



Ozone Generation by Pulsed Streamer Discharge in Air

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Abstract

Ozone is well established as an important oxidising agent in many industrial applications and the high-voltage pulsed streamer discharge technique has emerged as an efficient method for its production. The paper investigates the parameters that are important in this process, including the magnitude of the applied pulse voltage, the air flow rate and the electrode dimensions and spacing, when used in a plate-to-plate reactor configuration in which sheets of perforated aluminium are employed to enhance the degree of ionisation and soda lime glass dielectric barriers both to inhibit singular discharges and to promote the development of the streamer discharge. The highest ozone concentration achieved was 5995 ppmv and the greatest yield was 144 g/kWh.

Keywords: Ozone generation, Pulsed power, Pulsed streamer discharge

1. Introduction

Arising from the increased concern with environmental protection, ozone attracts widespread attention as an important oxidising agent, finding use in a wide range of industrial applications. With three oxygen atoms bonded together, it is a powerful but unstable oxidant, although having a half life of only between about 7 and 20 minutes. Nevertheless, when compared with alternatives such as the chlorination process (Samaranayake W., 2000; Buntat Z, 2003; Chalmers I D, 1994; Namihira T, 2001), it offers a number of significant advantages, in particular its lower energy consumption (Samaranayake W., 2000; Buntat Z, 2003; Chalmers I D, 1994; Namihira T, 2001; Buntat Z, 2005; Chalmers I D, 1996; Samaranayake, 2001). As a consequence of its attractive properties, many methods of enhancing the production rate of ozone have been investigated.

Ozone is produced during an electrical discharge at room temperature and pressure, with air assisting in the two main processes of dissociation and formation (Samaranayake, 2000). Initially, an oxygen atom is dissociated in collisions between high energy electrons and oxygen molecules (equations (1) and (2) below), before ozone itself is formed in a three-body reaction process (equation (3)). M, the third particle involved in the collision process, could in dry air be either O₂, O₃ or N₂ (Samaranayake, 2000; Simek, 2002).



Acceleration of excited electrons requires a high-voltage energy supply, which may however cause a spark breakdown to occur in a discharge chamber. Use of a pulsed streamer discharge [PSD] technique can alleviate this problem, since

the energy in the short duration fast rising pulse required to accelerate the electrons will be insufficient to lead to a breakdown (Chalmers, 1994; Chalmers, 1996; Samaranyake, 2001; Simek, 2002; Samaranyake, p.849-854). As a consequence, an intense streamer discharge is produced by the very effective ozone generation process. Dielectric material may be added inside the chamber to distribute evenly the micro-discharges over the dielectric layer area (Buntat, 2003; Samaranyake, 2001; Samaranyake, p.849-854) and to prevent the propagation of a single discharge.

High electric field strength in the vicinity of the electrodes is important in ionizing the gas molecules and in producing both more energetic electrons and more streamers. The ozone formation that occurs in the streamers is increased if they are more intense and wider. In the experiments described in this paper a perforated aluminium sheet with an array of sharp edged holes was located next to the aluminium foil electrodes, to produce an increased electric field strength in the vicinity of the holes and so lead to a greater ozone production (Buntat, 2005; Buntat, 2007).

The pulsed voltage applied throughout the present investigation was of positive rather than negative polarity, since it is known (Samaranyake, 2000; Samaranyake, p.849-854) that the streamers then produced have more branches and more channels per unit length, and are likely to lead to a greater dissociation of oxygen molecules. The pulse duration of 120 ns used in the experiments was sufficiently short not to accelerate significantly any ions, and as a result the chamber temperature remained at the ambient level (Tamaribuchi, 2007). This avoids any need for a cooling system and the ozone deformation rate is reduced. In addition, operation at a frequency of 50 pps promotes new streamer growth within every new pulse (Shuhai, 2000).

The paper studies maximisation of the oxygen yield in a PSD experiment by maximising the various parameters that are involved, including the applied pulse voltage, the air flow rate, the gaseous gap spacing between the dielectric barrier and the length of the electrode chamber.

2. Experimental arrangement

Figure 1 shows the constructional details of the discharge reactor that was purpose built for the PSD experimentation. A plane-to-plane electrode structure was used, with the commercial zero grade dry air flowing axially along the reactor having a composition of nitrogen 78.08%, oxygen 20.9%, carbon dioxide 0.03% and argon 0.93%, plus traces of neon, helium, methane, krypton, xenon, hydrogen and nitrous oxide. The flow rate of the gas was monitored before its entry into the reaction chamber, and was controlled to between 0.2 and 1 l/m at a constant pressure of 1 bar.

The gap spacing between the two dielectric barriers influences the electric field strength and the radius of the micro-discharge, and the electrode dimensions influence the gas ionisation and residence time. The variation of the gap spacing was therefore investigated between 1.5 mm and 3 mm and the electrode length between 100 mm and 220 mm, but at a constant width of 16 mm, since any variation in the electrode width has a similar effect to one in the length. The overall discharge volume thus ranged from $2.40 \times 10^{-6} \text{ m}^3$ to $10.56 \times 10^{-6} \text{ m}^3$, with the effective area being between $1.60 \times 10^{-3} \text{ m}^2$ and $3.52 \times 10^{-2} \text{ m}^2$.

Figure 2 shows diagrammatically the perforated aluminium sheet that was placed in the discharge chamber between the aluminium foil electrodes and the dielectric barrier, causing high-energy electrons to be produced by the enhanced electric field in their vicinity. The sharp edged circular holes were 1.2 mm in diameter, which resulted in the removal of about 23% of the area of the sheet. Both dielectric barriers were a 1mm sheet of soda lime glass having a relative permittivity of 7.75, with the high pulse voltage supply obtained from a cable generator and supplied at the centre of the electrodes, as shown in Figure 1.

Figure 3 shows the overall experimental assembly. The ozone monitor recorded the ozone concentration on the basis of the ultraviolet absorption (254 nm) properties. After measurement, the ozone was destroyed in a catalytic ozone destroyer before being exhausted to air. The high-voltage positive-polarity pulses for the reactor were provided by the combination shown in Figure 4 of a double Blumlein pulse generator using three coaxial cables together with a rotary spark gap switch, and could be varied between 1 and 20 kV at a fixed repetition rate of 50 pps. The pulse rise time was less than 50 ns and its duration was 120 ns. A high-voltage probe (Tektronix P6015A, 1000x) with a 0.22 μF capacitor connected in series to ground was used to measure the voltage supplied and a Rogowski coil with a Pearson current monitor (model 2877, 1:1 V/A) to measure the current. Both signals were displayed on a digital storage oscilloscope (LeCroy 9344) with a bandwidth of 500 MHz and a sample rate of 1 GS/s. The power and the energy input to the discharge per pulse were computed from the two digitised signals.

3. Experimental results and analysis

During the PSD experimentation all measurements were taken before the electrical discharge turned into the arc stage, after which ozone formation ceases. The discharge process was left to run continuously for at least five minutes, by which time the concentration level in the ozone analyser had always reached a steady state. The power supply was then removed until the ozone analyser indicated a zero level, when the power supply was restored and the cycle repeated.

Figure 5 shows waveforms of a typical positive polarity voltage pulse of about 14 kV peak and a current of about 8 A peak fed to the reaction chamber. The small oscillations evident in the current are attributed to both stray inductance and reflected waves due to a slight impedance mismatch between the power supply and the reactor.

In ozone generation either the ozone concentration in parts per million or the ozone yield in g/kWh is used as a figure of merit in assessing the efficiency of the process. Both are considered in the assessment below.

3.1 Effect of peak pulsed voltage on ozone concentration at different air flow rates

Figure 6 shows the variation of the ozone concentration as the peak pulsed voltage supplied to the maximum 3 mm long gap is increased from 13 kV, the lowest level at which ozone was produced, to 18 kV, beyond the normal breakdown voltage of the gap. This higher figure confirms the advantage of the higher voltage possible with the PSD approach in increasing the energy given to the electrons before a breakdown results. As the voltage is increased the higher electrical energy density in the discharge leads to more energy being transferred to the electrons, thereby increasing the possibility of collisions with the air in the chamber and giving an approximately linear increase in the ozone concentration. The effect of an increased flow rate on the ozone concentration is also evident in Figure 6, with the increased residence time producing a very much greater concentration at the lower flow rates.

3.2 Effect of air flow rate on ozone concentration at different peak pulsed voltages

The effect of the air flow rate on the ozone concentration is evident in Figure 7, with the concentration increasing with the voltage but decreasing with the flow rate. The residence time of the gas in the reaction chamber is obviously inversely related to the air flow rate, with an increased residence time providing more time for a reaction to occur and a correspondingly higher ozone concentration to be produced.

3.3 Effect of gaseous gap spacing on ozone concentration as a function of peak pulsed voltages

Figure 8 shows that the effect of reducing the gaseous gap length from 3 mm to 1.5 mm is clearly for the generation of ozone to begin at an appreciably lower voltage and to have produced a significantly higher concentration at any given flow rate.

This is clearly related to the increased energy density as the gap is decreased, together with the increased number of oxygen molecule/electron collisions and the correspondingly increased combination of oxygen atoms with oxygen molecules. However, for the 1.5 mm gap length the ozone concentration increases at a progressively slower rate than with the 3 mm gap length, particularly as the voltage approaches the maximum 16 kV that the shorter gap can sustain.

3.4 Effect of electrode chamber length on ozone concentration as a function of peak pulsed voltages

Figure 9 shows that the electrode chamber length has little effect on the ozone concentration at low peak pulsed voltages. In contrast, it has a marked effect at higher voltages above about 14 kV, with the increased residence time that accompanies the 60 mm increase in the chamber length being responsible for a 40% increase in the concentration at 18 kV. However, the longer the ozone spends in the air-filled chamber the greater is the rate at which it becomes deformed. Electrons and various undesirable products such as the nitrogen oxide compounds (NO_x) are also produced in the complex reaction processes, with these being the primary cause of the deformation.

3.5 Effect of peak pulsed voltage on ozone yield at different air flow rates

The ozone yield of a process clearly depends on both the concentration and the input electric energy density, with the efficiency η in g/kWh commonly being calculated from

$$\eta = \left[\frac{C(O_3) f_r}{P} \right] \quad (4)$$

where $C(O_3)$ is the ozone concentration (ppmv), f_r is the gas flow rate (l/min) and P is the discharge power (kW). Figure 10 shows that initially the yield increased for all flow rates, reaching a peak at about 15 kV before it decreased as the rate of ozone decomposition became higher than the rate of ozone formation at that particular energy input per cycle. Eventually, due to the greater and rapidly increasing energy input per cycle evident in Figure 11, the yield rises again, as shown in Figure 12 at voltages only the longer gap can withstand.

3.6 Effect of peak pulsed voltage on ozone yield at different gap spacing

Figure 12 shows that a maximum ozone yield is achieved, but that this is at the breakdown voltage for the smaller gap. For the larger gap the rapid increase in the energy input per pulse above about 15 kV eventually more than compensates for the increased decomposition rate, with the yield again increasing after having fallen to a minimum at about 17 kV.

3.7 Effect of peak pulsed voltage on ozone yield at different chamber length

Figure 13 demonstrates that at lower peak pulsed voltages the shorter chamber length produces a higher yield, but that the situation reverses at the higher voltages when the longer chamber produces an appreciably greater yield. Together

with figure 12, the results demonstrate that the maximum values of the yield are not obtained under conditions also giving the maximum concentration, and which one is optimised will depend on the particular application.

4. Conclusions

It has been confirmed that, as with other ozone production processes, concentration and yield are two different criteria that cannot be maximised simultaneously. The application for the ozone generation will be determined by which criteria is most important, and the PSD technique has been shown to be most useful when a high concentration rather than a high yield is required. It can also be concluded from the work that in general, the ozone concentration is increased by the use of a higher pulsed voltage, greater gaseous gap spacing and a longer electrode length but a slower air flow rate. On the other hand, the yield was generally found to be greatest at intermediate levels of these quantities with, for example the greatest concentration obtained when the electrode length was 220 mm whereas a length of 160 mm gave the greatest yield. Similarly, a greater gap spacing with higher input energy generates the highest yield of ozone at 15 kV. However, the ozone deformation due to the residence time at higher voltages also needs to be considered. Faster air flow rates (above about 1 l/m will be investigated in future research, as the ozone yield shows a linear relationship with an increasing air flow

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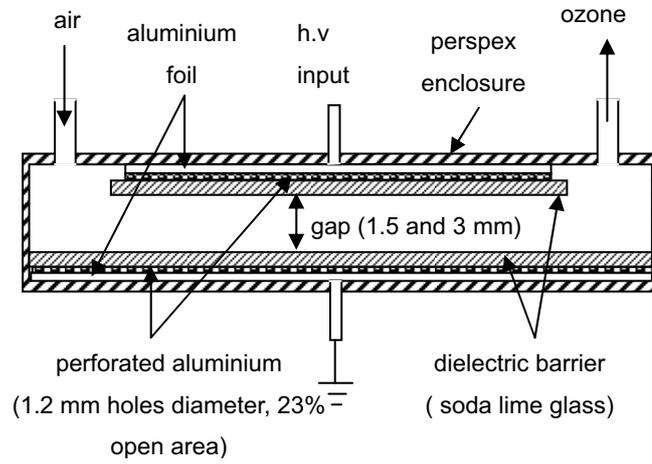


Figure 1. Reaction chamber (Buntat, 2005)

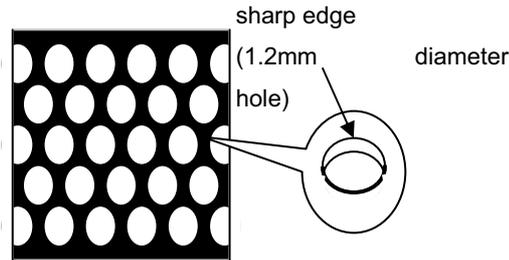


Figure 2. Perforated aluminium sheet (Buntat, 2007)

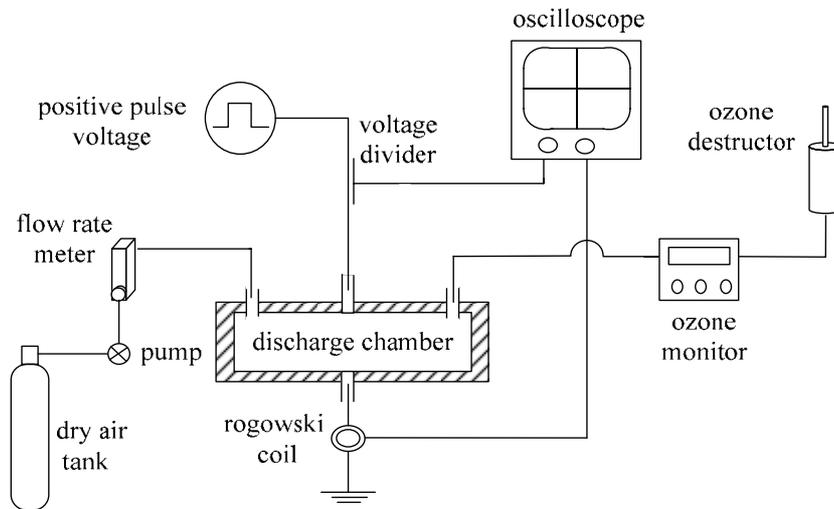


Figure 3. Block diagram of experimental assembly

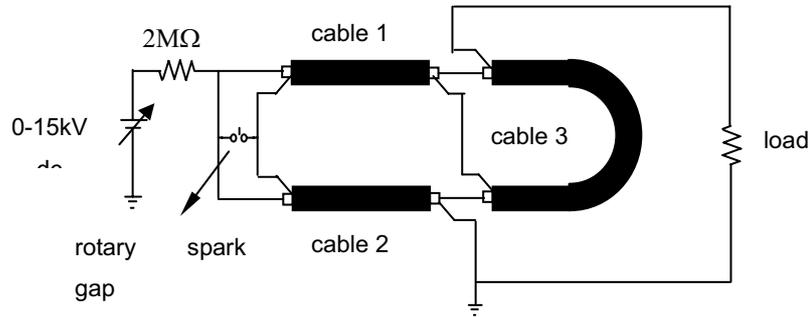


Figure 4. Pulse power supply of Blumlein configuration (Somerville, 1990)

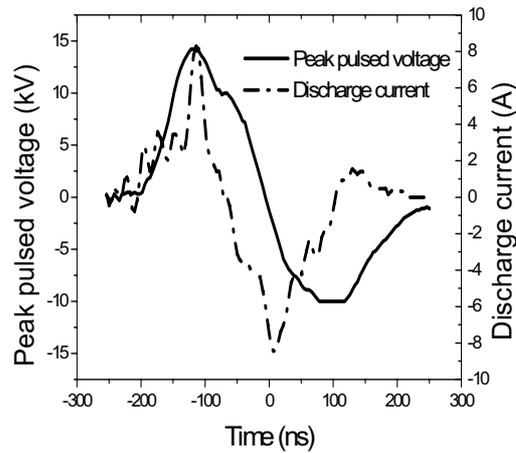


Figure 5. Typical voltage pulse and discharge current waveform. Gap spacing = 3 mm, peak pulsed voltage = 14 kV, air flow rate = 0.2 l/min, pulse repetitive rate = 50 pps, pulse duration = 120 ns, and pressure = 1 bar.

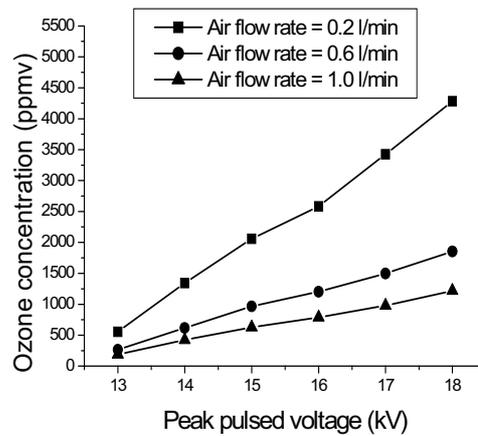


Figure 6. Effect of peak pulsed voltage on ozone concentration at different air flow rates. Gap length = 3 mm, chamber length = 160 mm, pressure = 1 bar

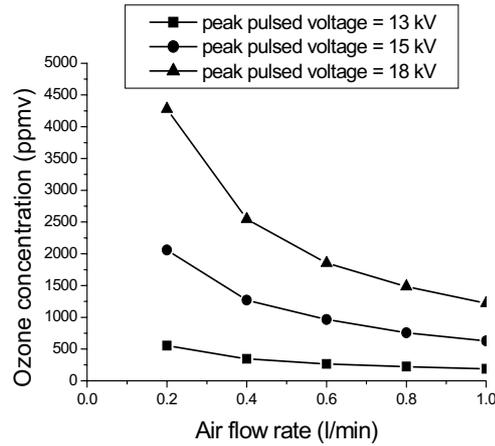


Figure 7. Effect of air flow rate on ozone concentration at different peak pulsed voltages. Gap length = 3 mm, chamber length = 160 mm, pressure = 1 bar

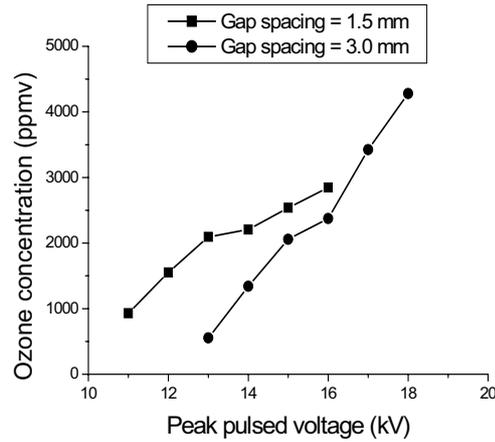


Figure 8. Effect of gap spacing on ozone concentration as a function of peak pulsed voltage. Chamber length = 160 mm, air flow rate = 0.2 l/min, pressure = 1 bar

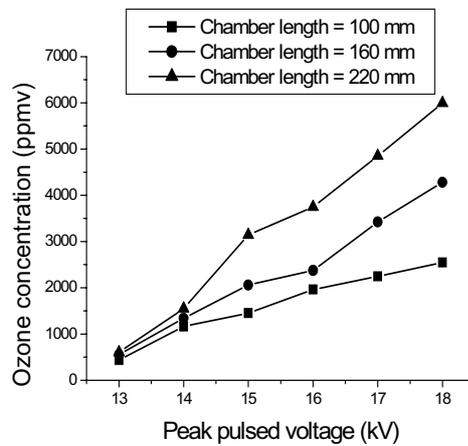


Figure 9. Effect of chamber length on ozone concentration as a function of peak pulsed voltage. Gap length = 3 mm, air flow rate = 0.2 l/min, pressure = 1 bar.

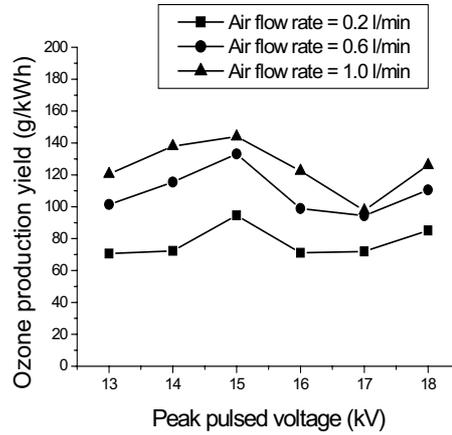


Figure 10. Effect of applied peak pulsed voltage on ozone yield at different air flow rates. Gap length = 3 mm, chamber length = 160 mm, pressure = 1 bar

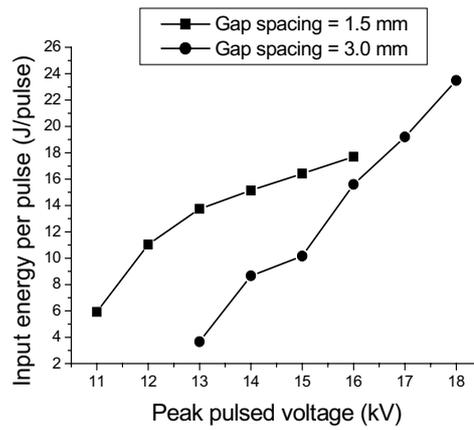


Figure 11. Effect of peak pulsed voltage on input energy into discharges/pulse.

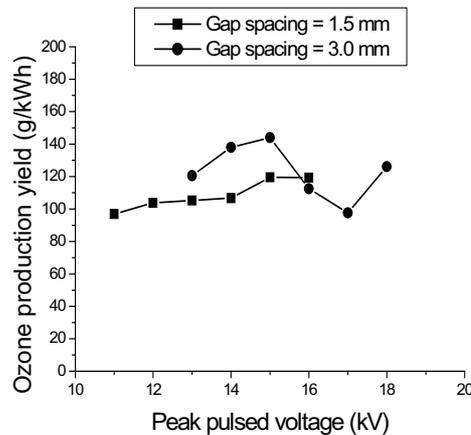


Figure 12. Effect of peak pulsed voltage on ozone yield at different gap spacing. air flow rate = 1.0 l/min, pressure = 1 bar.

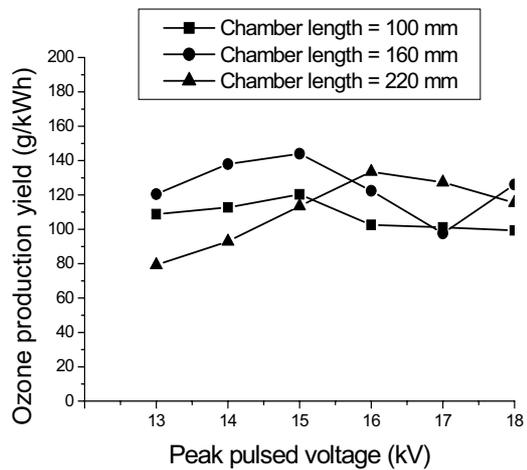


Figure 13. Effect of peak pulsed voltage on ozone yield at different. Gap length = 3 mm, air flow rate = 1.0 l/min, pressure = 1 bar.