

ELECTRODYNAMIC AND ELECTROCHEMICAL CHARACTERISTICS OF STREAMER CORONA DISCHARGE IN AN ELECTRODE SYSTEM WITH WATER CATHODE

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Experimental investigation of the electrodynamic and electrochemical characteristics of streamer discharge with a water cathode in the non-stationary burning mode has been carried out. It was found that the discharge current is a quasi-periodic sequence of pulses. It was obtained that with an increase in applied voltage the amplitude and frequency of current pulses also increase. Discharge current pulses have a time duration of 200...300 ns, while the frequency of current pulses is in the range of 9...25 kHz, and depends on the discharge gap distance. Investigation of the electrochemical parameters of the water cathode showed a change in pH level and electrical conductivity (EC) of water under discharge treatment. A decrease in pH level of water from 6.9 to 5 was fixed. At the same time an increase in EC from 10 to 70 $\mu\text{S}/\text{cm}$ was observed. Such changes are connected with the formation of new ions in water under streamer discharge treatment.

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INTRODUCTION

At the present time, studies of discharge with a liquid electrode are carried out in many laboratories around the world, which is due to the widespread use of this type of discharge in plasma-chemical technologies for disinfection of water solutions [1] and other technologies for solving environmental problems. One of the most important environmental problems which takes place in many countries of the world is the pollution of water supply sources, which leads to a progressive decrease in the efficiency of supplying people with drinking water. The relevance of research on discharges with a water electrode is due to the development and implementation of modern technologies for water purification and disinfection that are alternative to chlorination process [2, 3]. One of the perspective types of plasma-chemical technologies for water treatment is a corona discharge with a liquid electrode in a strongly inhomogeneous electric field [4]. Depending on the applied voltage value, its polarity and time duration, as well as the type of the gas, various modes of corona discharge burning occurs. In particular, for short discharge gaps (up to 10 cm) with a “needle-plane” type of electrode system, when a DC high-voltage is applied to the electrodes, a quasi-periodic mode of discharge current occurs. In electronegative gas, when positive DC high voltage applied to the needle electrode, the frequency of current pulses equals $\nu \sim 10^4$ Hz, and such a discharge is called streamer corona discharge [5]. In corona discharges, conditions for the formation of a strongly non-equilibrium gas state with a large difference between the electron temperature T_e and the gas temperature T_g ($T_e \gg T_g$) are easily realized [6], which is a necessary condition for increasing the rate of plasma-chemical reactions. In particular, this is necessary for the generation of active species and free radicals in low temperature plasma of gas discharge. The presence of such species significantly increases the efficiency of some plasma-chemical technologies. Thus, a streamer corona discharge in an electrode system with a liquid cathode is effectively used in technologies for disinfection

and purification of aqueous solutions and in hybrid “gas-liquid” plasma-chemical reactors for ozone synthesis [7]. It also used in technologies for changing the chemical composition of liquid solutions under discharge treatment. It is worth noting the work [8]. In that work the effectiveness of pulsed streamer discharge for water purification systems from harmful impurities has been investigated. Among the research results, the most important result is the effective purification of wastewater from phenol impurities.

In current work, the aim of the research is to identify and investigate stable discharge burning modes in a streamer discharge with water cathode. Only stable discharge burning modes, without transition to the spark breakdown, can be used for further implementation in plasma-chemical technology for water purification. Control, predictability and stability of discharge parameters in a plasma-chemical reactor are the main criteria for maintaining high efficiency of reactor, which is necessary for further implementation in a technological device. For this purpose, it is necessary to carry out experimental studies of the electrodynamic and electrochemical characteristics of a streamer discharge. Investigation of electrodynamic characteristics will include measurements of the current-voltage characteristics, registration of waveforms and amplitude-frequency characteristics of discharge current pulses. Investigation of electrochemical parameters will include measurements of the pH level and electrical conductivity (EC) of the water cathode under the discharge treatment.

Investigation all of these characteristics is necessary for more effective use of streamer discharge for disinfection of liquid solutions and their purification from harmful impurities.

EXPERIMENTAL SETUP

Experimental study of streamer discharge with a liquid cathode was carried out on the specialized research stand.

The experimental setup schematically shown in Fig. 1.

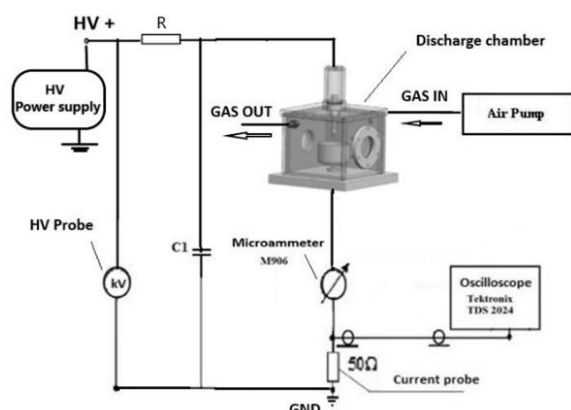


Fig. 1. Experimental setup. Air Pump – membrane compressor; C1 – capacitive voltage filter 1000 pF; R – ballast resistor 1 M Ω ; M906 – microammeter, oscilloscope – Tektronix TDS 2024, current probe 50 Ω ; C-196-HV voltmeter

To initiate discharge burning a stabilized high-voltage DC power supply with a maximum output voltage of 30 kV was used. The applied voltage was measured with a C-196 HV voltmeter. The average discharge current was measured using a M906 microammeter. To prevent voltage pulsations a high-voltage capacitor C1 with a capacity of 1000 pF was connected to the electrical circuit. An electrode system of the “needle-plane” type with a needle metal anode and a water cathode was used in the experiments. The anode was a thin tungsten needle with a radius of curvature at the apex $r \sim 30$ μm . The cathode was a metal disk with a diameter of $\varnothing 30$ mm, located at the bottom of a glass beaker, which was filled with distilled water. The volume of water was 66 ml. The cathode (liquid electrode) was at ground potential. The experiments were carried out at discharge gaps 5, 7 and 10 mm. The laboratory air was injected into the discharge chamber by using an AirPump membrane compressor. The gas flow rate was measured by a rotameter and was $Q = 2$ l/min. Measurements of electrochemical characteristics (pH level, electrical conductivity) of the water electrode were carried out during the experiment by using a pH meter PH-2603. Distilled water with an initial EC = 10 $\mu\text{S}/\text{cm}$ was used in the experiments.

To obtain waveforms and measure the temporal and amplitude characteristics of the discharge current pulses, current probe (shunt) 50 Ω was used. The signal from the shunt was analyzed with a digital oscilloscope Tektronix TDS-2024. The bandwidth of the oscilloscope was 200 MHz, sampling frequency – 1 GHz.

EXPERIMENTAL RESULTS

It is known, that streamer corona discharge in air at atmospheric pressure can burn in different modes [9]. It primarily depends on the applied voltage value. At the beginning the discharge burns in a stationary DC mode. This is so-called positive DC corona discharge. Further, with an increase of applied voltage, the discharge transits into a non-stationary mode, in which the discharge is characterized by the streamers that do not reach the cathode and become destruction in the discharge gap. The discharge current has a quasi-periodic type. With

further increase in the applied voltage, a non-stationary mode is also realized and streamers begin to cross the entire discharge gap. When streamers cross the entire discharge gap, a secondary ionization wave begins to propagate from the anode. Then with increase of applied voltage, the discharge passes into a spark breakdown. Thus, stable burning of the discharge in the streamer mode is possible only in a fixed range of applied voltage, at which the discharge does not pass into a spark breakdown.

When streamer discharge burns above the water cathode, it is possible to observe the implementation only non-stationary burning mode. In this mode, only streamers closing the discharge gap were observed. Below in Fig. 2, the current-voltage characteristics of a streamer discharge for different interelectrode distances are presented.

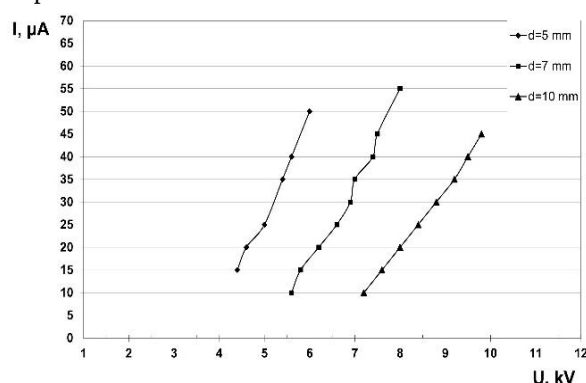


Fig. 2. Current-voltage characteristics of a streamer discharge with a water cathode. Discharge burns in non-stationary streamer mode

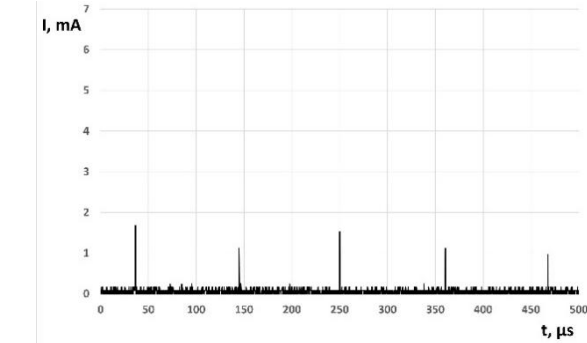
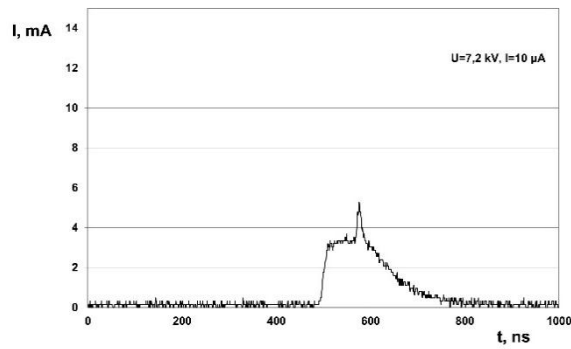
Fig. 2 shows that the streamer discharge with a water cathode burns in a limited applied voltage range (from the discharge ignition voltage to the spark breakdown voltage). That voltage range depends on the interelectrode distance, and its width is equal 1.6...2.6 kV. At the same time the discharge occurs only in the mode with streamers that closing the discharge gap. After reaching the threshold voltage value, the discharge passes to spark breakdown.

The current-voltage characteristic is an average characteristic of the discharge burning process. Therefore, to study the physical processes responsible for different modes of discharge burning, it is necessary to analyze the waveforms of current pulses for each point of the current-voltage characteristic.

To study the physical processes responsible for different modes of discharge burning, the waveforms of current pulses and the dynamics of streamers in the discharge gap were analyzed. At the same time, the applied voltage U and the average discharge current I_{av} were measured. Waveforms of streamer discharge current pulses in laboratory air are presented in Fig. 3. The needle anode has a special mechanical treatment, which provides a stable discharge burning above the water cathode. The discharge gap $d = 10$ mm.

Waveforms (1) and (2) in Fig. 3 correspond to the non-stationary mode of discharge burning with streamers that closing the discharge gap. The discharge current in the non-stationary mode of burning has a quasi-periodic type.

(1)



(2)

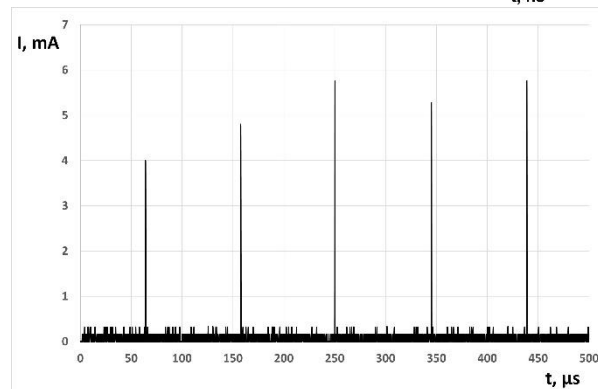
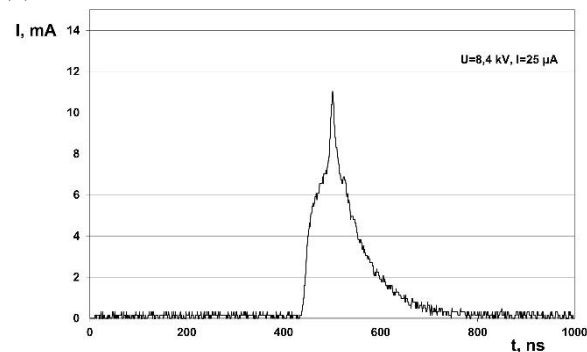


Fig. 3. Waveforms of current pulses and sequences of current pulses of the streamer corona discharge with water cathode. Discharge gap $d=10$ mm.

(1) Applied voltage $U=7.2$ kV, average discharge current $I_{av}=10$ μ A, current pulses frequency $\nu \sim 9$ kHz;

(2) $U=8.4$ kV, $I_{av}=25$ μ A, $\nu \sim 11$ kHz

The appearance of current pulses against the background of the DC current component and the establishment of a quasi-periodic pulse mode corresponds to transition to the streamer mode of discharge burning. At the same time, with an increase in the applied voltage, both the frequency of current pulses and their amplitude increase. It was also found that with a decrease in the inte-

re-electrode distance, the characteristic frequency of discharge current pulses increases. The characteristic range of frequencies of discharge current pulses that observed in the experiments is: 1) for discharge gap $d = 10$ mm, the frequency range $\nu \sim (9 \dots 13)$ kHz; 2) $d = 7$ mm, $\nu \sim (12 \dots 15)$ kHz; 3) $d = 5$ mm, $\nu \sim (20 \dots 25)$ kHz.

It should be noted that observed increasing of the frequency of current pulses with decreasing of discharge gap was also found in work [9]. In that work the non-stationary mode of positive corona discharge in the "needle-plane" electrode system with a metal cathode has been investigated.

Further, Fig. 4 shows the waveforms of streamer discharge current pulses for the discharge gap $d = 10$ mm at different applied voltages.

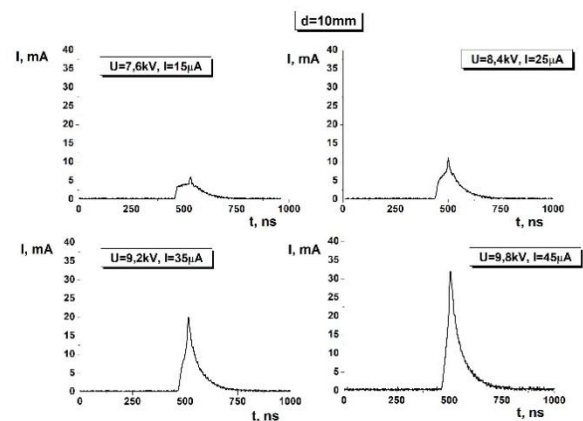


Fig. 4. Waveforms of streamer discharge current pulses. Discharge gap $d=10$ mm. Non-stationary discharge burning mode

From the waveforms of current pulses shown in Fig. 4, it is clear that the discharge is realized only in the burning mode with streamers that closing the discharge gap. The amplitude of the current pulses increases with increasing applied voltage. The waveforms also show that the characteristic time duration of the discharge current pulse is $\tau \sim (200 \dots 300)$ ns. The duration of a single current pulse is much shorter than the period of the current pulses $T \sim (50 \dots 100)$ μ s. This is due to the differences in the physical processes that occur during the pulse and in the time interval between the discharge current pulses. The current pulse is caused primarily by electron avalanche processes and the processes of formation and further development of the streamer. These processes occur in a time from 10 ns to several hundred nanoseconds. While the pulse period (~ 100 μ s) is determined by the time during which charged particles (mainly negative and positive ions that accumulated during the streamer development), are removed from the discharge gap. It is worth noting the identity of the discharge burning modes in both the electrode system with a metal cathode and the system with a liquid cathode.

In this work also investigated the influence of discharge on the electrochemical characteristics of the aqueous cathode. To do this, during the experiment, the electrical conductivity and pH level of the water cathode were periodically measured with a pH meter.

It is known that the acidity of aqueous solutions is measured by the pH indicator. The pH value shows the concentration of hydrogen ions $[H^+]$ in aqueous solu-

tion, which is expressed on a logarithmic scale. The pH value shows the degree of acidity of an aqueous solution. For convenience, a logarithmic scale is used to determine the concentration. For this purpose, the pH function was used, which has the expression:

$$\text{pH} = -\log [\text{H}^+],$$

where $[\text{H}^+]$ is the concentration of hydrogen ions, defined in mol/l. Since the pH scale is logarithmic, a change in the pH value by one unit corresponds to 10 times change in the concentration of hydrogen ions $[\text{H}^+]$. It is generally accepted that at $\text{pH} \sim 7$ the water solution corresponds to a neutral solution, at $\text{pH} > 7$ the solution is alkaline, and at $\text{pH} < 7$ the solution corresponds to an acidic solution. In the experiments, the pH level of the aqueous cathode was measured under the treatment by streamer corona discharge in the non-stationary burning mode. The time interval between measurements $\Delta t = 5$ min. The duration of the experiment is 25 min. Below in Fig. 5 the dependence of the pH level of water on the time of the discharge treatment is shown.

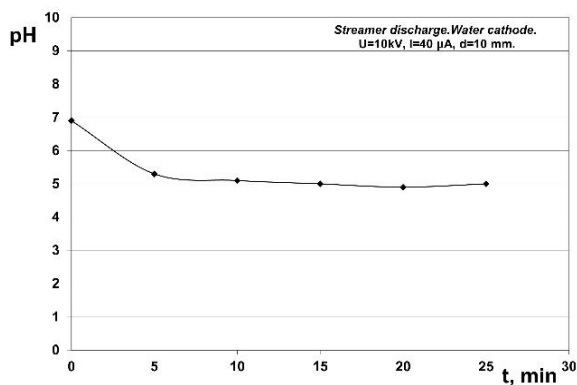


Fig. 5. pH level of the aqueous cathode under the streamer corona discharge treatment. Non-stationary burning mode. Discharge gap $d=10$ mm. Applied voltage $U=10$ kV, average discharge current $I_{av}=40$ μA .

Initial pH level of water cathode $\text{pH}=6.9$

From Fig. 5, it is clear that in the first 5 minutes of the discharge treatment, the pH level decreases from 6.9 to 5.3. After that, the pH level remained unchanged throughout the experiment, and equals $\text{pH} \sim 5$. Thus, a decrease in the initial pH level of water is observed, and the solution tends to become acidic. It can be assumed the formation of various nitrogenous species NO_x in the streamer discharge in air. That species are well soluble in water and causes formation of nitric acids solution. Observed decreasing in pH level during the discharge treatment of water solution is confirmed in other studies of the low-temperature plasma discharges with liquid electrodes. In work [10], a study of the low-temperature arc discharge (Non-Thermal Gliding Arc Discharge) with liquid electrode has been made. It was found that the decrease in the pH level of the solution is directly related to the increase in the concentration of nitric oxide NO and the formation of a solution of nitrous (HNO_2) and nitric (HNO_3) acids. This process also implements with observed increase in the electrical conductivity of the solution.

The electrical conductivity of an aqueous solution is the ability of a liquid to conduct an electric current, measured in $\mu\text{S}/\text{cm}$. This characteristic is an indicator

of the presence of ions in the water that can transfer an electric charge. Such conductivity is different from electronic conductivity because it occurs both with chemical transformations.

In the experiments, the EC of the water cathode was measured under streamer discharge treatment in the non-stationary burning mode. Below, Fig. 6 shows the EC value of water depending on the time of the streamer discharge treatment. The initial EC of water is 10 $\mu\text{S}/\text{cm}$.

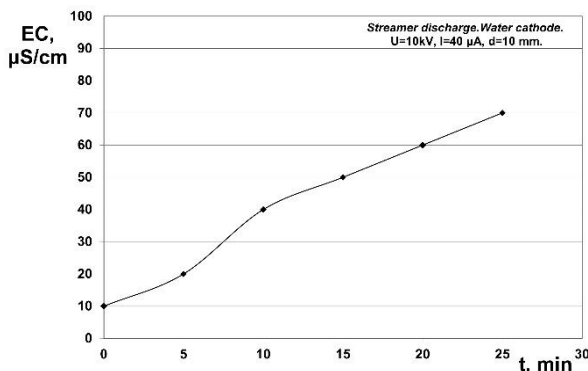


Fig. 6. Electrical conductivity of the water cathode under streamer discharge treatment. Discharge gap $d=10$ mm. Applied voltage $U=10$ kV, average discharge current $I_{av}=40$ μA

From Fig. 6, it is seen that the EC of the water cathode increases with time, and after 25 min of discharge treatment it reaches the value 70 $\mu\text{S}/\text{cm}$. That corresponds to an increase from the initial value by 7 times. Such an increase in electrical conductivity indicates that new ions are produced in water solution, and that the concentration of hydrogen ions $[\text{H}^+]$ becomes significant. The increase in EC can also be associated with the development of chemical reactions that produces nitrogenous species NO_x in the discharge and with the further formation of nitric acids solution.

CONCLUSIONS

The study of the current-voltage characteristics of a streamer corona discharge with a water cathode showed that the discharge burns in a limited applied voltage range. Obtained voltage range width is equal 1.6...2.6 kV and it depends on the discharge gap value. In this case, in the experiments it was observed only the non-stationary discharge burning mode. In this mode, streamers were close the discharge gap. This trend is established for all of discharge gaps that were investigated.

Analysis of the waveforms of the discharge current pulses showed that the discharge current in the non-stationary burning mode has a quasi-periodic type. Current pulses have a characteristic time duration 200...300 ns, and their formation is connected with the propagation of a positive streamer in the discharge gap. At the same time, with an increase in the applied voltage, both the frequency of the current pulses and their amplitude increase. The characteristic frequency range of the discharge current pulses is 9...25 kHz. It was found that with a decrease of discharge gap, the characteristic frequency of the current pulses increases.

Measurement the pH of water cathode showed a decrease in pH from 6.9 to 5 in the first 10 min of the dis-

charge treatment. After that, the pH value remains constant. Such a decrease in the pH value indicates that the solution under treatment tends to become acidic due to increase of hydrogen $[H^+]$ ions concentration.

Measurement of the EC of water cathode showed an increase from 10 to 70 $\mu S/cm$ within 25 min of discharge treatment. The increase in electrical conductivity indicates that new ions are produced in water solution.

The pH and EC measurements demonstrated that various nitrogen species NO_x are produced in the streamer discharge. This species are well soluble in water with the further formation of nitric acids solution. This leads to a decrease in pH to the acidic range and an increase in the EC value of the solution under discharge treatment.

The results of the research can be useful to optimize technological systems for plasma-chemical water treatment in a streamer discharge. Such systems can be used for disinfection and water purification from harmful impurities such as phenols, bacteria, spores and biogenic organics.

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ЕЛЕКТРОДИНАМІЧНІ ТА ЕЛЕКТРОХІМІЧНІ ХАРАКТЕРИСТИКИ СТРИМЕРНОГО КОРОННОГО РОЗРЯДУ В ЕЛЕКТРОДНІЙ СИСТЕМІ З ВОДНИМ КАТОДОМ

О.В. Болотов, Г.В. Таран, М.О. Єзоров

Досліджено електродинамічні та електрохімічні характеристики стримерного розряду з водним катодом у нестационарній стадії горіння. Встановлено, що струм розряду є квазіперіодичною послідовністю імпульсів. Відзначено, що зі збільшенням напруги на розрядному проміжку амплітуда та частота імпульсів струму збільшуються. Імпульси струму мають характерну тривалість 200...300 нс, при цьому частота імпульсів струму лежить у діапазоні 9...25 кГц і залежить від величини міжелектродної відстані. Дослідження електрохімічних характеристик водного катода показало зміну величини рівня pH і електропровідності води (ЕС) при дії стримерного розряду. Відзначено зниження рівня величини pH води від 6,9 до 5, при цьому одночасно спостерігалось збільшення електропровідності від 10 до 70 мкСм/см. Такі зміни можуть бути пов'язані з утворенням у воді нових іонів під дією розряду.