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DEVELOPMENT AND CHARACTERIZATION OF SURFACE DIELECTRIC BARRIER DISCHARGE-BASED REACTOR FOR OZONE PRODUCTION

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The aim of this work is to develop an efficient plasma reactor for ozone production. The electrical characterization of the developed reactor, including the current-voltage waveform and the consumed power, is investigated at different reactor configurations (including ground-electrode width, number of electrodes, and volume of the reactor) and operating parameters (including the air flow rates and energy density). The performance of the developed reactor for ozone production is evaluated. It is observed that the consumed power increased linearly with increasing the ground-electrode width and the number of electrodes, while the reactor volume and the flow rates have no noticeable effect on the consumed power. It is also found that the increase of the plasma area by increasing the width of the ground electrode is preferable than its increase by increasing the number of electrodes. This is attributed to the decrease of the power loss in the dielectric material. In addition, an optimum size of the reactor is obtained, as the higher and the smaller ratio of the reactor volume to the plasma area are not efficient.

1. INTRODUCTION

Dielectric barrier discharge (DBD) is one of the most efficient plasma type used for ozone generation in industry, due to its ability to produce a volume of non-thermal plasma at atmospheric pressure [1]. A typical DBD is characterized by the presence of an insulating material in the discharge path between two electrodes energized by an AC or pulsed high voltage. The insulting material plays the key role for the proper functioning of the discharge as it limits the current flow into the system, and prevents the glow-to-arc transition [2, 3]. In addition, the accumulated charges over the dielectric during one half cycle of the applied voltage favours the discharge breakdown in the next half cycle [4]. DBD can be classified based on the location of the electrodes with respect to the dialectic material into two types: volume-DBD (VDBD) and surface-DBD (SDBD) [5]. A typical volume DBD is characterized by the presence of at least one gas gap between one electrode and the dielectric material, where the discharge occurs in the volume of the gas gap and transferred to spread over the dielectric material. On the other hand, the insulating barrier in a SDBD is sandwiched between the two electrodes and the discharge develops along the surface of the dielectric plate. SDBD is widely investigated in different applications, such as airflow control [6], volatile organic compounds (VOCs) removal[7], ozone production [8, 9], and bactericidal agents [10]. The ozone, being a powerful oxidant [11], has many applications in industry. It also has a capability to inactivate microorganisms [12]. As regards ozone production, several studies showed that SDBD is more efficient compared to the volume DBD, due to the following reasons [13-15];(1) reduction of the applied voltage for its operation with a subsequent increase of the power efficiency, (2) preventing of the destruction of the produced ozone, which accumulates over the dielectric material without being destructed by the high temperature of the plasma channels, and (3) higher density of micro-discharges in surface DBD, because they occur over the entire length of the electrode. This produces more highly reactive species which suitable for various applications.

In electrical discharge, the ozone produced through the reaction of O atoms O2 by the following reaction [16]:

$$\mathrm{O(^3P)\ or\ O(^1D) + O_2 + Backround\ molecule \rightarrow O_3 + M} \qquad \text{(R1)}$$

The production of O atoms in the discharge is mainly due to the dissociation of oxygen molecules by the electronic dissociative excitations and collisions with excited states of nitrogen, as described in the following reactions [17]:

$$\mathrm{e^- + O_2 \rightarrow O(^3P) + O(^3P) + e^-} \qquad \text{(R2)}$$
$$\mathrm{e^- + O_2 \rightarrow O(^1D) + O(^3P) + e^-} \qquad \text{(R3)}$$
$$\mathrm{O(^1D) + N_2\ (or\ O_2) \rightarrow O(^3P) + N_2 (or\ O_2)} \qquad \text{(R4)}$$
$$\mathrm{N_2^* + O_2 \rightarrow O(^3P) + O(^3P) + N_2} \qquad \text{(R5)}$$

Although the huge advantages of the SDBD, there are still challenges in the SDBD need to be addressed to improve its chemical reactions. The main challenge is the occurrence of the discharge in a small area along the discharge electrode at the surface of the dielectric, which reduces the treatment volume and its practical usage. One of the succeeded attempts to increase the treatment volume and energy efficiency is using multi-electrodes SDBD [18, 19]. Yet, the correlations between the plasma area and the volume of the reactor along with the operating parameters have not been investigated before. Thus, the aim of this work is to develop a multi-electrodes SDBD based reactor, and optimize its operating conditions to maximize its efficiency for ozone production.

2. EXPERIMENTAL DETAILS

Figure 1(a) shows a schematic diagram of the experimental setup used in the present study. The reactor consists of a glass tube containing an SDBD source. The tube has a length of 220 mm, and thickness of 1.5 mm with different tube diameters (D_t =12.4, 21.7, 30.2 mm). The SDBD source consists of two copper tapes, each with length of 180 mm, fixed on both sides of the glass tube. One copper tape, with width (W_d) of 5 mm was fixed inside the tube and connected to a high voltage source. The other tape, with different widths (5, 10, 15, 20 mm), was grounded and fixed on the outer surface of the glass tube, where its center corresponds to the center of the discharge electrode, as shown in figure 1. The driving signal of the applied voltage is generated using a function generator (Key sight 33220A) and sent to a high-voltage amplifier (Trek, 20/20C-HS). The applied voltage was measured using a high-voltage probe (Tektronix P5210), which was connected to a digital oscilloscope (Tektronix DPO 3024, bandwidth: 200 MHz, 2.5 GS/s).

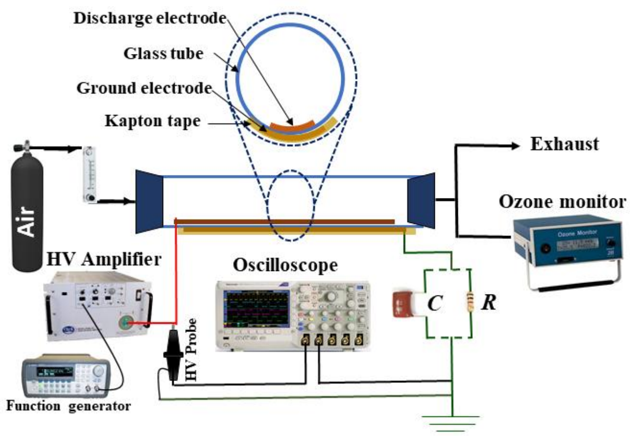

Figure 1: *Schematics of the experimental set-up*

Dry air from a compressed gas cylinder was used to feed the reactor at the atmospheric pressure, and its flow rate (Q) was controlled using a flow meter, as shown in figure 1. The ozone production of SDBD at the outlet of the reactor was measured using a UV ozone monitor (2B technologies). The measurements of ozone concentration were recorded at stable discharge with steady-state ozone levels as indicated by the O_3monitor.The ozone concentration values are presented as a function of the energy density, being defined as the energy per unit volume of gas consumed by the discharge reactor and changed with the input power to the reactor. The energy density can be determined using the following equation:

$$E_d = \frac{60\ P_d}{Q} \qquad (1)$$

where E_d, P_d, and Q are the energy density (J/L), consumed power (W), and gas flow rate (L/min), respectively. The energy density is changed with the applied voltage, when the frequency is fixed. The flow rate Q of inlet dry air through the reactor was controlled using a flow meter.

The consumed power in the reactor *(P_d)* was calculated using the charge-voltage Lissajous diagram technique[20], where a capacitor with capacitance of 100 nF was connected between the ground electrode and the ground. The area of the diagram represents the energy dissipated per cycle, and then the dissipated power is calculated by the following equation:

$$P = f \int_{cycle} V\, dq_c \qquad (2)$$

In addition to the energy dissipated, the diagram gives an interesting information about the characteristics of the discharge inside the reactor, including the onset voltage V_o, plasma on durations, and plasma off durations (where the displacement current flows only in the reactor), as well as the capacitance of the dielectric (C_d) and gas (C_g)[21], as shown in figure 2.

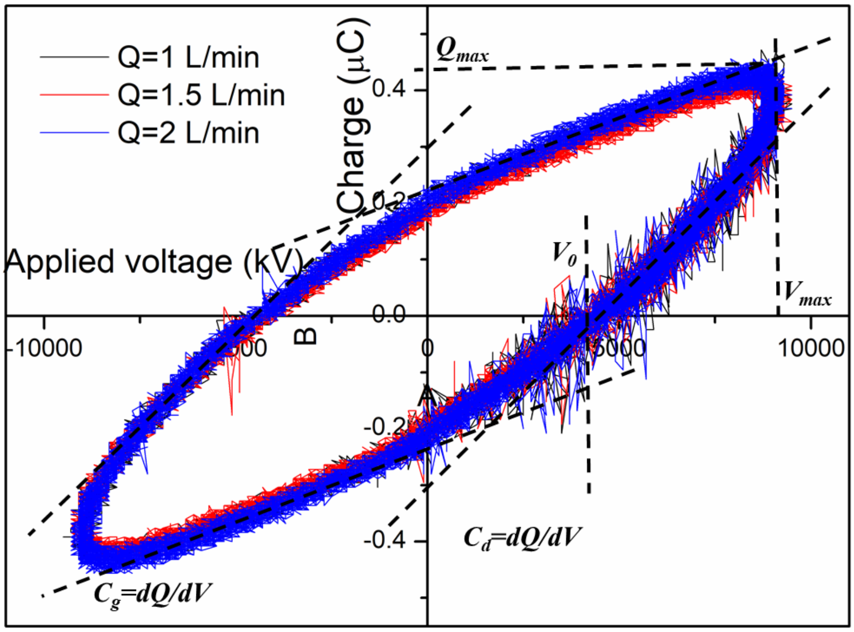

Figure 2: *q-V Lissajous diagram for the SDBD at different flow rates.*

The consumed power is confirmed by calculating the power in the reactor *(P_d)* by time integration of the product of the voltage and current waveforms over one cycle using the following equation:

$$P_d = f \int_0^\tau V(t) \cdot I(t)dt \quad (3)$$

where τ and f are the period and frequency of the applied voltage, respectively. *V(t)* and *I(t)* are the instantaneous values of the applied voltage and current, respectively. The current *I(t)*was measured using a shunt resistor technique, where a 1 kΩ resistor was connected in series with the reactor (i.e. between the grounded electrode and the ground).

The power values used in this study were the average of three power values corresponding to three measurements of the current–voltage waveform.

The energy yield (the production yield of ozone) (g/kWh) is defined as the amount of produced ozone per unit input energy and calculated as follows:

$$\eta = 60\ Q\ N\left(\frac{M}{22.4}\right)\left(\frac{10^{-3}}{P_d}\right) = 60\ Q\ N\left(\frac{48}{22.4}\right)\left(\frac{10^{-3}}{P_d}\right) = \frac{48 \times 3.6\ Q\ N}{22.4 \times 60\ P_d}$$

$$\eta = 7.7143 \frac{N}{E_d} \quad (4)$$

where *N* and *M* are the ozone concentration (ppm) and the molecular weight of ozone (48 g/mole), respectively. This equation is based on the fact that one mole of a gas occupies a volume of 22.4 L at a temperature of 0 °C and a pressure of 1 atm.

3. RESULTS AND DISCUSSION

3.1.Plasma Characterization

Figure 3 shows a typical applied voltage – current waveforms for the SDBD at different values of ground-electrode widths (W_g= 10, 15, and 20 mm) and a constant applied voltage (V_{pp}=16.8 kV). For all the widths values, the current waveforms composed of a series of pulses referred to the discharge current overlapped sinusoidal current that represent the capacitive current. Each of these current pulses corresponds to a set of microdischarge spread over the inner surface of the glass tube along the edges of the discharge electrode. This indicates that the discharge in the present reactor is in form of filamentary streamers. However, it is observed that the amplitude of the current pulses in case of W_g= 10 mm is lower than that of W_g=15 mm and W_g=20 mm. Increasing the grounded-electrode width provides a larger area for spreading the discharge[22], which leads to an increase of the discharge current. This explains the increase in the amplitude and the number of the current pulses with increasing the ground-electrode width. The increase in the current with increasing the ground-electrode width reflects on the behaviour of the consumed power, as shown in figure 4, wherethe consumed power increased linearly with the increasing of the width, but exponentially with the applied voltage. Figure 4also indicates that the effect of the ground-electrode width is more pronounced at higher applied voltages, where the slope of the linear relationship increased from 0.015 to 0.26 when the peak to peak of the applied voltage increased from 8.7 kV to 19 kV. This is due to the localization of the electric field at higher applied voltage in a small area when a narrow ground electrode width is used.

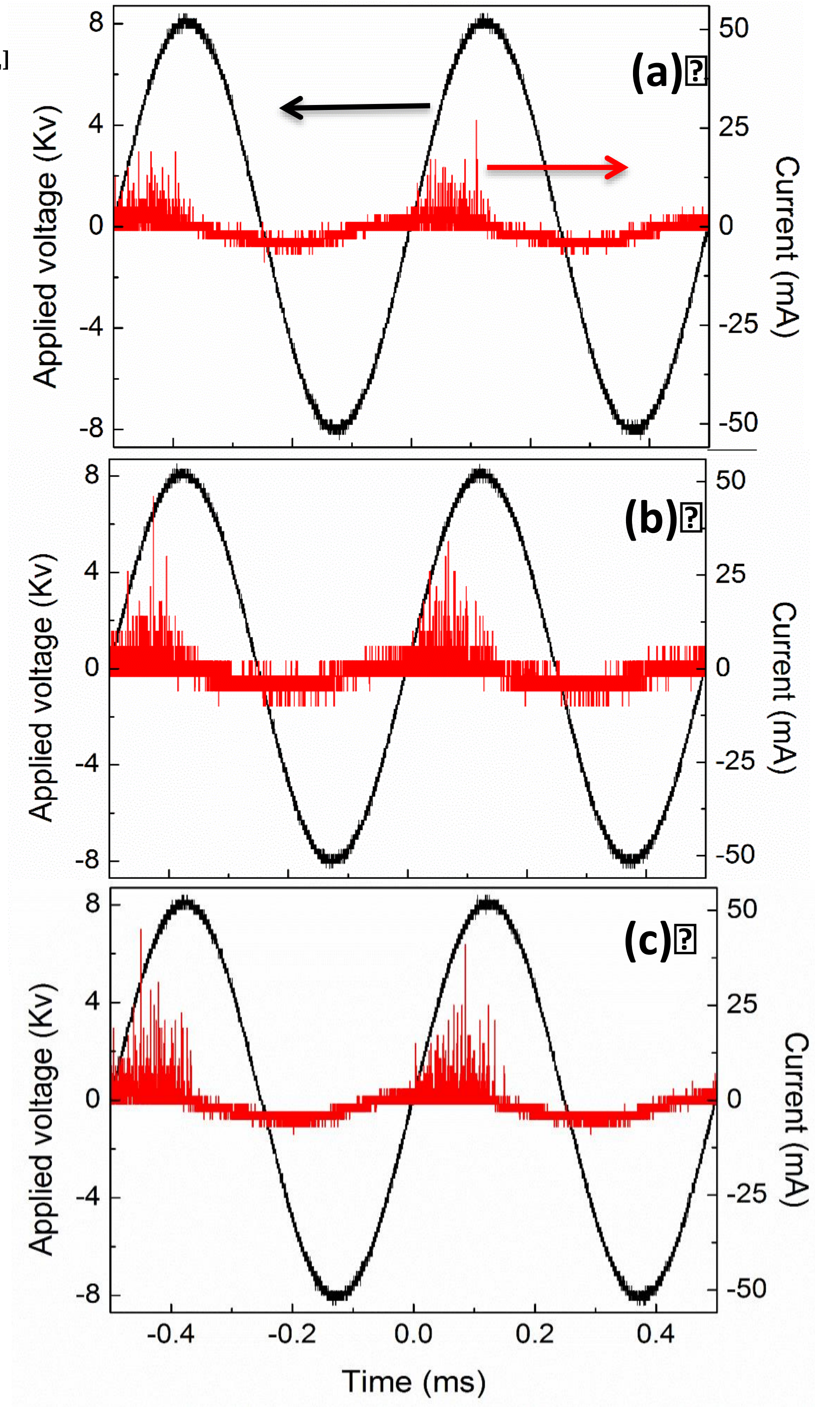

Figure 3: *Current–voltage waveforms of the SDBD at an applied peak-to-peak voltage of 16.8 kV and at a frequency of 2 kHz and ground electrode width (a) W_g=10 mm, (b) W_g=15 mm, and W_g=20 mm.*

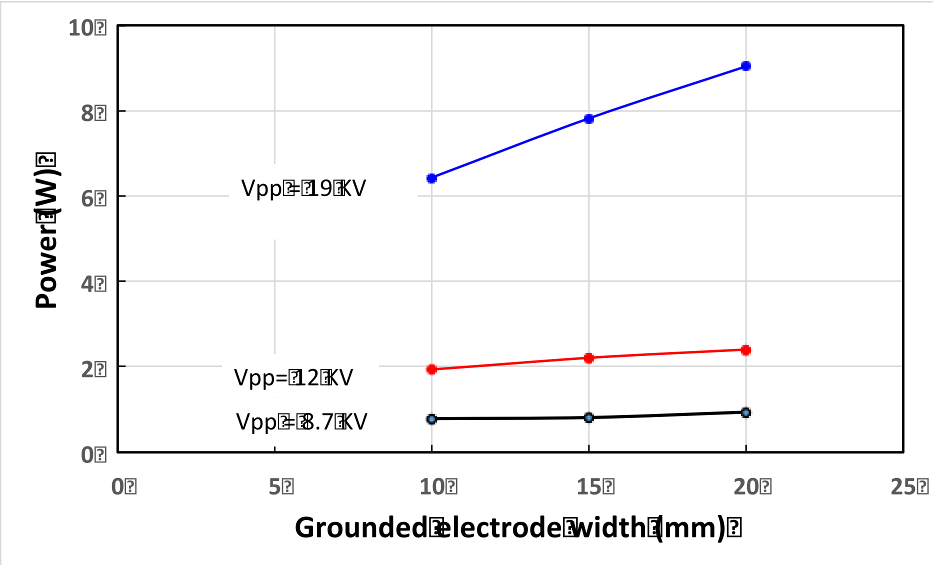

Figure 4*: Consumed power in the SDBD as a function of the ground electrode width at different applied peak-to-peak voltages.*

Figure 5 shows that the number of electrodes has an influence on the discharge current with a significant effect on the capacitive current, which increased remarkably with increasing the number of electrodes. Increasing the number of electrodes leads to increasing the discharge area due to the occurrence of the discharge at each electrode. As a result, the discharge current increases with increasing the number of the electrodes in the reactor. However, the capacitance and, subsequently, the capacitive current increases due to the increase of the dielectric area between the electrodes. This is clear from the Lissajou diagrams at different electrode numbers (figure 6), where the slope $(dV/dQ)_{on}$ of the "discharge on" side increases with increasing the number of electrodes. As a result of increasing the discharge current and the capacitive current, the consumed power increased with increasing the number of electrodes, as shown in figure 7.

Figure 8 shows that the volume of the reactor represented by the tube diameter has no noticeable effect on the consumed power, as the thicknesses of the glass tube was the same.

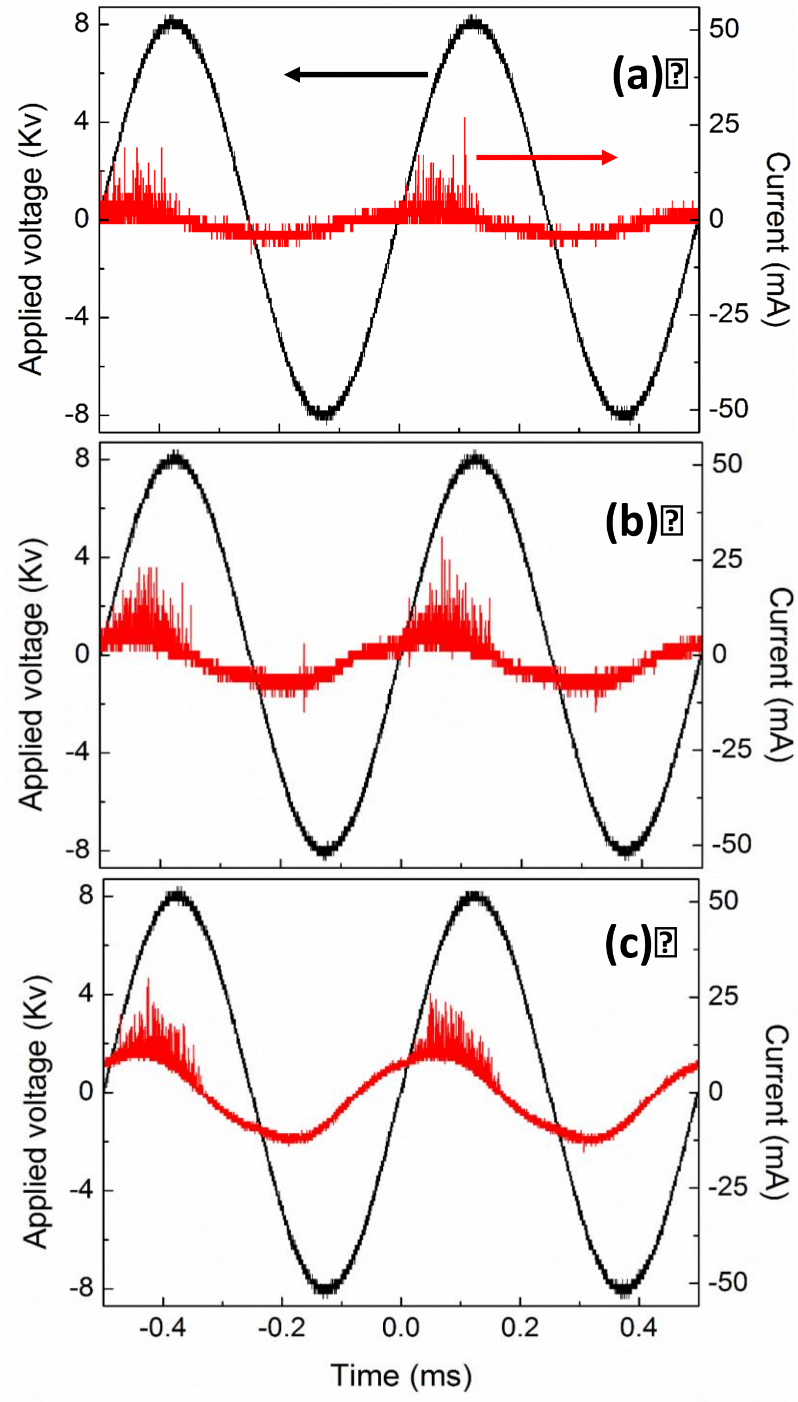

Figure 5: *Current–voltage waveforms of the SDBD at Vpp=16.8 kV, f= 2 kHz,* W_g*=10 mm and number of electrode (a) n=1, (b) n=2, and n=4.*

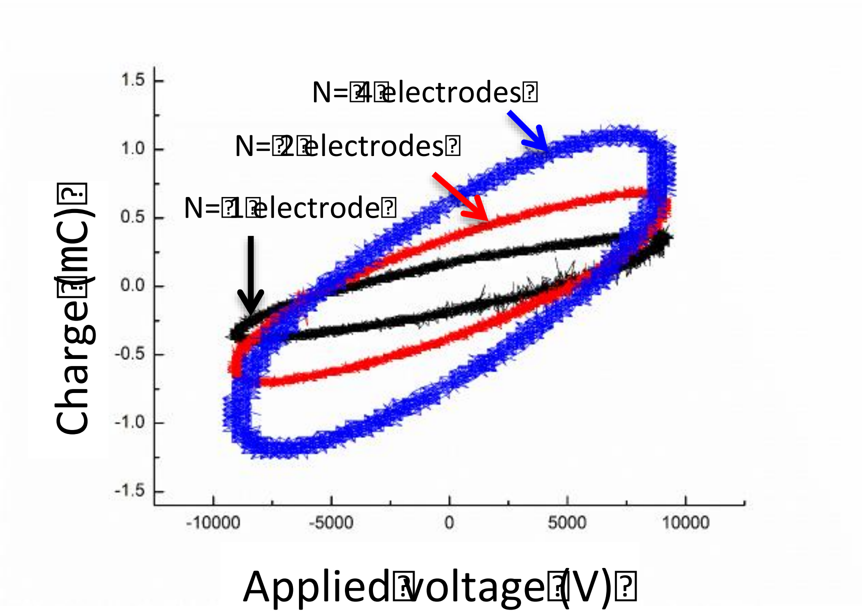

Figure 6: *q-V Lissajous diagram for the SDBD at different number of electrodes (N).*

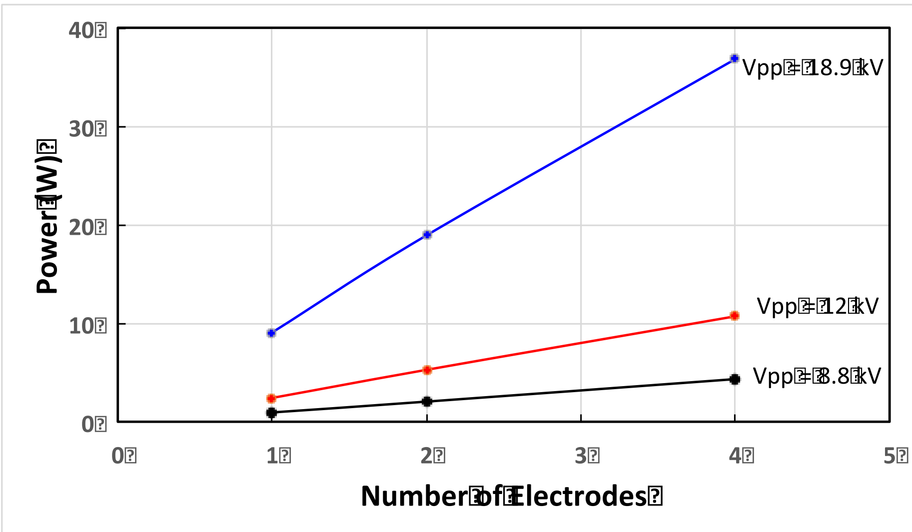

Figure 7: Consumed *power in the SDBD as a function of th number of electrodes at different applied peak-to-peak voltages.*

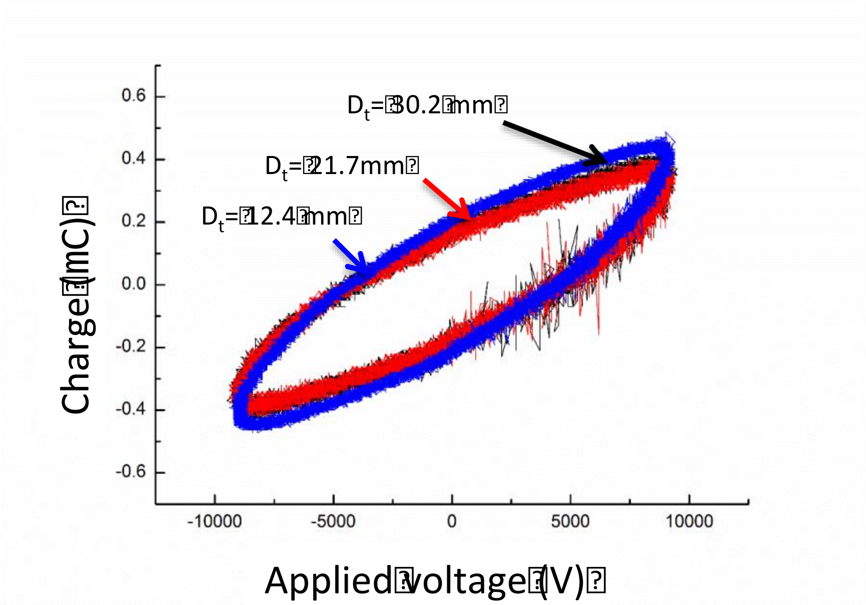

Figure 8: Consumed *power in the SDBD as a function of the tube diameter at different applied peak-to-peak voltages.*

3.2.Ozone production

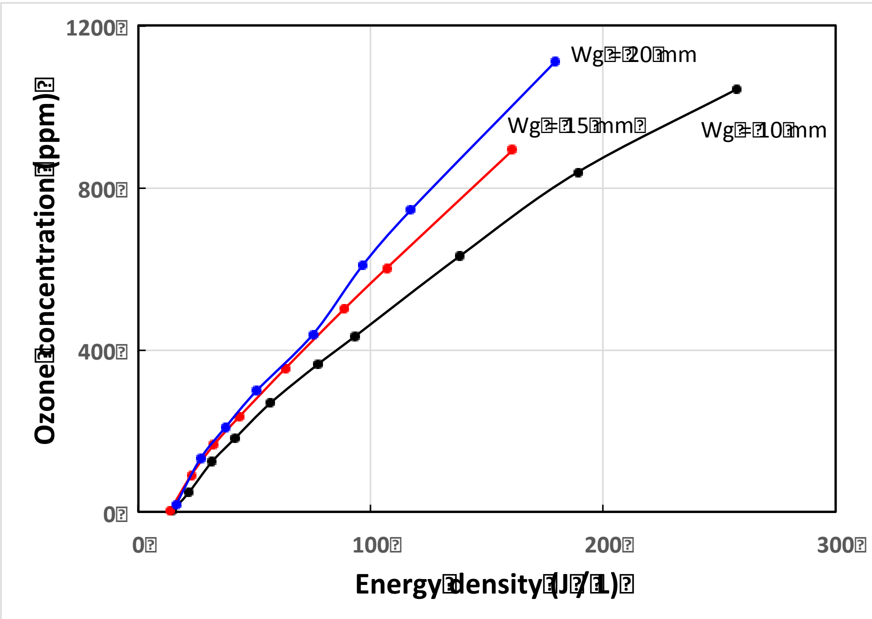

Figure 9: *Ozone concentration as a function of energy density at widths of the ground electrode (W_d=5 mm, W_g=10 mm, D_t=4 cm, f=2 kHz, and Q=1.5 L/min).*

Figure 9 shows the ozone production as a function in the energy density at different values of ground-electrode width. It is observed that the ozone production increases with increasing the energy density for all width values of the ground electrode. Moreover, the ozone production increased with increasing the grounded electrode area at the same energy density. This is because that more plasma area is available at the wider grounded electrode, which resulted in more chemical reactions lead to increasing of the ozone production. On the other hand, figure 10 shows that increasing the number of electrodes leads to a decrease in the ozone concentration at the same energy density value, despite the increase in the discharge area with increasing the number of electrodes. Although the consumed power in the reactor increased with increasing the number of electrodes, figure 10 indicates that this consumed is not utilized efficiently in the ozone production. This is could be due to increases the capacitance with increasing the number of electrodes and, subsequently, increases the power loss in the reactor on the expense of the ozone production.

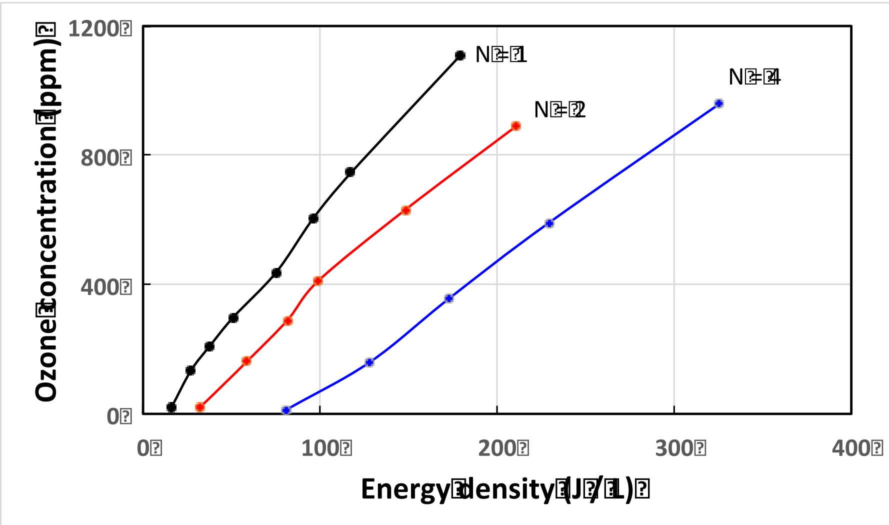

Figure10: *Ozone concentration as a function of energy density at different numbers of electrodes(W_d=5 mm, W_g=10 mm, D_t=4 cm, f=2 kHz, and Q=1.5 L/min).*

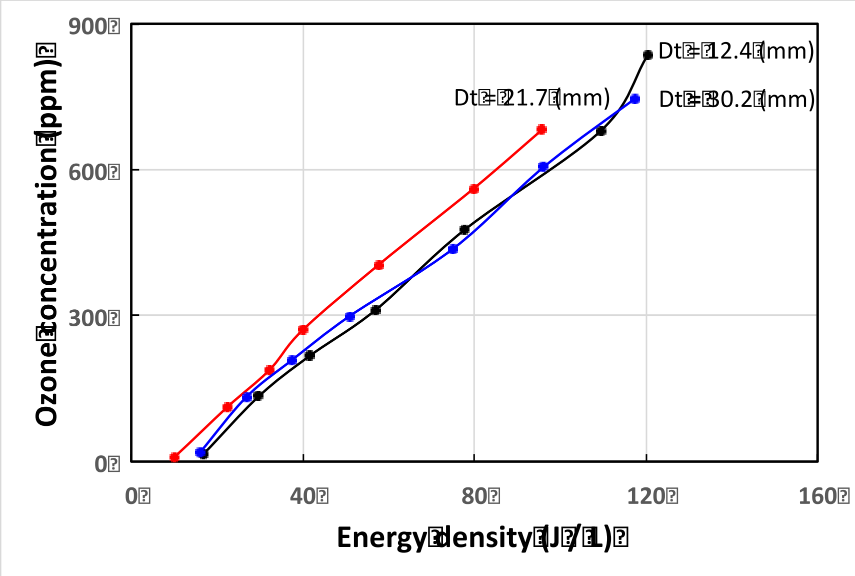

Figure11: *Ozone concentration as a function of energy density at different reactor diameters(W_d=5 mm, W_g=10 mm, f=2 kHz, and Q=1.5 L/min).*

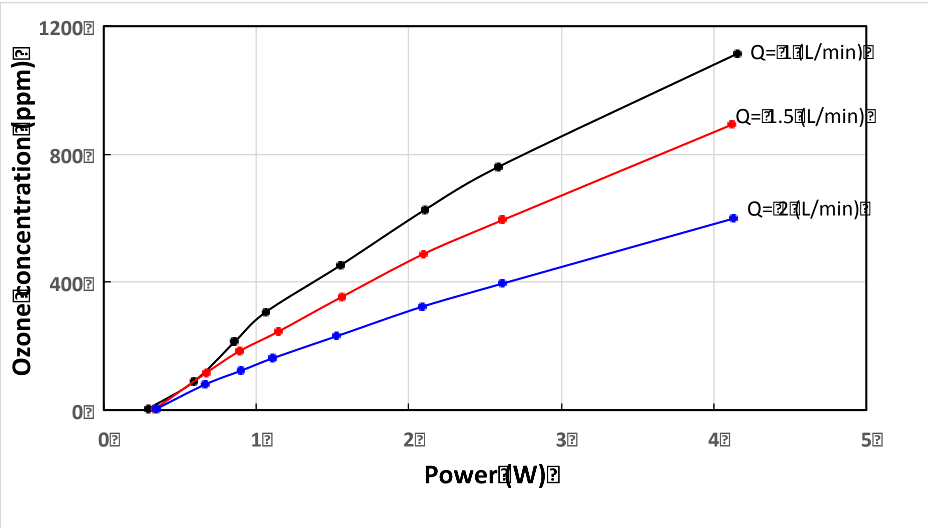

Figure 12: *Ozone concentration as a function of consumed power at different flow rates (W_d=5 mm, W_g=10 mm, D_t=4 cm, f=2 kHz, and Q=1.5 L/min).*

Increasing the volume of the reactor leads to increase of the residence time of the gas inside the reactor. As a result, the ozone increased with increasing the diameter of the reactor from 12.4 mm to 21.7 mm at the same energy density, as shown in figure 11. A further increase in the reactor diameter to 30.2 mm led to decrease in the ozone production at the same energy density. This might be attributed to more gas flow through the reactor with less reaction at the larger - diameter reactor. Thus, the medium size of the reactor is preferable for ozone production.

Decreasing the gas flow rate has no effect on the consumed power in the reactor, but its decrease leads to increase of the residence time of the gas inside the reactor, which, in turn, leads to more reactions between the reactive species together and with the background gas. As a result, the ozone production increases with decreasing the gas flow rate, as shown in figure 12.

3.3.Energy efficiency

The dependence of the energy efficiency of the ozone production on the ground-electrode width as a function of the ozone concentration is depicted in figure 13. As can be observed, the energy efficiency initially increases with the ozone concentration and reaches a maximum value at an ozone concentration about 300 ppm, which correspond to an energy density of approximately of 52 J/L. The energy efficiency started to decrease slightly at relatively high concentrations of ozone (> 500 ppm). The increase of the energy yield with the energy density confirms that the formation process of ozone is dominant and the decomposition process of ozone is almost negligible in this range of energy density. On the other hand, the stability of the energy efficiency after an energy density of 52 J/L indicates that the process of the ozone formation is encountered by the ozone decomposition via the formation of unfavorable species (such as NOx), which result in the decrease in the energy efficiency of the ozone production.

The highest energy efficiency of the proposed reactors 50 g/kWh (corresponding to ozone concentration of 1000 ppm), as achieved with ground electrode of width 20 mm.

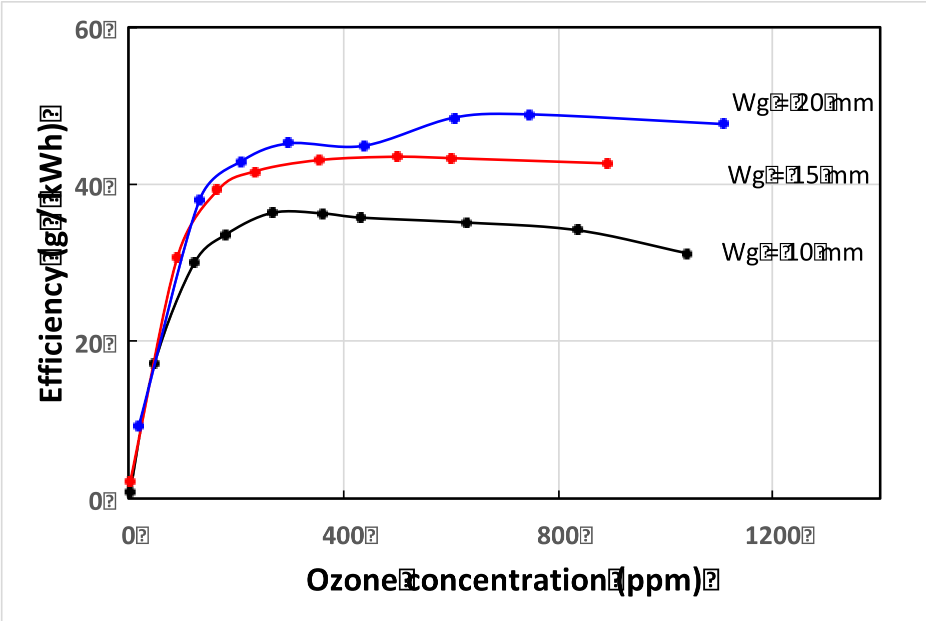

Figure 13: *Energy efficiency of the ozone production as a function of ozone concentration at different ground-electrode width.*

4. CONCLUSIONS.

In this study, a multi-electrode SDBD based reactor is developed and its characterization has been investigated at different reactor configurations. The performance of the developed reactor for the ozone production is evaluated, and the main findings are:

1. The ground-electrode width has a remarkable effect on the ozone production, where its concentration increased with increasing the width. This is due to increasing the discharge current with increasing the ground-electrode width.
2. Increasing the number of electrodes in the reactor has a negative effect on the efficiency of the ozone production due to increasing the power loss in the dielectric material.
3. An optimum size of the reactor was observed at a specific plasma area, despite of the decrease of the ozone production with increasing the gas flow rates.

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تطوير ودراسة خواص مفاعل التفريغ الكهربي السطحي على حاجز عازل لإنتاج الأوزون

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يهدف هذا البحث إلى تطوير مفاعل بلازما ذو كفاءة عالية لإنتاج الأوزون. ولقد تم دراسة الخواص الكهربائية للمفاعل (والتي تشمل تغير الشكل الموجي للتيار مع قيمة الجهد المسلط على المفاعل وكذلك القدرة المستهلكة بالمفاعل) عند تغيرات مختلفة في شكل المفاعل (ممثلاً في عرض القطب الأرضي وعدد الأقطاب في المفاعل وحجم المفاعل) وعند معاملات تشغيل مختلفة (كمعدل تدفق الغاز داخل المفاعل وكثافة الطاقة المستنفذة بالمفاعل). تم تقييم أداء المفاعل لإنتاج الأوزون، ولوحظ أن القدرة المستهلكة تزيد خطياً بزيادة عرض القطب الأرضي بينما لم يكن هناك تأثير ملحوظ لحجم المفاعل ومعدل تدفق الغاز وكذلك وجد أن زيادة مساحة البلازما عن طريق زيادة عرض القطب الأرضي أفضل من زيادتها عن طريق زيادة عدد الأقطاب. ويرجع ذلك إلى قلة قيم الفقد في القدرة المفقودة في المادة العازلة داخل المفاعل. بالإضافة إلى ذلك أمكننا استنباط الحجم المثالي للمفاعل باعتبار أن القيم العالية والمنخفضة لنسبة حجم المفاعل إلى مساحة البلازما وجد أنها غير مؤثرة.