INVESTIGATION OF OZONE GENERATION ON DIELECTRIC SURFACES

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Abstract The ozone generation has been investigated with test arrangements which consist of parallel wires on a dielectric and a counter electrode on its back side. The temporal development and local distribution of the ozone concentration between the electrodes on the surface have been measured polarity dependent. The efficiency as well as the production rate have been determined depending on electrode distance and pressure.

INTRODUCTION

Usually ozone is generated in devices where microdischarges in a gas gap and surface discharges on the surface of a dielectric treat oxygen containing gases. Instead of using a combination of volume and surface discharges it is possible to produce ozone with surface discharges only [1, 2]. Low ignition voltages and effective cooling properties of such arrangements are obvious advantages in comparison to usual ozone generators.

For the design and optimization of such ozone generators it is necessary to have knowledge of the behavior of surface discharges as well as the ozone formation at different parameters.

EXPERIMENTAL SET-UP

In order to understand the discharge behavior experiments with straight wires in parallel on a dielectric plate have been performed. The metallic wires with a length of 22 cm and a diameter of 2 mm are fixed on glass plates with relative dielectric constants of 4.8 and 8.4. They are electrically connected. Because of insulation measures dis-

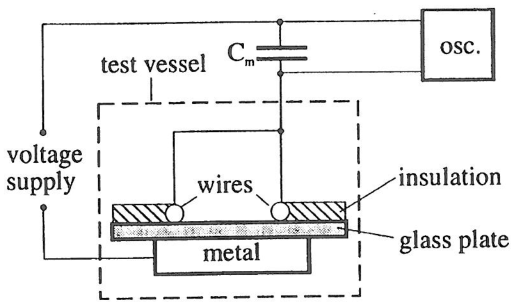

Fig. 1: Schematic diagram the plane arrangement

charges appear in the areas between the electrodes only. The counter electrode is a metallic block on the rear of the plate. The arrangement is fed by power supplies, which deliver single half cycles of voltage with 200 µs rise time and several cycles of ac voltage of 1 kHz frequency, respectively. The charge has been determined with a series capacitor (Fig. 1).

The local distribution of the ozone concentration has been measured by UV radiation absorption. A narrow UV beam at 253.7 nm with a diameter of about 0.8 mm in parallel to the wire is used. Half of the beam cross section is shaded by the dielectric. The sensitivity of the measuring system is about 8 $\mu g O_3$ /l. It is limited by the noise of the light source, a high pressure mercury lamp. The humidity was about 1 to 3 ppm.

For the investigation of the ozone yield a cylindrical device with a spring as surface electrode has been used. The prestressed metallic spring with a wire diameter of 1 or 2 mm and with pitches of 5 to 9 mm is pressed against the inner surface of a glass tube (ε = 4.8) with 0.64 m length, a thickness of 2.3 mm and an outer diameter of 44 mm. The outer dielectric surface of the tube is covered by an aluminum foil. The device is fed by ac voltages of 3 to 13 kV at a frequency of about 1 kHz. The gas flow through the tube is in the range of 1.7 to 17 l/min. The process gas is dried air at 10 ppm humidity. The device has been cooled by a water flow around the dielectric tube. The power of the ozone generator is determined by numerical integration of the time functions of the applied voltage and the measured charge.

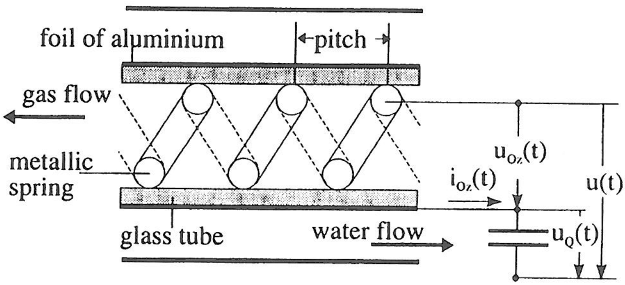

Fig. 2: Schematic diagram of the tubular arrangement

$$P = \frac{1}{T}\int_0^T u_{Oz}\, i_{Oz}\, dt = f \cdot C_m \int_0^T u_{Oz}(t)\, du_Q(t) \qquad (1)$$

MEASUREMENT RESULTS

Plane arrangement

The local distribution of the ozone concentration on the dielectric surface has been measured polarity dependent. In principle for the positive polarity the area where ozone can be detected is larger than in the negative case. This results from the different extensions of the discharges on the surface [3]. The temporal development of the ozone concentration at the wire electrode as well as the charge development following a generation of discharge channels at positive polarity is given in Fig. 3. The concentration behavior can be described by:

$$c(t) \;=\; c_0 \cdot \left(1 - \exp\left(\frac{t_0 - t}{\tau_1}\right)\right) \cdot \exp\left(\frac{t_0 - t}{\tau_2}\right) \qquad (2)$$

The time constant τ_1 describes the ozone formation process. It has been determined to be 45 ± 10 µs in air and 30 ±10 µs in oxygen. The constant τ_2 describes the local decrease of the ozone concentration. It is about 300 µs for air and 180 µs for oxygen. Increasing the voltage several generations of surface discharges with higher discharge channel density enhances the ozone concentration.

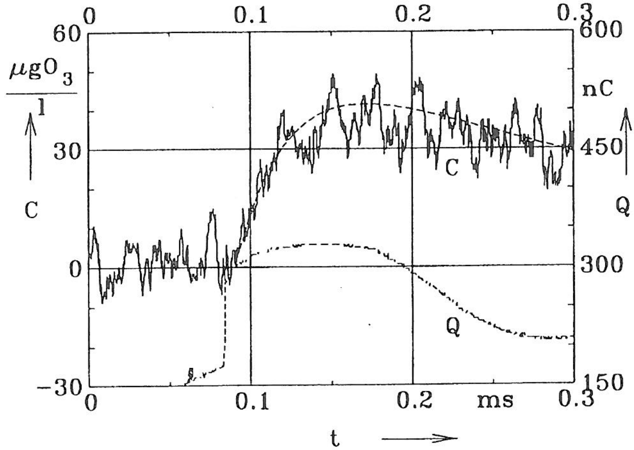

Fig. 3: Temporal development of ozone concentration near the wire electrode and of charge at positive polarity

Apart from using pulse voltages the influence of surface charges of preceding discharges has been studied with a sequence of ac cycles. For a single wire arrangement (wire Ø = 2 mm; plate thickness 2 mm; ε_r = 4.8) the temporal development of the ozone concentration in air at different humidity nearby the surface electrode is given in Fig. 4. At t = 0 the ac voltage rises with positive polarity and the amplitude is constant after two cycles. The first detected ozone belongs to a negative half cycle. For higher humidity the ozone concentration increase is more pronounced. However, after some cycles the averaged ozone concentration is decreasing in humid air and increasing in dried air. The pronounced increase in the second negative half cycle follows from an enlarged voltage amplitude.

The local distribution of ozone concentration after some cycles between parallel wires are nearly symmetric to the middle of the discharge arrangements [4]. The ozone concentration is enhanced nearby the electrodes. However, its value depends on the distance of the parallel electrode.

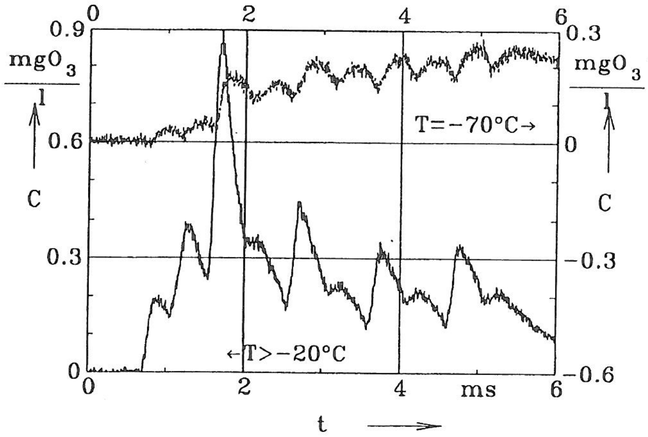

Fig. 4: The temporal development of the ozone concentrations nearby the wire electrode for different dew points T (single wire arrangement, $\varepsilon_r = 4.8$)

Tubular arrangement

With the tubular arrangement ozone concentrations have been measured at stationary conditions. In Fig. 5 the concentration is given for different spring pitches depending on voltage. With increasing pitch the ozone concentration rises at atmospheric pressure. In the high voltage range the concentration decreases. This decrease is shifted to higher voltages at an increased gas flow.

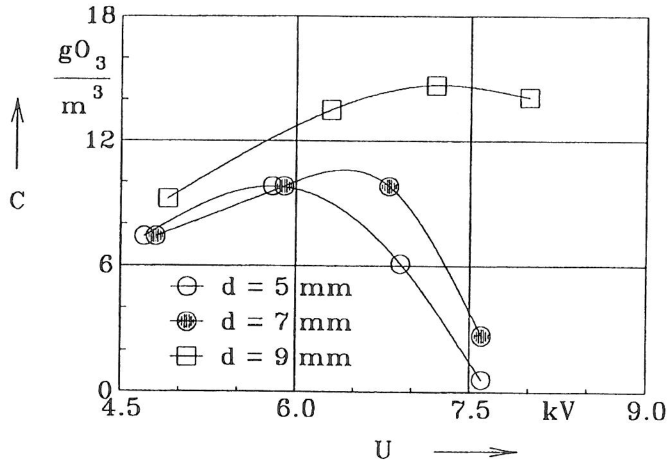

Fig. 5: Ozone concentration dependent on voltage for several spring pitches at p = 1 bar (tube ∅ = 44 mm; wire ∅ = 2 mm; air flow 1.7 l/min)

Higher values of ozone concentration could be achieved with increasing pressure. Fig. 6 shows the ozone concentration dependent on the power at 1 and 2.2 bar air. At low power input the concentrations are similar. At high power a considerable concentration increase is achieved at increased pressure only.

At power densities in the range of 0.15 to 2.5 kW/m^2 the ozone concentration is between 5 and 40 g O_3/kWh and the production rates between 10 and 70 g $O_3/h/m^2$.

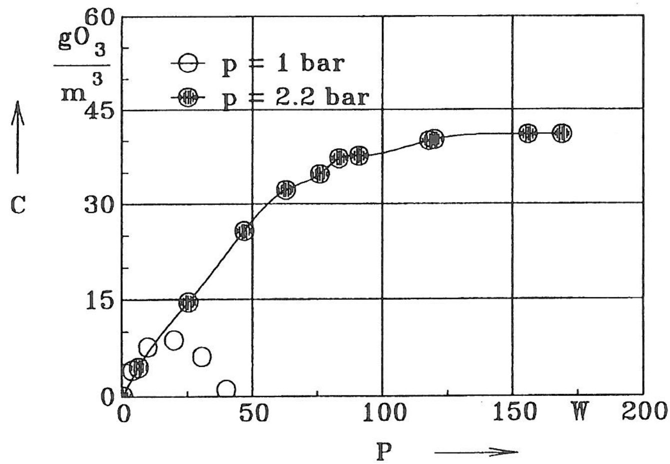

Fig. 6: Ozone concentrations in air dependent on the power for two pressures (tube ∅ = 44 mm; wire ∅ = 2 mm; pitch 7 mm; air flow 1.7 l/min)

DISCUSSION

Ozone concentration behavior

The ozone concentration is mainly influenced by the discharge behavior. The surface discharge consists of numerous discharge channels. Their extension and density increase with voltage. At negative polarity the degree of coverage with discharge channels is comparatively higher than at positive polarity [3]. At small electrode distance and high voltage the discharge channels of the facing electrodes interfere [4].

The measured time constants of the ozone formation process in air and oxygen are of the same order of magnitude as those measured elsewhere [5]. The time constant for the decrease of the ozone concentration τ_2 are determined by diffusion and convection processes.

From Fig. 4 it follows that within the negative half cycle the local concentration is higher than at positive polarity. This effect can be explained by the more homogeneous coverage of the surface with discharges. The concentration decay during each half cycle can be described by the time constant τ_2. In the case of enhanced humidity the polarity effect is more pronounced. The humidity effect has still to be explained.

Ozone yield

Running the surface discharge arrangement continuously steady state values have been measured. The mean ozone concentration rises first with voltage and decreases then with further rising voltage. The concentration increase is connected with the rise of the power and the discharge density while the decrease can be explained by a limited discharge area with interference of facing discharge channels and catalytic reaction chains with nitrogen oxides [6]. At small spring pitches the electrode length is longer at constant discharge area. In this case the effect is more pronounced and shifted to lower applied voltages (Fig. 5).

Increasing the gas pressure the ignition voltage rises and the discharged area is going down. Changing the pressure from 1 to 2.2 bar the maximum concentration is fourfold and the poisoning effect is shifted to higher power densities.

From the measurements in a limited range of parameter variation it follows that surface discharge ozone generators have at least similar efficiencies like generators with gas gaps. In addition to electrode distance and voltage the other design parameters as well as the process gas pressure and humidity have to be chosen carefully in order to get efficient surface ozone generation.

ACKNOWLEDGMENTS

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