

PARAMETERS INFLUENCING THE SYNTHESIS OF OZONE
IN ELECTRICAL DISCHARGES

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ABSTRACT

In this paper influences and interdependences of discharge parameters on the synthesis of ozone are shown and relations to the ozone yield at various discharge characteristics are considered. Surface effects at the solid dielectric are discussed on the basis of the local ozone distribution within the discharge gap. Aspects of the influence of the gap length and its splitting into several gaps by dielectrics are demonstrated.

1. INTRODUCTION

The application of fast rising voltages to an ozonizer can improve the efficiency of the ozone generation /1,2,3,4,5,6/. Furthermore it was demonstrated that in addition to a short rise time of the voltage an intensive energy input into the discharge is necessary for a high ozone yield /7,8/. It is possible to operate large ozonizers with fast rising voltages and improved ozone yields can be achieved /9,10/. In this paper the efficiency of the ozone generation and loss involving mechanisms are discussed on the basis of experiments.

2. CHARACTERISTICS OF THE DISCHARGE AND OZONE YIELD

The generation of ozone, the energy consumption and the efficiency of ozonizers are influenced by geometrical, electrical and several other operating parameters /2,7,9/. Of particular interest are the steepness of the applied voltage, the amplitude and the duration of the discharge current as well as the electrical charge absorption of the dielectric.

In fig. 1 the ozone yield A , the relative charge absorption of the dielectric $\Delta q / \Delta q_{\max}$ and the maximum of the discharge current i_{\max} are shown as a function of the steepness of the voltage for a small plate shaped model ozonizer with a gap length of $2 \times 3 \text{ mm}$ /7/. The improvement of the ozone yield by the increased steepness of the voltage is accompanied by an enlarged amplitude of the discharge current and a greater charge absorption of the dielectric. Both the improved ozone yield and the enlarged energy consumption due to the higher charge absorption lead to an increased ozone production. The experiments have demonstrated that a remarkable improvement of the ozone yield compared to sine waves of mains frequency can be achieved only for voltage steepnesses higher than some ten $\text{kV}/\mu\text{s}$. Only in this case an enlarged discharge current and electrical charge absorption is observed. Furthermore it can be seen that the steepness of the voltage provides for an increase of the relative charge absorption of the dielectric. This means a reduction of the remaining voltage over the discharge gap. This voltage is zero if the dielectric is charged completely. In this case $\Delta q = \Delta q_{\max}$.

The increase of the discharge current maximum in fig. 1 coincides with an important modification of the discharge characteristic of the model ozonizer investigated. At low steepnesses of some ten kV/ μ s the discharge consists mainly of a large number of small single current pulses similar to those at ozonizers supplied by sine wave voltages. With increased steepness an enlarged main current pulse can be observed and the number of the small pulses is reduced. The small pulses after the main one are vanishing and the amplitude of the main pulse is higher if the steepness is increased further. Finally the discharge is supplied only by one current pulse.

In fig. 2 the relations between the amplitude of the discharge current pulse, the relative charge absorption and the ozone yield are plotted. From these curves it can be seen that for a high ozone yield the discharge characteristic obtained at high current pulses and a complete charging of the dielectric is advantageous. But to achieve such discharge characteristics several boundary conditions have to be taken into account: a high current pulse and a complete charging of the dielectric require a homogeneous ignition of the discharge by a fast rising voltage. In addition the impedance of the supplying circuit has to allow a current flow which is able to charge the dielectric as high as possible. If the discharge circuit prevents such a complete charging a smaller ozone yield is achieved in spite of a homogeneous ignition.

Reasons for this behaviour can be the following phenomena. With an extended discharge duration the current transport is taken over more and more by ions producing no ozone /11/. If the dielectric is not charged completely, local differences on the surface can be responsible for loss involving mechanisms. A small charging of the dielectric implies remaining field strengths over the discharge gap causing the ignition of single discharge channels with high local energy density well-known from the discharge at sine waves /12/.

Running experiments have shown an improvement of the ozone yield could be achieved also if the dielectric is not charged completely. The voltage across the ozonizer has to be reduced to a value that the voltage over the gap is minimized.

Experiments with large ozonizer units have shown that the results achieved with small model ozonizers can be transferred in principle. Fast rising voltages allow a homogeneous ignition of the discharge and a nearly complete charging of the solid dielectric. But compared to the small model ozonizer the current amplitude and the current duration differ remarkably, see fig. 3. The duration of the current flow as well as the amplitude of the current is determined mainly by the elements of the circuit supplying the discharge. Current flow durations up to several 100 ns were obtained. Nevertheless a homogeneous discharge can be maintained leading to yields comparable to those achieved with the small ozonizer model. For a high steepness of the voltage a low inductance and resistance of the discharge circuit is necessary which means a compact set-up of the elements of the circuits.

3. LOCAL DISTRIBUTION OF THE OZONE PRODUCTION AND SURFACE EFFECTS

In fig. 4 the relative absorption E/E_{elec} of a light beam at a wave length of 254 nm is plotted versus the distance X from the grounded electrode as indicated in the figure. Relative absorption means the absorption E at the

distance X referred to that measured at the electrode E elec. All three curves of fig. 4 show an increase of the local ozone generation towards the dielectric. This increase is dependent on the voltage steepness applied to the ozonizer. A smaller voltage steepness reduces this effect, at sine waves the ozone concentration at the dielectric is remarkably decreased.

Obviously the voltage steepness and the involved change of the characteristic of the discharge have an influence on the effects near the surface of the dielectric. At steep voltages the whole area can be charged by a homogeneous discharge with a nearly endless number of simultaneous discharge channels which may--corresponding to $/12/$ - widen towards the surface of the dielectric resulting in a homogeneous charge distribution across the surface. If there are local differences in the charge distribution upon the surface of the dielectric, a charge transportation across the surface can be caused by the surface conductivity. This equalizing process results in additional losses without ozone formation and occurs, if the number of simultaneous discharge channels is reduced by an insufficient homogeneity of the discharge. If the charge transportation process across the surface takes place by surface discharges further losses are obtained. This is in agreement with $/3,12,13/$. These effects correspond to the correlation between the ozone yield and the discharge characteristics.

In fig. 5 the local absorption E is plotted versus the distance X from the grounded electrode. The distance of the electrodes was 8 mm, the thickness of the dielectric 2 mm resulting in a total length of the discharge gaps of 6 mm. The local arrangement of the dielectric was varied, for curve 1 it was arranged at the high voltage electrode, for curve 2 in a distance of 1 mm and for curve 3 in the middle between the electrodes. The comparison of the curves demonstrates that the shifting of the dielectric from one electrode towards the middle of the space between the electrodes leads to a reduced ozone production at the surface of the dielectric and a slight increase at the electrodes. In view of a nearly constant average production this means a better uniformity in the distribution of the ozone generation.

By comparison of curve 1 and curve 1(+) in fig. 5 the influence of the humidity on the ozone formation can be seen. The ozone generation is lowered in the whole discharge volume.

4. INFLUENCE OF GEOMETRICAL PARAMETERS ON THE OZONE YIELD

In fig. 6 the ozone yield is plotted versus the concentration for three different gap lengths of a tube ozonizer of 1 m length supplied with steep voltages. At small ozone concentrations the highest ozone yields are available at large gap lengths. With increasing concentration however higher yields can be obtained at smaller gap lengths. At gap lengths of 4 and 6 mm even there appears a certain saturation effect of the ozone concentrations. This behaviour is mainly due to the specific cooling conditions of the ozonizer. At larger gap lengths the energy input and the temperature of the discharge volume are increased resulting in a higher thermal destruction rate of ozone. At a certain ozone concentration the generation rate is equal to the destruction rate (curve 3, fig. 6).

Experiments with other ozonizer units have shown that the amount of the decrease of the yield and the observed saturation effect are strongly influenced by the specific design and set-up of the ozonizer itself and its cooling-conditions. Compared to fig. 6 much larger concentrations are achievable.

Another interesting point is the splitting of a large gap length into several small gaps. In fig. 7 the results of corresponding experiments with ozonizers of different size are given supplied by sine waves and operated in oxygen respectively in air. For the small model ozonizer (electrode area of 55 cm²) a comparison of curve 1 and curve 2 shows the well-known decrease of the yield at larger gap lengths. But if the large gap length is splitted into four gaps by inserting three dielectrics the same yields observed at the small gap length are achieved.

An equivalent behaviour was obtained with units where the electrode area of the plate shaped model ozonizer was enlarged up to 2500 cm² (compare curves 4 and 5). If the gap length of 8 mm is splitted by three dielectrics into four gaps of 2 mm each again a remarkable improvement is observed. This improvement even exceeds slightly the yields obtained from a 4 mm gap subdivided into two single ones (compare curves 3 and 5).

These results indicate that a splitting of a large gap into several small gaps improves the homogeneity of the discharge. In other words this means also that certain advantages of ozonizers operated with fast rising voltages can be achieved also in a sinusoidal operation mode.

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REFERENCES

- / 1 / L. M. L. F. Hosselet, Techn. Univ. Eindhoven, TH-Report 71-E-19, ISBN 90 6144 019X (1971)
- / 2 / J. Salge, P. Braumann, proceedings 4th Int. Symp. on Plasma-Chemistry, Zürich (1979)
- / 3 / P. Braumann, thesis, TU Braunschweig (1981)
- / 4 / B. Eliasson, U. Kogelschatz, proceedings 4th Int. Symp. on Plasma-Chemistry, Zürich (1979)
- / 5 / B. Eliasson, U. Kogelschatz, proceedings XIVth Int. Conf. on Phenomena in Ionized Gases, Grenoble (1979)
- / 6 / L. A. Rosocha, thesis, University of Wisconsin-Madison (1979)
- / 7 / J. Salge, H. Kärner, M. Labrenz, K. Scheibe, P. Braumann, proceedings Sixth Int. Conf. on Gas Discharges and their Applications, Edinburgh (1980)
- / 8 / M. Labrenz, "Elektrische Gasentladungen zur Ozonerzeugung", TU Braunschweig, to be published
- / 9 / J. Salge, H. Kärner, M. Labrenz, K. Scheibe, P. Braumann, Congress report, Wasser Berlin 81, Berlin (1981)
- / 10 / K. Scheibe, "Über den Betrieb von Ozonisatoren mit steilflankiger Wechselspannung", TU Braunschweig, to be published
- / 11 / United States Patent 4, 016, 060 (Apr. 5, 1977)
- / 12 / M. Hirth, thesis, TH Karlsruhe (1979)
- / 13 / G. Pietsch, C. Heuser, Congress report, Wasser Berlin 81, Berlin (1981)

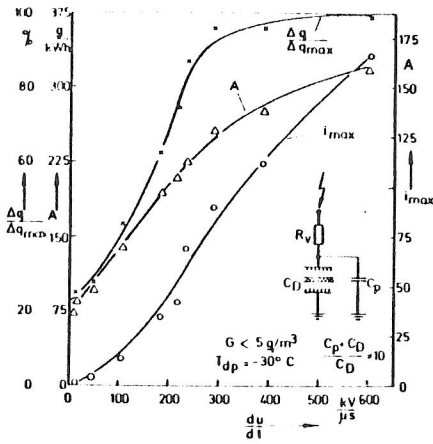


Fig. 1: Ozone yield A , relative charge absorption $\Delta q / \Delta q_{max}$ and maximum of the discharge current i_{max} versus the steepness of the voltage du/dt

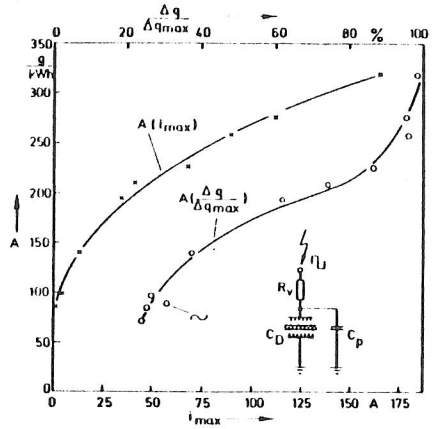


Fig. 2: Ozone yield A versus the maximum of the discharge current i_{max} and the relative charge absorption $\Delta q / \Delta q_{max}$

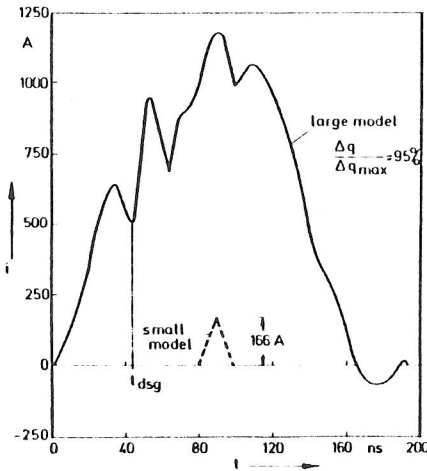


Fig. 3: Discharge current i for a large tube ozonizer in comparison with the small model

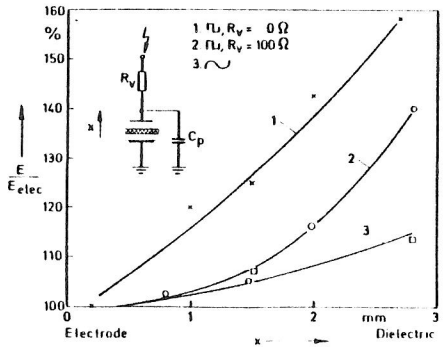


Fig. 4: Relative absorption E/E_{elec} in the discharge gap versus the distance x from the grounded electrode

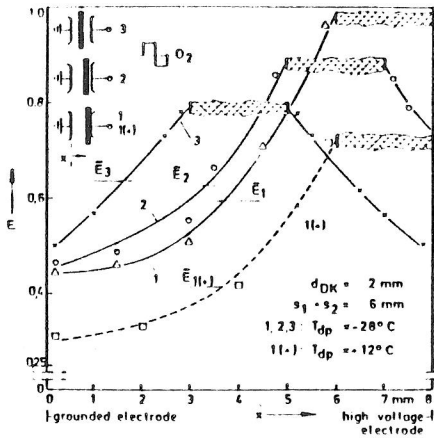


Fig. 5: Local absorption E in the discharge gaps versus the distance X from the grounded electrode

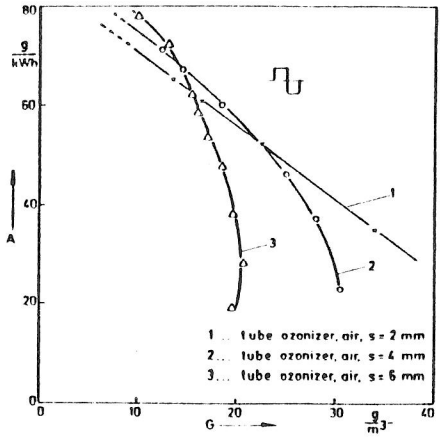


Fig. 6: Ozone yield A versus the concentration G for different gaps for a cooled tube ozonizer

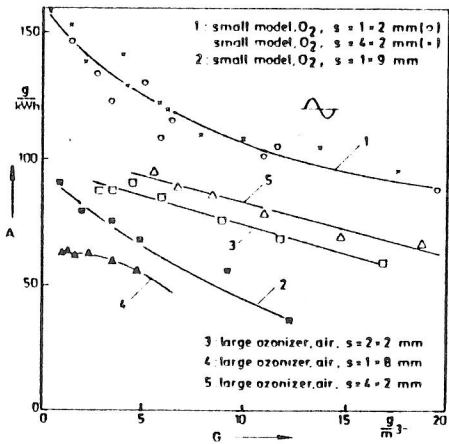


Fig. 7: Ozone yield A versus the concentration G for different gaps and arrangements of the dielectric