Analysis of processes in the supply chain of ozone generator by sinusoidal and pulse voltage

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Abstract. Based on an analysis of the processes in the supply circuit of ozone generators with periodic sinusoidal and pulsed voltages of high duty cycle, it is revealed that when powered by a sinusoidal voltage, a significant part of the energy consumed is spent on heat loss. Only in a small part of the sinusoidal voltage, namely in the zone of reaching the amplitude value, a barrier discharge occurs with the generation of ozone. Therefore, the efficiency ozone generators at sinusoidal voltage do not exceed 2%. When powered by a pulsed voltage, ozone generation occurs almost during the application of the entire voltage pulse; in addition, ozone generation also occurs in the pause between pulses, due to oscillatory processes in the pause between pulses. In addition to a significant increase in ozone yield, there is practically no process of heating the dielectric barrier.

1. Introduction

Since its discovery in the late 18th century, ozone has been of constant interest to specialists in various fields and researchers due to its unique properties, primarily due to its high oxidizing and disinfecting ability. According to its oxidizing abilities, ozone (oxidation potential of 2.07 V) takes the third place among the known oxidizing agents, chlorine (1.73 V) - the eighth, and oxygen - the thirteenth. In industrialized countries, ozone is widely used in the chemical and petrochemical, metallurgical, electronic, pulp and paper, paint and varnish, microbiological, food industries, as well as in mechanical engineering, agriculture, medicine and municipal services. It is known that by-products formed during chlorination of water are toxic substances and cannot be completely removed from drinking water. During ozonation, in contrast to chlorination, oxidation reactions occur in which nontoxic final compounds are formed. These compounds are easily removed by filtration. In industrial ozone generators, a barrier discharge is applied when powered by a sinusoidal voltage. The main disadvantage of these devices is the preliminary preparation of air, cooling the ozone generator with running water, low efficiency - no more than 2%.

To assess the limits of increasing the efficiency of ozone electrosynthesis in a barrier discharge (BR), kinetic models are used that take into account a number of the most important processes that form the balance of O3. Examples of calculating the efficiency of the conversion of O2 to O3 are given in [1-3].
However, in experiments [4-6], a yield exceeding 300 g/kW×h was achieved. These results were obtained with a discharge in oxygen at an ozone concentration of less than 10 g/m³. However, according to [7, 8], industrial equipment from Osmonics Inc. (USA) produces ozone with a specific cost of 4 kW×h/kg at an ozone concentration of 150 g/m³, which exceeds the previous data in this area by about two times.

It is known that the energy yield of ozone in a discharge depends on many factors, including the material of the dielectric barrier and its thickness, as well as the magnitude of the interelectrode gap. A discussion of the role of these factors is given in [1, 7-15].

However, information sources do not contain information on the influence of the voltage form on the process of ozone electrosynthesis. This disadvantage was considered in this article.

2. Methods

The development of a more efficient method of electrosynthesis will increase the ozone output, efficiency, eliminate the heating of the dielectric barrier and simplify the process of preparing the air before being fed to the ozone generator, which will simplify the technological scheme of ozone electrosynthesis and expand the scope of their use, in particular, the creation of household appliances for disinfection of drinking water, which especially relevant for rural areas, farms and others.

The need for research and development work is also due to the lack of small-sized, high-performance, economical plants for the electro-synthesis of air ozone on the Uzbek market. In addition to the well-known technological processes using ozone, it is of interest to use ozone in greenhouses to disinfect irrigation water, to control pests, plant diseases, putrefactive bacteria and spores and others.

In existing ozone electrosynthesis devices, sinusoidal voltages with a frequency of 500 Hz are used. When the ozone generator is powered by a sinusoidal electromotive force (emf) source, the current $i$ in the circuit has a complex harmonic composition. The averaged curve of the instantaneous value of current $i$ contain discontinuities (Figure 1) at the moments when the discharge occurs. The discharge on the period of the supply voltage $u$ twice occurs and stops. The existence of a discharge is possible if the operating voltage $U_{s.w.a.}$ on the ozonizer exceeds a certain minimum voltage $U_{cr}$. In this case, $U_{s.w.a.}$ is selected less than the voltage of the total electrical breakdown of the discharge gap. When powered by a sinusoidal voltage, the dielectric barrier is heated, which leads to a decrease in ozone output.

![Figure 1. Averaged curves of instantaneous voltage and current through an ozone generator when powered by a sinusoidal voltage](image-url)
Therefore, ozone generators provide for cooling with a running water of an electrode covered by a barrier.

Let us analyze the process of electrosynthesis of ozone when supplied with sinusoidal voltage using the well-known theory of sinusoidal currents with the substitution of parameters corresponding to the process of barrier discharge [1]. The total power circuit of the ozone generator (GO) is

\[
P_{\text{wind}} = \left( \frac{1}{T} \int_0^T u^2 \, dt \right)^{0.5} \times \left( \frac{1}{T} \int_0^T i^2 \, dt \right)^{0.5}. \tag{1}
\]

Circuit power during barrier discharge

\[
P_{\text{allow}} = \left( \frac{1}{T} \int_{t_1}^{t_2} u^2 \, dt \right)^{0.5} \times \left( \frac{1}{T} \int_{t_1}^{t_2} i^2 \, dt \right)^{0.5}. \tag{2}
\]

In equations (1) and (2)

\[
u = U_{s.w.a} \sin (\omega_s t - 0.5), \tag{3}
\]

\[
i = I_{s.w.a} \sin \omega_s t, \tag{4}
\]

where \( \omega_s \) – is the angular frequency of the sinusoidal voltage;

\( u, i \) – instantaneous current and voltage;

\( U_{a.c} \) - sine wave amplitude of voltage;

\( I_{a.c} \) – discharge current amplitude;

\( T \) – sine wave period;

\( t_1 \) – discharge start time;

\( t_2 \) – discharge completion time.

Power loss is

\[
P_{\text{los.}} = P_{\text{tot.}} - P_{\text{los.}}. \tag{5}
\]

From the analysis of formulas (1 ... 5) it follows that a significant power loss occurs at a time at which there is no discharge in the ozone generator. The working hypothesis follows from here: it is possible to increase the efficiency of the ozone electrosynthesis process by using periodic voltage pulses in the form similar to the shaded region of the sinusoidal voltage, i.e. rectangular with a large duty cycle (Figure 2). In this case, it is possible to increase the amplitude of the supply voltage \( U_{p.a} \) above the breakdown threshold of the sinusoidal voltage \( U_{s.w.b.} \), which is characterized by an overvoltage coefficient

\[
K = \frac{U_{p.a.}}{U_{s.w.b.}}. \tag{6}
\]

With pulse power, the capacitance of the ozone generator is charged to the amplitude value of the voltage, which is higher than the amplitude of the sinusoidal voltage. In the time interval 0 - t1 (Figure 2), the discharge power is determined by the dependence
\[ P_{0-t_1} = I_{p,a} \cdot U_{p,a}. \]  

(7)

In the pause between pulses (time from \( t_1 \) to \( t_2 \)), the discharge power will be determined by the charge accumulated in the \( OG \) capacitance. The charge will be discharged at the loss resistance \( R_{los} \), the \( OG \) capacitance, the resistance \( R_{wind} \) and the inductance \( L \) of the secondary winding of the step-up transformer, which are elements of the oscillating circuit (Figure 3). Moreover, due to the fact that \( R/2L < (CL)^{0.5} \) [1], we have a small attenuation in the circuit, which is the sum of the loss currents at \( R_{los} \) (the first term of the right side of the equation) and the process in the circuit

\[
P_{t_1-t_2} = \frac{U_{p,a}^2}{R_{los}} e^{-\frac{2t}{R_{los}C_{OG}}} + \frac{U_{p,a}^2}{R_{tot}} e^{-\frac{tR_{los}}{2L}} \left( \cos \omega_{o.c.} t + \frac{R_{wind}}{2\omega_{o.c.}} \sin \omega_{o.c.} t \right).
\]  

(8)

In formula (8), the angular frequency \( \omega_k \) to the oscillatory circuit is determined by the relation

\[ \omega_{x:k} = \left( \frac{R_{wind}^2}{4L^2} - \frac{1}{LC_{OG}} \right)^{0.5}. \]  

(9)

Starting from time \( t_2 \), the voltage of the discharge gap decreases below the critical \( U_{cr} \), the discharge ceases, and the capacitance is discharged by \( R_{wind} \), which is the loss power
An analysis of equations (7-10) shows that the bulk of the energy of periodic voltage pulses is spent on the discharge process, which is accompanied by ozone synthesis.

$$P_{t2-t3} = \frac{U_{p.d.}^2}{R_{obm}} e^{-\frac{tR_{tot}}{2L}} \left( \cos \omega_{o.c.} t + \frac{R_{tot}}{2\omega_{o.c.}} \sin \omega_{o.c.} t \right) \quad (10)$$

An analysis of equations (7-10) shows that the bulk of the energy of periodic voltage pulses is spent on the discharge process, which is accompanied by ozone synthesis.

**Figure 3.** The equivalent circuit of the power circuit of the discharge gap of the ozone generator in the pause between pulses.

In high voltage ozone generators power supply circuits, the resistance of the secondary winding of the step-up transformer can be in the range from 2 to 20 kΩ, inductance from 50 to 200 H, the capacity of ozone generators is 10-7 to 10-9 F. In this case, the condition \( (R_{wind}/2L) < (LC) \) and in the circuit the discharge flows with low attenuation. To transfer the circuit to critical attenuation mode, in parallel with an \( OG \), you can turn on a capacitor with a capacity exceeding the \( OG \) capacity, which will increase the frequency of periodic voltage pulses and, accordingly, the energy indicators of the ozone electrosynthesis process.

**Figure 4.** Oscillogram of sinusoidal voltage (upper) and current (lower) when powered by an ozone generator (sweep 0.01 s/cm)

Oscillography was carried out when the ozone generator was powered by a step-up transformer. A voltage of 10 kV was applied to the ozone generator. For voltage oscillography, an ohmic voltage divider was connected to potential \( OG \) electrodes, and an ohmic shunt was connected between the grounded electrodes and ground to oscillate the current.
3. Results and Discussions
The waveform of voltage and current when powered by a sinusoidal voltage is shown in Figure 4. An oscillogram of the discharge current clearly shows high-frequency discharges. For an approximate determination of the discharge frequency, a current waveform was taken at a sweep of 0.25 ms/cm. The oscillogram approximately determines the frequency of these discharges, which lies in the limit of 400 kHz. This is the main reason for the heating of the dielectric barrier. Similar studies were conducted for pulsed voltages. In studies, a frequency of 50 Hz was chosen. Periodic voltage pulses were supplied from a machine generator. This is due to the stability of the type of voltage. As studies of the electronic generator have shown, the shape of the voltage pulse curves is unstable, and is subject to frequency fluctuations and the amplitude in the form during operation. According to the waveform, the discharge current has rapidly decaying oscillations with a frequency of 400 Hz. The practically oscillatory process ends after the first oscillation period. To reduce the frequency of oscillations, and accordingly to reduce the damped oscillatory process of the current, a capacitor was connected in parallel to the latter in the supply circuit of the ozone generator.

![Oscillogram of a periodic voltage pulse (upper) and current (lower) when connected in parallel with a condenser ozone generator (sweep 0.01 s/cm)](image)

**Figure 5.** Oscillogram of a periodic voltage pulse (upper) and current (lower) when connected in parallel with a condenser ozone generator (sweep 0.01 s/cm)

The capacitance of the connected capacitor is twelve times greater than the capacity of the ozone generator (respectively 40 and 470 pF). In this case, the current in the circuit oscillates at a frequency of 200 Hz (Figure 5.). In this case, the discharge current has stable amplitude with attenuation. In this case, the energy accumulated in the supply circuit of the ozone generator will be used for additional ozone synthesis.

4. Conclusion
- When powered by a sinusoidal voltage, a barrier discharge occurs for a short period of time when the voltage reaches its amplitude value. The discharge flows with a frequency of the order of 400 kHz, while the amplitude of the discharge current and the frequency varies over large limits, i.e. has a random character. This determines the process of heating the dielectric barrier.
- When powered by a pulsed voltage of large duty cycle, high-frequency discharges are absent, and in the pause between pulses an oscillatory process with a large attenuation takes place.
When a capacitor with a capacity of 10 or more times larger is connected in parallel to the ozone generator, the oscillatory process in the circuit oscillates at a frequency of 200 Hz. In this case, the discharge current has stable amplitude with attenuation.

The results of experimental studies of the energy indicators of the ozone electrosynthesis process showed that when feeding a tubular ozonizer with a sinusoidal voltage, the ozone output was 88 g/kW hour, and when feeding a pulse voltage with an amplitude coefficient of more than 5, the ozone output was 287 g/kW× hour. These data were taken at a surface temperature of the ozone generator of 240°C. At a temperature of 450°C, the output at a sinusoidal voltage decreased by 3 times, and the temperature at a pulsed voltage practically does not increase. A similar phenomenon requires additional basic research.

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