Pulsed Power Production of Ozone Using Nonthermal Gas Discharges

Key Words: Pulse character, reactor geometry, ozone yield, dielectric barrier

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Ozone is increasingly being used in a large number of diverse applications as an alternative to other oxidants such as chlorine, peroxides, permanganates and dichromates, due to its better environmental compatibility. Generally, ozone is generated with high voltage ac in a corona gas discharge. A dielectric barrier made of a glass tube is usually placed adjacent either to the inner or the outer electrode in a concentric coaxial geometry.

The energy efficiency in conventional generators operating either at the power frequency or using thyristor controlled converters at frequencies up to a few kilohertz is rather low, because a portion of the applied electrical energy is converted into heating the gas and the electrodes and therefore wasted. Ozone production by pulsed streamer non-thermal discharges has been shown to be very effective without significantly raising the gas temperature or inducing arc breakdown between the electrodes at room temperature and atmospheric pressure. Experimental investigations of high voltage, short-pulsed streamer discharges in oxygen, and dry air under various operating conditions are reviewed. Ozone concentration and ozone production yield (efficiency) depend on the peak pulsed voltage, pulse repetition rate, input energy density, gaseous gap spacing, reactor length, type of gas, flow rate and the type of dielectric material used to form the barrier. The electrode configuration, and in particular the central electrode design, is important as in its vicinity the electric field is high. Therefore, most of the radicals that lead to the production of ozone are formed there. A spiral copper wire made to a cylindrical configuration in a concentric coaxial electrode system of various dimensions, with and without dielectric barriers, is preferable to a straight wire. In addition, the production yield of ozone (g/kWh) in a reactor without a dielectric barrier is compared with that using polyvinyl chloride (PVC) as a dielectric barrier.

Ozone is a very effective and potent oxidant and, therefore, there is a wide interest currently in an energy efficient production of ozone for numerous and diverse practical applications. These may include, as shown in Fig. 1, sterilization (germicide, viricide, and bactericide for drinking water, waste water, swimming pools, fish farms,), deodorization and decolorization (treatment of industrial wastes including dyes, phenolic, and cyanide compounds), bleaching processes (to get white kaolin, for textiles, for wax, and for paper pulps), chemical synthesis (for the oxidation of oleic acid and the production of certain hormones, antibiotics, vitamins, flavors, perfumes and fragrances), and gas treatment

The industrial requirements of the concentration of ozone depend on the applications to which the ozone is put to and this in turn determines the operational conditions of the ozonizer.

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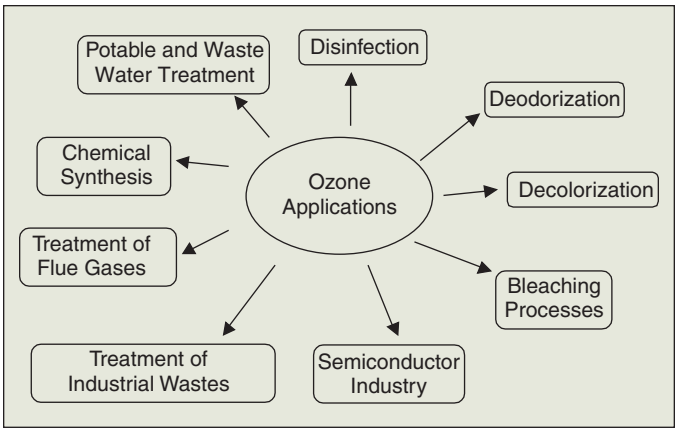

Fig. 1. Typical industrial applications of ozone.

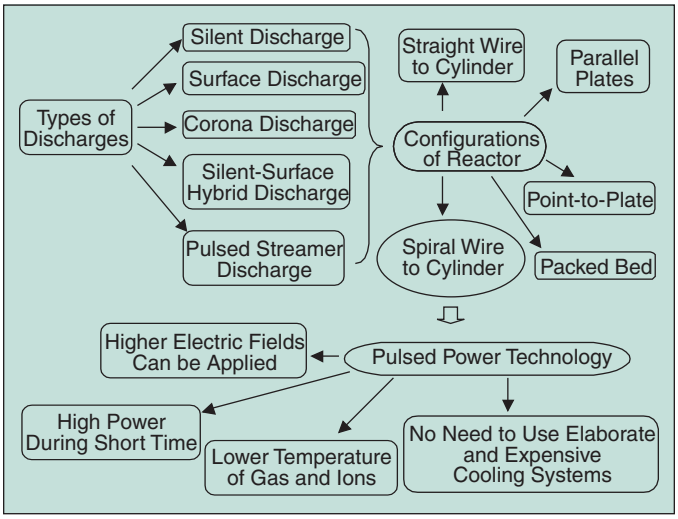

Fig. 2. Types of discharges, reactors employed in the production of ozone and merits of pulsed power technology.

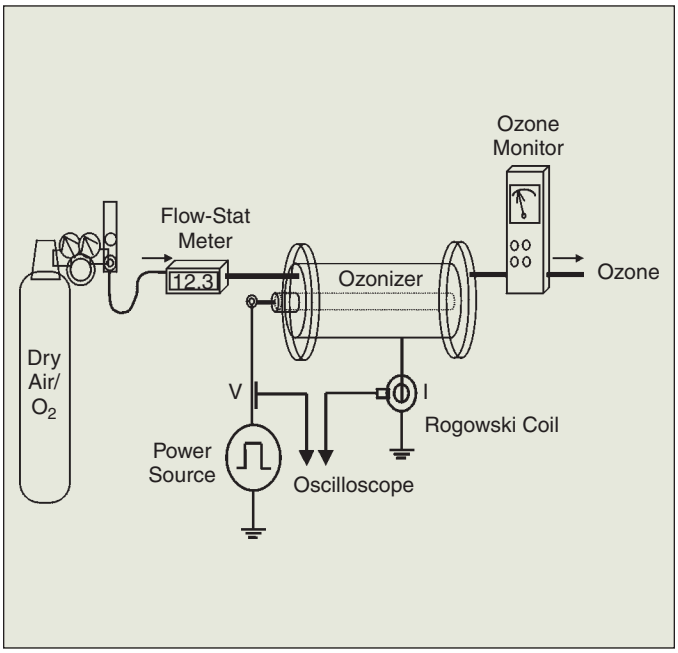

Fig. 3. Reactor and associated instrumentations using a spiral wire-to-cylinder configuration.

(to deodorize the gases containing hydrogen sulfide and formaldehyde, and to remove NO_x from flue gases) [1]-[4]. Ozone has little detrimental effect on the environment, as the natural decay product of ozone is oxygen. It has the added advantage of less energy consumption than other alternatives, such as the chlorination process [1]. Because of the hazards of storage, handling, and transportation due to its inherent instability [1], ozone is usually generated on the site of its application.

Ozone synthesis employing a dielectric barrier discharge in oxygen and in air has been extensively studied using ac or dc applied voltages [1], [4]-[8]. Different types of discharges have been used, as shown in Fig. 2. These include silent, surface, corona, a hybrid of silent and surface and pulsed streamer discharges. However, pulsed power either with or without a dielectric barrier offers substantial advantages [9, 10]. The main merits of using very short durations of the order of 10s of ns of pulsed power are that the temperature of the ions and of the neutral gas does not increase much above the ambient [11], homogeneous discharges at atmospheric pressure can be created and high energy electrons can be generated, which produce the radical O that is necessary for the production of ozone. Further, a breakdown leading to an arc across the electrode gap does not readily form [9], [12]-[16]. Streamers are created in atmospheric pressure of either oxygen or dry air, when a sufficiently high pulsed voltage is applied to the central electrode of a coaxial concentric cylindrical electrode geometry. The outer electrode is usually grounded.

The use of very short high-voltage pulses combined with a dielectric layer placed adjacent to the outer electrode results in a short lifetime of the streamers. This results in less energy transferred into the ions and the neutral gas, which obviates the need to use an elaborate cooling system to remove the heat from the electrodes, with a consequent decrease in the energy cost of generating the ozone. The presence of a dielectric layer in the discharge zone mitigates against the development of an arc discharge, and thus promotes the development of streamer discharges [17]. The dielectric layer reduces the electron emission from the cathode, which further inhibits the streamer-arc transition [1]. The dielectric layer reduces the charge transported by a single streamer and distributes the streamers over a wide area near the dielectric layer [1]. The high-energy electrons in the streamers dissociate the oxygen molecules into atoms, which are prerequisite for the subsequent production of ozone by collisions with oxygen molecules and third particles [1], [18]. The generation of ozone using air instead of oxygen is advantageous due to the readily available air, which obviates the requirement of cryogenic systems for producing oxygen on site. However, the production of ozone in air is substantially less than when oxygen is used. Further, ozone synthesis in oxygen acts to mitigate against the production of NO and NO_2, nitrogen oxides (NO_x), which are not desirable.

The present paper discusses ozone synthesis by employing a short duration (~120 ns) of pulsed power and using

positive voltages in dry air and in oxygen with and without a dielectric barrier. Ozone concentration and ozone production yield (efficiency) are measured at various peak pulsed voltages, pulse repetition rates, varying input energy densities and different gaseous gap spacings at a pressure of 1.01 x 10^5 Pa and a temperature of 26 ± 4 °C. A spiral copper wire made to a cylindrical configuration in a concentric coaxial electrode system of various dimensions, with and without dielectric barriers, is employed. In addition, the concentration of ozone in parts per million of the gas (ppm) and the production yield of ozone (g/kWh) without a dielectric barrier are compared with that of a reactor having a dielectric barrier made of PVC using a fixed reactor length of 1 m. The industrial requirements of the concentration of ozone depend on the applications to which the ozone is put to and this in turn determines the operational conditions of the ozonizer.

Reactors for Production of Ozone and Procedure

Figure 2 shows a wide range of reactor configurations that have been employed in the production of ozone. These include wire-cylinder, point-plate, parallel plates, packed bed with dielectric beads and spiral wire-cylinder geometries. A typical reactor using a spiral wire-to-cylinder configuration with the associated instrumentations for the generation of ozone is shown in Fig. 3. The discharge gap without a dielectric barrier is formed between a spiral copper wire, typically of 1 mm in diameter, coiled on vinyl chloride tubes and, in this case, having diameters in the range 18 to 38 mm and a copper tube 58 mm in internal diameter. Thus, the gaseous gap between the electrodes is varied in the range 9 to 19 mm. Another reactor with a 4 mm thick PVC dielectric barrier and 58 mm in outer diameter is also employed with a spiral wire coiled on a PVC tube 26 mm in diameter. A copper foil 0.1 mm thick is wrapped on the outside of the PVC dielectric barrier. A fixed gaseous gap of 11 mm between the central helical wire electrode and the inner surface of the PVC dielectric barrier is thus formed.

Either oxygen or dry air is obtained from gas cylinders and they are fed axially into the reactor (Fig. 3) and in this case using a constant flow rate of 1.5 or 3.0 l/min. The concentration of ozone is measured using an ultraviolet (UV) absorption meter at 253.7 nm, which is close to the maximum of the Hartley absorption band of ozone [1], [19]. The measurements are instantaneous and are not affected by the presence of nitrogen oxides, which are produced when dry air is used [1]. The output

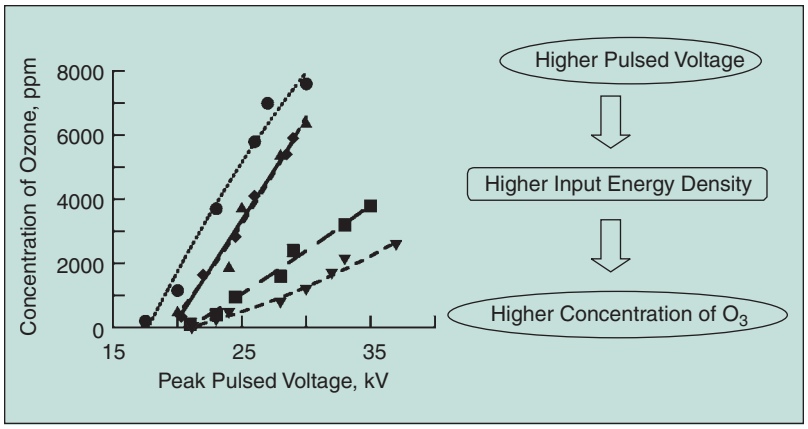

Fig. 4. Dependence of ozone concentration on applied peak pulsed voltage using 1 m long reactor without a dielectric layer in dry air.
Conditions: flow rate, 3.0 l/min; pressure, 1.01 x 10^5 Pa; temperature, 26 ± 4 °C; gaseous gap, 15 mm; pulse repetition rates: ▼25 pps; ■50 pps; ◆100 pps; ▲200 pps; ●400 pps.

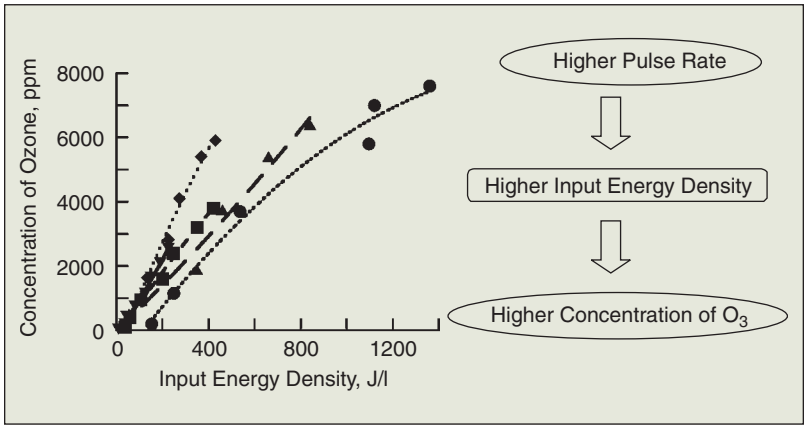

Fig. 5. Dependence of the concentration of ozone on the input energy density in dry air using a reactor without a dielectric barrier.
Conditions: flow rate, 3.0 l/min; pressure, 1.01 x 10^5 Pa; temperature, 26 ± 4 °C; gaseous gap, 15 mm; pulse repetition rates: ▼25 pps; ■50 pps; ◆100 pps; ▲200 pps; ●400 pps.

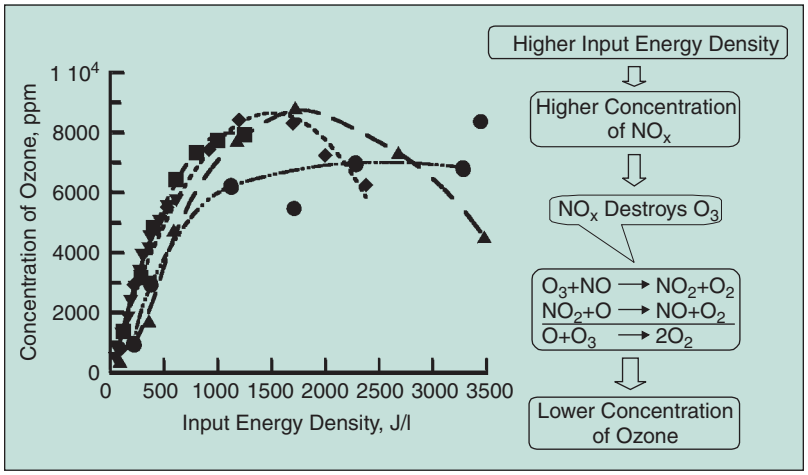

Fig. 6. Dependence of the concentration of ozone on input energy density into the discharge using a reactor with a PVC dielectric barrier, 4 mm thick in dry air.
Conditions: gaseous gap, 11 mm; pulse repetition rates: ▼25 pps; ■50 pps; ◆100 pps; ▲200 pps; ●400 pps.

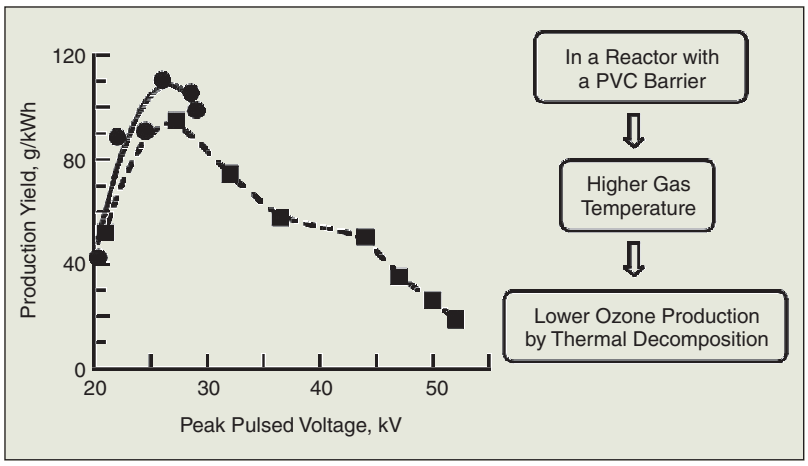

Fig. 7. Dependence of the production yield of ozone on peak pulsed voltage for 1 m long reactors with and without a dielectric barrier in dry air.
Conditions: pulse rate, 100 pps; electrodes gap length, 15 mm; type of reactor, ● *reactor without a dielectric layer,* ■ *reactor with a PVC dielectric barrier.*

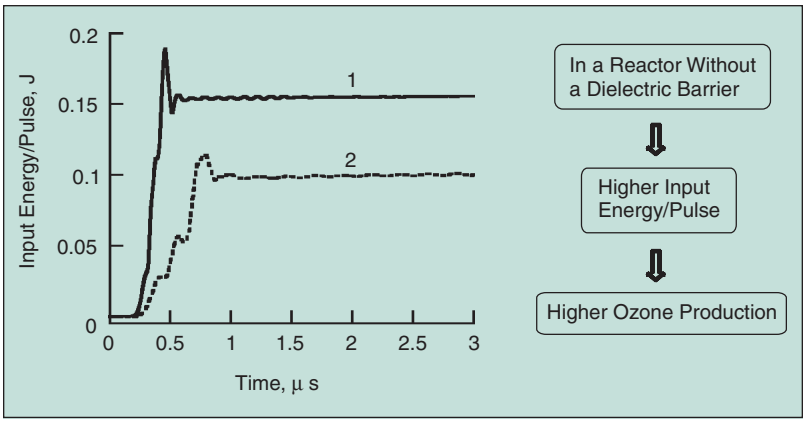

Fig. 8. Typical input energy plots per pulse to the discharge as a function of time at 26 kV peak pulsed voltage for 1 m long two types of reactors in dry air.
Conditions: pulse rate, 100 pps; electrodes gap length, 15 mm; curve 1, reactor without a dielectric barrier; curve 2, reactor with a PVC dielectric barrier.

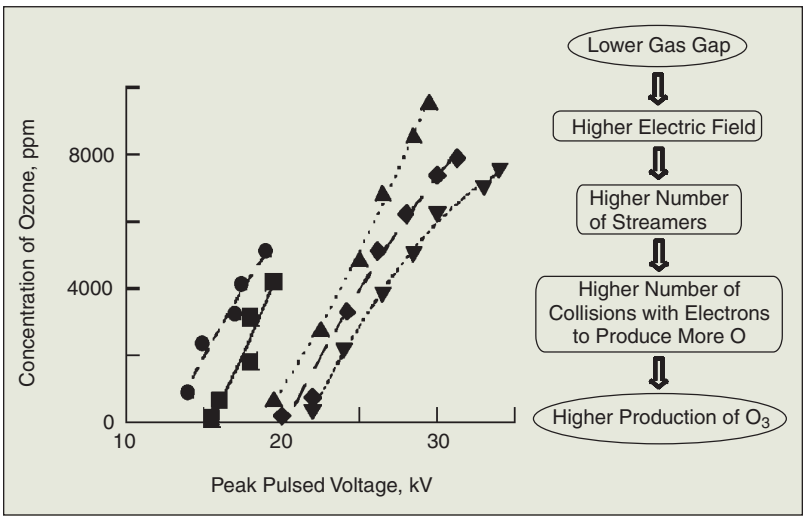

Fig. 9. Production of ozone as a function of peak pulsed voltage for different gap lengths without dielectric layer in dry air.
Conditions: pulse repetition rate, 100 pps; flow rate, 1.5 l/min; gaseous gap lengths: ● *9 mm;* ■*12 mm;* ▲ *15 mm;* ◆ *17 mm;* ▼ *19 mm.*

gas from the ozonizer can be put directly into use as an oxidizing agent. The gas pressure is typically at 1.01×10^5 Pa and the temperature is at 26 ± 4 °C. The waveforms of the applied pulsed voltage V (t) and the discharge current I (t) are monitored continuously via a high voltage capacitive divider and a Pearson Rogowski coil, respectively. The energy ($\int VIdt$) input to the discharge per pulse is determined from the digitized signals of the voltage (V), current (A) and time (s). The production yield of ozone in mol/kWh or (g/kWh) is determined from

$$\eta = F\ (l/60\ s) \times N\ (O_3) \times 3.6 / \{22.4\ (l/mol) \times f\ (pps) \times E\ (J)\}\ (in\ mol/kWh) \quad (1)$$

where F is the gas flow rate in l/min, $N(O_3)$ the concentration of ozone in ppm, f the pulse repetition rate (pulses/s) and E the input energy to the reactor per pulse (J). To obtain units of $g(O_3)$/kWh, equation (1) should be multiplied by 48. High voltage (HV) pulses of positive polarity are applied to the central electrode from a magnetic pulse compressor (MPC) with a repetitive rate capability of up to 500 pulses per second (pps) and up to 62 kV [18]. The positive polarity of the central electrode has been shown to be very effective for pulsed ozone production, as the streamers propagate to longer distances in the radial direction, and hence, the volume of the reaction zone will be larger, leading to more production of O_3 with more branches and more streamer channels per cm length than using the negative polarity [15], [17], [20, 21]. The positive polarity also tends to postpone the development of the breakdown of the gap due to lower secondary ionization emission from the negative outer electrode, where the electric field is lower [22, 23]. A typical electrical circuit diagram of the MPC and a brief description of its operation can be found in [18].

Ozone Production With and Without a Dielectric Barrier in Dry Air

The concentration of ozone in dry air without a dielectric barrier is shown in Fig. 4 as a function of the pulsed voltage for different pulse repetition frequencies. A flow rate of 3.0 l/min and a gaseous gap of 15 mm were employed. The concentration of ozone increases with increasing pulse repetition rate at a fixed peak pulsed voltage. At a lower pulse repetition rate, higher voltage can be applied before a streamer to arc transition occurs. This is because a higher pulse rate leads to an increase in the discharge current with a higher electron density thus leading to breakdown of the gap.

The dependence of the ozone concentration on the energy density input into the discharge (in J/l) at different pulse rates is shown in Fig. 5 without, and in Fig. 6 with, a dielectric barrier. It will be seen from Fig. 5 that the concentration of ozone increased steadily with increasing input energy density at all repetition rates. For the reactor with a dielectric barrier, the ozone concentration increases steadily, with increasing input energy density up to about 1700 J/l and then saturation is reached followed by a reduction in the concentration at < 400 pps (Fig. 6). Increasing the energy input into the discharge above this level for pps < 400 is counter productive as the destruction reactions of ozone are more predominant than its formation. For 400 pps, a saturation in the concentration is reached at about 1000 J/l (Fig. 6).

The production yield against peak pulsed voltage for two types of reactors with and without a dielectric barrier is shown in Fig. 7. It will be observed that the reactor without a dielectric barrier has a slightly higher production yield than the reactor with a PVC dielectric barrier. This is due to the higher input energy into the discharge in the former reactor for the same applied voltage (Fig. 8). Further, the temperature of the reactor containing a PVC dielectric barrier is slightly higher than that in the reactor without a dielectric barrier, and hence, a thermal decomposition of ozone takes place in the former reactor. These reasons lead to a higher concentration of ozone giving a higher production yield than for the reactor with a dielectric barrier as shown in Fig. 7.

Ozone Production Using Different Gaseous Gap Lengths in a Reactor Without a Dielectric Barrier in Dry Air

The dependence of the concentration of ozone on the applied peak pulsed voltage is shown in Fig. 9 for different gaseous gaps in dry air. It will be observed from Fig. 9 that as the gap length decreases, the concentration of ozone becomes higher for a fixed applied pulsed voltage. This is attributed to the higher electric field at the spiral wire with decreasing gap length, which results in decreasing the average density of the low energy electrons that decompose the generated ozone [24]. Thus, the higher electric field indirectly increases the production of atomic oxygen, which is a prerequisite for the generation of ozone. Further, at the shorter gap length, the production of ozone started at a lower pulsed voltage (14 kV at 9 mm gaseous gap, Fig. 9). A production yield of ozone in dry air in a reactor without a dielectric barrier of 109.1 g/kWh was obtained at 12 mm gaseous gap and 1.5 l/min flow rate using 18 kV peak pulsed voltage [18].

All yields (g/kWh) of ozone in dry air using a reactor without a dielectric barrier peaked at a certain pulsed voltage and then decreased with further increase in the peak pulsed voltage. The dependence of the production yield is related to the dependence of the concentration of ozone (Fig. 9), and the dependence of the energy input into the discharge [25] on the peak pulsed voltage. The energy input into the discharge increased with decreasing gaseous gap of the reactor at a fixed voltage and a constant flow rate [25]. This is because at a fixed pulsed voltage, the discharge current increases with decreasing gaseous gap. This results in an increase in the conductivity of the plasma discharge due to the higher electric field at short gaps resulting in an increase in the number density of streamers.

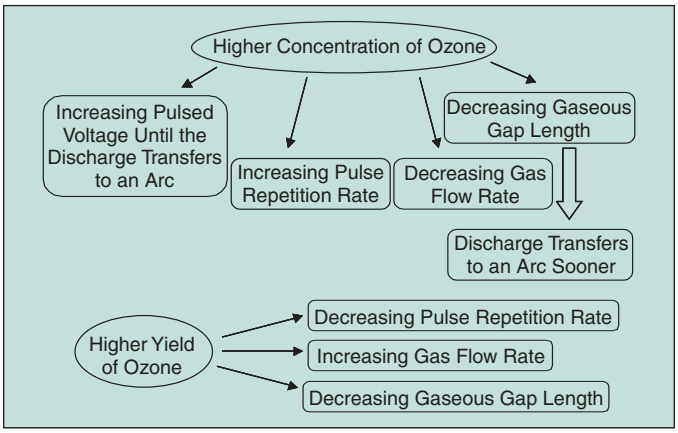

Fig. 10. Conclusions for dry air using a reactor without a dielectric layer.

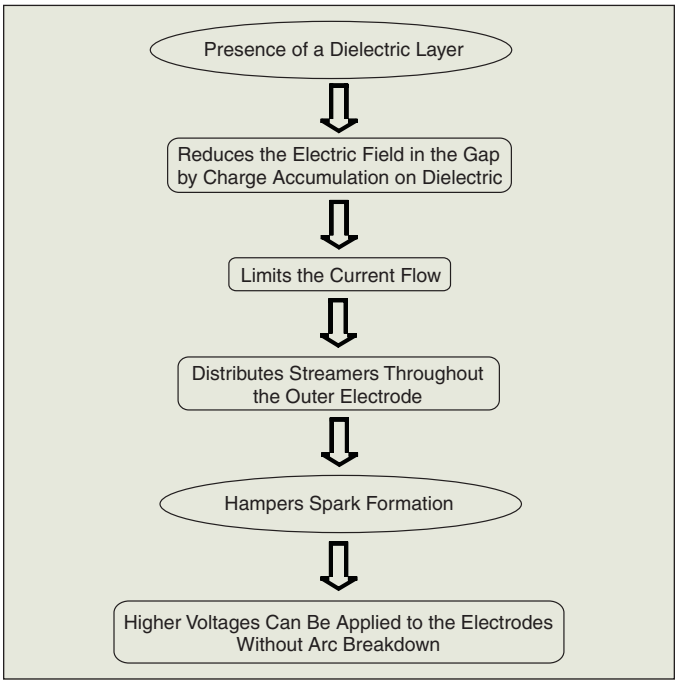

Fig. 11. A summary of the effects of introducing a dielectric layer in the discharge reactor.

Figure 10 summarizes the effects of the variation of different discharge parameters on the concentration of ozone produced in dry air in a reactor without a dielectric barrier. A higher concentration of ozone can be obtained with increasing the pulsed voltage until a threshold level is reached, and thereafter the discharge transfers into an arc. Increasing the pulse repetition rate, decreasing both the gas flow rate and the gaseous gap length also result in increasing the concentration of ozone (Fig. 10). However, a higher energy yield can be obtained with increasing the gas flow rate and de-

creasing of both the gap length and the pulse repetition rate in dry air (Fig. 10).

Ozone Production in a Reactor with a PVC Dielectric Barrier in Oxygen

Figure 11 summarizes the effects of introducing a dielectric layer into the discharge reactor. The presence of the dielectric layer is very useful, as it enables the application of a higher pulsed voltage to the reactor before a breakdown of the gap occurs. Therefore, a larger amount of energy can be fed into the discharge resulting in a higher production of ozone [26].

The dependence of the ozone concentration on the applied peak voltage in oxygen is shown in Fig. 12 and as a function of input energy density in Fig. 13. It will be observed from Fig. 12 that the concentration of ozone increases with increasing applied pulsed voltage at a fixed pulse rate. The rate of rise of the concentration of ozone with increasing voltage increases with increasing pulse repetition rate (Fig. 12). Thus, a higher repetition rate gives a higher concentration of O_3 at a fixed pulsed voltage (Fig. 12). In order to get increased amounts of ozone, it was necessary to apply higher pulsed voltages with a lower pulse rate, or alternatively to apply lower pulsed voltages with a higher pulse rate (Fig. 12). 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 (Fig. 13). It will also be seen from Fig. 13 that the concentration of ozone increased steadily with increasing input energy density at all repetition rates. However, the lower pulse rates are preferable to produce the same amount of concentration of ozone with lower input energy density, as shown in Fig. 13. The measurement of ozone concentration is restricted in this case to 10,000 ppm in oxygen due to the ozone monitor used, and not by the breakdown voltage of the gaseous gap, which is the case in dry air without a dielectric barrier (Figs. 4 and 9).

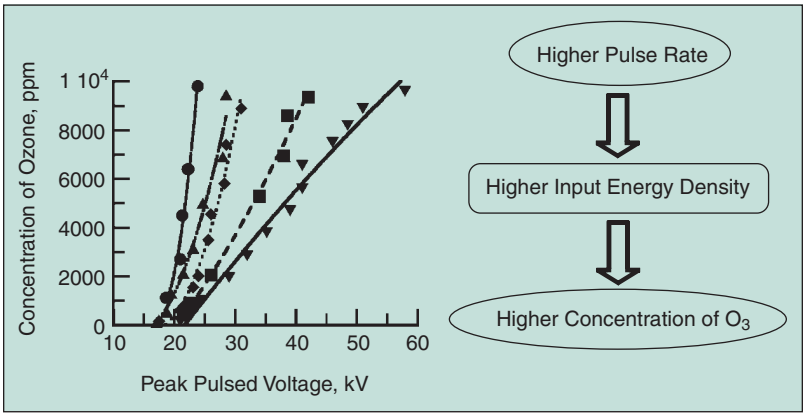

Fig. 12. Dependence of ozone concentration on peak pulsed voltage with PVC dielectric barrier in oxygen at various pulse repetition rates; ▼ 25 pps; ■ 50 pps; ◆ 100 pps; ▲ 200 pps; ● 400 pps.

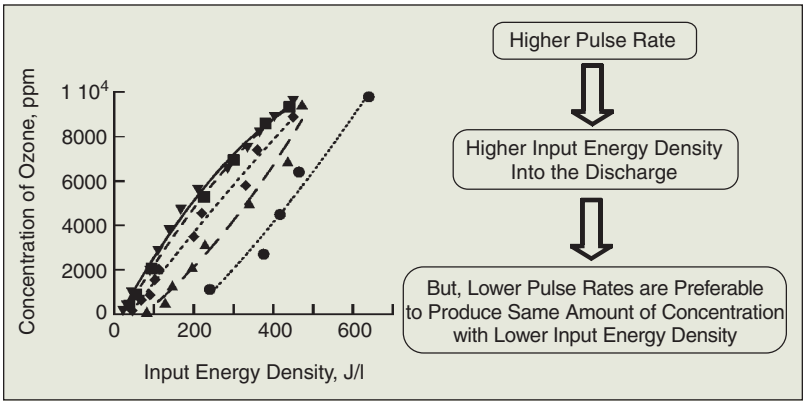

Fig. 13. Dependence of the concentration of ozone on the input energy density into the discharge in oxygen at various pulse repetition rates; ▼ 25 pps; ■ 50 pps; ◆ 100 pps; ▲ 200 pps; ● 400 pps.

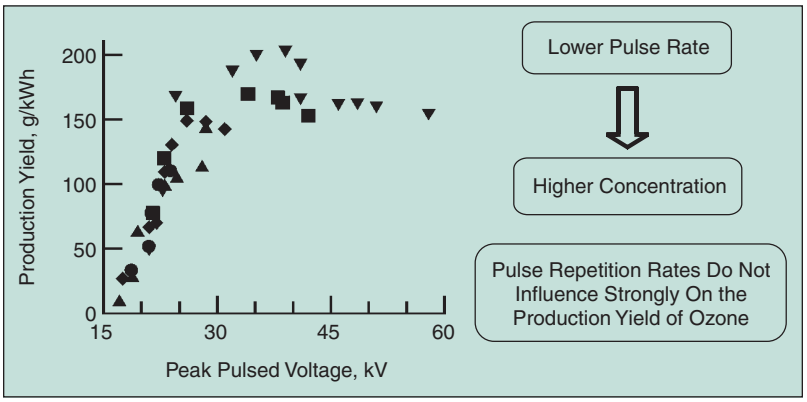

Fig. 14. Dependence of ozone yield on peak pulsed voltage in oxygen at various pulse repetition rates; ▼ 25 pps; ■ 50 pps; ◆ 100 pps; ▲ 200 pps; ● 400 pps.

The dependence of the production yield of ozone on the peak pulsed voltage is shown in Fig. 14. The production yield increases to a maximum value of 202 g/kWh (4.21 mol/kWh) with increasing voltage up to about 39 kV and then reaches a saturated level. Figure 14 shows that the pulse repetition rate does not influence strongly the production yield. Above about 39 kV, the input energy per pulse increases with increasing voltage at a faster rate than linearly while the concentration of ozone increases linearly [26]. This results in the reduction of the yield with further increasing voltage. The non-linear increase in the input energy at high voltages is because the discharge current increases much higher than linearly with increasing voltage. The large increase in the discharge current is due to the non-linear increase in the conductivity of the discharge arising from the simultaneous increase in the electron density at higher applied voltages as well as due to higher electron mobilities at the higher electric fields.

Oxygen-fed ozonizers are preferable for ozone production as they consume lower energy density than in dry air. The latter requires more than twice the input energy density under the same experi-

mental conditions. For example, 490 J/l input energy density is required to produce a concentration of ozone in oxygen of about 6000 ppm at 400 pps (Fig. 13) while in dry air, it requires 1120 J/l (Fig. 6). This is 2.3 times that in oxygen for producing the same amount of ozone, using the same reactor, with a PVC dielectric barrier and under the same conditions.

Figure 15 summarizes the effects of varying different parameters on the production and the energy yield of ozone in a reactor with a dielectric barrier filled with oxygen. A higher concentration of ozone can be obtained with decreasing the gas flow rate and increasing of both the pulse repetition rate and the applied pulsed voltage. A higher yield can be obtained with decreasing pulse rate and increasing gas flow rate. Figure 16 gives a summary of the effects of various parameters on the concentration of ozone and its energy yield using dry air in a reactor with a dielectric barrier. Thus, a higher concentration of ozone can be achieved by increasing the parameters of pulsed voltage, pulse repetition rate, reactor length, pitch length of anode wire, decreasing the gas flow rate and with including a ceramic dielectric layer [27]. A higher yield of ozone can be obtained with increasing both of the reactor length and pitch length of anode wire, decreasing pulse rate and with including a ceramic dielectric barrier [27].

Figure 17 shows a comparison between the energy yield of ozone in oxygen and in dry air under the same conditions of applied voltage and using the same discharge reactor. It will be observed that the production yield of ozone is substantially higher in oxygen than in dry air. This is primarily due to the presence of a higher concentration of oxygen when oxygen is used than in dry air, at the same gas pressure. The higher concentration of oxygen results in the production of a higher concentration of the radical O. This leads to a higher production of ozone, as shown in Fig. 17.

It should be noted that the concentration and the production yield of ozone cannot be maximized simultaneously, and the choice of which is to be maximized should be made depending upon the application. The required level of ozone concentration depends on its specific application, and the results discussed here are applicable to a wide range of 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 up to 10,000 ppm [28].

Chalmers, et al., reported that in oxygen there are strong dependent values of the yield on the pulse width from 300 g/kWh at 20 ns to 147 g/kWh at 120 ns without a dielectric barrier [9, 10]. Using a surface discharge in oxygen at 10 kHz, best result reported by Masuda et al. in pure oxygen was a yield of 170 g/kWh [29] with a high level of ozone production of 5 to 10 %. Pietsch, et al., reported yield values of 122 and 27.07 g/kWh

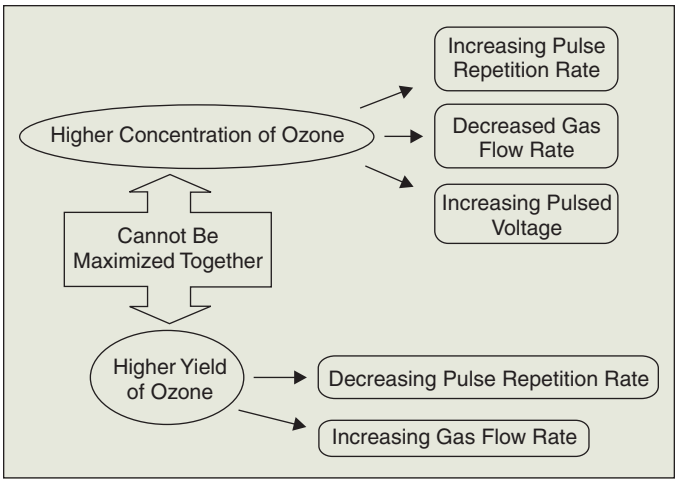

Fig. 15. Summary of the effects of varying various parameters on the production and yield of ozone in a reactor with a dielectric barrier filled with oxygen.

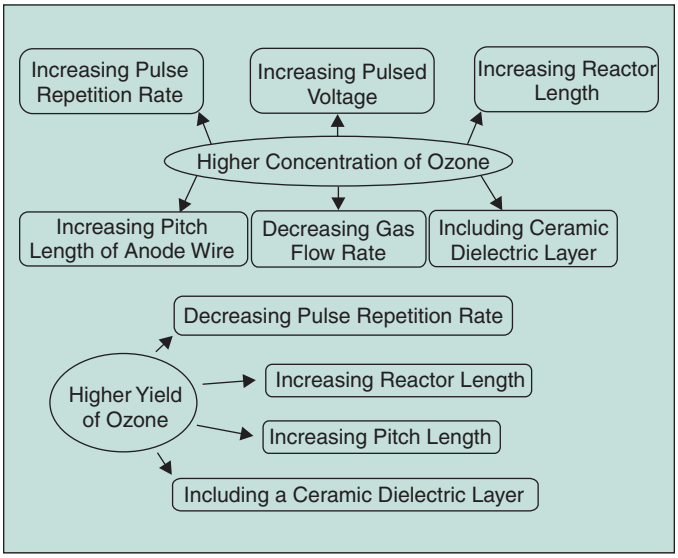

Fig. 16. Summary of the effects of various parameters on the concentration of ozone and its energy yield using dry air in a reactor with a dielectric barrier.

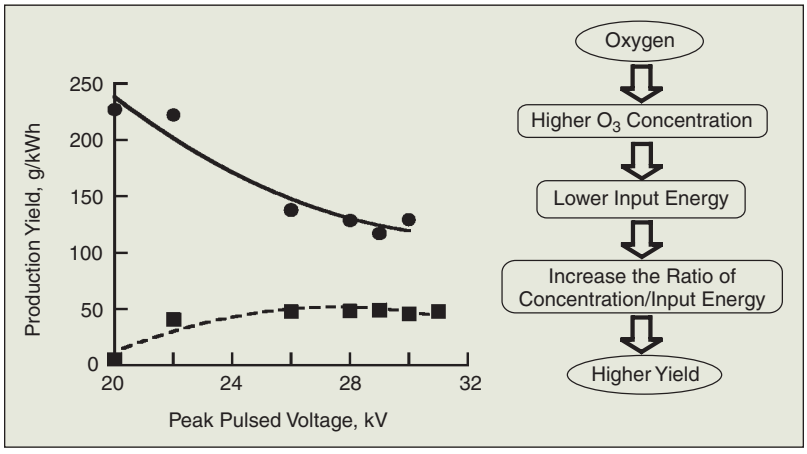

Fig. 17. Dependence of ozone yield on peak pulsed voltage for 1 m long reactor without a dielectric barrier in oxygen and in dry air.
Conditions: ■ dry air; ● oxygen; flow rate, 3.0 l/min; pulse repetition rate, 100 pps; electrodes gap length, 17 mm.

in O_2 and in air respectively, using dielectric barrier discharges [30].

The theoretical yield of ozone is 1226 g (O_3)/kWh. In commercial systems, the production yield of ozone in air was reported to be 50 to 55.6 g/kWh (18-20 kWh/kg) [31]. Masuda, et al. [21] 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. The results using pulsed power are compared very favorably with the previously reported values of the production yield of ozone in dry air (Fig. 7) and in oxygen (Fig. 14).

Conclusions

1. Positive pulsed corona in a dielectric barrier discharge has shown promising results that can readily be implemented in industrial applications using coaxial cylindrical ozonizers.

2. The concentration of ozone increases with increasing applied peak pulsed voltage at a constant pulse rate, a fixed gap length and a constant gas flow rate with and without PVC dielectric layer in oxygen and in dry air.

3. The concentration of ozone increases with decreasing gaseous gap length at a fixed applied pulsed voltage, a fixed pulse rate and a constant gas flow rate in a reactor without a dielectric barrier.

4. In dry air, the ozone concentration in a reactor without a dielectric barrier increases steadily with increasing input energy density at all pulse repetition rates while in the reactor with a PVC dielectric barrier, the concentration of ozone initially increases with increasing input energy density, reaches saturation and then declines with further increase in the input energy density at a fixed flow rate and a pulse repetition rate.

5. The production yield of ozone is higher without a dielectric barrier than with a PVC dielectric barrier in dry air.

6. A pulse repetition rate of 25 pps using a fixed gaseous gap of 11 mm and a constant flow rate of oxygen of 3.0 l/min produces the highest production yield of ozone (202 g/kWh).

7. Oxygen fed ozonizer is preferable to produce ozone with lower energy consumption than in dry air, under the same experimental conditions.

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