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Analysis of Double Dielectric Barrier Discharge (DDBD) Reactor as Ozone Generator Influence of Pulse Frequency and Duty Cycle

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Abstract:- Double Dielectric Barrier Discharge (DDBD) generator is a reactor that is considered more qualified as an ozone reactor for medical applications because there is space between two barriers that function as a place for the flow of pure oxygen. Research on medical ozone production with Double Dielectric Barrier Discharge (DDBD) technology with variate pulse frequency and duty cycle configuration has been carried out. The purpose of this study is to obtain the optimum dose of ozone for applications in the medical field. The DDBD reactor is made of Pyrex glass length of 19 cm, a outter diameter of 4.23cm and inner diameter of 1.95 cm. Stainless mash are used as the inner and outer electrode materials with a length of 13.5 cm and 5 cm wide. Both variations in the pulse frequency, namely 30 Hz, 60 Hz, 90 Hz and variations in the duty cycle, namely 20%, 40%, 60% used with voltage variations from 200 V to 12 kV and flowrate used is 0.6 L/minute. The results showed that the higher the pulse frequency make the higher current and the higher the voltage reached so that the ozone concentration maximum reached was also higher. The optimum of duty cycle is 60% for mobility, however this mobility is unstable duty cycle. The power required to produce ozone at a frequency of 30 Hz is around, 60 Hz around and 90 Hz around. The maximum ozone concentration is 1416 mg/L in the 90Hz 60% configuration with a voltage of 11.9 kV. The highest efficiency to produce high ozone concentration with low power is in the 30 Hz 60% configuration, which is 708 mg/L ozone concentration with 1.21 Watt of power.

Keywords: *DDBD, ozone, pulse frequency, duty cycle*

1. Introduction

Ozone (O_3) is a gas that naturally occurs in the Earth's atmosphere and has strong oxidizing properties and functions as a disinfectant agent. In the medical field, ozone is used for ozone therapy which utilizes the antimicrobial and wound healing properties of this gas [1]. Ozone production for medical applications requires an efficient and reliable method, one of which is through plasma technology using Double Dielectric Barrier Discharge (DDBD) with a spiral-spiral electrode configuration [2].

DDBD technology has shown significant potential in achieving the ozone concentrations required for medical applications. Double Dielectric Barrier Discharge (DDBD) works by using two electrodes separated by two dielectric layers and a gas (usually oxygen) in between. When a high AC voltage is applied, a strong electric field is generated between the electrodes, triggering the ionization of the gas and producing a non-thermal plasma. Reactions within this plasma produce ozone from molecular oxygen (O_2) [3]. The spiral-spiral electrode configuration in DDBD helps improve ozone production efficiency and ensures even distribution in therapeutic applications. The process involves a plasma reactor that uses pure oxygen as the gas source and variations in AC high voltage between 0.5 to 3 kV. Parameters such as the number of spiral windings and the length of the mesh electrode also affect the concentration and dose of ozone produced [4].

The use of ozone in medical therapy has grown rapidly, especially in cancer applications. Ozone shows great potential in cancer therapy through oxidative mechanisms that can damage cancer cells without damaging normal cells. Studies show that ozone can enhance the effects of chemotherapy by increasing oxygen levels in cancer tissues, which can make cancer cells more susceptible to conventional treatments [5, 6]. In addition, ozone also plays a role in reducing pain and inflammation in cancer patients, and improving their quality of life [3].

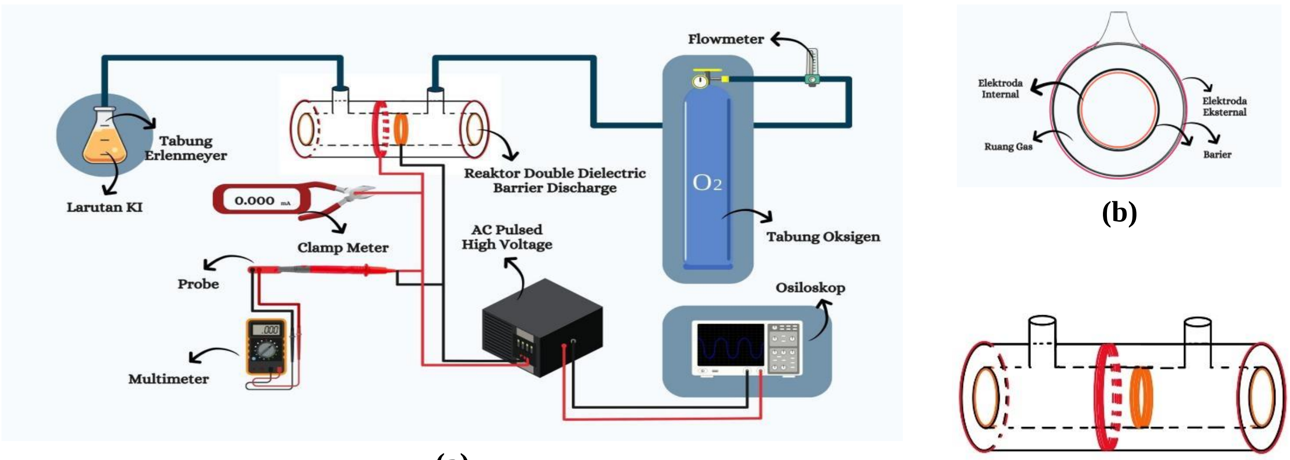
2. Objective

The objective of this research is to obtain a stable medical ozone generator. The stability of the medical ozone generator will be transformed into the utilization of the production scale. The stability of the ozone generator is also needed to ensure the safety and effectiveness of medical applications of ozone produced by the DDBD reactor. To obtain the performance of the DDBD reactor, a variation of the frequency pulse of the operating voltage and duty cycle was carried out. This is important for the further development of medical ozone.

3. Methods

3.1. Materials

This study uses a pulsed AC high-voltage source produced by PT Dipo Technology which can vary the frequency of the voltage pulse. The voltage pulse frequency is adjusted with the help of the GW INSTEK GDS 1102-U oscilloscope. AC high voltage was measured using a SANWA CD 772 digital multimeter connected to the SEW PD-28 1000x high voltage probe. Electric current was measured using Kyoritsu KEW SNAP 2433 amperage pliers. The gas source uses oxigen gas (O2) and the flow rate is regulated using a WIEBROCK flowmeter. The double-barrier reactor (Double Dielectric Barrier Discharge Development of Ozon is made of Pyrex glass with a length of 19 cm and a diameter of 4 .23cm, stainless mash are used as the inner and outer electrode materials with a length of 13.5 cm, respectively (figure 1.c). The outer electrode is 13.5 cm long and 5 cm wide, while the inner electrode is 13.5 cm long and 5 cm wide. Ozone concentration was measured using the titration method, the material used was Potassium Iodide Analysis (KI) EMSURE and the titrant Sodium Thiosulfate. Titration using a GILSON P-10 micropipette and a HERMA Erlenmeyer.



(a) (b) (c)

Fig 1. (a)Experimental Set Up(b) Configuration of DDBD Reactor(c) DDBD Reactor

3.2 Ozone generator preparation

The research procedure is shown in (Figure 1.a). The input gas source is free to air with 0.6 liters per minute (lpm). The AC pulse voltage source is set with the pulse frequency with variations of 30, 60, and 90 Hz. The operating voltage is given from 0 to the maximum voltage, and the amperage is recorded. Ozone production comes out of the output hose. The resulting ozone concentration was measured using the titration method with the equation below. The research procedure is shown in (Figure 1.a). The input gas source is air with a flow rate of 0.6 liters per minute (lpm). The AC pulse voltage source is set with a pulse frequency of 30, 60, and 90 Hz. The operating voltage is given from 100 Volts to 12,000 volts and this depends on the pulse frequency. I-V characterization is done by obtaining the current as a function of voltage for several frequencies. The concentration of ozone produced is measured by the titration method with the equation [7].

$$C_{O_3} = \frac{R\ x\ V_t x\ N_t}{V_{gas}} \tag{1}$$

$$C_{p_{O_3}} = C_{O_3} x\ Q \tag{2}$$

C_{O_3} is the concentration of ozone (mg/l), R is the ratio of analytical moles and reactants in stoichiometric equilibrium, V_t is the volume of titrant (l), N_t is the normality of sodium thiosulfate (mol/l), $V_{gas}$is the volume of air (l), and Q is flowrate of O_2(mg/L). The volume of air can be calculated from the product of the flow rate and the time of ozonation into the KI. The titrant volume is the volume needed to titrate the KI solution which is usually brownish-yellow to clear.

3.3 Robinson equation

In 1971 Choelo performed an analysis of the relationship corona current, voltage, distance between electrodes of an electrode system which generates a field is not symmetric. Under conditions of corona discharge ion flow is one type of ion (unipolar). Unlike the arc discharge conditions can charge flowing positive ions, negative ions and electrons (bipolar). Unipolar ion current flowing to the mobility μ by charge density ρ (r,t) and current density $j = \rho\ v$ without experiencing diffusion in an electric field E (r,t) , changes in the charge density (ρ) along the flow is below [8]:

$$\frac{d\rho}{dt} = \frac{\partial \rho}{\partial t} + \nu.\nabla\rho \tag{3}$$

$$\frac{\partial \rho}{\partial t} + \nabla(\nu\rho) = \frac{\partial \rho}{\partial t} + \nu.\nabla\rho + \rho\nabla.\nu = 0$$

$$\frac{\partial \rho}{\partial t} + \nu.\nabla\rho = \rho\nabla.\nu$$

Substituting equation (4) into equation (3), the equation becomes:

$$\frac{d\rho}{dt} = \rho\nabla.\nu \tag{5}$$

$$\rho\nabla.\nu = \rho\nabla.(-\mu E) = -\mu\rho\nabla.E$$

$$E = \frac{\rho}{\varepsilon_0}$$

$$-\mu\rho\nabla.E = \frac{-\mu\rho^2}{\varepsilon_0}$$

$$\frac{d\rho}{dt} = \frac{-\mu\rho^2}{\varepsilon_0} \tag{6}$$

Solve equation (6) using the integral

$$\int_{\rho_0}^{\rho} \frac{d\rho}{\rho^2} = -\frac{\mu}{\varepsilon_0} \int_{t_0}^{t} t\, dt$$

$$\frac{1}{\rho(t)} - \frac{1}{\rho_0} = \frac{\mu}{\varepsilon_0}(t - t_0)$$

$$\frac{1}{\rho} - \frac{1}{\rho_0} = \frac{\rho - \rho_0}{\rho\rho_0} = \frac{\mu}{\varepsilon_0}(t - t_0) = \mu T$$

$$\frac{\Delta\rho}{\rho} = -\frac{\rho_0 \mu T}{\varepsilon_0} = \frac{\rho_0}{\rho_s}$$

$$\rho_s \equiv \frac{\varepsilon_0}{\mu T} \tag{7}$$

If the ion distance (L) in the Electric field (E) then the flow time can be defined $T = \frac{L}{\mu E}$. It is known that the average flow rate of an ion is $v = \mu E = \mu \frac{V}{L}$ and ρ_s can be changed to $\rho_s = \varepsilon_0 \frac{E}{L}$. The saturation current density in Robinson's equation:

$$J_s = \frac{\mu \varepsilon_0 V^2}{L^3}$$

$$j_s = \rho_s . \mu_s = \frac{\mu \varepsilon_0 E^2}{L} \tag{8}$$

The saturation current in the corona plasma will be:

$$I_s \approx \frac{2\mu\varepsilon_0 V^2}{d} \tag{9}$$

If you calculate the corona voltage limit V_i, the saturation current equation will become:

$$I_s = \frac{2\mu\varepsilon_0}{d}(V - V_i)^2 \tag{10}$$

Where I_s is a saturation current in mA, μ is avarage charge carrier mobility in $cm^2/V.second$, ε_0 is a permittivity, V is operating voltage, and Vi corona threshold voltage in volts. According to our results in DBD plasma, where we find that I-V characteristics are follow Robinson’s formulation [9]. It has to harmonise of Robinson’s formula with our experimental condition where there are three types of dielectric materials between two electrodes. Robinson’s formula must be modified, and the relationship between Is (V) follow [10].

$$I_s = \frac{2\mu_{RT}\varepsilon_t S}{d^3}(V - V_i)^2 \tag{11}$$

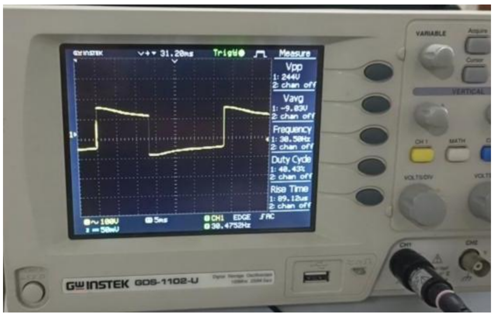
Where S is a surface area of passive electrode in cm^2 and d is the distance between the electrodes in cm. Into the space between the electrodes inserted dielectric materials eg air/gas, liquid and solid, with each of the dielectric permittivity is ε. Futhermore, $\varepsilon_t$vis an effective permittivity for some dielectric materials. By using the formula (11) average mobility μ of charge carriers can be determined.

4. Results

4.1 Pulse Frequency and duty cycle

Frequency can be represented by the number of alternating voltage waves period on the oscilloscope screen, the frequency is directly proportional to the number of alternating voltage waves, shown by (figure 2). Then the duty cycle can be represented by the composition or comparison of the width of the upper alternating voltage wave periode compared to the width of one full wave period ($T_{on}/(T_{on} + T_{off})$). Where T_{on} is transmitter on and T_{off} is transmitter off [11]. Duty cycle 20% means that the system voltage has a composition of 20% transmitter on and 80% transmitter off. At 30 Hz with duty cycle approximately 20% (figure 2.a) there are 6.4 blocks in 1 wave with 1.28 (20% of 6.4) blocks owned by the width of peak wave. Difference with figure 2.b has duty cycle 40% (2.56 of 6.4). The difference of frequency shown in (figure 2.c) has frequency 60 Hz and (figure 2.d) has frequency 90 Hz.

(a)



(b)

(c)

(d)

Fig 2. Measurment of frequency and duty cycle with oscilloscope, (a) f= 30 Hz, DC=20%, (b) f= 30 Hz, DC=40%, (c) f= 60 Hz, DC=60%, (d) f= 90 Hz, DC=20%,

4.2 Current Characterization

Figure 3 is a graph of the electric current formed as a function of voltage at various pulse frequencies (figure 4.a-4.c) and different duty cycle (figure 4.d-4.f). The current characteristics in this study have relatively similar trends at different pulse frequencies and different duty cycles. Changes in frequency affect the shift in ozone production area and maximum operating voltage. Not only that, higher frequencies tend to produce higher currents (figure 4.d-4.f). The higher frequency causes more flameout waves per second so that the maximum output voltage produced also increases. In addition, the addition of voltage can produce a larger potential difference, so the electric field strength generated is also greater. The electric field accelerates the movement of charged particles and collisions occur between particles which then trigger excitation, deexcitation, ionization, and recombination to produce electric charges. Electric charges that move every unit of time form an electric current [12]. As the pulse frequency increases, the maximum voltage required to maintain the discharge also increases, the plasma becomes more continuous, and the energy input needs to be higher to sustain the ionization level. The maximum voltage limit at a frequency of 30 Hz is 2700 volts, at a frequency of 60 Hz is 3800 volts, and at a frequency of 90 Hz is 11900 volts. There are two regimes at a pulse frequency of 90 Hz, namely the $I \sim V^2$ regime (before 4000 volts) and the linear $I \sim V$regime (after 4000 volts).

The difference in Duty Cycle does not really affect the graph trend, but the 40% duty cycle looks different relative to the 20% and 60% duty cycle trends. At 40% duty cycle, it has a spike trend at voltages over 2000V (figure 4.a) and 3300 Volts (figure4.b) which tends to be smoother than the 20% and 60% duty cycle. And at 90 Hz with a duty cycle of 60%, the current measurement when the voltage is more than 4000 Volts shows a less stable value (figure 4.c). Duty cycle that is too high can cause a decrease in system stability and plasma stability because it will cause excessive heat and damage to the dielectric material[11].

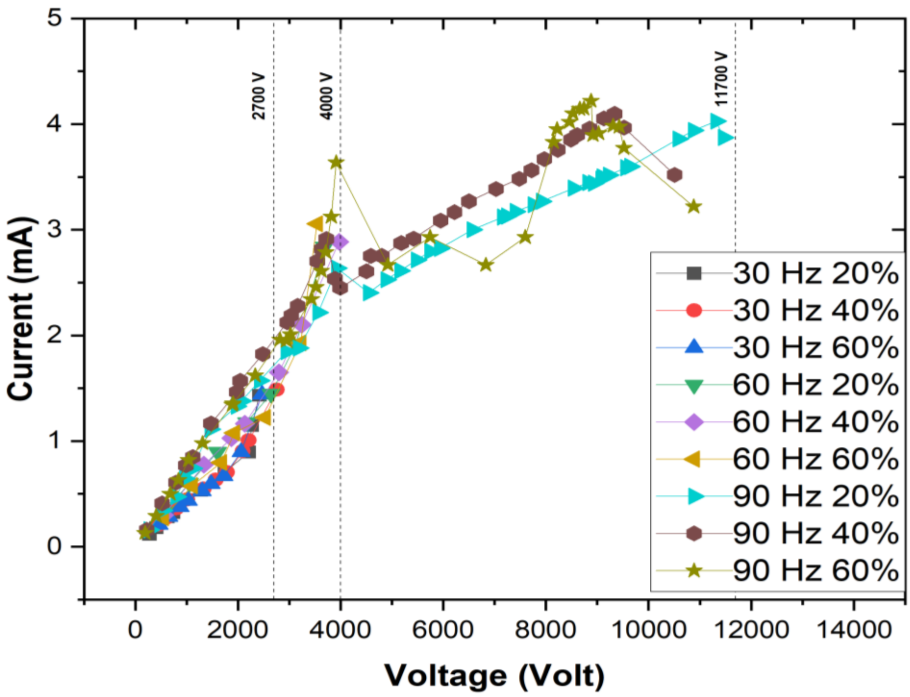
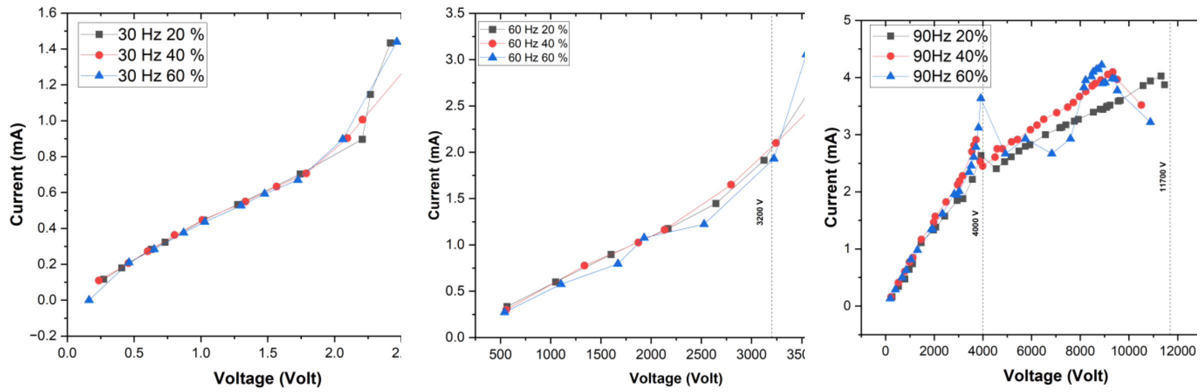
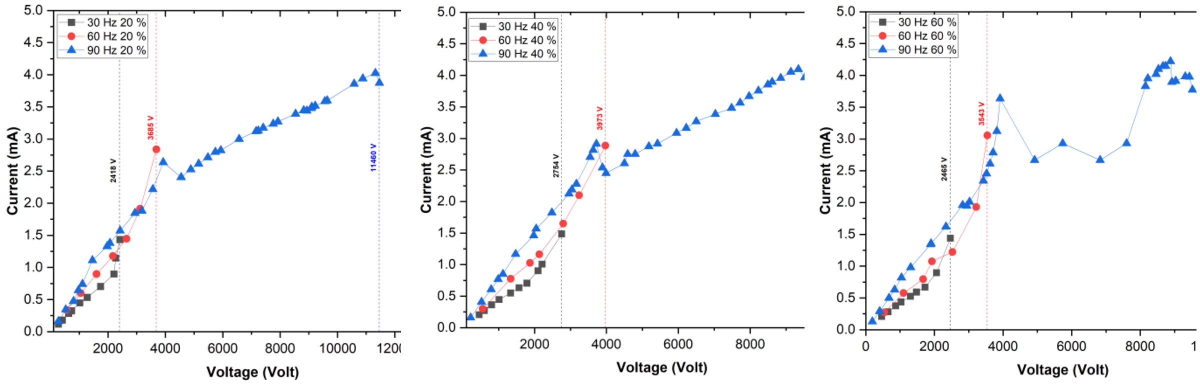


Fig 3. Electrical Current as Voltage Function at all configuration



(a) **(b)** **(c)**



(d) (e) (f)

Fig 4. Electrical Current as voltage function at **(a)** 30 Hz **(b)** 60 Hz **(c)** 90 Hz **(d)** 20% **(e)** 40% **(f)** 60%

3.3 Mobility

Mobility is the ability or degree of freedom of ions to move[13]. In this study the reactor is filled with oxygen gas and will ionize into O^{2-}. In the Robinson equation for ion wind mobility, the current value is directly proportional to the voltage squared. Therefore, to calculate the mobility value, the first step is to find the value of (I/V^2) or, with slope being the fitting value g $(\sqrt{I}/V)$.

$$I_s = \frac{2\,\mu\,\varepsilon_{total} S}{d^3} V^2 \tag{12}$$

$$\mu = \frac{d^3}{2\,\varepsilon_{total}\,S}\,\frac{I}{V^2}$$

$$\mu = \frac{d^3}{2\,\varepsilon_{total}\,S}\,(slope)^2, \qquad \text{slope} = \frac{\sqrt{I}}{V} \tag{13}$$

Relative permittivity is obtained from both types of materials, namely pyrex ($\varepsilon_{r_{pyrex}} = 4.7$) with a thickness of 2 mm for the bottom and 1.6 mm for the upper. and oxygen ($\varepsilon_{r_{oxygen}} = 1$) with a thickness of 19.2 mm arranged in series. So the Total of

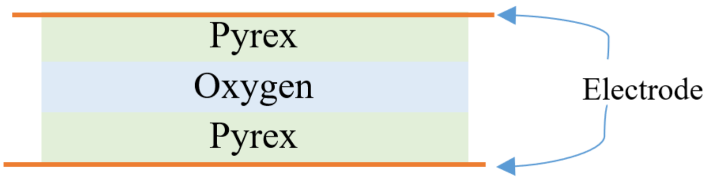
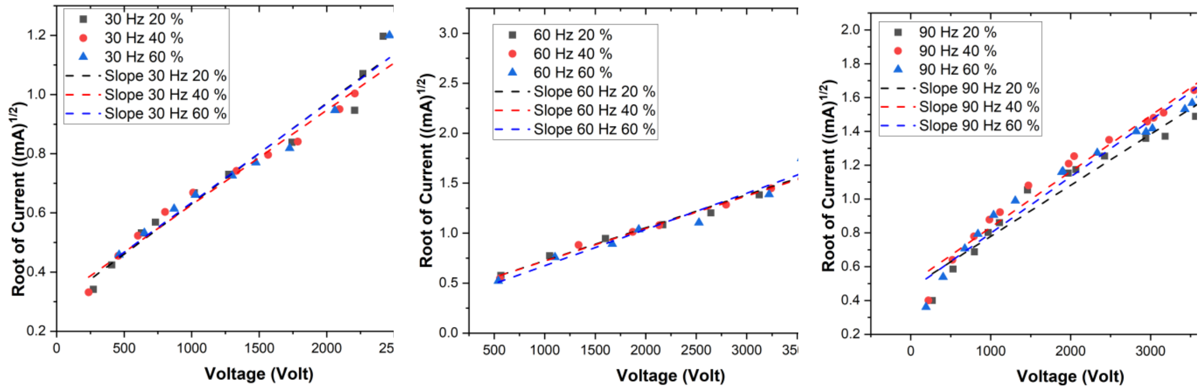


Fig 5. Schema of dielectric in DDBD reactor

$$\varepsilon_{relative} = \left(\frac{d_{pyrex_{up}}}{\varepsilon_{r_{pyrex}}} + \frac{d_{oxigen}}{\varepsilon_{r_{oxygen}}} + \frac{d_{pyrex_{down}}}{\varepsilon_{r_{pyrex}}}\right)^{-1} \tag{14}$$

$$\varepsilon_{total} = \varepsilon_0\,\varepsilon_{relative} \tag{15}$$

where ε_0 is the permittivity of a vacuum which is 8.85 x10-12$(C^2/N.m^2)$, $\varepsilon_{relative}$ is the permittivity relative which is 50.085 and ε_{total} which is 4.43455E-10. The calculation of ion wind mobility uses a slope graph of the root current (√I) as a function of voltage (V). Below is the slope graph as a function of voltage.



(a) **(b)** **(c)**

Fig 6. Root Current Slope (√I) as A Function of Voltage (V) at (a) f=30 Hz (b) f=60 Hz (c) f=90 Hz

Table1. Slope Root of Current as Voltage Function

Frekuensi (Hz)	*Duty Cycle* (%)	Slope (Ampere$^{1/2}$/Volt)	R^2
	20	1,07581 x 10^{-5}	0,9682
30	40	1,01161 x 10^{-5}	0,9858
	60	1,30602 x 10^{-5}	0,893
	20	1,04229 x 10^{-5}	0,9834
60	40	1,02078 x 10^{-5}	0,9918
	60	1,14696 x 10^{-5}	0,9497
	20	9,54692 x 10^{-6}	0,9549
90	40	1,04482 x 10^{-5}	0,9737
	60	1,05525 x 10^{-5}	0,9689

The slope values in table 1 represent the slope values on each graph in figure 6. Table 1 shows that the highest slope value at each frequency value is when the duty cycle is 60%. However, the best R^2 value at each frequency value is when the duty cycle is 40%. This shows that duty cycle values above 50% can ionize more gas, but the level of system stability or R^2 is relatively low because duty cycles above 50% can cause the system to overheat

(). From the slope calculation value and the Robinson equation that has been modified [Nur et al, 2017]. the mobility value will be obtained.

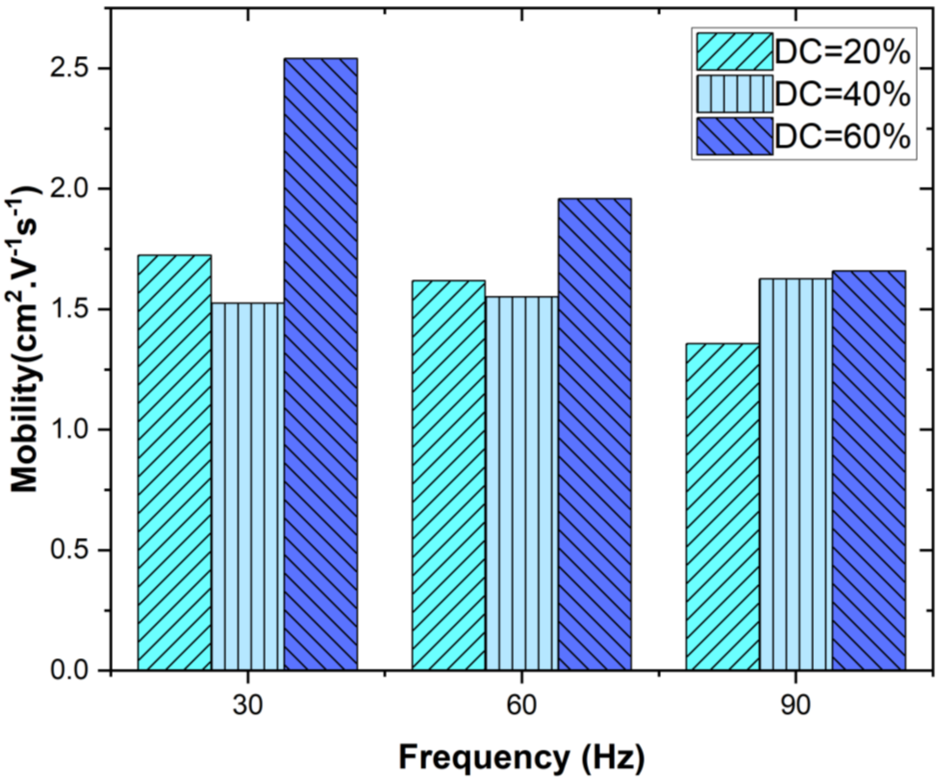


Fig 7. Graph of Mobility

Table 2. Mobility value for all configuration frequency and duty cycle

Frekuensi (Hz)	*Duty Cycle* (%)	Mobility (cm^2/V. s)
	20	1.723554443
30	40	1.523999653
	60	2.540133202
	20	1.617822212
60	40	1.551756024
	60	1.959076969
	20	1.357321142
90	40	1.625685249
	60	1.658321767

The DDBD reactor has a surface (S) which is 0.017947286 m^2, with distance between two electrodes which is 0.0228 m, and ε_{total} which is 4.43455E-10 $(C^2/N.m^2)$. Figure 7 and Table 2 show that the highest mobility value occurs when the frequency is 30 Hz and duty cycle 60% with a mobility value of 2.54013 cm^2/V.s, then the smallest mobility value is when the frequency is 90 Hz and duty cycle 20% with a mobility value of 1.35732 cm^2/V. s. Duty Cycle 60% has the highest mobility value at each frequency value.

4.4 Power and Specific Initial Energi (SIE)

Power is the amount of energy needed every second. Variations in the voltage provided can affect the value of the input power produced as shown in Figure 7. The greater the voltage, the greater the input power and the SIE. This trend applies to all configuration of frequency and duty cycle with flow rate 0.6L/minute. Input power (P) can be determined based on current and voltage measurements or I-V characteristics, besides that it can also be determined by multiplying the voltage (V) by the current (I) using the equation below

$$P = I.V \tag{16}$$

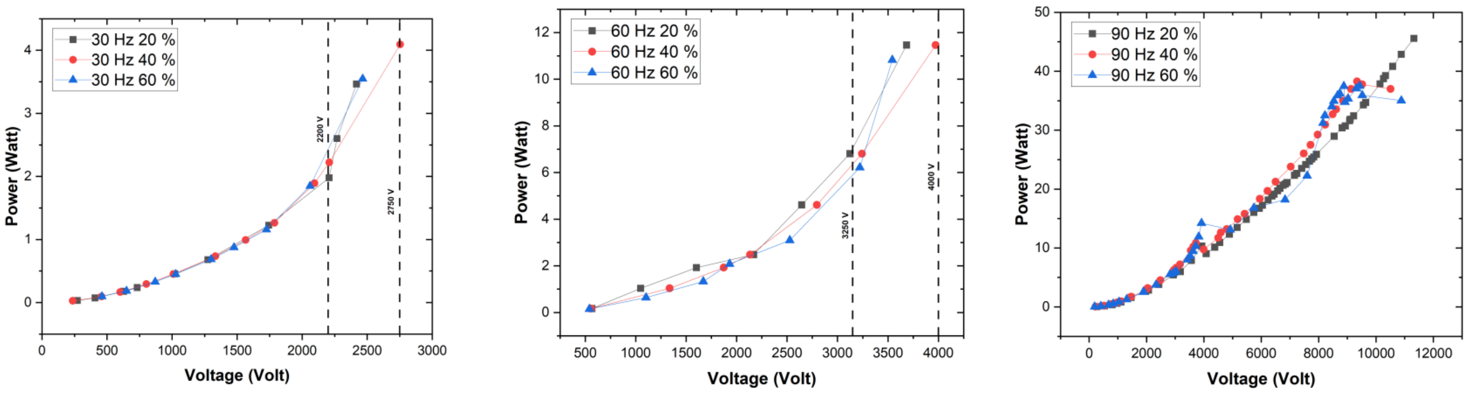
In equation (10) presents that $I \approx V^2$, so

$$P \approx V^3 \tag{17}$$

Specific Input Energy (SIE) of the discharge was calculated by following equation [14]:

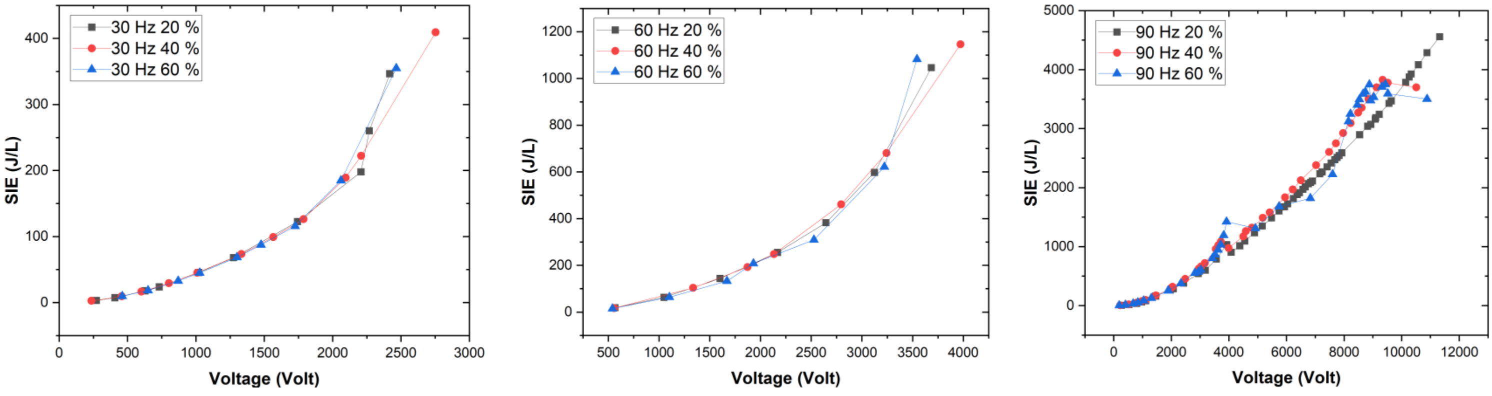
$$SIE = \frac{P}{Q} \tag{18}$$

Where P is the discharge power (Watt), Q is flow rate of oxygen (L/sec) and SIE (JL^{-1}).



(a) **(b)** **(c)**

Fig 7. Input Power Value as a Function of Voltage at frequency **(a)** 30 Hz **(b)** 60 Hz **(c)** 90 Hz



(a) **(b)** **(c)**

Fig 8. SIE Value as a Function of Voltage at frequency **(a)** 30 Hz **(b)** 60 Hz **(c)** 90 Hz

The measured power ranges from 0.02 - 4.09 Watt at a frequency of 30 Hz, 0.014 - 11.47 Watt at a frequency of 60 Hz and 0.02 - 45.58 Watt. This shows that Robinson's formula [11] (equation 10) can be used for double dielectric barrier discharge according to the modification (equation 11) done by Nur et.al.

4.5 Ozon Concentration and Ozon Capacity

Figure 9 below shows the ozone concentration produced by the DDBD reactor as a function of operating voltage for three groups of pulse frequencies 30 Hz, 60 Hz and 90 Hz and three groups of duty cycles 20%, 40% and 60%.From the graph in the picture it is clearly seen that DDBD requires higher operating voltage for higher pulse frequency. At high frequency and high pressure it will be able to produce high concentration as well.

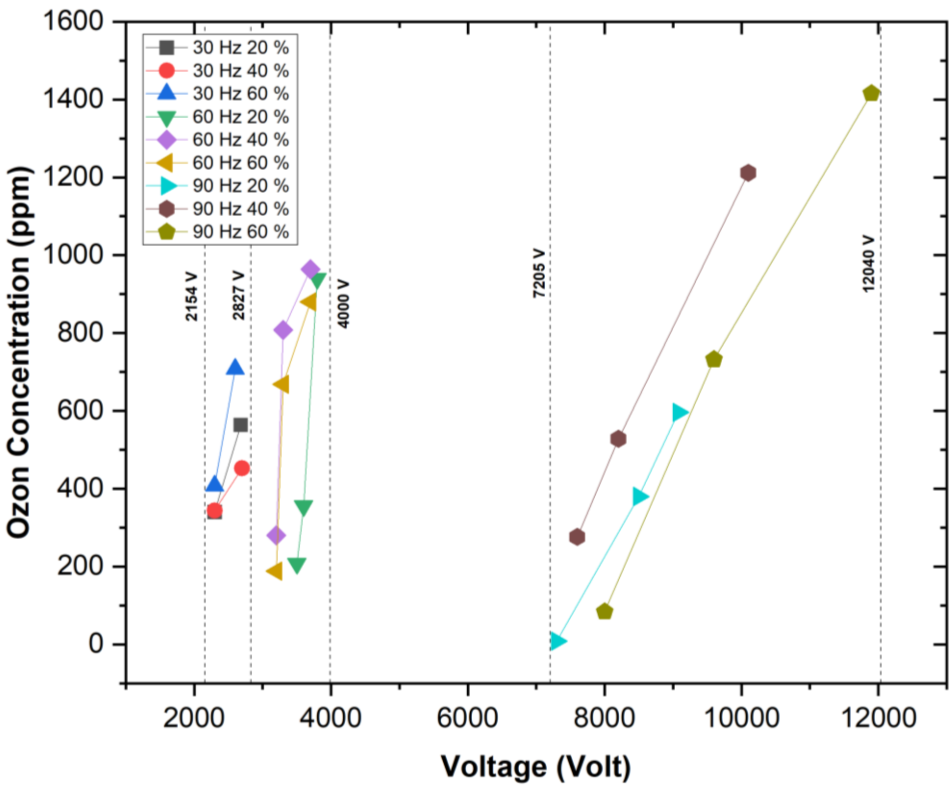


Fig 9. Ozon Concentration as Function of Voltage

Figure 10 shows the dependence of ozone capacity on operating voltage. Like ozone concentration, ozone capacity is also greatly influenced by the pulse frequency of operating voltage. The figure shows that there are three main clusters that are influenced by frequency. The frequency clusters are 30, 60 and 90 Hz. The highest capacity ozone production is obtained for a frequency of 90 Hz and a duty cycle of 60%. The first ozone produced for different frequencies is also different. For a frequency of 30 Hz, 2200 volts and the operating voltage is limited to below 3000 volts. For a pulse frequency of 60 Hz, the operating voltage that produces the first ozone is 3000 volts and cannot be increased beyond 4000 volts.

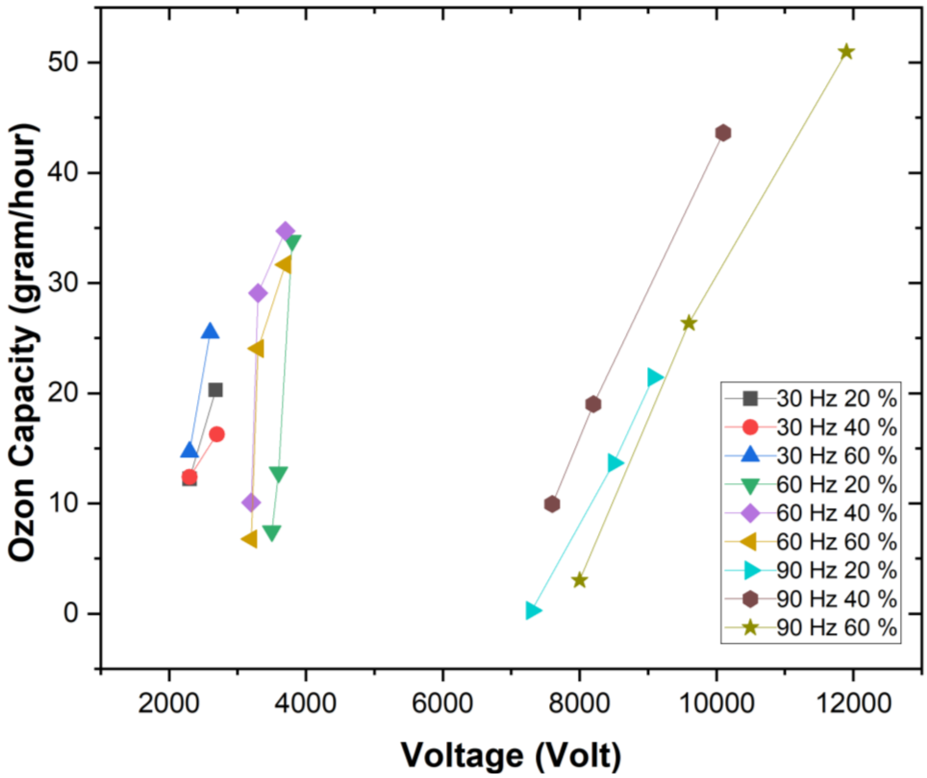


Fig 10. Ozon Capacity as Function of Voltage

The process of ozone formation is when the reactor provides high voltage, the initial electrons will be accelerated and collide with oxygen atoms in the reactor. These collisions result in a doubling of electron folds and produce ions and free radicals. Then the oxygen radicals will react with oxygen to produce ozone (E. 20) with the help of neutral molecules as a catalyst. as follows [15]

$$O_2 + e \rightarrow O + O + e \tag{19}$$

$$O + O_2 + M \rightarrow O_3 + M \tag{20}$$

Each frequency has its own ozone formation zone as seen in figure 10[12]. The ozone formation zone starts from a voltage of 2.3 - 2.7 kV at a frequency of 30 Hz, 3.3 - 3.8 kV at a frequency of 60 Hz and 7.3 -11.7 kV at a frequency of 90 Hz. The higher the voltage at a fixed frequency configuration, the more intense the color change in KI which can be seen in figure 9. The more intense the KI color represents the more ozone concentration that comes out [16, 12].

5. Conclusions

This research has succeeded in characterizing the development of a ozone generator using DDBD technology. Ozone production is influenced by pulse frequency, operational voltage, and duty cycle. This research obtained the highest pulse frequency make the highest current and the highest voltage can be reached. The highest voltage make produce the highest ozone concetration, so the highest pulse frequency can be reached the highest ozone concentracion. The pulse frequency make zone of ozone concentration as a function of voltage. The highest duty cylce make the highest mobility, but electrical system was unstable at duty cycle 60%. The highest efficiency velue at frequency 30 Hz.

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