# Method for increasing the efficiency of ozone electrosynthesis process with periodic voltage pulses

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**Abstract.** This article presents the results of a scientific study to improve the efficiency of ozone electrosynthesis. The issues of applying another voltage as a periodic voltage pulse are considered. The analysis of the process of electrosynthesis of ozone when fed with periodic high-voltage pulses is given, as a result of which the stability of discharges in the discharge gap was achieved. Transient processes in the discharge gap in the pause between voltage pulses are considered. The results of studies of the energy indicators of pulsed and sinusoidal voltages are also presented. Where the analysis of the equations shows that the main part of the energy of periodic voltage pulses is spent on the discharge process, which is accompanied by ozone electrosynthesis, which leads to an increase in the efficiency of ozone electrosynthesis with a periodic pulse voltage.

## 1. Introduction

Studies on improving the efficiency of the ozone electrosynthesis process through the use of various types of periodic and pulsed voltages are given in [1, 2, 3]. The disadvantages of these studies include the fact that the voltage conversion is carried out at a high voltage, which greatly complicates the circuits with a significant increase in the dimensions of the devices. It should also be noted that there are no reliable semiconductor converters, and the use of vacuum tubes drastically reduces the service life (often not exceeding 500 hours).

The disadvantages of these devices are also unreasonable range of the slew rate of the negative part of the pulse and cannot fluctuate in this range. According to the description, it is not clear what the pulse generator is and the shape of the pulses at its output is not given, and the stability of the barrier discharge cannot be achieved at a frequency of 1.5 Hz.

Despite the numerous patents for inventions, they have not found wide industrial use. For the electrosynthesis of ozone on an industrial scale, sinusoidal voltages with a frequency of about 50 Hz and high-frequency ozonizers with a frequency of 1000 Hz and higher are mainly used. These devices were accepted as a prototype [3].

In such processes, ozone electrosynthesis is heavily dependent on the electric field strength in the technological discharge gap, and in practice, a barrier or so-called barrier discharge is used to raise the electric field strength above the breakdown threshold of the gas being processed [4, 5]. However, this situation causes the dielectric barrier to heat up and necessitates cooling. The temperature and humidity of the processed gas must meet strict standards. As a result, in the currently employed electrosynthesis of ozone technologies, the gas is first prepared by chilling and drying it before processing. Efficiency of present ozone electrosynthesis systems does not surpass 2%. The instability of the discharge current in frequency and amplitude distinguishes the barrier discharge from all other types of independent discharges in gases [6, 7]. Efforts to automatically control the discharge current in ozone generators did not give the intended results and are not currently employed.

In industrial ozone generators, as a rule, a barrier discharge in the volume or a surface (sliding) barrier discharge is realized. A barrier discharge is a special form of an electric discharge that occurs in a medium between the reactor electrodes, separated by a dielectric layer (barrier) and a discharge gap filled with particles of the medium, if a potential difference is applied to the electrodes that is sufficient (for a discharge) to occur [8, 9]. The discharge current is limited by the dielectric layer. The dielectric layer stabilizes the discharge current of the ozone generator and imparts a uniform character to the barrier discharge in the reactor volume. Studies have shown that the dielectric layer plays an important role in the process of ozone electrosynthesis, and its properties generally determine the quality, reliability and performance of the generator [10, 11].

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Existing ozone electrosynthesis devices mainly use high sinusoidal voltages with a frequency of 500 Hz.

The energy efficiency of the established technological procedures of electrosynthesis does not surpass 1–2%. The majority of energy is converted into heat, which causes the dielectric barriers to heat up too much and necessitates cooling.

The current leads the voltage by 90 electrical degrees as a result of the supply of a capacitive load. In the discharge gas gaps, in which independent discharges occur, space charges (ions) are generated. These charges turn out to be sufficient to stop the current from growing at the same time that the voltage of the counter voltage half-wave increases [12–15].

It should be noted that self-sustained discharges are characterized by instability in the discharge current's amplitude; as a result, the time it takes for space charges to neutralize will vary. This explains why the ozone electrosynthesis process is unstable. As a result, it is possible to use various types of voltage, such as periodic pulses with a high duty cycle, to improve the efficiency of the ozone production process.

## 2. Materials and Methods

### 2.1. Analysis of the process of ozone electrosynthesis via periodic high voltage pulses

One of the low-cost ways to improve the efficiency of electron-ion technology devices is the use of pulse modulation of the power supply. At present, there is a long experience in operating pulsed microsecond power supplies at large power units, for example, in Denmark and Japan.

The development of a new method for the electrosynthesis of ozone is due to the shortcomings of existing devices. As has been repeatedly pointed out [8, 9] impulses of discharge currents of independent discharges are randomly distributed in time, the realization of which is random in amplitude and frequency. In a barrier discharge, as in electric discharges of all types, various elementary processes occur simultaneously: the emergence, movement, and destruction of charged particles. It has so far been possible to quantitatively derive the properties of discharges on the basis of laws only in some cases, and then only under the condition that essential assumptions are introduced. This is due to mathematical difficulties that arise when combining various patterns. Such types of discharges are called independent. The listed properties of the barrier discharge of alternating voltage lead to the instability of the discharge currents in amplitude, to the locking of the discharge, the transition to a spark or arc form. Obviously, by ensuring the stability of discharge processes, it is possible to increase the efficiency of ozone electrosynthesis.

The specificity of barrier electric fields is the presence of a flow of space charges in the discharge gap. The electric field created by these charges is directed opposite to the main electric field:

$$E(x) = E_0(x) + \Delta x, \tag{1}$$

where: $E(x)$ – true electric field strength;

$E_0(x)$ – main field strength;

$\Delta x$ – field strength created by space charges.

The value of $\Delta x$ is determined based on the laws of electrostatics:

$$\Delta x = 4\pi q_t \{[exp\alpha x/(exp\alpha d\text{-}1)]\text{-} 1/\alpha d\}, \tag{2}$$

where: $q_t$ - is the time-dependent total number of positive ions per unit surface of each of the electrodes of the discharge gap.

The voltage on the electrodes also changes by the value:

$$\Delta U = \int_0^d \Delta x \cdot dx \tag{3}$$

Since $E(x)$ depends on the distance, $\alpha$ also depends on the distance. Let's mark it with index $x$:

$$\alpha_x = Apexp[-Bp/(E_0(x)+\Delta x)] = Apexp\{-Bp/E_0(x)[1/1+\Delta_x/E_0(x)]\}, \tag{4}$$

where: $A$ and $B$ are constants equal to 14.5 and 365, respectively, for air at the application area $E/p$ = 160 ... 600 *V/cm * mm. Hg.*

After expanding into a series up to the number $\Delta/E_O$ squared, we get:

$$\alpha_x = \alpha_0 \{1 + (Bp\Delta/E^2_0) + [Bp\Delta^2/E^4_0 (0,5Bp\text{-}E_0)]\} \tag{5}$$

For reduction, let $\alpha_0$ denote $\alpha = Apexp(-Bp/E0)$. Accordingly, the total number of ionizing collisions of the primary electron in the distorted field will be:

$$\int_0^d \alpha_0 \cdot dx = \alpha_0 d + \left(\frac{\alpha_0 Bp\Delta U}{E_0^2}\right) + [\alpha_0 Bp/E_0^4 (0,5Bp - E_0)] \cdot \int_0^d \Delta^2 dx \tag{6}$$

The integral in the last term is positive in all cases, since $\Delta^2$ must always be positive. Thus, the last term is positive if $0,5Bp>E0$, and vice versa. If the field distortion occurred at a constant voltage on the electrodes $(\Delta U=0)$, then:

$$\int_0^d \alpha_x \cdot dx - \alpha_0 d > 0 \text{ for } E_0/p < 0,5B, \tag{7}$$

$$\int_0^d \alpha_x \cdot dx - \alpha_0 d < 0 \text{ for } E_0/p > 0,5B, \tag{8}$$

If, on the contrary, we look for such changes in the voltage on the electrodes, which again return the ionization in the distorted field to its original value in the undistorted field

$\int_0^d \alpha_x \cdot dx - \alpha_0 d = 0$, то:

$$\Delta U = (E_0 - 0{,}5Bp)1/E_0^2 \int_0^d \cdot \Delta^2 dx \qquad (9)$$

from here

$$\Delta U < 0 \quad \text{for} \qquad E_0/p < 0{,}5B, \qquad (10)$$
$$\Delta U > 0 \quad \text{for} \qquad E_0/p > 0{,}5B \qquad (11)$$

Equations (7) and (11) show that for a constant voltage on the electrodes, a uniform field E0 less than the critical value of 0.5 V, due to field distortion, becomes advantageous for ionization. In this instance, a lower voltage on the electrodes is necessary to keep the ionization at the same level in the distorted field as in the undistorted field.

Field distortion is detrimental to ionization if the undistorted field is greater than the critical value, as demonstrated by equations (8) and (11), respectively. The conclusions from equations (8) and (11) are completely independent of how the field is distorted in each specific case, which is one feature of those equations.

According to the analysis done, it is possible to achieve stability of discharge processes when the supply of the next voltage pulse of opposite polarity occurs without space charges in the discharge gap or when their number has little effect on the main electric field's distortion.

### 2.2. Analysis of transient processes in the discharge gap in the pause between voltage pulses

The process that takes place in the discharge gap while there is a pause between pulses under the influence of the charge that has collected in the discharge gap is interesting.

By selecting a pulse repetition rate at which all processes of movement and recombination of charged particles will be reduced to the point where they practically have no impact on the conductivity of the discharge gap during the pause between pulses, the problem of stabilizing the discharge process in terms of current values (in our case, the currents arising under the influence of a voltage pulse and currents in the pause between pulses) can be resolved.

Space charges in the discharge gap cause the main electric field's pattern to change, which is what causes independent discharges to become unstable.

Space charges with a density of are created in the discharge gap after delivering a voltage pulse of one of the polarities, which led to a barrier discharge [10]. Due to recombination and the transfer of space charges, the density of the gas volume decreases, and as a result, the current density through the discharge gap decreases [10]:

$$j = e \, b \, E \, \tau(t) \qquad (12)$$

where: $e$ – is the electron charge, $1{,}6 \times 10^{-19}$ C;

$b$ – ion mobility, $m^2$/V s;

$E$ – electric field strength, V/m;

$\tau(t)$ – volume charge density, $C/m^3$.

As a result, the current density in the discharge gap when there are no ionization processes is proportional to the field intensity, i.e., conductivity is linear. The variation in will influence the type of change in current density (t). The specifications of the electrical circuit to which the discharge gap is attached will in turn determine the latter.
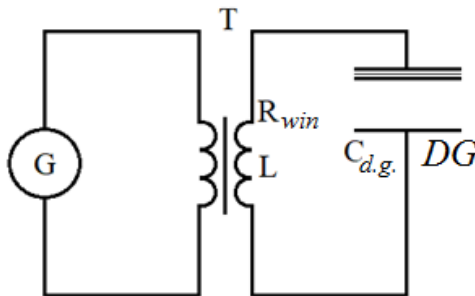


**Fig. 1.** Schematic diagram of the ozone electrosynthesis process: G – periodic voltage pulse generator; T – step-up transformer; $C_{d.g.}$ – capacity of the discharge gap; DG - discharge gap

Based on the aforementioned, the transient process in the power circuit may be used to assess the process in the discharge gap in the time between pulses. The discharge gap in this instance can be viewed as a component of the electrical circuit.

According to the plan illustrated in Figure 2, which increases high voltage pulses by the transformer T and feeds them to the technical discharge gap, high voltage pulses in the form of Figure 1 can be created. The circuit includes a control element CE and a discharge current control sensor S.

The ozone generator's technological discharge gap, which has its own capacitance Cd.g., is connected to the circuit's output (Figure 1). The ability to take into account the technological discharge gap as a component of the supply circuit is one of the benefits of the proposed method of electrosynthesis of ozone over the current method of electrosynthesis of ozone with a sinusoidal voltage [10].

In the time interval between pulses, the transient process in the electrical circuit of the source of high-voltage voltage pulses was examined. According to the diagram in Figure 2 we have a closed loop consisting of the capacity of the discharge gap $C_{d.g.}$, active $R_{win.}$ and inductive $\omega L$ resistances of the secondary winding of the step-up transformer [10].

Let us analyze the process in the circuit in the pause between pulses. After the application of the pulse, the capacitor $C_{d.g.}$ is charged up to the amplitude value of the voltage pulse.

To find the law of change in voltage and current of the discharge gap in the pause between pulses, we compose the circuit equation in complex form [11]:
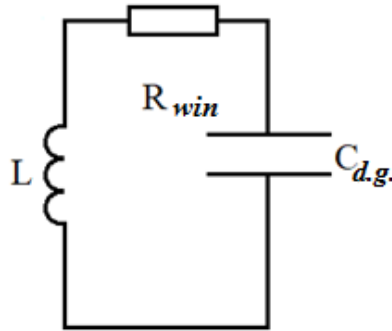
$$Z = R_{win.}+j\omega L+1/(j\omega\, C_{d.g}) \tag{13}$$

The factor $j\omega$ is replaced by the operator p and the resulting expression $Z(p)$ is set equal to zero:

$$Z(p) = R_{win.}+pL+1/(pC_{d.g.}) = 0, \tag{14}$$

or

$$p^2(C_{d.g}L)+pC_{d.g}\, R_{win} +C_{d.g} = 0 \tag{15}$$



**Fig. 2.** Equivalent circuit of the high voltage circuit of the ozone generator: *L* – inductance of the secondary winding of the step-up transformer; $C_{d.g}$ – capacity of the discharge gap; $R_{win.}$ – active resistance of the secondary winding of the transformer

The roots of the characteristic equation (2.15) are determined by the equality:

$$p_{1,2} = \{-C_{pn}R_{обм}\pm[(C_{d.g}R_{win.})^2-4(C_{d.g}L)(C_{d.g})]^{0,5}\}/(2C_{d.g}L), \tag{16}$$

From equation 2.16, we determine the change in voltage across the capacitor $C_{d.g}$

$$U_{Cсв} = (A_1e^{p1t} +A_2e^{p2t} ), \tag{17}$$

and current in the circuit:

$$i_{св} = [C_{d.g.}d(U_{Csv})]/dt = C(A_1p_1e^{p1t} +A2p_2e^{p2t}), \tag{18}$$

Initial conditions for calculations:

$$U_{Cd.g.} = U_a ,i_a = i_{sv.o}, t = 0, \tag{19}$$

where:   $U_{c2}$ – voltage amplitude in the discharge gap;
$U_a$ - voltage amplitude on the transformer;
$i_a$ - discharge current amplitude;
$i_{sv.0}$ – initial circuit current;
$t$ - integration step.

Taking into account the assumptions and initial conditions, we obtain:

$$U_a =A_1 +A_2, \qquad\qquad i_a =A_1p_1 +A_2p_2, \tag{20}$$

from here

$$A_1 = (p_2U_a\, i_a)/(p_2 -p_1), \tag{21}$$
$$A_2 = (p_1\, U_ai_a)/(p_2 -p_1) \tag{22}$$

With these values of the integration step, the laws of voltage and current changes in the pause between pulses take the form:

$$U_{C2.sv} = [1/(p_2-p_1)]\, [(p_2U_a-i_a)p_1e^{P1t}-(p_1U_a-i_a)p_2e^{P2t}], \tag{23}$$
$$I_{sv} = [C_{d.g.}/(p_2-p_1)]\, [(p_2U_a-i_a)p_1e^{P1t}-(p_1U_a-i_a)p_2e^{P2t}] \tag{24}$$

The problem was solved on a PC for the following values of the circuit parameters: $L$=7 Гн; $R_{win.}$= 3,5x$10^3$ Ом; $C_{d.g.}$= $10^{-9}$, Ф; $U_a$= 2x$10^4$В; $i_a$ = $10^{-4}$ A. Integration step 0.001 s. According to the results of calculations, graphs of the change in voltage and current of the discharge gap in the pause between pulses were plotted (Figure 3) [10].

The result of the analyzes of the curves indicates the same law of change in $U_{C2.sv}$ and $i_{sv}$. The transient process depends on the parameters of the circuit $R_{win.}$, $L$, $C_{d.g.}$, i.e. from the roots of the characteristic equation. At the same time, the value of the capacitance of the discharge gap $C_{d.g.}$ is decisive.

The charge compensation time increases with increasing capacity of the discharge gap. So, with the capacitance $C_2$=10-9 F, the charge compensation time is 0.006 s. The maximum pulse repetition rate was properly 166 imp$^{-1}$.



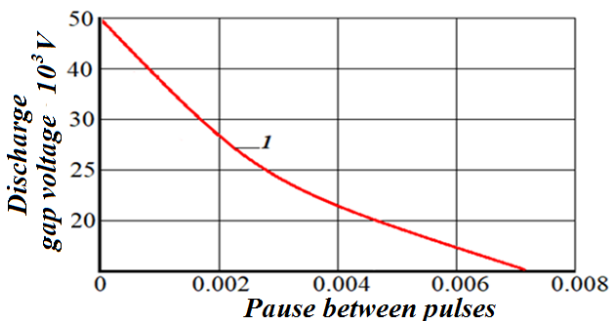**Fig. 3.** Characteristics of the change in the voltage of the discharge gap in the pause between pulses:1 – $C_{d.g.}$ = $10^{-9}$ F

An analysis of the change in the periodic impulse and free voltages of the discharge gap (Figure 4) shows that the barrier discharge proceeds in an area of 1,2,3,4.
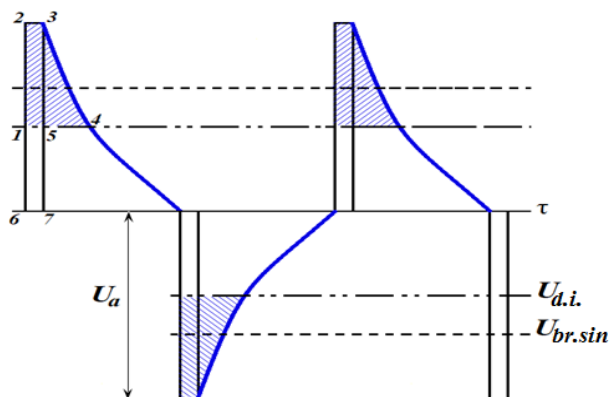


**Fig. 4.** Changes in the periodic pulsed and free voltages of the discharge gap

The ignition voltage of the discharge $U_{i.d}$ is the starting point for this website. When the ozone generator is powered by a sinusoidal voltage, $U_{d.g.sin}$, it is noticed that the pulse amplitude surpasses the breakdown voltage. The area bounded by points 1,5,6,7 describes the energy of the impulse voltage. This area is smaller than the area that the barrier discharge takes up. As a result, the efficiency of periodic voltage pulses can be much greater than that of sinusoidal voltages.

## 3. Results and Discussion

### 3.1 Results of studies of energy indicators of pulsed and sinusoidal voltages

Existing ozone electrosynthesis devices are powered by a sinusoidal voltage with a frequency of 500 Hz. The supply of the ozone generator with a sinusoidal voltage leads the current i in the circuit to a complex harmonic composition. At the times of the discharge, the average curve of the instantaneous value of the current I exhibits discontinuities (Figure 5). On the supply voltage period, the discharge happens twice and then stops. The existence of a discharge is

possible if the operating voltage $U_{a.s}$ on the ozonizer exceeds a certain minimum voltage $U_{cr.}$ In this case, $U_{a.s}$ is chosen to be less than the voltage of the total electrical breakdown of the discharge gap.
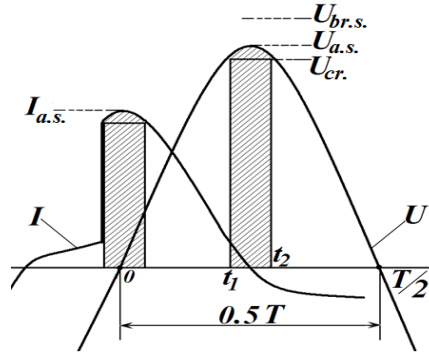


**Fig. 5.** Average curves of instantaneous voltage and current through the ozone generator

When powered by a sinusoidal voltage, the dielectric barrier heats up, which leads to a decrease in the ozone output. Because of this, the electrode in ozone generators is cooled by flowing water [12].

By substituting parameters that match to the barrier discharge process, the well-known theory of sinusoidal currents is used to examine the electrosynthesis of ozone when fed with a sinusoidal voltage. The ozone generator (GO) power supply circuit has a total power of :

$$P_{\text{gen}} = \left( \frac{1}{T} \int_0^T u^2 dt \right)^{0.5} \times \left( \frac{1}{T} \int_0^T i^2 dt \right)^{0.5} \tag{25}$$

Circuit power during barrier discharge

$$P_{paз} = \left( \frac{1}{T} \int_{t1}^{t2} u^2 dt \right)^{0.5} \times \left( \frac{1}{T} \int_{t1}^{t2} i^2 dt \right)^{0.5} \tag{26}$$

In equations (25) and (26)

$$u = U_{a.s} \sin\left( \omega_c t - 0.5 \right), \tag{27}$$

$$i = I_{a.s} \sin \omega_c t, \tag{28}$$

where: $\omega_s$ – angular frequency of sinusoidal voltage;

$u, i$ – instantaneous values of current and voltage;

$U_{a.s.}$- sinusoidal voltage;

$I_{a.s.}$ – discharge current amplitude;

$T$ – period of sinusoidal voltage;

$t_1$ – discharge start time;

$t_2$– discharge completion time;

The power loss is

$$P_{\text{los.}} = P_{gen} - P_{los.} \tag{29}$$

According to the examination of formulas (1–5), the period of time during which there is no discharge in the GO is when there is a substantial power loss. This leads to a working hypothesis: periodic voltage pulses having a shape comparable to the shaded area of a sinusoidal voltage, i.e. rectangular with a high duty cycle, can boost the efficiency of the ozone electrosynthesis process (Figure 6). In this case, it is possible to increase the amplitude of the supply voltage $U_{i.a}$ above the breakdown threshold of the sinusoidal voltage $U_{br.s}$, which is characterized by the overvoltage factor [13, 14, 15].

$$K = U_{u.a} / U_{np.c}. \tag{30}$$

With pulsed power, the GO capacitance is charged to the amplitude value of the voltage, which is higher than the amplitude of the sinusoidal voltage. In the time interval $0 - t_1$ (Figure 6), the discharge power is determined by the dependence

$$P_{0-t1} = I_{a.i} \cdot U_{a.i}. \tag{31}$$
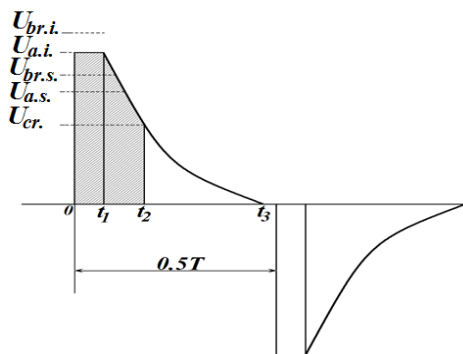
**Fig. 6.** The process of changing the voltage across the discharge gap when powered by periodic voltage pulses

In the pause between pulses (time from $t_1$ to $t_2$), the discharge power will be determined by the charge accumulated in the GO tank. The charge will be discharged on the loss resistance $R_{br.}$ GO, resistance $R_{win.}$ and inductance $L$ of the secondary winding of the step-up transformer, which are elements of the oscillatory circuit. At the same time, due to the fact that $R/2L<(CL)^{0.5}$, we have low attenuation in the circuit, which is the sum of the loss currents on $R_{br.}$ (the first term on the right side of the equation) and the process in the circuit $R_{win.}LC_{GO}$:

$$P_{t1-t2} = \frac{U_{a.i}^2}{R_{br}} e^{-\frac{2t}{R_{br}C_{TO}}} + \frac{U_{a.u}^2}{R_{win}} e^{-\frac{tR_{win}}{2L}} \left( \cos \omega_{o.c} t + \frac{R_{win}}{2\omega_{o.c}} \sin \omega_{o.c} t \right), \qquad (32)$$

where: $\omega_{o.c.}$ – angular frequency of the oscillatory circuit.

Starting from time $t_2$, the voltage of the discharge gap decreases below the critical $U_{k.d.}$, the discharge stops, and the capacitance is discharged to $R_{win.}$ what is the power loss:

$$P_{t2-t3} = \frac{U_{a.i}^2}{R_{win}} e^{-\frac{tR_{win}}{2L}} \left( \cos \omega_{o.c} t + \frac{R_{win}}{2\omega_{o.c}} \sin \omega_{o.c} t \right). \qquad (33)$$

Analysis of equations (2.31-2.33) shows that the bulk of the energy of periodic voltage pulses is spent on the discharge process, which is accompanied by ozone electrosynthesis.

In high-voltage ozone generator power supply circuits, the active resistance of the secondary winding of the step-up transformer can be in the range from 2 to 20 kOm, inductance from 50 to 200 H, capacitance of ozone generators $10^{-7}$ to $10^{-9}$ F. In this case, the condition $(R_{win}/2L)<(LC)$ and the discharge in the circuit proceeds with low attenuation [15]. To transfer the circuit to the critical attenuation mode, in parallel with the GO, a capacitor with a capacity exceeding the capacity of the GO can be switched on, which will increase the frequency of periodic voltage pulses and, accordingly, the energy indicators of the ozone electrosynthesis process.

## 4. Conclusions
a)  Discharge processes in technological discharge gaps can be stabilized under the condition of full compensation of space charges in the discharge gap in the pause between pulses.
b)  Periodic high voltage pulses should have a duty cycle of more than 5 to provide sufficient time for the recombination of space charges in the discharge gap.
c)  Efficiency ozone generators when powered by a sinusoidal voltage is 1 ... 2%. The rest of the energy is converted into heat, for the removal of which the electrodes are cooled by running water.
d)  Due to the instability of the discharge processes at a sinusoidal voltage, the existing ozone generators do not provide for control and automatic regulation of the process. In the case of stabilization of the discharge current, by the value of which it is possible to control and automatically regulate the process.
e)  An analysis of the processes in the discharge gap revealed that due to the excess of the amplitude of the voltage pulses of the electrical strength of the discharge gap at a sinusoidal voltage and the supply of the next pulse after the complete recombination of volume charges, the energy associated with discharge processes exceeds the energy converted into heat. Therefore, the expected efficiency may exceed 50%.
f)  Depending on the inductance and active resistance of the secondary winding of the step-up transformer and the capacitance of the ozone generator, the frequency of periodic high voltage pulses should not exceed 1000 Hz.

g) To confirm the results of theoretical studies, it is necessary to conduct experimental studies. These studies must be carried out with a source of high-voltage voltage pulses with a frequency control range of 100 ... 1000 Hz, a pulse duty cycle of 5 ... 20, and a maximum amplitude of voltage pulses of 30 kV. In addition to the above parameters, it is necessary to provide for a separate regulation of the amplitude of both polarities within (0.5 ... 1) Ua.

h) To generate periodic high-voltage voltage pulses, a circuit was adopted that operates on the principle of generating low-voltage periodic voltage pulses with subsequent increase.

i) The use of periodic high-voltage pulses makes it possible to increase the ozone concentration at the GO outlet by a factor of 3.9. In this case, there is practically no process of heating the dielectric barrier.

j) When using pulsed periodic voltages, less heat-resistant materials can be used and the required dielectric properties can be imparted to them.

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