INFLUENCE OF A PULSE DURATION OF HIGH-VOLTAGE SUPPLY ON THE EFFICIENCY OF OZONE SYNTHESIS IN THE “NEEDLE — PLANE” ELECTRODE SYSTEM

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We present the results of studies of the electrodynamic characteristics of a barrierless discharge with electrodes of the “ needle — plane” type and a high-voltage pulse of positive polarity, being applied to the edge electrode. The efficiency of ozone synthesis is determined as a function of the pulse duration and repetition rate. It is shown that the electrodynamic characteristics of the discharge and the effectiveness of ozone synthesis in oxygen-containing gas mixtures essentially depend on the parameters of the pulse supply. As a high-voltage commutator, we used a high-speed high-voltage semiconductor switch of the HTS-300 line produced by BEHLKE Electronic GmbH (Germany).

1. Introduction

Active application of ozone technologies has stimulated the appearance of a large number of investigations devoted to the optimization of characteristics of a classical barrier ozonator and the examination of the processes of ozone synthesis in it [1,2]. Along with these works, the investigations of gas discharges of other types aimed at the application to the ozone production are also expanded [3—6]. Among the alternative gas discharges, a special attention is paid to the barrierless discharge in the “needle — plane” electrode system [7]. This is conditioned by a high reliability of plasma-chemical reactors using such electrode systems, especially under hard service and climatic conditions [8]. Due to the absence of an insulator in the area between the electrodes, the incident breakdown of a discharge gap doesn’t result in a fatal damage of the reactor. In addition, the use of a pulse unidirectional high-voltage supply allows one to essentially increase the effectiveness of ozone synthesis and to decrease the mass/volume characteristics of the high-voltage power supply. Thus, the optimization of the pulse supply parameters can serve as a decisive factor of the wide application of ozonators of barrierless type.

2. Experimental Apparatus

The given paper presents the results of investigations of the discharge in a reactor of ozone synthesis with electrodes of the “needle — plane” type under pulse high-voltage supply. We investigated the electrodynamic characteristics of a discharge, as well as the effectiveness of ozone generation in a barrierless ozonator, as functions of the pulse duration and the pulse repetition frequency under various flows of the working gas. A coaxial construction of the plasma-chemical reactor was used, where the cathode represented a thin-walled electroconductive calibrated cylinder made of 12X18H stainless steel of 350 mm in length having an internal diameter of 34 mm and a wall thickness of 1 mm, while the anode system consisted of 32 wheel spiders made of stainless steel of 0.4 mm in thickness arranged on the anode rod 10 mm apart. Each wheel spider had 25 electrode-beams, and its maximal diameter amounted to 22 mm. Thus, the anode system consisted of 800 needle electrodes coaxially arranged along the axis of the tubular electrode with the help of edge isolators. The gap between the cathode and the peaks of the beam electrodes amounted to 6 mm. The pulse supply of the plasma-chemical reactor was realized with the help of a

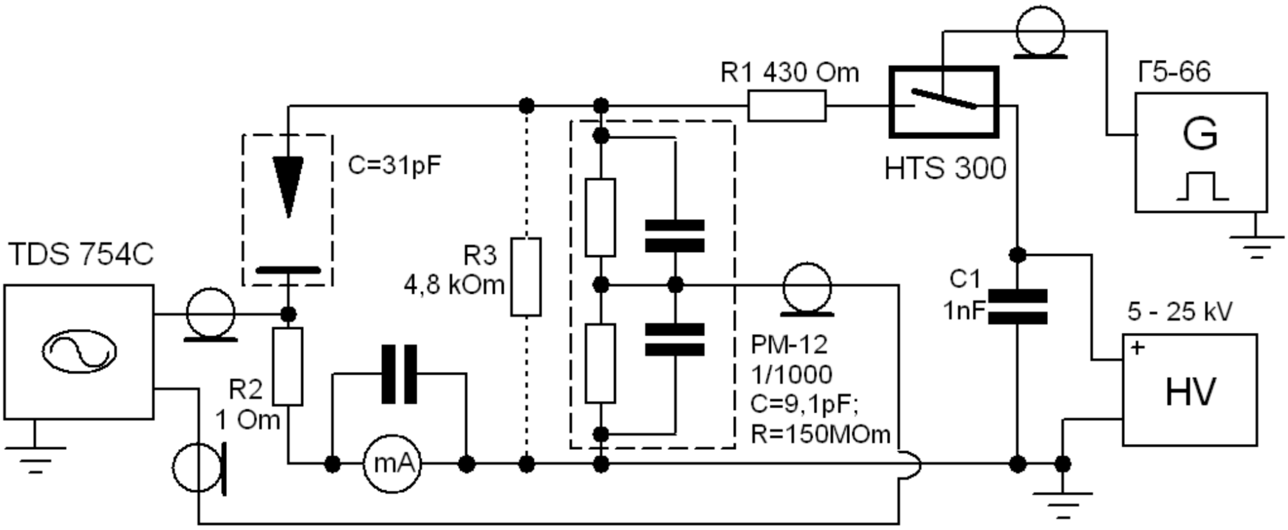

Fig. 1. Basic electric circuit of the experimental setup

high-voltage pulse commutator of a HTS-300 line made by BEHLKE Electronic GmbH company (Germany). The impulse front time of the commutator was of the order of 10 ns, the time of the on-state equaled 200 ns, and the maximal voltage was 30 kV. As a source of working gas, we used an oxygen generator AS-12 produced by "AirSep" (USA) that generated enriched oxygen with a concentration of 90-95%. The measurements were performed at two flows of the working gas through the reactor: 0.7 l/min and 5 l/min. The gas flow rate was controlled by a PM-II flowmeter with a relative accuracy of 2.5%. For the measurements of the ozone concentration, we used an ozone concentration meter "MiniHicon" produced by "IN USA, Inc." (USA).

Figure 1 presents a basic electric circuit of the experimental setup.

As a high-voltage source of direct current, we used a stabilized power unit with a controllable output voltage in the range of 4—25 kV. The maximal average current of the supply was equal to 10 mA. In the mode of high-amplitude current pulses, the voltage at the input of the commutator was stabilized with the help of capacity $C1$, while resistor $R1$ restricted the maximal current flowing through the commutator. The switching frequency of the HTS-300 commutator was controlled by means of a G5-66 pulsed oscillator. In the experiments, the pulse repetition rate amounted to 100 Hz, 333 Hz, 1 kHz, 3.33 kHz, and 10 kHz. The electric parameters of a pulse were measured by a high-voltage divisor Pm-12 produced by NORTH STAR (USA) having a dividing coefficient of 1:1000 and a bandpass of 75 MHz. The form and amplitude of a current pulse passing through the reactor was measured by a high-frequency current shunt with a nominal of 1 Ohm and a bandpass of 250 MHz. The signals from the current and high-voltage probes were digitized with the help of a TektronixTDS754C digital oscillograph with a bandpass of 500 MHz.

The rise time of a high-voltage pulse on the reactor amounted to 27 ns. This is associated with the presence of a charging RC-circuit, where R is a current-limiting resistor $R1 = 430$ Ohm, while $C1 = 40.1$ pF is a summary capacity of the reactor and the high-voltage divisor.

After the commutator is turned off, the voltage across the reactor decreases according to the law

$$U\left(t\right) = U_0 \exp^{-t/\tau}, \tag{1}$$

where U_0 stands for the voltage across the reactor at the moment when the commutator is turned off, $\tau = RC$ denotes the characteristic time of the capacity discharge, $R = 150$ MOhm is the input resistance of the high-voltage divisor, $C = 40.1$ pF is the summary capacity of the reactor and the high-voltage divisor. Hence, for the circuit presented in Fig. 1, τ =6 ms and, correspondingly, the total time of the drop of the reactor voltage approximates 20 ms, that is, the drop time of the reactor voltage is comparable to the pulse period, and the reactor turns out to be connected to the source of a slowly varying high voltage, at whose background the high-voltage pulses with a steep leading edge are supplied ("long" mode).

In order to reduce the duration of a high-voltage pulse ("short" mode), we connected an additional discharge resistance $R3$ (a resistor of TVO-10 type with a nominal of 4.8 kOhm) in parallel to the reactor, which resulted in a decrease of the fall time of the voltage across the reactor to 250 ns. Thus, in this mode, the reactor was subjected to nanosecond high-voltage pulses with a half-amplitude duration of 300 ns. It's worth noting that, in such a circuit, a sufficiently large part of the power of the high-voltage supply is dissipated at the discharge resistor. That's why this method can't be considered optimal for the use in industrial ozone plants. In order to realize the advantages of the pulse supply for ozone synthesis and to obtain high values of the ozonator efficiency, the circuit of a pulse high-voltage power supply should provide the recuperation of energy supplied for the charge of the reactor capacity.

Thus, in the given paper, we investigated the characteristics of a barrierless plasma-chemical reactor with an electrode system of the "needle — plane" type in three modes of high-voltage power supply:

— long high-voltage pulses with a steep leading edge ("long" mode);

— short high-voltage pulses with a half-amplitude duration of 300 ns ("short" mode).

— direct-current voltage ("DC" mode). The "DC" mode, in which a reactor is supplied with direct-current high voltage, is well studied [9—11] and was used for the comparison and analysis of the experimental results.

In the course of our investigations, we measured the form and amplitude of both the pulse current and voltage at various values of the average current flowing through the reactor varying from the minimal current (one tenth of the highest average current) up to the maximal one (the breakdown of the discharge gap), as well as the ozone concentration at the reactor output.

3. Experimental Results

The characteristic oscillograms of the voltage and current pulses in the "short" and "long" modes are presented in Fig. 2.

These oscillograms reflect the dynamics of variation of the electrodynamic characteristics of the reactor at various values of the average discharge current. It's worth noting that the first impulse on the oscillogram of the current flowing through the reactor (CH4 channel) is conditioned by the charging of the reactor capacity and doesn't contribute to the average current. The second impulse on the current oscillogram corresponds, in fact, to the discharge current in the reactor. Moreover, its form essentially changes depending on the operating mode of the reactor and the average current flowing through it. It is important to note that, in the "long" mode, the current oscillograms reflect only that part of the discharge current, which corresponds to the pulse stage of the discharge, while the portion of the current flowing at the stage of a slowly varying voltage was measured separately.

The comparison of the current and voltage oscillograms in Fig. 2 demonstrates that, in the "short" mode, the discharge in the reactor starts, when the voltage across the latter reaches 9.5—11 kV. In the "long" mode, the discharge is initiated at a higher voltage of 10—15 kV. With increase in the pulse repetition rate, we observed the tendency to a decrease of the ignition voltage of the discharge in the both modes. It's worth noting that, in the "long" mode, the discharge in the reactor started with a larger time delay with respect to the edge of the high-voltage pulse than that in the "short" mode. The delay time reduced with decrease in the average current flowing through the reactor and increased with rise in the repetition rate of high-voltage pulses and can reach several μs. In addition, in the "long" mode, the form of the current pulse of a discharge and the time delay with respect to the edge of the high-voltage pulse have no stable characteristics from pulse to pulse. One can suggest that the observed differences of the electrodynamic characteristics in the "short" and "long" modes are conditioned by the volume charge influence. Indeed, in the "long" mode, the volume charge from the discharge gap is dispersed during the time of the order of 50 μs due to the drift of charged particles in the electric field formed by the slowly decreasing voltage across the reactor. That's why, by the moment of arrival of the next high-voltage pulse, insulating properties of the gap completely recover. Thus, in the "long" mode, the conditions for the origin of the next streamer don't depend on the preceding impulse.

In the "short" mode, due to the application of an additional shunting resistance $R3$ (see Fig. 1), the voltage across the reactor in the period between the pulses is absent, which impedes the process of dispersal of the volume charge from the discharge gap. That's why, by the arrival time moment of the following high-voltage pulse, the discharge gap still contains charged particles. These charged particles facilitate the origin of a new streamer in the field of the following voltage

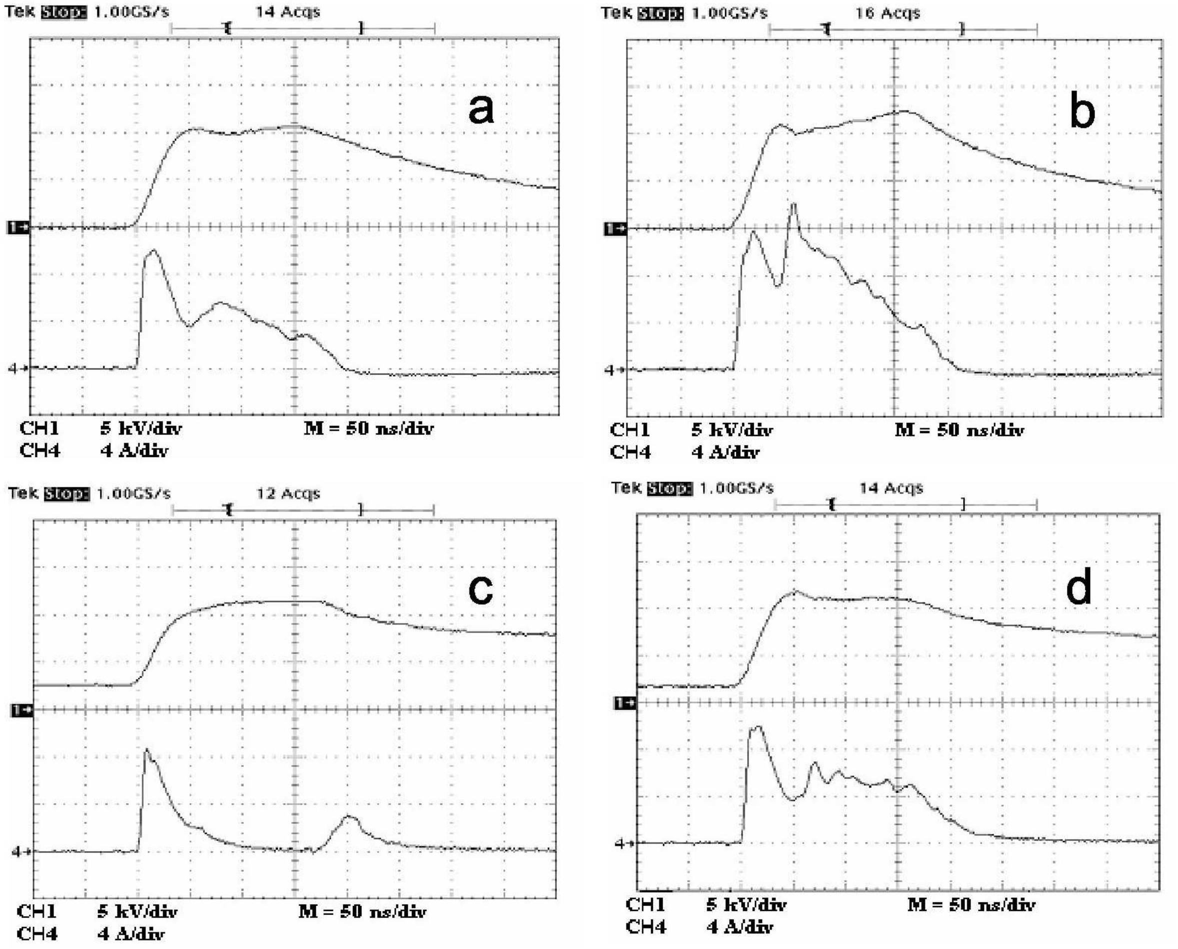

Fig. 2. Oscillograms of the voltage across the reactor (the first oscillograph channel CH1) and current flowing through it (the fourth oscillograph channel CH4) for the working gas flow of 5 l/min and the repetition rate of high-voltage pulses of 333 Hz: *a* — "short" mode, average current through the reactor 0.2 mA; *b* — "short" mode, average current through the reactor 0.4 mA; *c* — "long" mode, average current through the reactor 0.2 mA; *d* — "long" mode, average current through the reactor 0.4 mA

impulse, which results in the reduction of the discharge ignition voltage as compared to the "long" mode.

The experimental apparatus allowed us to determine the pulse power supplied to the discharge to a rather high accuracy using the method of point-by-point multiplication of the instantaneous values of the current and voltage across the reactor:

$$W = \frac{\Delta t}{T} \sum U_n \cdot I_n, \tag{2}$$

where Δt is the time step of measuring the instantaneous values of the voltage across the reactor, U_n, and the current flowing through it, I_n, while T is the repetition rate of high-voltage pulses.

It's worth noting that the practically full power in the "short" mode is supplied to the discharge during the transit time of a current pulse, whereas the power supplied to the discharge in the "long" mode can be presented as a sum of the contributions of two terms:

$$W_{\text{long}} = W_{\text{pulse part}} + W_{\text{DC part}}. \tag{3}$$

Here, $W_{\text{pulse part}}$ is the pulse power properly that corresponds to the pulse portion of the discharge current (the second impulse on the oscillogram of the current flowing through the reactor, see CH4 channel in Fig. 2), and $W_{\text{DC part}}$ is the power corresponding to the constant component of the current that flows after the passage of the pulse portion of the discharge current when the high voltage across the discharge gap slowly decreases. Over a sufficiently long period, this voltage remains higher than

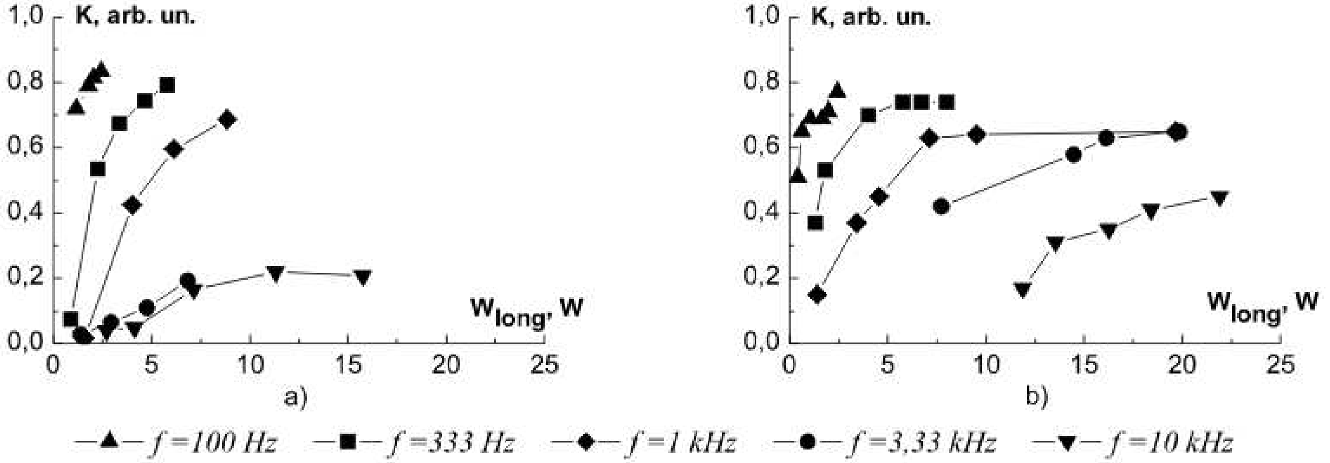

Fig. 3. Dependence of the relative value of the pulse power component on the total power supplied to the discharge in the "long" mode for various repetition rates of high-voltage pulses: *a* — the flow of the working gas 0.7 l/min; *b* — the flow of the working gas 5 l/min

the ignition potential for the given reactor in the "DC" mode. That's why, during this time interval, the conditions for the discharge burning analogous to the "DC" mode are satisfied. Using the experimental data, $W_{\text{pulse part}}$ and W_{long} for the "long" mode were determined according to expression (2). The constant component of the power $W_{\text{DC part}}$ for the "long" mode was calculated as a difference between W_{long} and $W_{\text{pulse part}}$. The diagram in Fig. 3 presents the dependence of the relative pulse power $K = W_{\text{pulse part}}/W_{\text{long}}$ on the total power supplied to the discharge in the "long" mode at various repetition rates of high-voltage pulses and flows of the working gas.

As one can see from the diagrams in Fig. 3, the pulse component of the power in the "long" mode increases with the total power supplied to the discharge.

Moreover, while $W_{\text{pulse part}}$ is higher than $W_{\text{DC part}}$ at a low repetition rate of pulses supplying a reactor, this relation changes to the converse one with increase in the frequency.

By the experimentally measured values of the power supplied to the discharge and the ozone concentration at the reactor output, we determined the power inputs η for ozone synthesis for a given reactor:

$$\eta = \frac{W}{n_{\text{O}_3} \cdot Q}, \tag{4}$$

where n_{O_3} denotes the ozone concentration at the reactor output in g/m^3, and Q is the gas flow through the reactor in m^3/h.

The power inputs for ozone synthesis as functions of the ozone concentration at the reactor output at various gas flows and pulse repetition rates for the "short" and "long" modes of the reactor supply are depicted in Fig. 4. For the sake of comparison, we also present here the dependence of the power inputs for ozone synthesis in the "DC" mode.

The analysis of the diagrams given in Fig. 4 allows one to make the following conclusions concerning the power inputs for ozone synthesis:

1) In all the investigated pulse modes, the power inputs for ozone synthesis are much lower than in the "DC" mode; moreover, in the "DC" mode, one can observe an essential increase of the power inputs for ozone synthesis with increase in the ozone concentration.
2) In the "short" mode, the power inputs are minimal and weakly increase with the ozone concentration.
3) In the "long" mode with increase in the ozone concentration, the power inputs decrease firstly and then increase.
4) The variation of the pulse repetition rate practically doesn't influence the power inputs in the "short" mode except for the case where the repetition rate of high-voltage pulses amounts to 10 kHz and the gas flow equals 0.7 l/min.
5) In the "long" mode, the increase in the pulse repetition rate results in the essential growth of the power inputs, which correlates with the increase of the contribution of the constant component of the power with respect to the total power supplied to the discharge (see Fig. 3).

Generalizing these conclusions, we may suppose that the efficiency of ozone synthesis for the reactor in the "long" mode P_{long} can be presented, like the power, as a sum of two independent processes,

$$P_{\text{long}} = P_{\text{pulse part}} + P_{\text{DC part}}, \tag{5}$$

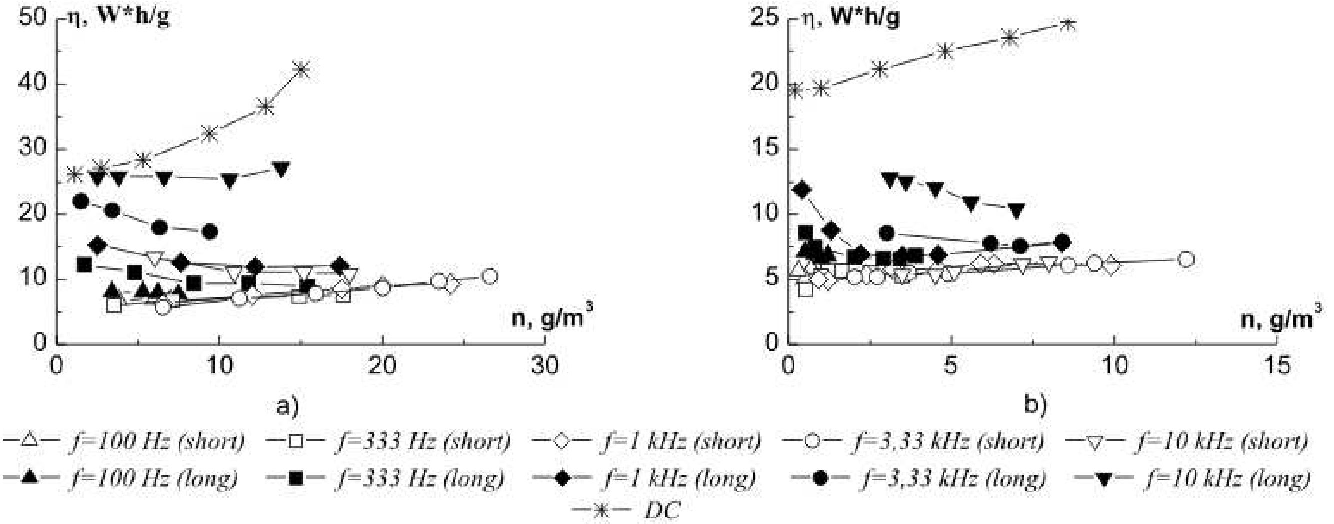

Fig. 4. Power inputs for ozone synthesis versus the ozone concentration at the reactor output: *a* – the flow of the working gas 0.7 l/min; *b* – the flow of the working gas 5 l/min

where $P_{\text{pulse part}}$ is the efficiency of ozone synthesis during the passage of the pulse component of the power $W_{\text{pulse part}}$, when the ozone generation is similar to that in the "short" mode. Respectively, the power inputs for ozone synthesis are the same as in the "short" mode. After the passage of the pulse current component in the "long mode", the voltage across the reactor slowly decreases remaining sufficiently high over a long period, so that the discharge in the reactor continues similar to the "DC" mode.

Thus, the efficiency of ozone generation $P_{\text{DC part}}$ corresponds to the contribution of the constant component of the power $W_{\text{DC part}}$ with high power inputs for ozone synthesis characteristic of the "DC" mode.

In order to confirm this assumption, we will use the known experimental values of the pulse and constant components of the power for the "long" mode as well as the values of the power inputs for ozone synthesis in the "short" and "DC" modes. Based on these data, we will calculate the summary efficiency of ozone synthesis P'_{long} and compare the obtained result with the ozone efficiency P_{long} determined experimentally in the "long" mode.

The ozone synthesis efficiency P_{long} is determined by the expression

$$P_{\text{long}} = \frac{W_{\text{long}}}{\eta_{\text{ long}}\left(n_{\text{O}_3}\right)}. \tag{6}$$

We now can present the calculated efficiency of ozone synthesis P'_{long} as

$$P'_{\text{long}} = \frac{W_{\text{pulse part}}}{\eta_{\text{ long}}\left(n_{\text{O}_3}\right)} + \frac{W_{\text{DC part}}}{\eta_{\text{DC}}\left(n_{\text{O}_3}\right)}. \tag{7}$$

Here, $\eta_{\text{ long}}\left(n_{\text{O}_3}\right)$ and $\eta_{\text{DC}}\left(n_{\text{O}_3}\right)$ are the measured values of the power inputs for ozone synthesis in the "short" and "DC" modes, correspondingly, taken from the diagram in Fig. 4, while $W_{\text{pulse part}}$ and $W_{\text{DC part}}$ are the experimentally measured values of the pulse and constant components of the power in the "long" mode. In this case, all the quantities in expression (7) are taken at the same value of the ozone concentration n_{O_3} and a fixed flow of the working gas Q.

Figure 5 shows the calculated P'_{long} and experimental P_{long} efficiencies of ozone synthesis in the "long" mode for various flows of the working gas Q and pulse repetition rates.

The diagrams in Fig. 5 demonstrate a good agreement of the calculated and experimental values of the efficiency of ozone generation for all the investigated values of the repetition rate of high-voltage pulses and flows of the working gas. This confirms the assumption made above that, in the "long" mode, the summary efficiency of ozone synthesis in a plasma-chemical reactor can be presented in the form of the contributions of two independent processes: the pulsed stage characterized with low power inputs for ozone synthesis and the stage of a slowly decreasing voltage characterized with high power inputs for ozone synthesis. In addition, as was mentioned above, with increase in the repetition rate of

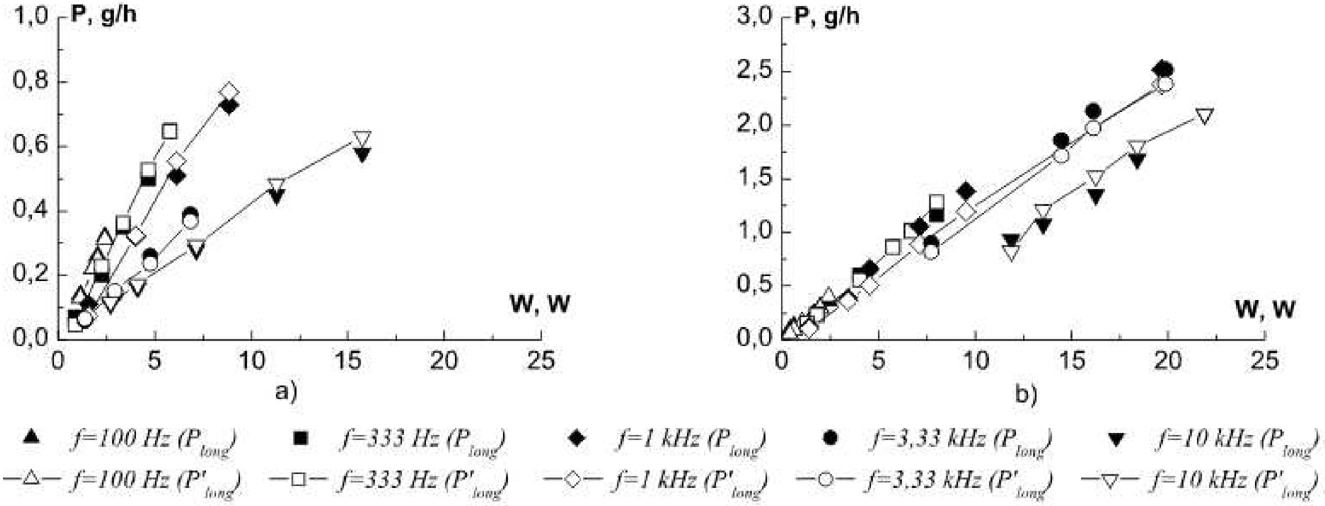

Fig. 5. Experimental P_{long} (black symbols) and calculated P'_{long} (white symbols) values of the efficiency of ozone synthesis at the reactor output versus the supplied power: *a* — the flow of the working gas 0.7 l/min; *b* — the flow of the working gas 5 l/min

high-voltage pulses, the magnitude of the residual voltage across the reactor grows by the moment of arrival of the following pulse, which results in the rise of the portion of the constant component of the power. In turn, the increase in the portion of the constant power component results in the growth of the summary power inputs for ozone synthesis in the "long" mode.

4. Conclusions

The performed investigations of a pulsed barrierless discharge in enriched oxygen at atmospheric pressure with electrodes of the "needle — plane" type and a high-voltage pulse supply of positive polarity being applied to the edge electrode have demonstrated that the electrodynamic characteristics of a discharge strongly depend on the parameters of a pulse supply. For example, in the case of a pulse supply with the duration of a high-voltage pulse being equal to 300 ns, the discharge in the reactor starts at a voltage lower by 20–25% than that in the case where the discharge is supplied with long pulses characterized with a steep leading edge and a slow fall (of the order of 50 μs). This is conditioned by the influence of the residual volume charge in the interelectrode gap to the moment of arrival of the following high-voltage pulse for short pulses. At the same time, for long impulses, the isolating properties of the discharge gap completely recover to the moment of arrival of the next pulse.

It is shown that, if the pulse supply with a pulse repetition rate in the range of 100 Hz — 10 kHz is used, the power inputs for ozone synthesis increase with the duration of pulses. This is associated with the fact that, for a voltage pulse longer than 300 ns, the current through the reactor includes not only a pulse component but also a rather large constant one, whose contribution increases with the growth of the pulse repetition rate and decreases with the rise of the average power supplied to the reactor.

We have experimentally confirmed the assumption that, under a pulse supply of a plasma-chemical reactor, the ozone synthesis can be represented in the form of two independent processes: the pulsed stage with low power inputs for ozone synthesis and the stage of a slowly decreasing voltage across the reactor that corresponds to the constant component of the discharge current flowing through the reactor and high power inputs for ozone generation.

Thus, the larger the magnitude of the discharge current flowing through the reactor, the higher the power inputs for ozone synthesis are.

For short high-voltage pulses, whose duration is less than 300 ns, the power inputs for ozone synthesis amounted to 5—6 W·h/g.

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ВПЛИВ ТРИВАЛОСТІ ІМПУЛЬСУ ВИСОКОВОЛЬТНОГО ЖИВЛЕННЯ НА ЕФЕКТИВНІСТЬ СИНТЕЗУ ОЗОНУ В СИСТЕМІ ЕЛЕКТРОДІВ ВІСТРЯ—ПЛОЩИНА

В.І. Голота, Л.М. Завада, В.І. Карась, О.В. Котюков, О.В. Поляков, С.Г. Пугач

Резюме

Наведено результати дослідження електродинамічних характеристик безбар'єрного розряду з електродами типу вістря—площина при прикладанні високовольтного імпульсу позитивної полярності до вістря. Визначено ефективність синтезу озону в залежності від тривалості і частоти проходження імпульсів. Показано, що електродинамічні характеристики розряду й ефективність синтезу озону в кисневмісних газових сумішах істотно залежать від параметрів імпульсного живлення. Як високовольтний комутатор використовувався швидкісний високовольтний напівпровідниковий ключ серії HTS-300 фірми BEHLKE Electronic Gmb (Germany).