Ozone Production Using Pulsed Dielectric Barrier Discharge in Oxygen

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ABSTRACT

The production of ozone was investigated using a dielectric barrier discharge in oxygen, and employing short-duration pulsed power. The dependence of the ozone concentration (parts per million, ppm) and ozone production yield ($g(O_3)/kWh$) on the peak pulsed voltage (17.5 to 57.9 kV) and the pulse repetition rate (25 to 400 pulses/s, pps) was investigated. In the present study, the following parameters were kept constant: a pressure of 1.01×10^5 Pa, a temperature of 26 ± 4℃, a gas flow rate of 3.0 l/min and a gaseous gap length of 11 mm. A concentric coaxial cylindrical reactor was used. A spiral copper wire (1 mm in diameter) was wound on a polyvinylchloride (PVC) cylindrical configuration (26 mm in diameter) and placed centrally in a concentric coaxial electrode system with 4 mm thick PVC dielectric layer adjacent to a copper outer electrode of 58 mm in internal diameter. HV and current pulses were provided by a magnetic pulse compressor power source.

1 INTRODUCTION

THERE is wide interest in an energy efficient production of ozone for practical applications. These include the treatment of drinking and waste water; bleaching of kaolin and paper pulp, where ozone serves as strong oxidizing agent; in the semiconductor industry as oxidizer; and it is a potent germicide, viricide, bactericide and a sterilizer [1–3]. Ozone synthesis in oxygen and in air employing a dielectric barrier discharge has been studied extensively using ac or dc applied voltages [1, 4–8]. Studies of ozone production incorporating dielectric barriers include high frequency corona discharges using either glass [3, 9] or ceramic [10–12]. The production of ozone without a dielectric barrier has been reported with pre-ionization [13], using point-plane [14, 15] and wire-to-plate [16, 17] electrodes configurations. In the wire-to-plate configuration both positive and negative pulse streamers were used [16, 17]. However, there are relatively few studies of ozone synthesis with dielectric barriers using very short duration HV pulsed discharges [18].

Streamers or microdischarges are created in atmospheric pressure of oxygen when a sufficiently high voltage is applied to the central electrode of a coaxial concentric cylindrical electrode geometry with a dielectric sheet placed on the inner surface of the outer electrode. The dielectric sheet forms a barrier, which impedes the development of the arc discharge and thus a complete breakdown of the gap is also impeded. A short pulsed streamer discharge has been shown to be very effective in producing an intense corona but not long enough to initiate an arc breakdown [19, 20]. The high-energy electrons in the streamers dissociate the oxygen molecules into atoms and these collide with an oxygen molecule and a third particle to produce ozone [1]

$$e + O_2 \rightarrow O + O + e \quad (1)$$

The reaction rates at E/N=40, 70, 100, $150T_d$ (10^{-17} Vcm^2) were reported to be k_1=1.1×10^{-12}, 7.4×10^{-11}, 3.1×10^{-10} and 7.9×10^{-10} cm^3/s, respectively [21]. Ozone is then produced by a three-body reaction with oxygen

$$O + O_2 + O_2 \rightarrow O_3 + O_2 \quad (2)$$

with reaction rate 6.9×10^{-34} cm^6/s at 300 K [22]. The latter reaction rate is in reasonable agreement with that of 6.3×10^{-34} cm^6/s reported in [23].

Unlike the production of ozone using air, its synthesis in oxygen mitigates against the production of NO and NO_2, which are not desirable. The natural decay products of ozone are oxygen and CO_2 in the presence of organic substances [3].

The present paper reports on ozone synthesis employing a short duration (120 ns) of pulsed power using positive voltages in a dielectric barrier discharge in oxygen. The effects on the production of ozone of the peak pulsed voltage (17.5 to 57.9 kV) and pulse repetition rate (25 to 400 pulses per second, pps) were studied. In the present work the following parameters were kept constant: the gas flow rate (3.0 l/min), the gaseous gap separation (11 mm), the gas pressure (1.01×10^5 Pa) and the temperature (26±4℃).

2 EXPERIMENTAL PROCEDURE

Figure 1 depicts the experimental setup for the generation of ozone. Oxygen was obtained from a gas cylinder having a purity of 99.5% (Kumamoto Sanso, Japan). The gas flow rate in the discharge reactor

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was set at 3.0 l/min and was monitored using a flow-stat meter (Floline, model SEF-1 R, made by Stec Inc, Japan). The ozone concentration in parts of ozone per million parts of oxygen molecules (ppm) was determined using an ultraviolet absorption meter (Ebara model DOA 300). The gas was exhausted to the atmosphere via an activated carbon (charcoal) trap, which largely absorbed the ozone. The coaxial cylindrical configuration shown schematically in Figure 2 was driven by very short pulses of HV to create short-lived streamer discharges. The central electrode was in the form of a copper wire 1 mm in diameter, which was coiled on a PVC tube having an outer diameter of 26 mm. The outer electrode was a copper foil 58 mm in internal diameter and 0.1 mm thick. The copper foil was wrapped on the outside of a PVC tube, 4 mm thick. This configuration formed a gaseous gap of 11 mm between the central helical wire electrode and the inner surface of the PVC dielectric barrier.

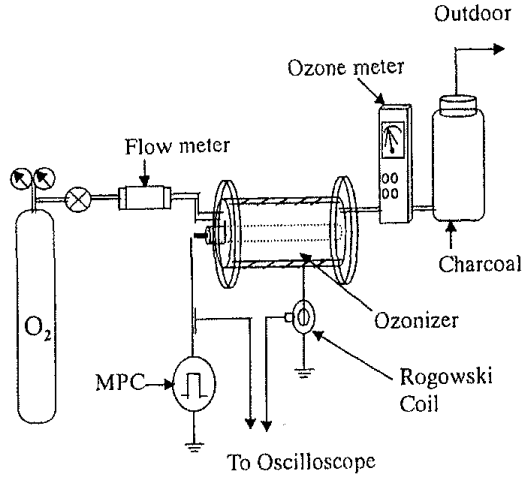

Figure 1. Experimental setup for generation of ozone MPC.

HV pulses of positive polarity were provided by a magnetic pulse compressor (MPC) with a repetitive rate capability of $\leqslant$ 500 pulses per second (pps) and $\leqslant$ 60 kV [24]. The HV pulses were applied to the central electrode. The electrical circuit diagram of the MPC and a brief description of its operation were reported recently [24]. A typical duration of 120 ns was measured at a peak pulsed voltage of 30.5 kV (Figure 3). The pulse length is defined as the full-width half maximum (FWHM) of the pulse voltage. In the present work, the production of ozone reached a saturation level during 3 to 5 min after the application of pulsed voltage. Therefore, artificial cooling of the discharge reactor was not employed as it was not necessary.

The gas pressure was 1.01×10^5 Pa and the temperature was at 26±4°C. The waveforms of the applied pulse voltage $V(t)$ and the discharge current $I(t)$ were obtained via a HV capacitive divider and a Pearson Rogowski coil, and were displayed using a Hewlett Packard digital oscilloscope (HP 54542 A) (Figure 1). The latter had a maximum bandwidth of 500 MHz. The energy ($\int VIdt$) input to the discharge per pulse was determined from the digitized signals of the voltage (V in V), current (I in A) and time (t in s). The production yield of ozone

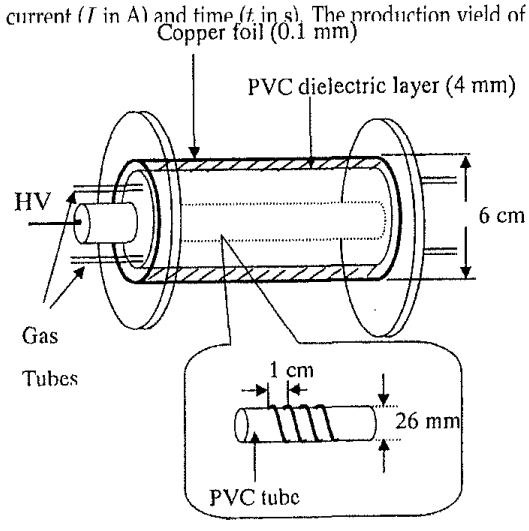

Figure 2. Discharge reactor configuration with a dielectric barrier. Wire diameter, 1 mm; reactor length, 1 m.

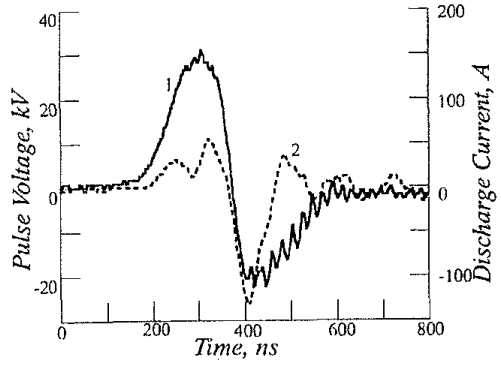

Figure 3. Typical pulse voltage (1) and discharge current (2) waveforms produced by a MPC in a dielectric barrier discharge. Conditions: Oxygen flow rate, 3.0 l/min; peak pulsed voltage, 30.5 kV; pulse repetition rate, 100 pps; gap length, 11 mm.

in mol/kWh or (g/kWh) was determined (in mol/kWh) from

$$\eta = 3.0\frac{l}{60}\times N(O_3)\times\frac{3.6}{22.4}\times f\times E \qquad (3)$$

where $N(O_3)$ is the concentration of ozone in ppm, f and E are the pulse repetition rate (pulses/s) and the input energy (J) to the reactor per pulse (J/pulse), respectively. It should be noted that 0.048 kg of O_3 is equivalent to 1 mol and 0.0224 m^3 at 1.01×10^5 Pa and 273 K. Equation (3) can also give the yield in g/kWh by multiplying η (mol/kWh) by 48.

In the present work only the energy dissipated in the discharge is used, which excludes the energy dissipated by the power supply. This is because pulsed power sources in the nanosecond range are currently undergoing intense development to produce high efficiency power sources. The present MPC power source has an efficiency of $\sim$ 60%.

3 RESULTS AND DISCUSSION

3.1 CONCENTRATION OF OZONE

A positive pulse voltage was used because it was shown that in air the streamers extended much further in the radial direction and the production of ozone was higher in the wire-cylinder geometry than for negative polarity [18, 25]. In a wire-plane geometry in air, there were more streamer channels for the positive compared to the negative wire, and the former had more branching [16]. Furthermore, the positive streamer discharges in air generated slightly more than twice the total ozone density that was generated using negative streamer discharges [16]. Masuda *et al.* [18] also reported that in air-filled wire-cylinder geometry, short pulsed positive streamers extended much further in the radial direction than negative streamers. The negative streamers were restricted to the region close to the wire and the effective volume of ozone production was thus smaller than for the positive streamer [18]. A more spatially extended and branched streamer discharge leads to a higher production of ozone.

Figure 3 shows a typical pulsed voltage and the discharge current waveforms produced by the MPC in a dielectric barrier discharge at 3.0 l/min flow rate of oxygen, 100 pps repetition rate, and a fixed gaseous gap length of 11 mm. The gas flow in the reactor tube was axial. The initial current ($i = CdV/dt$) was due to the charging of the capacitance (C in F) between the electrodes but this was small, because this capacitance was also small ($\sim$ 20 pF) and subsequently the current was determined by the conductivity of the streamer discharge. The oscillations present in the peak of the voltage were due to oscillations with the stray inductance of the circuit because a capacitive voltage divider was employed.

When a resistive divider was used, the oscillations near the peak voltage were eliminated completely. The oscillations at the tail of the wave were due to multiple reflections arising from impedance mismatch between the MPC source and the discharge reactor. When a resistive divider was employed, the oscillations at the tail of the pulsed voltage continued to be present as the high impedance divider had only a small influence on the impedance mismatch, which was difficult to eliminate in the present setup. All voltages shown in this work were determined at the peak of the wave. The largest amplitude of the oscillations in the region of the peak of the pulsed voltage was 3.7% of the peak voltage.

The development of the discharge current is influenced by the presence of the dielectric layer. The dielectric layer restricts the amount of charge transported by a single streamer and distributes the streamers over a wide area in the vicinity of the outer electrode [3]. In a dielectric barrier discharge, the fast electrons transfer their energy by collision to the surrounding atoms and molecules at the high pressure used. The formation of a spark across the gaseous gap is hampered by the presence of the dielectric layer due to the reduction of the field in the solid dielectric and the limitation on the current flow [10]. Thus the streamers that develop in a dielectric-barrier discharge self extinguish when sufficient charge builds up on the dielectric layer, which reduces the local electric field below its sustainable level. The production of ozone largely occurs in the vicinity of the central wire electrode [16, 17] where the electric field is high.

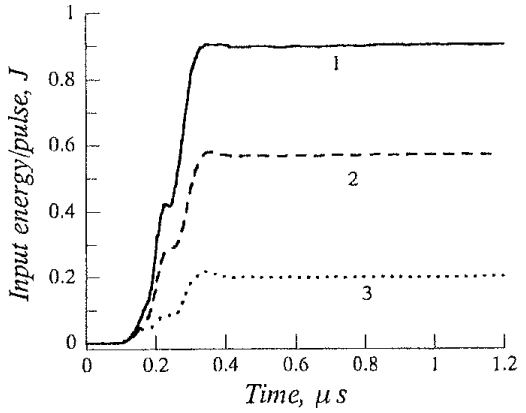

Figure 4. Typical input energy plots to the discharge as a function of the pulse total time. Peak voltages: curve 1, 57.9 kV; curve 2, 41 kV; curve 3, 32 kV; pulse repetition rate, 25 pps. Other conditions as in Figure 3.

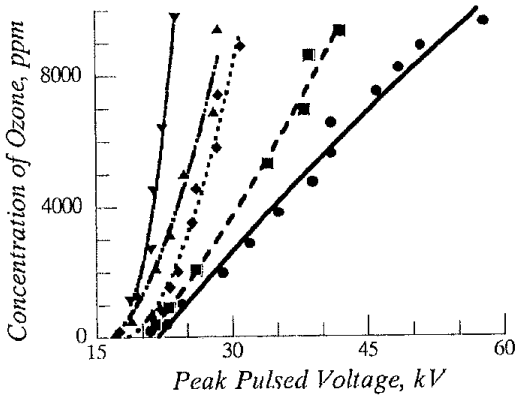

Figure 5. Dependence of ozone concentration on peak pulsed voltage. Pulse repetition rates: ● 25 pps; ■ 50 pps; ♦ 100 pps; ▲ 200 pps; ▼ 400 pps. Other conditions as in Figure 3.

The dependence of the input energy to the discharge on time for different pulsed voltage peaks is shown in Figure 4. It will be observed that the energy input to the discharge per pulse reaches a steady level in all cases. The energy pulse increases with increasing peak voltage (Figure 4). Figure 5 shows the production of ozone in ppm as a function of the pulse voltage for different pulse rates in the range 25 to 400 pps. The concentration of ozone at each applied peak voltage is reported here after it has reached a steady state value. In the application of very short duration of the HV pulses, the ions and neutrals are not heated significantly above the ambient [19]. This was confirmed using spectroscopic measurements, which showed that for streamer pulses lasting <100 ns, the gas temperature in atmospheric nitrogen remained in the range 300 to 350 K [26]. It will be observed from Figure 5 that the concentration of ozone increased with increasing applied pulse voltage at a fixed pulse rate. The rate of rise of the concentration of ozone with increasing voltage increased with increasing the pulse repetition rate. In the present work the ozone monitor was restricted to 10000 ppm. Therefore, using a given pulse rate, the maximum voltage that was used was limited by the ozone monitor and not by the breakdown voltage of the gaseous

gap, as was in the case in dry air using a discharge reactor without a dielectric barrier [24]. Figure 5 shows that to get increased amounts of ozone, it was necessary to apply higher pulse voltages with a lower pulse rate or alternatively to apply lower pulse voltages with a higher pulse rate. For example, a concentration of ozone of ~ 9800 ppm could be achieved using 23.9 kV peak voltage with 400 pps and ~ 9600 ppm could be obtained using 57.9 kV with 25 pps (Figure 5). This is due to the increased energy input to the streamer discharge, which can be attained either with increasing voltage or increasing pulse repetition rate.

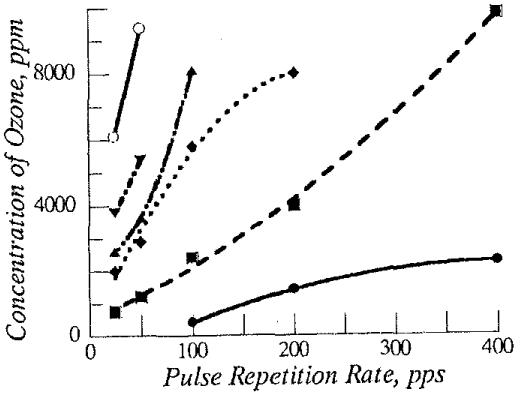

Figure 6. Dependence of ozone concentration on pulse repetition rate. Peak pulsed voltages: ● 20 kV; ■ 24 kV; ♦ 28 kV; ▲ 30 kV; ▼ 34 kV; ○ 42 kV. Other conditions as in Figure 3.

Figure 6 shows the dependence of the concentration of ozone on the pulse repetition rate using different pulsed voltages. At a fixed applied voltage, the concentration of ozone increases with increasing pulse rate due to the higher energy injected into the discharge per unit time at a constant gas flow rate. The increase in the concentration of ozone is larger at higher voltages with increasing pulse rate due to the higher injected energy than at lower voltages.

3.2 PRODUCTION YIELD OF OZONE

The variation of the production yield of ozone with peak pulsed voltage at different repetition rates and a constant pressure of 1.01×10^5 Pa and flow rate of 3 l/min is shown in Figure 7. The influence of the pulse repetition rate was not strong. Figure 7 shows that the present ozonizer gave a maximum production yield of 202 g/kWh (4.21 mol/kWh). The production yield increased with increasing voltage to ~ 39 kV, and then reached a saturated level (Figure 7). The reason for this dependence can be gleaned from Figure 8, where it can be seen that at above ~ 39 kV the input energy to the discharge per pulse increases with increasing peak pulsed voltage at a faster rate than linearly while the concentration of ozone increased linearly with increasing voltage (Figure 5). At peak voltages below 39 kV the input energy per pulse increased linearly with increasing voltage (Figure 8) while the concentration of ozone increased faster than linearly with increasing voltage (Figure 5).

The reason for the relatively steep increase in the energy input to the discharge at higher voltages (>39 kV) was because the discharge

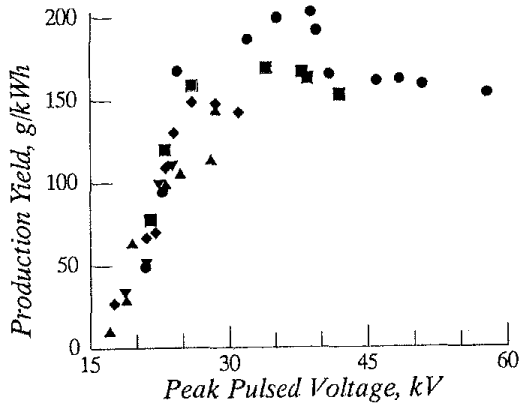

Figure 7. Dependence of ozone yield on peak pulsed voltage. Pulse repetition rates: ▲ 25 pps; ■ 50 pps; ♦ 100 pps; ● 200 pps; ▼ 400 pps. Other conditions as in Figure 3.

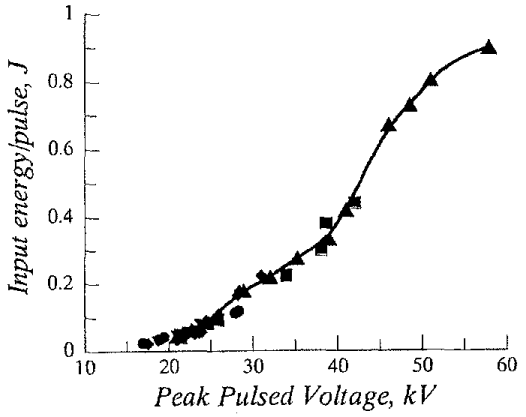

Figure 8. Dependence of the input energy to the discharge per pulse on the applied peak pulsed voltage. Symbols and conditions as in Figure 7.

current (not shown) increased much higher than linearly with increasing voltage. Typically at 25 pps, the peak discharge current at a peak pulsed voltage of 21 kV was 20 A while at 57.9 kV (an increase of a factor of 2.8), the peak discharge current became 201 A (an increase of a factor of 10.1). This was due to the nonlinear increase in the conductivity ($\sigma = nek_e$, in S/m) of the discharge at HV (>39 kV) arising from the simultaneous increase in electron density (n, in m^{-3}) with higher ionization, as well as higher electron mobilities (k_e, in m^2/sV) at the higher electric fields.

The cross sections for the dissociation of O_2 have two peaks near 10 and 20 eV [27] and therefore in order to obtain an optimum production yield it is necessary to optimize the formation of the radical O (reaction 1), which is a prerequisite for the production of ozone (reaction 2). The production of O depends strongly on the energy of the electrons and therefore, on the peak of the pulsed voltage.

Figure 9 shows the dependence of the production yield on the pulse repetition rate at different applied peak voltages. At a fixed peak voltage the yield decreased with increasing pulse repetition rate. This is because the energy input to the discharge increased at a faster rate

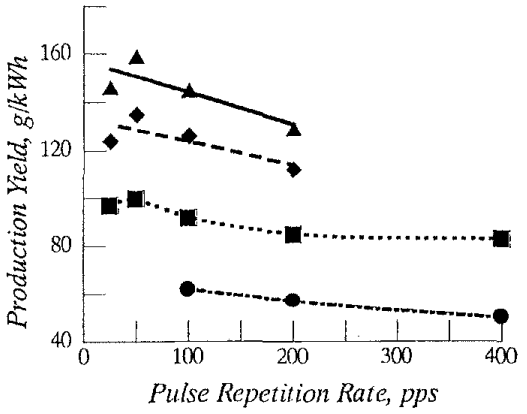

Figure 9. Dependence of ozone yield on pulse repetition rate. Peak pulsed voltages: ● 20 kV; ■ 22 kV; ♦ 25 kV; ▲ 28 kV; Other conditions as in Figure 3.

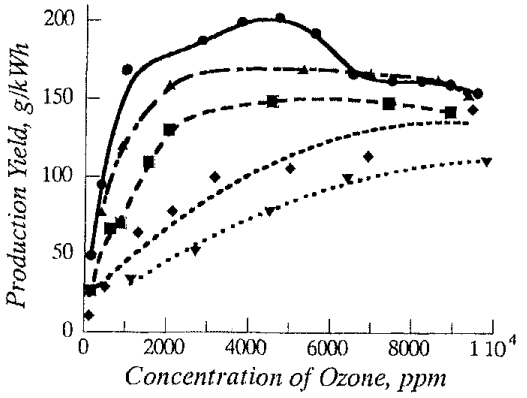

Figure 10. Production yield of ozone *vs.* its concentration. Pulse repetition rates: ● 25 pps; ▲ 50 pps; ■ 100 pps; ♦ 200 pps; ▼ 400 pps; Peak pulsed voltages, 17.5 to 57.9 kV; Other conditions as in Figure 3.

with increasing pulse rate (figure not shown) than the concentration of ozone (Figure 6). The excess energy resulted in a slight heating of the electrodes and the flowing gas.

3.3 PRODUCTION YIELD FOR DIFFERENT OZONE CONCENTRATION

Figure 10 shows the production yield of ozone as a function of the concentration of ozone at different pulse rates for a fixed flow rate of oxygen of 3.0 l/min and various applied peak pulsed voltages from 17.5 to 57.9 kV. It will be observed that the highest yield of 202 g/kWh was achieved at the low pulse rate of 25 pps with 4700 ppm concentration of ozone. Figure 10 shows that for 25 pps, the maxima values of both the yield and the concentration of ozone cannot be obtained simultaneously. Therefore, a choice must be made as to which one of these quantities should be maximized. According to Figure 10, the pulse rate of 25 pps gave the best yield of the oxygen-fed ozonizer using a dielectric barrier in the present coaxial cylindrical geometry. For pulse rates $\geqslant$100 pps, the yield initially increased with increasing the concentration of ozone until it reached a maximum and then slightly declined (Figure 10). This reduction in the yield was due to the faster growth of the energy input to the discharge than that of the production of ozone. For 200 and 400 pps the yield increased steadily to the limit of 10^4 ppm of the ozone monitor used in this work. The yield of ozone in the present work compares favorably with the recently reported literature results. These are briefly reviewed here for comparison purposes. It was stated that the highest reported yield of ozone generation in laboratory experiments with dielectric barrier discharges had been 250 g/kWh in pure oxygen and 100 g/kWh in dry air [28]. In commercial systems the yield in oxygen was 66.7 to 76.9 g/kWh (13 to 15 kWh/kg) and in air 50 to 55.6 g/kWh (18 to 20 kWh/kg) [28].

The theoretical yield of ozone can readily be calculated. The dissociation energy of O_2 (reaction 1) is 5.084 eV [29] and therefore, 2.542 eV is expended to produce a radical O. Since reaction (2) is exothermic it results in an excess energy of 1.08 eV and thus the required energy to generate O_3 is 1.462 eV. This leads to 140.93 kJ/mol (O_3) and 1226 g (O_3)/kWh. Masuda *et al.* [18] reported in dry air varying yield values from 100.6 to 106 g/kWh using cylindrical ozonizers without a dielectric barrier and 77.4 to 200 g/kWh using strip plates with dielectric barriers. Using a surface discharge in oxygen at 10 kHz Masuda *et al.* best reported result in pure oxygen was a yield of 170 g/kWh [10] with a high level of ozone production of 5 to 10%. Pietsch *et al.* reported yield values of 122 g/kWh in O_2 and 28 g/kWh in air using dielectric barrier discharges [30]. Chalmers *et al.* reported in oxygen strongly dependent values of the yield on the pulse width from 300 g/kWh at 20 ns to 47 g/kWh at 120 ns [31, 32].

4 CONCLUSIONS

1. Positive pulsed corona in a dielectric barrier discharge has shown promising results that can be implemented readily in industrial applications using coaxial cylindrical ozonizers.
2. The concentration of ozone increased with increasing peak pulsed voltage at a fixed gap length and a constant gas flow rate.
3. The rate of increase of the ozone concentration with increasing peak pulsed voltage increased with increasing pulse rate at a fixed gap length and a constant gas flow rate.
4. A pulse repetition rate of 25 pps using a fixed gap length of 11 mm and a constant flow rate of oxygen of 3.0 l/min produced the highest production yield of ozone (202 g/kWh).
5. The g/kWh yield of ozone was found to strongly depend on the concentration of ozone and therefore on the applied peak pulsed voltage and the pulse repetition rate.

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