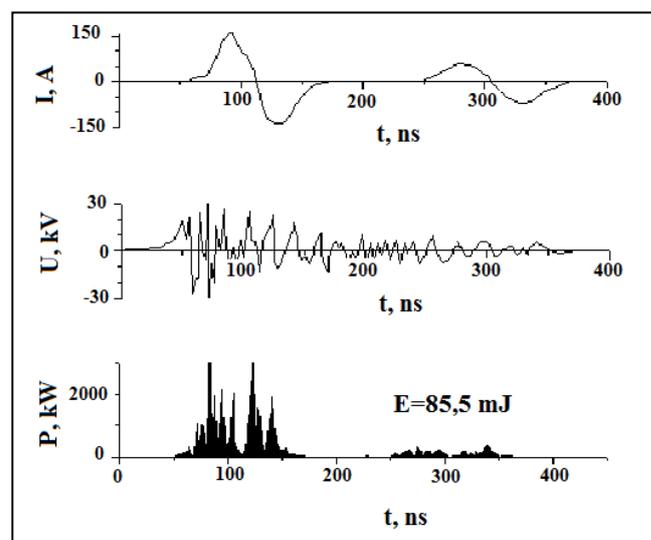


Figure 2. The dependence of the pulsed electric power of the discharge on time, oscillograms of voltage and current of a bipolar nanosecond discharge above the surface of a 5% solution of $CuSO_4$ salt in distilled water at the air pressure in the discharge chamber equal to 6.5 kPa.

The dependence of the pulsed electric power of the discharge on time was obtained by graphically multiplying the oscillograms of voltage and current, and the magnitude of the energy input to the discharge per pulse was determined by integrating the discharge power of the discharge over time.

For the discharge of atmospheric pressure over the surface of distilled water, the analysis of the voltage and current waveforms showed that the current pulse has an amplitude of up to 50 A and a duration of about 50 ns and is formed under the action of a voltage pulse with an amplitude of 30 kV, which had a duration of 30 ns. Due to the mismatch of the output impedance of the pulsed voltage generator on the voltage oscillograms in the conditions of these experiments, an oscillatory structure was observed. The maximum value of the pulse electric power of the discharge during the first 30 nanoseconds since the moment of ignition of the discharge reached 1 MW. In one discharge pulse, the energy input to the plasma was 30 mJ.

With a decrease in air pressure in the discharge module to 6.5 kPa, the energy input to the plasma per pulse-increased threefold (**Figure 2**). At the same time, the amplitude of the current pulse reached 150 A for a duration of 70 ns. The voltage pulse had an amplitude of 20 kV and a duration of 25 ns. The pulsed electric power of the discharge reached 3 MW. **Figures (3 and 4)** show images of a nanosecond discharge on the surface of distilled water at an air pressure of -101 kPa and on the surface of a 5% solution of a copper sulfate at air pressure of 6.5 kPa.



Figure 3. The image of the discharge glow above the surface of distilled water at an air pressure of 101 kPa.

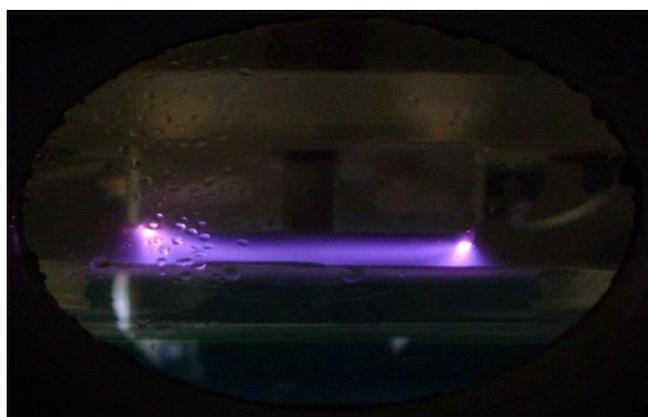


Figure 4. Image of the discharge above the surface of a 5% solution of a CuSO_4 salt at an air pressure in the discharge chamber of 6.5 kPa.

In the first case, the discharge consisted of a fairly uniform set of streamer channels, which were observed against a weak background of a diffuse surface discharge. The nanosecond discharge in air of atmospheric pressure in the system of electrodes "blades - surface of distilled water" had a similar appearance [15]. The brightness of the plasma glow slightly decreased in the direction from the tip of the steel blade to the center of the discharge gap, which is probably due to this distribution of the specific electric field strength of the bipolar discharge. When the air pressure in the discharge chamber was reduced to 5–25 kPa (**Figure 4**), the surface discharge acquired a diffuse appearance and completely covered the liquid surface between the steel blades, which were installed parallel to each other.

Figure 5 shows the emission spectrum of the discharge plasma above the surface of distilled water at an air pressure of 101 kPa. The results of identification of this emission spectrum are presented in **Table 1**. As can be seen from **Table 1**, the bands of the second positive system of the nitrogen molecule prevail in the plasma emission spectrum, and the main part of the radiation was concentrated in the spectral range of 280–390 nm.

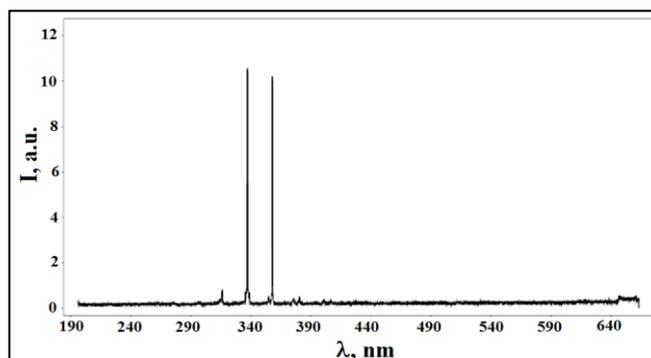


Figure 5. The emission spectrum of the discharge above the surface of distilled water at an air pressure in the discharge chamber equal to 101 kPa.

Table 1. The results of the interpretation of the emission spectrum of the discharge formed above the surface of distilled water ($p = 101$ kPa).

Wavelength, nm	Molecule	v'	v''	$E(v')$, eV
297.68	N_2	2	0	0,128
315.93	N_2	1	0	0,376
337.13	N_2	0	0	0,128
357.69	N_2	0	1	0,128
371.05	N_2	2	4	0,619
375.54	N_2	1	3	0,376
380.49	N_2	0	2	0,128
394.3	N_2	2	5	0,619
399.84	N_2	1	4	0,376
405.94	N_2	0	3	0,128

The absence of hydroxyl radicals bands in the plasma emission spectrum indicates that the density of water vapor in the plasma is low, that is, the plasma heats the water surface a little and can be considered “cold” (not more than 50° C). This is important for biological and medical applications of the investigated discharge. Compared with the known plasma jets [10], this source can simultaneously process relatively large surfaces of non-metallic solutions.

The characteristic plasma emission spectrum of a nanosecond discharge above the surface of a 5% solution of CuSO₄ salt in distilled water at an air pressure of 6.5 kPa (Figure 6) differs fundamentally from the spectrum for discharge above the water surface by the presence of relatively intense spectral lines of the copper atom that enters the plasma from the solution.

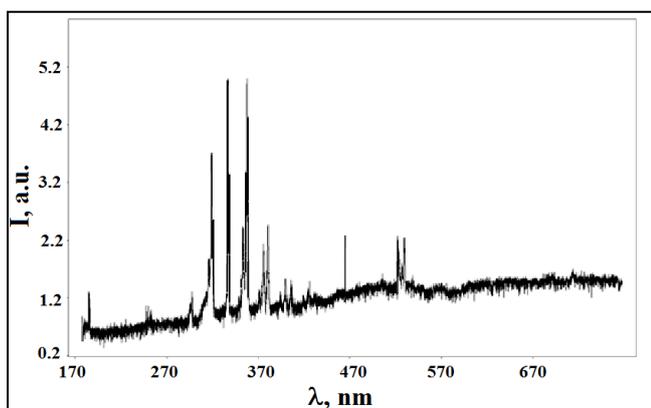


Figure 6. The emission spectrum of the discharge above the surface of a 5% solution of CuSO₄ salt in distilled water at an air pressure in the discharge chamber equal to 6.5 kPa.

The results of the identification of the spectrum shown in Figure 6 in part of the radiation objects entering the discharge from the surface of the solution are summarized in Table 2. But even in this case, the bands in the second positive system of the nitrogen molecule, located in the spectral range of 280-390 nm, were major in the spectrum. After treatment the surface of the copper sulfate solution for 2-3 hours at a pulse repetition rate of 100-150 Hz, the solution changed its color from blue to green. This indicates the complete transformation of copper cations and anions

Table 2. The results of the identification of objects of the radiation of the discharge plasma above the surface of a 5% solution of copper sulfate at air pressure of 6.5 kPa.

Wavelength, nm	Atom	E _{low} , eV	E _{up} , eV	Lower term	Upper term
249.215	Cu I	0	4,97322	3d ¹⁰ 4s ² S _{1/2}	3d ⁹ (² D)4s4p(³ P°) ⁴ P _{3/2}
319.41	Cu I	1,642133	5,52246	3d ⁹ 4s ² ² D _{3/2}	3d ⁹ (² D)4s4p(³ P°) ⁴ D _{3/2}
353.038	Cu I	1,642133	5,15286	3d ⁹ 4s ² ² D _{3/2}	3d ⁹ (² D)4s4p(³ P°) ⁴ F _{5/2}
405.112	Cu I	5,724385	8,7842	3d ⁹ (² D)4s4p(³ P°) _{3/2}	3d ⁹ 4s(³ D)4d ² P _{3/2}
525.20	Cu I	5,52246	7,88305	3d ⁹ (² D)4s4p(³ P°) ⁴ D° _{3/2}	3d ⁹ 4s(³ D)5s ⁴ D _{3/2}
529.25	Cu I	5,394745	7,73659	3d ⁹ (² D)4s4p(³ P°) ⁴ D° _{7/2}	3d ⁹ 4s(³ D)5s ⁴ D _{7/2}

(SO₄)⁻ in solution into a colloidal solution based on copper oxide nanoparticles (Figure 7).



Figure 7. Images of copper sulfate solutions in distilled water before and after plasma treatment of a nanosecond discharge in air at atmospheric pressure.

The green color of the solution corresponds to the radiation at the wavelength of the plasmon resonance of copper oxide nanoparticles. This makes it possible to recommend the introduction of small copper sulfate additives in a variety of biologically active solutions in order to enhance the bactericidal effect of UV illumination of the surface of the solution and of copper oxide nanoparticles with a strong bactericidal effect [20, 21].

Conclusion

Thus, the conditions for obtaining a uniform discharge in the air above the surface of non-metallic liquids are established; the maximum pulsed discharge power is in the range (1-3) MW with an energy input to the plasma in one pulse (30-900) mJ; in the emission spectra of the plasma, the emission of the second positive system of nitrogen molecule bands in the spectral range of 280-390 nm prevails, but for the plasma on the surface of the copper sulfate solution, a relatively intense emission of copper atoms entering the discharge from the surface of the solution was detected; treatment of the salt of copper sulfate with surface-discharge plasma allows the synthesis of colloidal solutions based on nanoparticles of copper oxides, promising for use in nanotechnology, microbiology and medicine.

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