

Pulsed Streamer Discharge Characteristics of Ozone Production in Dry Air

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ABSTRACT

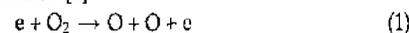
Experimental investigation of HV short pulsed streamer discharges in dry air-fed ozonizers under various operating conditions are reported. Ozone concentration, energy input and ozone production yield (efficiency) were measured at various voltages (14 to 37 kV), pulse repetition rates (25 to 400 pulses per second, pps), flow rates (1.5 to 3.0 l/min) and different gap spacings (10 to 20 mm) at a pressure of 1.01×10^5 Pa in dry air. A spiral copper wire (1 mm in diameter) made to a cylindrical configuration (18 to 38 mm in diameter) in a concentric coaxial electrode system of various dimensions was employed. A magnetic pulse compressor provided the HV and current pulses. Higher voltage and higher repetition rates yielded higher concentrations of ozone at a fixed air flow rate. The present investigation was extended to assess the performance of this pulsed ozone generator using dry air under desired conditions of high concentration and high yield of ozone for industrial applications.

1 INTRODUCTION

OZONE is being used increasingly as an alternative to chlorination of drinking water, in the treatment of industrial waste, in bleaching processes of textiles and paper pulp, in chemical synthesis of vitamins and perfumes, and in the processing of semiconductor devices. It has little detrimental effect on the environment because the natural decay product of ozone is oxygen, and in the presence of organic substances, carbon dioxide also may be created. It has the added advantage of less energy consumption than other alternatives, such as the chlorination process [1].

Historically, the major application of ozone has been in the treatment of drinking water: ozone is known as a potent bactericide and viricide [2]. Ozone is an unstable molecule, which decays into oxygen. So, it must be generated where it is required to be used, and it cannot be shipped or stored in a gaseous form [1]. Currently major efforts are being expended world wide to increase the efficiency of ozone production in order to reduce costs. Ozone synthesis from oxygen in dielectric barrier discharges has been studied [1-4]. However, there are relatively few studies of synthesis without a dielectric barrier using HV pulsed power although this offers substantial advantages [5, 6]. Usually ozone is generated by silent discharges using a dielectric barrier placed adjacent to the outer cylinder [1, 2, 3, 7] where very short durations of micro discharges prevail at the surface of the dielectric. A dc applied voltage has been used also for ozone generation in a wire-cylinder geometry [8].

Generally, the purpose of the ozonizer is to produce intense streamer discharges without inducing arc breakdown between the electrodes. A pulsed streamer discharge has been shown to be very effective for this purpose if it is long enough to produce intense corona but not too long to induce arc breakdown [5, 9]. The streamer discharge energized by a fast rising pulse voltage can produce an intense plasma, which creates atomic oxygen by dissociation [1]



and



Ozone is formed via



where M is a third collision particle which could be O_2 , O_3 and N_2 in air [1]. A HV pulse can produce an intense streamer discharge at room temperature and atmospheric pressure. Although the electrons are accelerated to a high level in the high field if the pulse duration is very short, it does not readily lead to a spark breakdown [5, 6, 9-12]. The industrial requirements of the concentration of ozone depend on the applications in which the ozone is used, and this in turn determines the operational conditions of the ozonizer.

In the present paper, most of the parameters affecting the ozone production were examined. These include pulse voltage (14 to 37 kV), pulse repetition rate (25 to 400 pps), gas flow rate (1.5 to 3.0 l/min) and gap spacing (10 to 20 mm) at a pressure of 1.01×10^5 Pa of dry air. A figure of merit of an ozonizer is the concentration of ozone in parts per

million (ppm) and the ozone production yield given in g/kWh, and both of these quantities have been determined at different discharge conditions.

2 EXPERIMENTAL PROCEDURE

A schematic diagram of the apparatus to generate ozone is shown in Figure 1. The reactor in which ozone was produced consisted of a discharge tube which contained a spiral copper wire of 1 mm in diameter, made into a cylindrical configuration as shown in Figure 2. The wire was coiled on formers, vinyl chloride tubes, having diameters in the range 18 to 38 mm.

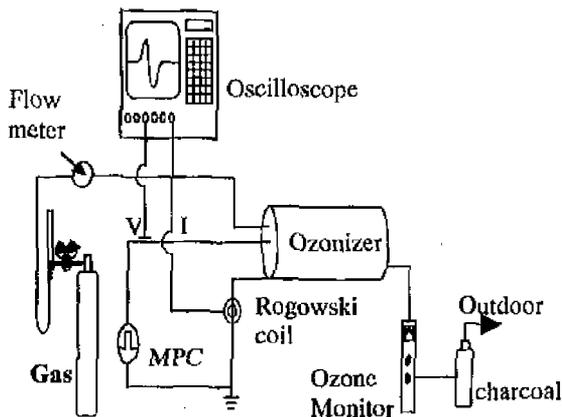


Figure 1. Experimental setup for generation of ozone.

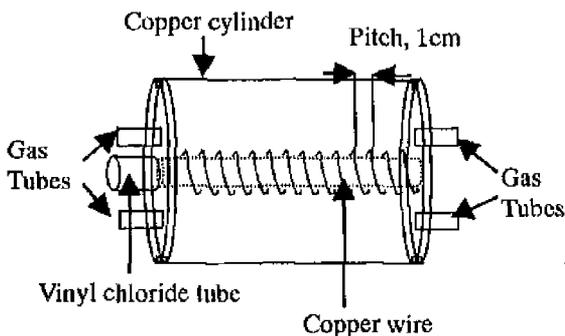


Figure 2. Reactor configuration. Wire diameter 1 mm; reactor length 1 m.

The inner diameter and the length of the outer copper cylinder were 58 mm and 1 m, respectively. Therefore, the gap spacing between the electrodes was varied over the range 10 to 20 mm by using different formers.

If the electric field is made highly non-uniform, then high-energy electrons are generated close to the wire where the field is high. The high-energy electrons dissociate the oxygen molecules into oxygen atoms (reaction 1) and thus the production of ozone is largely produced in the vicinity of the HV wire [11, 12]. Dry air was used which had concentrations of nitrogen 78.08%, oxygen 20.95%, carbon dioxide 0.03%, argon 0.93% and traces of neon, helium, methane, krypton, xenon, hydrogen and nitrous oxide [13]. The gas flow in the discharge tube was axial. The flow rate and the ozone concentration were monitored by

means of a flow-stat meter (Floline, model SEF-1 R, made by Stec Inc, Japan) and a UV ray absorption ozone meter (Ebara model DOA300), respectively. The UV absorption measurements were carried out at 253.7 nm where the absorption cross section is large at $1.14 \times 10^{-21} \text{ m}^2$ [1, 14]. This method has the advantage that it is instantaneous and does not suffer from errors arising from the presence of other substances such as nitrogen oxides in the gas [1].

The pulsed voltage and the discharge current were measured using an oscilloscope via a HV capacitive divider and a Pearson Rogowski coil, respectively. A Hewlett Packard digital oscilloscope (HP 54542A) with a maximum bandwidth of 500 MHz and a maximum sample rate of $2 \times 10^9 \text{ s}^{-1}$ recorded the signals. The power VI and the energy $VI dt$ input to the discharge per pulse were determined from the digitized signals, using a computer. The flow rate of the air, fed into the reactor tube was varied in the range 1.5 to 3.0 l/min at a pressure of $1.01 \times 10^5 \text{ Pa}$. The output gas from the ozonizer was exhausted to the outdoors through charcoal (activated carbon).

A magnetic pulse compressor (MPC) [15] provided the voltage and the current pulses with a repetition rate of up to 500 pps. A typical duration of 110 ns, defined as the full-width half maximum (FWHM) of the pulse voltage was measured at 35 kV output voltage. The circuit diagram of the MPC is shown in Figure 3. The MPC consisted of a high speed gate turn off (GTO) thyristor and a single stage pulse compression element. Following the charging of the capacitor C_1 , GTO is turned on. At the beginning, the current in the GTO is reduced by the saturable inductor (SL, SL1). After the saturation of SL1, the stored charge on C_1 is stepped up to C_2 through GTO, the step-up pulse transformer (PT) and the saturable transformer (ST). The ST compresses the current pulse and steps up the voltage. After the saturation of ST, the charge in C_2 is transferred to the peaking capacitor C_3 . Finally C_3 is charged to the desired voltage. The pulses obtained were directly applied to the coaxial electrodes.

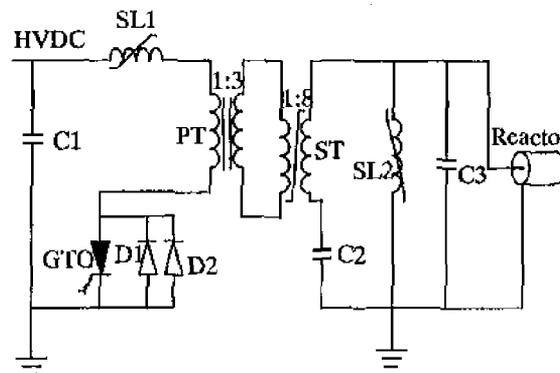


Figure 3. Circuit diagram of the MPC. Gate turn off thyristor H10 D33 YFH (Meidensha Co., Japan); C_1 , primary energy storage capacitor; SL1, saturable inductor; PT, step-up pulse transformer; ST, saturable transformer; C_2 , secondary capacitor; SL2, saturable inductor; C_3 , peaking capacitor.

3 RESULTS AND DISCUSSION

Throughout the present study, a positive polarity of the HV pulses was applied to the wire forming the central electrode. It has been shown

that using this polarity, the production of ozone was higher than that with the negative polarity in the wire-cylinder configuration [8]. It was shown also that in a positive wire-plane geometry in 1.01×10^5 Pa air, there were more streamer channels (8 channels/cm) compared to the negative wire (1 to 2 channels/cm) and the latter had a reduced branching compared to the positive polarity [12]. Further, using applied dc voltage, the positive wire had a higher breakdown voltage than the negative wire in the pressure region above the Paschen minimum [16, 17]. This was attributed to the higher total secondary ionization coefficient for the case when the wire was made a cathode, because the electric field was high in its vicinity. For positive wire the electric field is lower near the negative outer cylinder, resulting in a lower total secondary ionization coefficient and thus higher breakdown voltage [16, 17].

A diffused streamer channel leads to a lower electron density and this tends to postpone the development of the complete breakdown of the gap and therefore to a higher production of ozone via reactions (1) to (3). In general, when the pulse voltage was raised, the discharge became more stable.

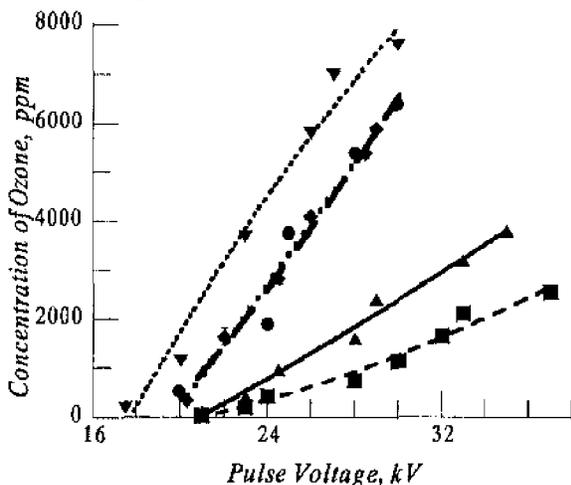


Figure 4. Dependence of ozone concentration on applied pulse voltage. Conditions: gas, dry air; pressure, 1.01×10^5 Pa; flow rate, 3.0 l/min; gap length, 16 mm; pulse repetition rates: ■ 25 pps; ▲ 50 pps; ◆ 100 pps; ● 200 pps; ▼ 400 pps.

3.1 CONCENTRATION OF OZONE

The production of ozone as a result of the application of pulsed power into dry air in parts of ozone per million parts of air molecules (ppm) is shown in Figures 4 and 5 as function of pulse voltage and pulse repetition rates, respectively. 1 ppm of ozone in 1.01×10^5 Pa of air at 293 K is equal to a density of 2.69×10^{19} molecules/m³ and to a mass density of 2.14 mg/m³. The ozone concentration increased with increasing pulse voltage at a fixed repetition rate, and increasing repetition rates at a fixed pulse voltage as shown in Figures 4 and 5, respectively. At a constant pulse voltage in the range 21 to 29 kV, the concentration of ozone increased linearly with increasing repetition rate, but only to ~ 100 pps (Figure 5). The concentration then increased less than linearly with further increase in the repetition rate (100 to 400 pps). The

transition from a streamer discharge into an arc reduces the production of ozone because the arc is narrow and not diffused at the high pressure employed here. All the measurements reported here were taken in the presence of streamers and before the discharge transferred into an arc. Figure 4 shows that it was not possible, for example, to apply a voltage >30 kV for 400 pps while it was possible to operate the reactor at $\lesssim 37$ kV for 25 pps. These observations are in general agreement with those reported by Chalmers *et al.* [5].

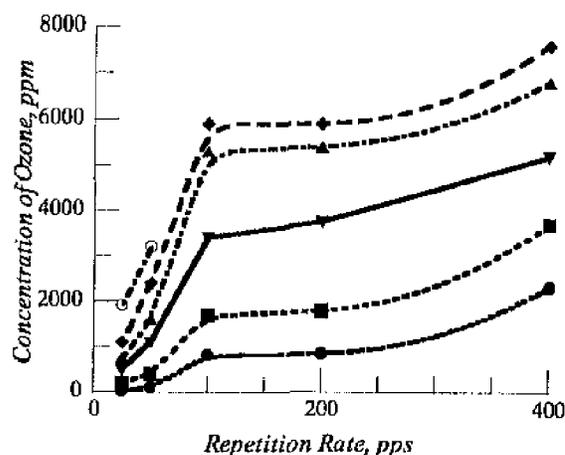


Figure 5. Dependence of ozone concentration on repetition rate at different pulse voltages. Conditions: gas, dry air; pressure, 1.01×10^5 Pa; flow rate, 3.0 l/min; gap length, 16 mm; pulse voltages: ● 21 kV; ■ 23 kV; ▼ 25 kV; ▲ 28 kV; ◆ 29 kV; ○ 33 kV.

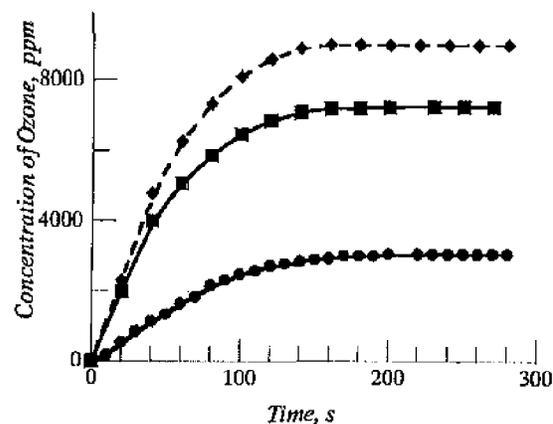


Figure 6. Dependence of ozone concentration on time of application of different pulse repetition rates. Conditions: gas, dry air; flow rate, 1.5 l/min; pressure, 1.01×10^5 Pa; pulsed voltage, 32 kV; gap length, 16 mm; length of the reactor, 1 m; pulse repetition rates: ● 50 pps; ◆ 200 pps; ◆ 400 pps.

Figure 6 shows typical times, needed for the ozone production to reach steady state values at 1.5 l/min flow rate in dry air. The observed longest saturation time of ~ 230 s (for 50 pps, Figure 6) was influenced by the volume of the discharge vessel, the gas flow rate, the inner diameter (5 mm) and the length (1.5 m) of the connecting polyethylene or Teflon™ tube which carried the gas to the UV monitor detecting the ozone.

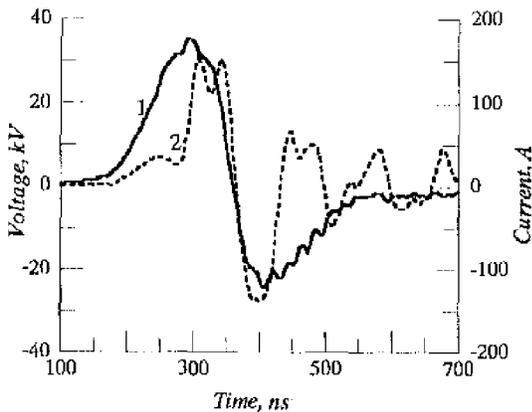


Figure 7. Typical pulse voltage (1) and discharge current (2) waveforms produced by a MPC. Conditions: flow rate of dry air, 3.0 l/min; pulse repetition rate, 50 pps; gap length, 16 mm.

For example at 3.0 l/min gas flow rate (not shown), the longest saturation time decreased to 90 s. In this work the concentration of ozone is reported after it had reached a steady state value.

Figure 7 shows typical pulse voltage and discharge current waveforms produced by the MPC of Figure 3 at 3.0 l/min flow rate of dry air, 50 pps repetition rate and a fixed gap length of 16 mm. It will be observed that the FWHM of the applied voltage is 110 ns, while that of the current is 60 ns. The overshoot in the voltage waveform may be attributed to the circuit stray inductances and the reflections due to the impedance mismatch between the power supply and the ozonizer. The dependence of the current i on time t was determined by the impedances of the reactor tube and the discharge in the gas. The impedance was largely capacitive before the onset of the discharge

$$i = C_1 \frac{dv}{dt} \quad (4)$$

where v is the applied voltage and C_1 is the capacitance of the reactor before the onset of the streamer discharge. During the discharge the impedance became complex

$$V = Ri + L \frac{di}{dt} + \frac{1}{C} \int i dt \quad (5)$$

where R is the resistance, L the inductance and C the capacitance of the reactor during the discharge.

Figure 8 shows the dependence of the concentration of ozone on the pulsed applied voltage for varying gap lengths in air at a pressure of 1.01×10^5 Pa. The gap length between the electrodes was varied from 10 to 20 mm. This was done in an attempt to vary the electric field in the gap, in order to influence the generation of ozone. It will be observed from Figure 8 that as the gap length decreases, the concentration of ozone increases for a fixed applied voltage. This is attributed to the higher electric field at the spiral wire with decreasing gap length. The higher electric field results in decreasing the average density of the low-energy electrons, which decompose the generated ozone [18]. Thus the higher electric field indirectly increases the production of atomic oxygen (reactions (1) and (2)) and this results in a larger generation of ozone according to reaction (3). Further, at the shorter gap length, the

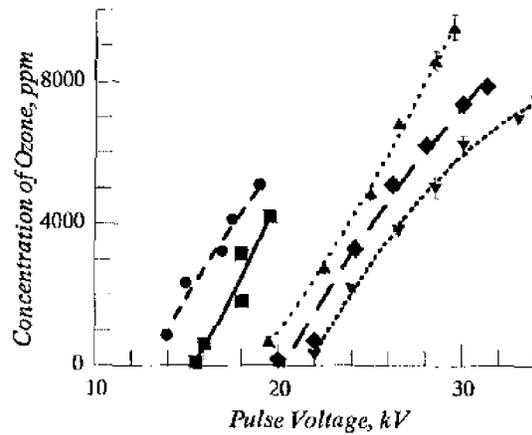


Figure 8. Production of ozone as a function of pulse voltage for different gap lengths. Conditions: gas, dry air; pressure, 1.01×10^5 Pa; repetition rate, 100 pps; gas flow rate, 1.5 l/min; gap lengths: ● 10 mm; ■ 13 mm; ▲ 16 mm; ◆ 18 mm; ▼ 20 mm.

production of ozone started at a lower pulse voltage (14 kV at 10 mm gap length, Figure 8).

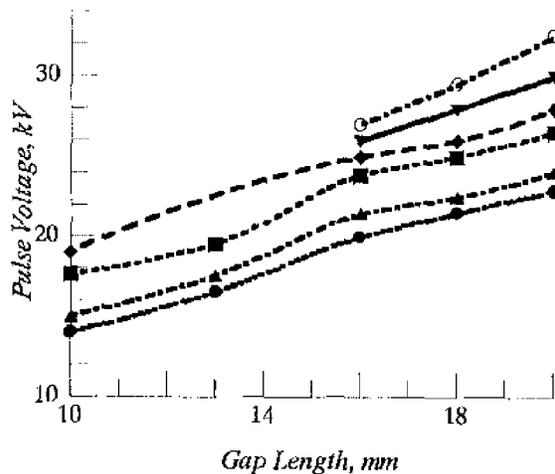


Figure 9. Dependence of the applied pulse voltage on the gap length to generate different concentrations of ozone. Conditions: gas, dry air; pressure, 1.01×10^5 Pa; repetition rate, 100 pps; gas flow rate, 1.5 l/min; concentrations of ozone: ● 1000 ppm; ▲ 2000 ppm; ■ 4000 ppm; ◆ 5000 ppm; ▼ 6000 ppm; ○ 7000 ppm.

Figure 9 shows the dependence of the applied pulse voltage on the gap length to generate various ozone concentrations in dry air. It will be observed that in order to attain a given concentration of ozone, the applied voltage must be increased with increasing gap length (Figure 9). Figure 10 shows the effect of varying the gas flow rate on the production of ozone. It will be realized that for a fixed condition of applied voltage and gap length, the slower flow rate of 1.5 l/min gave much larger concentrations of ozone than at 3.0 l/min (Figure 10). This occurred for all gaps in the range 13 to 20 mm and applied voltages in the range 15.5 to 34.5 kV. This was to be expected because a slower flow rate allowed an increased production (ppm) of ozone in constant con-

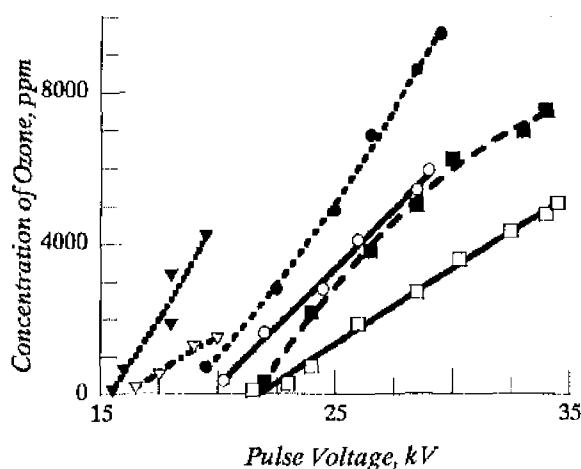


Figure 10. Concentration of ozone vs. pulse voltage showing the effect of varying the gas flow rate at different gap lengths. Conditions: gas, dry air; pressure, 1.01×10^5 Pa; repetition rate, 100 pps; gas flow rates and gap lengths: \blacktriangledown 1.5 l/min and 13 mm; ∇ 3.0 l/min and 13 mm; \bullet 1.5 l/min and 16 mm; \circ 3.0 l/min and 16 mm; \blacksquare 1.5 l/min and 20 mm; \square 3.0 l/min and 20 mm.

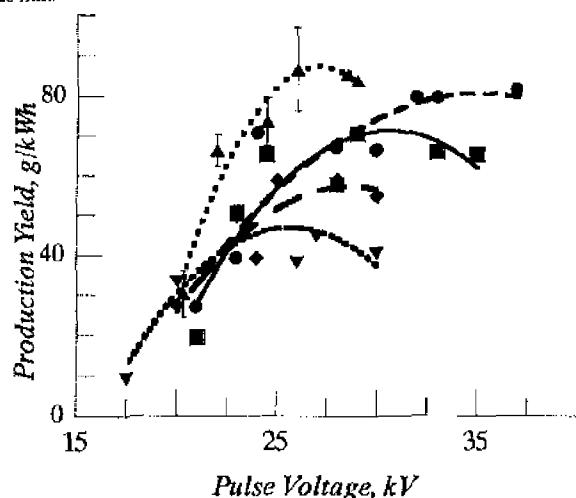


Figure 11. Dependence of ozone yield on pulse voltage for different pulse repetition rates. Conditions: gas, dry air; pressure, 1.01×10^5 Pa; flow rate, 3.0 l/min; gap length, 16 mm; pulse repetition rates: \bullet 25 pps; \blacksquare 50 pps; \blacktriangle 100 pps; \blacklozenge 200 pps; \blacktriangledown 400 pps.

ditions of gas pressure, electrode gap and applied pulse voltage, and is in general agreement with the previous work [5, 6, 19]. This is because of the longer residence time of gas molecules at lower flow rates for the effective formation of ozone in the reactor.

3.2 PRODUCTION YIELD OF OZONE

The ozone production yield in g/kWh of input energy to the gas is shown in Figure 11 as a function of pulse voltage at a fixed gap length of 16 mm, 3.0 l/min dry air flow rate and different repetition rates in the range 25 to 400 pps. It will be observed from Figure 11 that the production yield of ozone initially increased with increasing pulse voltage for all pulse repetition rates. The highest yield of 87 g (O_3)/kWh

(1.81 mol/kWh) at this flow rate and gap length was obtained at 100 pps and 26.5 kV pulse voltage. Figure 11 shows that at a certain voltage, depending on the pulse rate (≥ 50 pps), the yield started to decrease with further increase in the applied voltage. For example, at 50 pps the yield started to decrease above 30.5 kV while at 400 pps, it started to decrease above 25.5 kV (Figure 11). The reduction in the yield was also observed with increasing repetition rate (≥ 100 pps) at a fixed voltage (≥ 21 kV), see Figure 12.

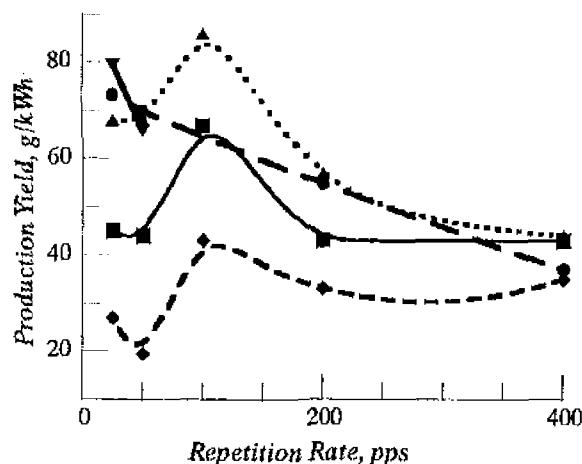


Figure 12. Dependence of ozone yield on the pulse repetition rate for different discharge voltages. Conditions: gas, dry air; pressure, 1.01×10^5 Pa; flow rate, 3.0 l/min; gap length, 16 mm; pulse voltages: \blacklozenge 21 kV; \blacksquare 23 kV; \blacktriangle 28 kV; \bullet 30 kV; \blacktriangledown 33 kV.

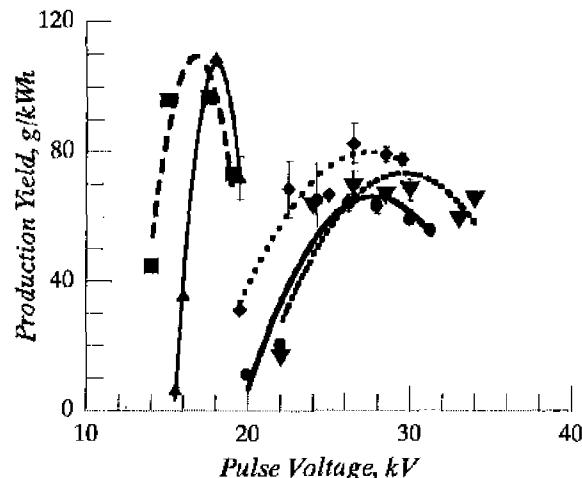


Figure 13. Dependence of ozone yield on the pulse voltage for different gap lengths. Conditions: gas, dry air; pressure, 1.01×10^5 Pa; flow rate, 1.5 l/min; repetition rate, 100 pps; gap lengths: \blacksquare 10 mm; \blacktriangle 13 mm; \blacklozenge 16 mm; \bullet 18 mm; \blacktriangledown 20 mm.

Figure 13 shows the ozone yield as a function of pulse voltage at a fixed repetition frequency of 100 pps and varying gap lengths in the range 10 to 20 mm. It will be observed from Figure 13 that the highest yield of 110 g/kWh was achieved at 10 mm gap length between the coaxial electrodes, 1.5 l/min flow rate and 16.9 kV pulse voltage. All yields peaked at a certain pulsed voltage and then decreased with

further increase in the pulse voltage. The bars shown in the experimental results represent the variations for two repetitions throughout this work.

3.3 DEPENDENCE OF MAXIMUM OZONE YIELD ON DISCHARGE PARAMETERS

The dependence of the applied pulse voltage at which the yield of ozone peaked on different gap lengths is shown in Figure 14. It will be observed from Figure 14 that this voltage increased with increasing gap length. Higher voltages, >29.6 kV, which correspond to a higher energy input to the discharge per pulse, are associated with the reduction of the production yield of ozone. Another possibility that might explain the reduction in the yield at higher voltages (Figure 13) was that the input energy was increased at a higher rate than the concentration of ozone as shown in Figure 8, due to the dissipation of heat.

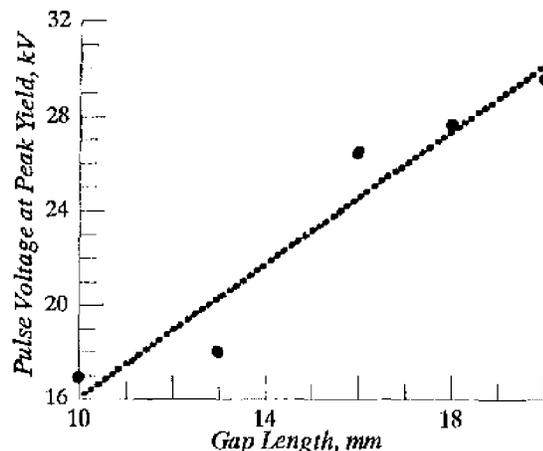


Figure 14. Dependence of the applied pulse voltage at which the ozone production yield peaks on the gap length. Conditions: gas, dry air; pressure, 1.01×10^5 Pa; flow rate, 1.5 l/min; repetition rate, 100 pps.

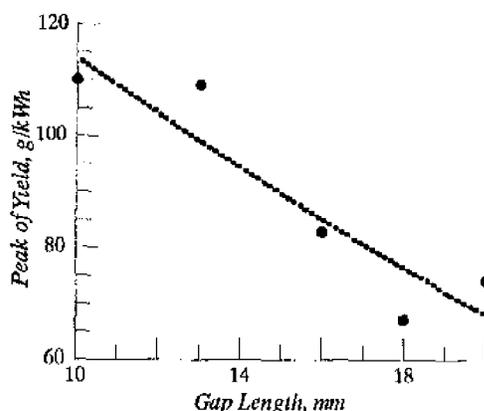


Figure 15. Peak yield vs. gap length for different pulse voltages (16.9 to 29.6 kV). Conditions: gas, dry air; pressure, 1.01×10^5 Pa; flow rate, 1.5 l/min; repetition rate, 100 pps.

Figure 15 shows the peak yield vs. gap length for different applied pulsed voltages (16.9 to 29.6 kV, Figure 14). Figure 15 shows that the

maximum yield that could be obtained from the ozonizer was strongly dependent on the gap length and decreased with increasing gap length of the coaxial geometry. This is attributed to the complex dependence of the development of the streamers, their size and diffusivity on the non-uniform electric field.

3.4 PRODUCTION YIELD FOR DIFFERENT OZONE CONCENTRATION

Figure 16 shows the production yield of ozone vs. the concentration of ozone at different gap lengths for a fixed flow rate of air of 1.5 l/min, pulse repetition rate of 100 pps and applied pulse voltage of 14 to 34 kV. It will be observed that the highest yield of 107 ± 4 g/kWh was obtained at short gaps of 10 and 13 mm for ozone concentrations of 3150 ± 150 ppm. According to Figure 16, the gap length of 16 mm at 100 pps repetition rate yielded the optimum condition of air-fed ozonizers by giving the largest amount of ozone concentration (9550 ppm) with a good yield value of ~ 77.9 g/kWh. In all cases the yield initially increased with increasing concentration until it reached a maximum and then declined with a further increase in the concentration of ozone. This is because, and as shown in Figure 8, the increase in the concentration with increasing voltage becomes less than linear at higher voltages. Therefore, the input energy is also increased at a higher rate than the concentration of ozone as mentioned above, leading to a lower yield at higher voltages and therefore at higher concentration.

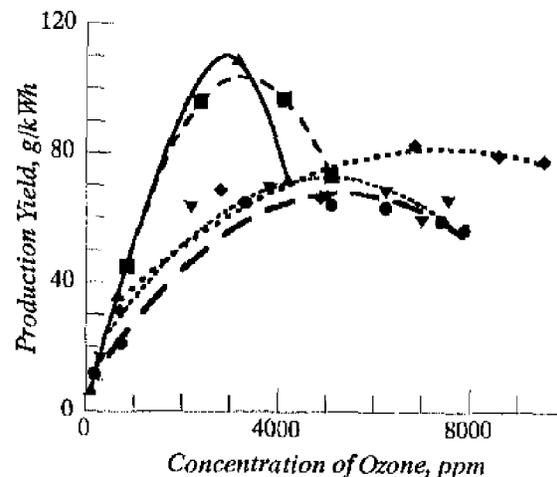


Figure 16. Production yield of ozone vs. its concentration at different gap lengths. Conditions: gas, dry air; pressure, 1.01×10^5 Pa; flow rate, 1.5 l/min; repetition rate, 100 pps; applied voltage, 14 to 34 kV; gap lengths: ■ 10 mm; ▲ 13 mm; ◆ 16 mm; ● 18 mm; ▼ 20 mm.

The required level of ozone concentration depends on its specific application. For example, the requirements for the concentration of ozone in drinking water and/or waste water treatments are different from the requirements of ozone use in the chemical and pharmaceutical industries, for bleaching kaolin and paper pulp [3]. Therefore, the results reported here are applicable to a wide utilization including deodorization (food hygiene, 6 ppm), purification of water (semiconductor industry, 1200 ppm), water treatment (nursery, 7000 ppm), air purification (air

cleaner, 40 ppm) and all applications which need a concentration of ozone in the range of $\lesssim 9550$ ppm [20].

4 CONCLUSIONS

1. Positive polarity of applied pulsed voltage to the central electrode was found to be effective for the production of ozone in dry air using a spiral wire in a coaxial geometry.

2. The pulse repetition rate of 100 pps, and 16 mm gap length produced the highest concentration of ozone with a considerable production yield.

3. The concentration of ozone increased with increasing applied pulsed voltage at a constant pulse rate, a fixed gap length and a constant gas flow rate.

4. The concentration of ozone increased with increasing pulse rate at a fixed pulsed voltage, a constant gas flow rate and a fixed gap length.

5. The production of ozone reached a saturated level within about 230 s for a gas flow rate of 1.5 l/min. At 3.0 l/min the saturation time decreased to 90 s for all experimental conditions specified in this study.

6. The production of ozone increased with decreasing gas flow rate.

7. The concentration of ozone increased with decreasing gap length at a fixed applied voltage, a fixed pulse rate and a constant gas flow rate.

8. The production yield of ozone was found to strongly depend on the concentration of ozone and therefore on the applied voltage, pulse rate and gas flow rate.

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