Analysis of Low Frequency on Dielectric Barrier Discharge Plasma Reactor for Ozone Production

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Abstract

The current study investigates the effects of pulse frequency and magnitude of AC voltage applied on a cylindrical dielectric barrier discharge (DBD) plasma reactor with mesh electrodes, operated at atmospheric pressure, for ozone production have been systematically investigated. Results showed that increasing the discharge current and the operational power consumed by the DBD plasma reactor, which were proportional to the square and the cubic polynomials of the operational voltage, respectively, were essential for enhancing the concentration of ozone produced by the DBD plasma reactor. With pulse frequency being in the range of 500 - 900 Hz, the AC voltage of 9, 8 and 7 kV applied to the DBD reactor produced ozone with the concentration in the range of 140 - 180, 110 - 150 and 95 - 110 mg/L, respectively. The effect of pulse frequency is reflected by different frequency dependencies; i.e. in the ranges of 300 - 600, 600 - 900 and 1,000 - 1,200 Hz, which produce ozone with concentrations of 10 - 185, 10 - 220 and 10 - 225 mg/L based on a power consumption of 1.5 - 30, 10 - 80 and 15 - 200 W, respectively. The best performance, in terms of the efficiency, capacity and stability of ozone production as well as the input power efficiency and the specific energy input, was achieved at pulse frequency of 500 Hz. Overall, this study provides insights into the construction of an efficient DBD plasma-based ozone generator with low power input requirements.

Keywords: DBD reactor, Low frequency, Ozone production, Efficiency

Introduction

Cold atmospheric plasma (CAP) has been of interest to scholars and medical practitioners due to its ability to generate a variety of reactive oxygen and nitrogen species to be applied in a wide range of medical applications, while the temperature of CAP can be maintained below 40 °C [1,2]. For instances, CAP has been applied in decontamination of food, such as fruits, vegetables and fish [3], dermatology and skin care [4]. This type of plasma can be generated at normal atmospheric pressure by corona discharge (CD), glow discharge (GD), dielectric barrier discharge (DBD) or surface barrier discharge (SBD). The most important feature of DBD is that non-thermal equilibrium plasma can be obtained more easily than the other methods [5-7]. Moreover, CAP can be generated in a common planar, cylindrical or surface discharge configuration of DBD plasma reactor ranging from small laboratory scales with low power to large manufacturing setups possessing an extremely high power [8,9]. The applied electric field, however, should be abundantly high to

accelerate the electrons and obtain sufficient energy for ionization, dissociation and excitation of a gas source including air, oxygen, nitrogen, helium, argon or their combinations in the plasma formation [10,11].

An applied electric field can generate electric dipoles in dielectric barriers of DBD plasma reactors, which are often composed of soda-lime glass, silica glass, ceramic materials and thin layers of enamel or polymers. DBD operates at approximately 10 MHz, while CAP can be generated using an AC voltage source and plate or cylindrical electrodes covered with a dielectric barrier. This type of CAP is a good plasma technology for producing ozone [12] or breaking down covalent bonds of reactant molecules [13]. An interesting advance has been done [14] by connecting 1 electrode of DBD with grounding water, where it can drive the production of ozone with high concentrations. Therefore, DBD plasma technology has been extensively investigated for potential applications in many areas [15,16], including material processing, energy production, medical treatment, environmental remediation and to modify optical properties of metal oxide semiconductor thin films [17], due to its ability to form highly reactive oxygen and nitrogen species near room temperature with low energy consumption using a simple reactor system at atmospheric pressure [15]. In particular, hybrid catalytic DBD plasma reactors are promising in the energy sector for the production of synthesis gas and hydrocarbons [18]. Recently, the development of DBD plasma may be applied in detection platform of cancer screening techniques [19,20].

The development of DBD plasma-based ozone production systems focuses on optimizing operational factors including electricity efficiency, ozone production stability, power consumption and ozone concentration as well as on applications of the generated ozone [21]. Despite the aforementioned efforts, plasma systems frequently face several obstacles, such as efficiency losses and significantly high power consumption, emphasizing the need for performance enhancement by finding appropriate frequency and voltage of input power, so that the resulting ozone has higher concentrations, while the use of the applied electric power is highly efficient [22,23]. Additionally, the temperature of the gas in the DBD plasma system should be maintained as low as possible to avoid the breakdown of ozone [24]. Therefore, this study is aimed to develop a characterization method suitable for cylindrical DBD plasma-based generators. The objectives are (i) to optimize the power supply pulse frequency along with the operating voltage in the ozone production, and (ii) to provide insights into the constructions and applied electric parameters for an efficient DBD plasma-based ozone generator.

Materials and methods

In this study, the experimental setup consisted of a cylindrical DBD plasma-based ozone reactor and a mesh electrode featuring 10 holes per square inch. An electric, as a power source, applied to the DBD reactor was an AC voltage with frequency in the range between 300 and 1,200 Hz. The reactor system consisted of a dielectric barrier, which is constructed from borosilicate glass (also known as Pyrex) tube: Diameter 4 cm, length 19 cm and thickness 0.2 cm, with an outer electrode component being an aluminum foil; length 19 cm and diameter 4 cm. The inner electrode comprised a mesh with the same length and diameter 2.8 cm. Oxygen, as an input gas, was flowed into the reactor through a Resun LP-20 pump at a flow rate of 10 L/min. A Wilebrock flow meter was used to regulate the flow rate of dry air input.

The high voltage source applied to the reactor was in the range from 0.5 to 13 kV (20 kV AC; Dipo Technology, Indonesia). The high voltage was measured with the SEW PD-28 series HV probe, functioning as a voltage order divider from kilovolts to volts which was then read using a Sanwa CD 772 voltmeter. Moreover, the electric current passing through the circuit was measured using a Kyoritsu 2433-type AC/DC digital clamp device. The diagrammatic representation of DBD plasma-based ozone generator is shown in **Figure 1**. The ozone produced by the DBD plasma reactor was monitored using an ozone monitor (2B Technologies An InDevR Company 106-M Serial 1182M) which can measure its concentrations from 0 to 1.000 mg/L. The concentration of ozone was further estimated the titration method.
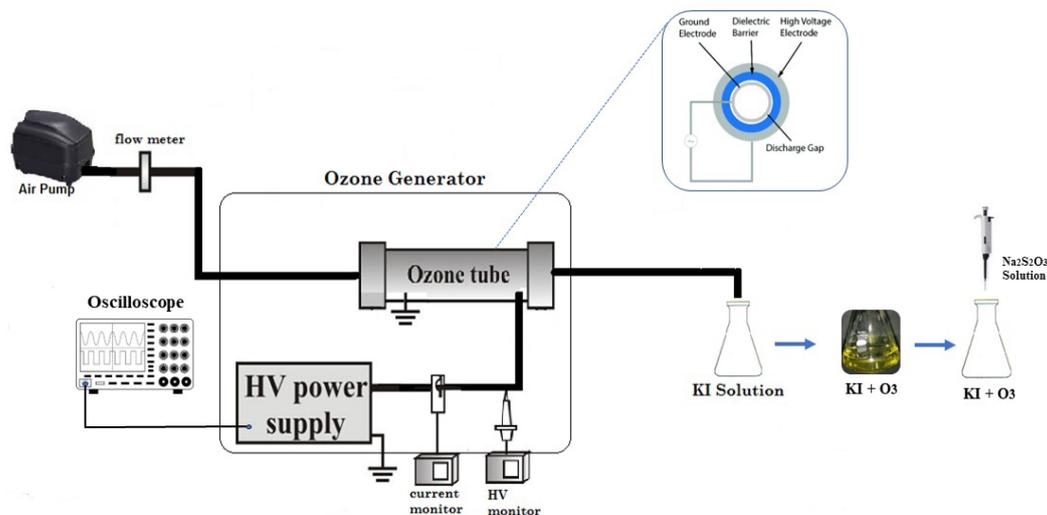
Figure 1 Schematic experimental setup.

Concentration and capacity of ozone

The concentration of ozone produced by the DBD plasma reactor is influenced by several parameters, including voltage, distance between the 2 electrodes, oxygen flow rate and pulse frequency of the AC power supply. In order to take the effect of oxygen flow rate into account, the ozone capacity, $C_{ap,O3}$ (in mg/min), was calculated based on the following equation [2];

$$C_{ap,O3} = C_{O3} \times Q \qquad (1)$$

where C_{O3} represents ozone concentration (in mg/L), and Q denotes the oxygen flow rate (in L/h).

Operational power and specific energy input

Additionally, the operational power (P_{oz}) consumed by the DBD reactor to generate ozone was calculated as;

$$P_{oz} = I_{dis} \times V_{op} \qquad (2)$$

where I_{dis} is the discharge current and V_{op} denotes the operational voltage.

However, the specific energy input (SEI) directly related to the energy consumption and the formation of reactive species in the discharge process was calculated as [25];

$$SEI = P/Q_{O2} \times 3.6 \qquad (3)$$

where P signifies the input electric power (in W), and Q_{O2} represents the air/O_2 flow rate (in L/min).

Results and discussion

Discharge characteristics

Figures 2 and **3** show the average output current and power of DBD plasma as a function of voltage for AC pulse frequencies between 300 and 1,200 Hz at intervals of 100 Hz. Although the electric current was found to increase with the applied voltage, as it has been well documented in literature [6,12], the plots of the average output current as a function of voltage show that there are 2 groups of frequency-dependent *I-V* characteristics. The first group was observed for the frequencies in the range of 300 - 600 Hz and the second group was for the frequencies in the range of 700 - 1,200 Hz. This can be rationalized by considering that the formation of ozone from the reaction of oxygen gas in the DBD plasma reactor exhibits at least 2 groups, related to the pulse frequency-dependent impedance of the electrical circuit of the DBD plasma reactor, as it will be discussed in detail in the next sections.
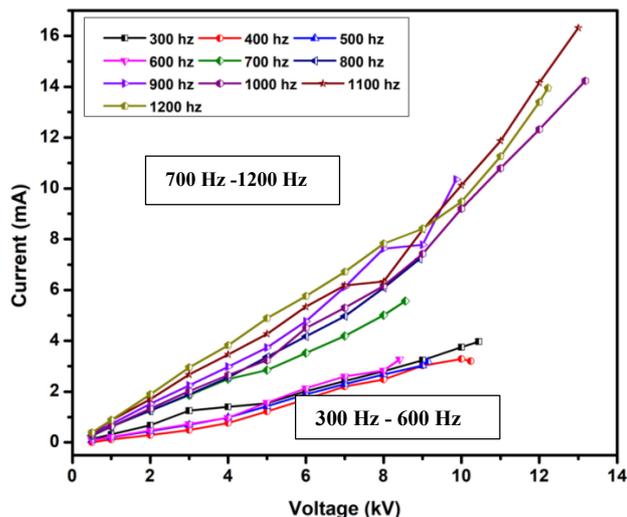
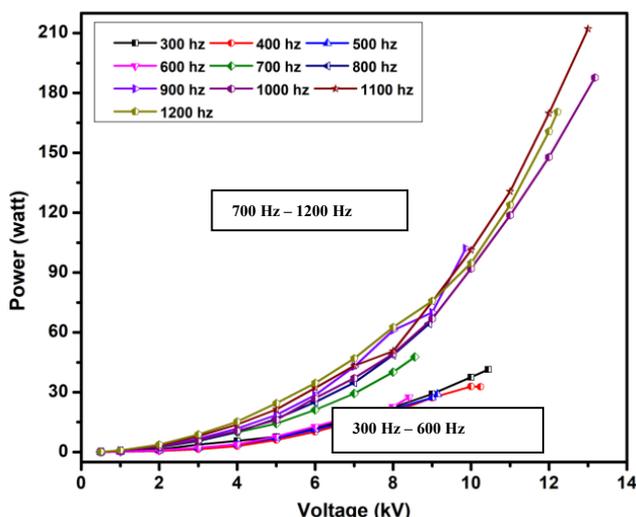**Figure 2** Current as a fuction of voltage for different AC frequencies.

Figure 3 Power as a function of voltage for several frequencies.

Influence of AC frequency on electrical current and power

Figures 4 and 5 show the average output current and power as a function of the pulse frequency for different operating voltages at 7, 8 and 9 kV. For each pulse frequency, the average discharge current is always greater at higher operating voltages. A proportional increase in discharge current with the operating voltage for a certain pulse frequency has reported in literature [12,26]. Investigation on the *I-V* characteristic of a DBD reactor has also been conducted using a pulsed DC voltage [27]. In the current study, at low frequencies in the range of 300 - 600 Hz, the average discharge current was between 2.5 and 3.5 mA, which was nearly unchanged with the pulse frequency. When the pulse frequency was higher than 600 Hz, the average discharge current abruptly and continuously increased until it reached the highest value at 900 Hz, and then tended to saturate at higher pulse frequencies. The value of the discharge current at the pulse frequency of 600 - 900 Hz was in the range from 3.5 to 8 mA.
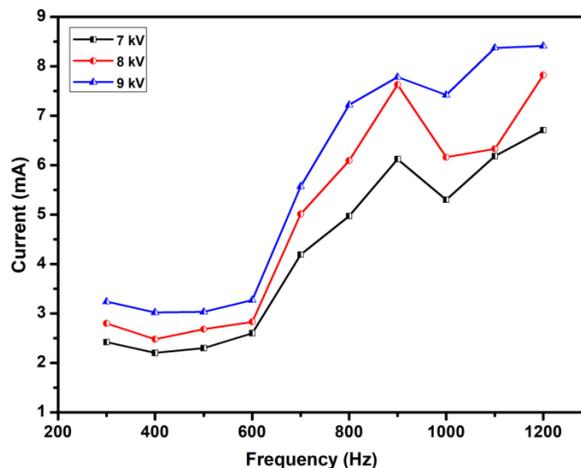
Figure 4 The average output current as a function of AC frequeny for several different operation voltages.

As mentioned above, the output current increases until reaching the maximum value at pulse frequency of 900 Hz, then decreases or tends to saturate. This is caused by the impedance of the electrical circuit $|Z|$ of DBD. The reactor had a minimum value of $|Z|$, equal to circuit resistance and DBD discharge (R). At higher pulse frequencies, $|Z| > R$ due to the contribution of inductance $X_L = 2\pi fL$ (where L is the inductance of the output coil of the step-up transformer as well as other circuits, and f is frequency) and capacitance $X_c = 1/2\pi fC$ (where C is the reactor capacitance). A previous investigation showed a maximum point of the discharge current, known as the resonant frequency point, which was not visible in this present study, but the frequency f_0 of 900 Hz was observed from the existing data. For the $f > f_0$, $|Z|$ increases, and consequently, the discharge current decreases or saturates [23].

The high electric current, when using a higher pulse frequency, can be explained by the exchange of electric dipole directions in the dielectric material utilized as a barrier in the DBD plasma reactor. The change in direction of the electric field due to changes in electric dipoles leads to a greater breakdown voltage in the dielectric material. In DBD, a buildup of charge in the dielectric material causes opposing electric fields that equalize the electric field induced by AC voltage applied during the initial half period as well as prevent the formation of a electric field in the same position at the second half period [8,28]. Therefore, streamers are statistically distributed across the discharge gap, leading to more rapid plasma formation and ionization occurring in the gas in the reactor tube. The capacitive current increases, and this is the reason for the current elevation consistently at higher frequencies.
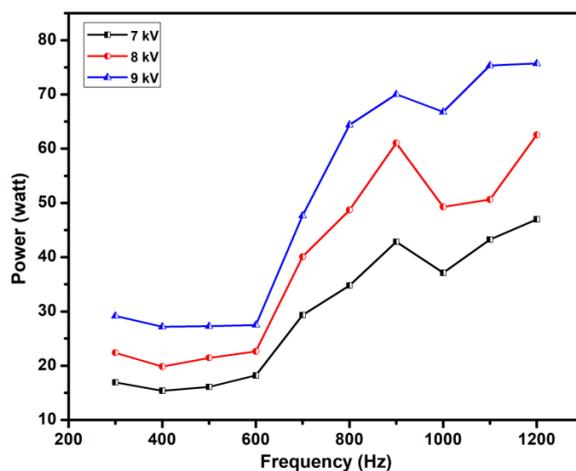

Figure 5 The average output power as a function of AC frequency for several different operation voltages.

Influence of AC pulse frequency on the concentration of ozone

Figure 6 shows the effect of frequency on the ozone concentration produced by the DBD reactor in this study. The data show that the ozone concentration was minimal at a pulse frequency of 400 Hz and reached the maximum at 800 - 900 Hz. This trend reflects a pulse frequency-dependent DBD plasma-based ozone production system. Changes in the frequency of the applied high voltage are assumed to affect the collisions between the oxygen molecules. The collision leads to high molecular separation, thereby increasing the ozone

formation. An increase in the pulse frequency should decrease the total impedance of DBD reactor and enhance the current between electrodes. Therefore, when the total impedance reaches a minimum value, the discharge current achieves a maximum value, with the microdischarge and ozone production attaining the maximum values [23]. As shown in **Figure 6**, for effective pulse frequencies at 500 - 900 Hz, the ozone concentrations produced by the DBD plasma rector are in the range of 140 - 180, 110 - 150 and 95 - 110 mg/L at operational voltages of 9, 8 and 7 kV, respectively.
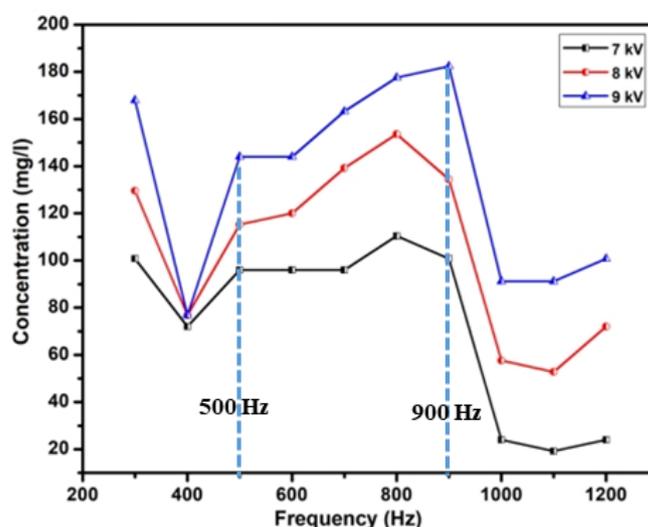

Figure 6 The ozone concentration as a function of pulse frequency for several operation voltages.

Figure 7 demonstrates the similar pattern of pulse frequency-dependent ozone concentrations produced at different applied voltages of 7, 8 and 9 kV. It should be noted that the dissociation energy of a nitrogen molecule is 10 eV, while it is 5.2 eV for oxygen, signifying the need for a specific energy to separate the oxygen molecules for high ozone production. When the energy provided by DBD reactor was much higher than the dissociation energy of the oxygen molecules, nitrogen gas also dissociates, leading to a decrease in the ozone formation. According to **Figure 6**, the AC voltage with

pulse frequency in the range of 500 - 900 Hz was found to generate highest ozone capacity. Similarly, **Figure 7** presents that, for effective pulse frequencies at 500 - 900 Hz, ozone capacities of 88 - 110, 70 - 90 and 58 - 65 g/h can be produced at operational voltages of 9, 8 and 7 kV, respectively. However, at frequencies higher than 1,000 Hz where the concentrations of the O-atom dissociation fragments, ozone and electrons are extremely high, the reactions of these species efficiently reduce the ozone capacity, as presented in Eqs. (R1) - (R4).
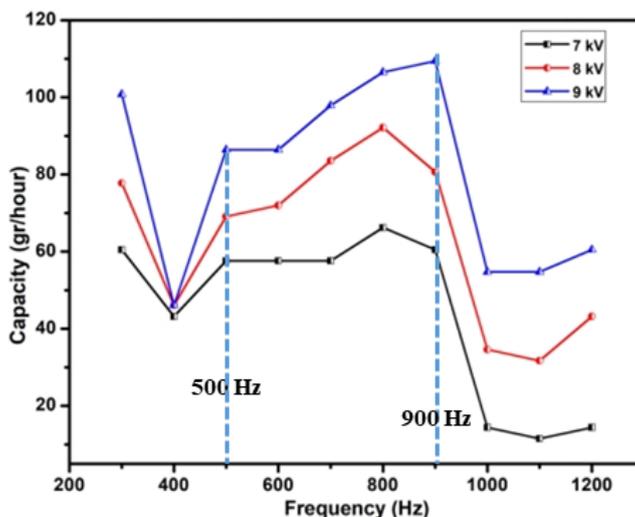
Figure 7 The ozone capacity as a function of pulse frequency for several operation voltages.

Influence of frequency on power and specific energy input

Figure 8 shows the effect of frequency on specific energy input (SEI) in this study. SEI was directly related to the energy consumption and the formation of reactive oxygen and nitrogen species in the discharge process [26,29]. It is clearly observed that SEI remained relatively unchanged at different pulse frequencies in the range of 300 - 600 Hz and abruptly increased at pulse frequencies higher than 600 Hz until reaching the respective maximum values at 900 Hz. In the interval of 600 - 900 Hz, SEI presented a linear upward trend, then decreased and/or fluctuated unstably.
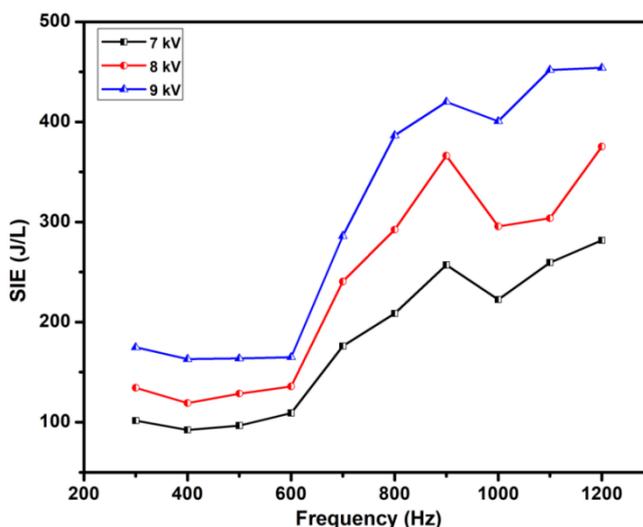

Figure 8 The maximum SEI as a function of frequency for several voltages.

Input power efficiency

Figure 9 presents the logarithmic plots of concentration of ozone produced at different input powers for various pulse frequencies. Interestingly, the plots showed that, except for the frequency at 400 Hz, the concentration of ozone was linearly related to the logarithmic input power provided to the reactor. It is clearly evidenced that there are 3 groups of frequency dependence. Group A (300 - 600 Hz), group B (600 - 900 Hz) and group C (1,000 - 1,200 Hz) produce ozone with concentrations of 10 - 185, 10 - 220 and 10 - 225 mg/L based on a power consumption of 1.5 - 30, 10 - 80 and 15 - 200 W, respectively. In all the groups A, B and C, an increase in ozone concentration was observed after a certain threshold of input power, and the concentration starts from 10 mg/L with a capacity of 6 g/h. The power

threshold for ozone production corresponds to the breakdown voltage to initiate the discharge current. The integrated discharge current during the pulse period increases with increasing voltage which is confirmed from the results of this study, as presented in **Figures 2** and **3**. The existence of a threshold power or voltage

applied to DBD plasma reactor to produce ozone is also consistent with the report by Sung *et al.* [12]. The next aspect of this section is the influence of dielectric materials and the effect of pulse frequency of the applied AC voltage.
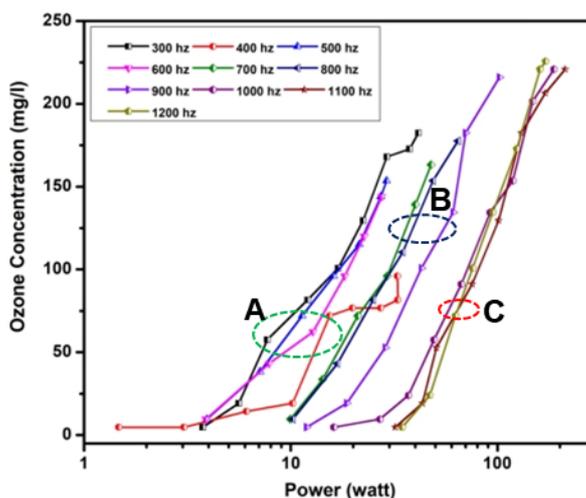

Figure 9 Ozone concentration as a function of power for several frequencies.

As shown in **Figure 9**, 3 groups of pulse frequency dependence are found to provide different ozone concentrations. Dielectric materials are the core components of DBD reactors, and the materials have a significant influence on the ozone formation [29]. This study used quartz, which can be explained through the following aspects. First, application of effective permittivity (ε_t), representing the electrical ability of a dielectric material to store charge. A dielectric material with a higher ε_t has a greater electrical conductivity, leading to a reduction in the reactor impedance and an increase in the electrical conductivity of the discharge current. The discharge current in this dielectric material is a phenomenon where space charge plays an important role. According to research conducted by Wang *et al.* [30], the spatial distribution of charges on dielectric materials is greatly influenced by polarity, pulse frequency and duty cycle of the applied electric voltage. Second, in DBD process, electric charges are accumulated on the dielectric surface. Therefore, at higher voltage pulse frequency, more electrical charge is accumulated, leading to a greater degree of gas dissociation in the plasma formation chamber. The accumulated electric charge can be distributed over a

large area on the surface of the dielectric material, contributing to an increase in electric discharge [29]. Based on **Figure 9**, group C comprising the highest pulse frequency provides the greatest ozone concentration.

The ozone formation is triggered by the free-electron impact dissociation of oxygen molecules [2],

$$e^- + O_2 \rightarrow 2O^* + e^- \tag{R1}$$
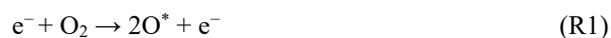
The O-atom dissociation fragments then react with oxygen molecules to form ozone through the following reaction [10]:

$$O^* + O_2 + M \rightarrow O_3 + M \tag{R2}$$

where M is another gas molecule. Simultaneously, O-atom dissociation fragments and electrons can react with ozone to form oxygen molecules [2].

$$O^* + O_3 \rightarrow 2O_2 \tag{R3}$$

$$e^- + O_3 \rightarrow O_2 + O^* + e^- \tag{R4}$$

Based on the reactions in (R1) - (R4), the ozone formation has many influencing factors, including oxygen content, input gas temperature, contaminants in the input gas and power density in the discharge [6].

To determine the best frequency for ozone production, the efficiency of the DBD reactor (η) is to produce ozone is estimated using [24,25],

$$\eta = C_{ap,O3}/P \tag{4}$$

where $C_{ap,O3}$ represents the ozone capacity (in g/h) calculated by multiplying the concentration (in g/L) with the airflow (in L/h), and P denotes the input power

(in W). The plots of η for the ozone concentration produced as a function of the pulse frequency at applied voltages of 7, 8 and 9 kV are shown in **Figure 10**.

These plots revealed that the η values generally decreased with the pulse frequency, hence high efficiency was obtained at 500 Hz. However, under these conditions, the generator was unstable in producing ozone, leading to a reduction in the optimum performance. This finding generally confirmed that the efficiency of ozone production depends strongly on pulse power characteristics such as pulse frequency, polarity and duty cycle of the applied AC voltage [12], in addition to the oxygen flow rate.
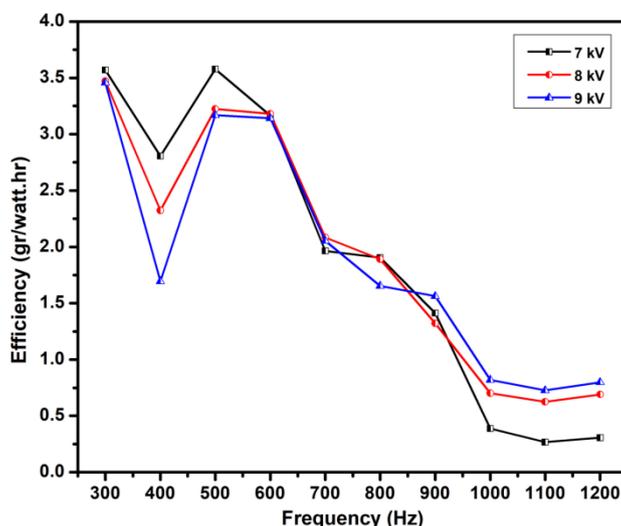

Figure 10 Efficiency of ozone generation as a function of frequency.

Discussion

The graph of discharge current as a function of voltage in **Figures 2** and **3** was correspond to the *I-V* characteristics of DBD plasma. These have been well described by the modified Robinson equation [7,31]:

$$I_S = \frac{2\,\mu_{RT}\varepsilon_t\,S}{d^3}(V - V_i)^2 \tag{5}$$

where I_s is the saturation current (mA), μ_{RT} denotes the mobility of electric charge carriers, ε_t represents the effective permittivity, S is the surface area of the passive electrode (cm^2), d signifies the distance between the electrodes (cm), V is the operating voltage and V_i signifies the corona threshold voltage (V).

This modified Robinson equation has been successfully applied to analyze the *I-V* characteristics of

DBD plasma produced in a reactor with a variety of electrode areas and different media between the 2 electrodes [7]. Taking into account Eq. (5), for the I_{dis} would be equal to I_s, the relationship between the operational power consumed by the DBD reactor and operating voltage, which is expressed in Eq. (2), can be rewritten as,

$$P_{oz} \approx V_{op}{}^3 \tag{6}$$

This equation underlines a cubic relationship between the operational power consumed by the DBD reactor and operating voltage (**Figures 2** and **3**).

It is also interesting to highlight that, as shown in **Figures 6** and **7**, the concentration and capacity of ozone produced by DBD plasma reactor depend on the voltage pulse frequency, where they reach high values when

pulse frequency of the AC voltage was in the range of 500 - 900 Hz and decreased at frequencies higher than 900 Hz. It can be rationalized that the high pulse frequency is associated with high specific energy input, allowing energetic electrons to dissociate nitrogen gas in the dry air. In this regard, energetic electrons at higher energies are capable to dissociate nitrogen molecule, which has a dissociation energy of 10 eV, higher that of oxygen molecule (5.2 eV). Therefore, a DBD plasma reactor operating with high frequency AC voltage and dry air as a gas source can produce reactive oxygen and nitrogen species, which are very useful in controlling microorganisms and in the production of nitrate-based fertilizers [23-25,32-34]. On the other hand, the operating voltage of the DBD reactor also significantly determined the concentration and capacity of ozone, where they are increased with the operating voltage. Considering overall the concentration, capacity and stability of ozone production as well as the input power efficiency, the most prominent pulse frequency of AC voltage applied on the DBD plasma-based ozone reactor is 500 Hz.

Conclusions

In this study, the effects of several operating parameters, including pulse frequency and magnitude of AC voltage applied on dielectric barrier discharge (DBD) plasma-based ozone production have been systematically investigated. The results emphasize that the *I-V* characteristics of the DBD reactor with a cylindrical discharge configuration and mesh electrodes were fitted with the modified Robinson equation. Increasing the applied voltage enhances both current and power within the DBD reactor, which are correlated to the higher concentrations of the generated ozone. The effect of the applied AC voltage with effective pulse frequencies at 500 - 900 Hz is reflected by the concentration of ozone produced by the DBD plasma rector which is in the range of 140 - 180, 110 - 150 and 95 - 110 mg/L at operational voltage of 9, 8 and 7 kV, respectively. On the other hand, the effect of pulse frequency of the applied AC voltage is related to impedance of the electrical circuit of the DBD plasma reactor. Nevertheless, there are 3 groups of frequency dependence, i.e. in the ranges of 300 - 600, 600 - 900 and 1,000 - 1,200 Hz, which produce ozone with the concentrations of 10 -185, 10 - 220 and 10 - 225 mg/L

based on a power consumption of 1.5- 30, 10 - 80 and 15 - 200 W, respectively. The pulse frequency in the range of 500 - 900 Hz was established to be the most effective for stable and efficient ozone production, while higher frequencies led to instability. The best performance, in terms of the efficiency, capacity and stability of ozone production as well as the input power efficiency and the specific energy input, was achieved at pulse frequency of 500 Hz. Overall, this study provides insights into the construction of an efficient DBD plasma-based ozone generator with low power input requirements. The findings suggest that careful adjustment of voltage and pulse frequency can lead to an improved ozone generation, which has potential applications in medical, decontamination of food, environmental remediation and industrial processes. This research emphasizes that, for optimal performance, DBD reactors should operate at well-defined voltage and frequency settings to maximize ozone production while minimizing power consumption.

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