Development of ozone reactor for medicine base on Dielectric Barrier Discharge (DBD) plasma

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Abstract. The development of the reactor for a lower concentration of ozone need to be done. The type of reactor can be used for the application of medical ozone therapy. Medical ozone therapy can be produced by using double DBD plasma reactor with oxygen as source and variation of voltage. The influence of flow rate to ozone capacity was investigated using double DBD plasma reactor with cylinder-cylinder configuration. The high AC voltage was applied in the range of 0-2000 Volts and the frequency of 50 Hz. The pure oxygen inserted into the reactor with several variations in flow rate, i.e. 2 to 10 L/min. The results showed that the ozone concentration decreased with increasing gas flow rate. I-V characteristic shows that reactor has two regime current as a function of voltage. This characteristic is different with single dielectric DBD Plasma reactor. The suitable dose of ozone therapy for medical other than useful in numerous pathological conditions could be a powerful therapeutic resource to prevent the damages of aging and to improve many functions in human bodies without deleterious effects.

1. Introduction

Ozone (O₃) is a gas naturally present in the earth’s atmosphere, has a strong smell, strong oxidizing and disinfecting agent [1,2]. The ozone molecule is very reactive and has very short lifetime. It will be going back into oxygen in 20-30 minutes [3]. Ozone also has a unique biological ability that is so much studied for use in the medical world [4]. It promotes the treatment of diabetes mellitus by reducing blood sugar levels and increasing oxygen supply into tissues [5,6]. Christopher Friedrich Schoonbein first used ozone to treat wounds and infections caused by anaerobic bacteria [5]. Ozone can be delivered by EBOO (Extracorporeal Blood Oxygenation and Ozonation) method for wound healing therapy [7]. Ozone can also be delivered by ozonized oils to the wound [8]. In some countries, including in Cuba, olive oil ozone has been tested to overcome skin diseases [9] but the orthodox medicine has not been widely accepted for its use [1]. So that need further analysis about ozone concentration, ozone capacity, ozonated oil, and ozone therapy.

Ozone can be produced by dielectric barrier discharge (DBD) reactor [10, 11]. DBD is generated by AC high-voltage and cylindrical DBD reactor. A layer of the dielectric barrier as a DBD thermalization barrier [12]. The oxygen gas flows into the gap between two electrodes which are affected by the high-
energy electrons resulting in the dissociation of the oxygen molecule. Then Ozone formed due to the reaction between oxygen radicals with oxygen [13] ozone concentrations increase with increasing of voltage, the length of the reactor and small flow rate [14]. Two reactor configurations experimentally and theoretically analyzed are single dielectric barrier discharge (DBD) and double dielectric barrier discharge (2DBD) [15,16]. Double DBD is used in this study because the decay of discharge is faster so that the discharge voltage is lowered. In double DBD, the external electric field is reduced additionally by the accumulated surface charges on the barrier at the cathode [15] and this reactor is safer because there is no direct contact between the input gas and the active electrode.

To produce medical ozone safely and in proper dosage, the medical ozone generator and the channeling system as the source of oxygen must have a degree of medical purity so as to avoid nitrogen and other impurities because nitrogen can produce NO which is toxic to tissues [17] In this study, using AC HV with the voltage of 0-2000 Volts and the frequency of 50 Hz. The pure oxygen inserted into the reactor with several variations in flow rate, i.e. 2 to 10 L/min to know its effects on ozone concentrations, ozone capacity, and ozone dose. The suitable dose of ozone therapy for medical other than useful in numerous pathological conditions could be a powerful therapeutic resource to prevent the damages of aging and to improve many functions in human bodies without deleterious effects.

2. Experimental Methods

In this research, we used double DBD reactor with cylinder-cylinder geometry to produced ozone (figure 1). The inner electrodes (positive) with length of 12.56 cm (coated to a pyrex tube forming a cylinder) and the outer electrodes (negative) with the length of 25.12 cm (coated to a pyrex tube forming a cylinder). Both are made of copper plate with the width of 12.5 cm. The diameter of pyrex tubes is 2 cm and 4 cm and the distance between pyrex is 0.5 cm. The length pyrex on the outer electrode and inner electrode (double DBD) is 16.5 cm.

Figure 1. Double DBD reactor Scheme [18] (a) the geometry of the electrode looks ahead, (b) the geometry of the electrode side view

Figure 2 shows the experimental set up of this research. Double DBD was generated by AC HV with the voltage of 0-2000 Volts and the frequency of 50 Hz. The pure oxygen inserted into the reactor with several variations in flow rate, i.e. 2 to 10 L/min. The gas flow rate is measured using a flowmeter (WIEBROCK). At both sides of the reactor, there are each two small pipes which act as input and output of gas. Both electrodes are connected to a high voltage AC source that has been coupled with the amperemeter and probe. The HV probe has been connected to the voltmeter so that it can know the value of the applied voltage. And Ozone concentration is measured using Iodometric titration method. Ozone concentration measurements begin with making a solution of KI (kalium iodide) 33 gr with a concentration of 0.2 M into 1 liter of aquadest. Then prepare a solution of Na2S2O3 (sodium thiosulphate) 6.32 gr with a concentration of 0.4 M to 100 ml of aquadest. The ozone has been produced from double DBD reactor was given to Erlenmeyer with 50 ml of KI solution for 2 minutes. Aqueous KI will change color to yellow because of ozone. Then titrated with Na2S2O3 using a micropipet (10-
100 thoroughness) until the solution is clear colored back. The calculation of the concentration of ozone is as follows [19,20]:

\[ O_3 (\text{mg/L}) = \frac{24000V_tN_t}{V_g} \]  

(1)

\( O_3 \) is ozone concentration (mg/L), \( V_t \) is Na2S2O3 volume (ml), \( N_t \) is Normality Na2S2O3 (mol/L), and \( V_g \) is the volume of gas input (L). Volume gas input is the same as the flowrate x time of ozonated KI.

Then use the best ozone concentration to ozonated olive oil during 3 hours. The rate of ozone production was determined by:

\[ N_{Prod} (O_3) = 0.5 V_{Blank} (S_2O_3^{2-})c(S_2O_3^{2-}) \]

(2)

Where \( V_{Blank} (S_2O_3^{2-}) \) is the titrated volume thiosulfate solution in the blank experiment and \( c(S_2O_3^{2-}) \) is the concentration of the thiosulfate solution.

And The amount of ozone consumed by the oils is given by:

\[ N_{consumed} O_3 = 0.5 [V_{Blank} (S_2O_3^{2-}) - V_{ozonation} (S_2O_3^{2-})] c(-S_2O_3^{2-}) \]

(3)

Where \( V_{ozonation} (S_2O_3^{2-}) \) is the volume thiosulfate solution used to titrate the non-reacted ozone from the ozonation of the oils [21].

![Figure 2. Experimental Set up](image)

3. Results and Discussion

3.1 Current-voltage characteristics

In this study, AC High Voltage with the low frequency of 50 Hz has been used. The variation of voltage are 1.6 kV; 1.7 kV; 1.8 kV; 1.9 kV, dan 2.0 kV. Figure 3 shows that the electric current will increase with the increase of voltage. It is caused by the charge accumulated in the reactor. When the double DBD reactor was given by voltage, there was the potential energy between the electrodes. The strong electric fields caused the ion surrounding positive electrodes to collide and trigger the excitation, deexcitation, ionization, and recombination process. Those process made free electrons go through the electrode base on ion charge (negative ion to the positive electrode and positive ion to the negative electrode. Figure 3 also shows that there is the difference between up current and down current at the same voltage. In this case, the currents are not always linear but there is current saturation. It because the current replaces between positive and negative such as AC voltage cycle. This is called the hysteresis
curve. The characteristics of I-V at oxygen with the flowrate of 2L/min, 4L/min, 6L/min, 8L/min, and 10L/min shows the same trend.

**Figure 3.** The characteristic of electric current as a function of voltage

![Figure 3](image1)

**Figure 4.** Graphic of ozone concentration (at the flowrate of 2 L/min, 4 L/min, 6 L/min, 8 L/min, and 10L/min) as the function of Voltage

![Figure 4](image2)

### 3.2 The Effects of voltage to ozone concentration

Figure 4 shows that ozone concentration will increase with the increase of voltage. This research has the same result as Fang, et al 2008. Because of the increase of voltage, the collision between the electrons and ions move occurred. And figure 5 shows the change of KI color at any voltage variation when delivered ozone. The result shows that the increase of voltage can change the color of KI solution from a clear becomes a rich orange color. Its indicates the ozone concentration is higher in orange color. By chemical reaction, when KI solution reacts with ozone (O3) produces KIO3 solution with old orange color. To ensure this, we calculated an ozone concentration using equation (1) to know the ozone concentration in each voltage. The results of ozone concentration calculation have been shown in Figure 4.
3.3 The Effects of Flowrate to Ozone Concentration

The ozone concentration decreases with the increase of flow rate as it shows in figure 6. It is caused by gas residence time decreases with the increase of flow rate. Reaction to the formation of ozone in the discharge is from the O2 dissociation due to collide with electrons (R1), followed by three body reaction (R2)

\[ \text{O}_2 + e \rightarrow 2\text{O} + e \]  
\[ \text{O} + \text{O}_2 + \text{O} \rightarrow \text{O}_3 + \text{O}_2 \]

Where at a constant flow rate, based on the number, three body reaction rates (R2) is much slower compared to the dissociation reaction of electron collision impact (R1). The ozone production gradually suppressed when given flow rate increases because atom O generated by R1 runs out more quickly following the flow rate of the gas that comes out more quickly, without causing the ozone production (R2) so the concentration decreases with the increase of flow rate [22].

3.4 Application for Medical Therapy

Ozone has a unique biological ability that can be used in the medical world [23]. Ozonated oils have been attributed to antibacterial and fungicidal effects with applications pharmaceutical industry [24]. In
some countries, including in Cuba, olive oil ozone has been tested to overcome skin diseases [7] but the orthodox medicine has not been widely accepted for its use [1]. So need a proper dose so that it can be safe for the human body. Based on our experiment, we can produce ozonated oil with the ozone dose of 421 mg/mL in 60 mL olive oil with the ozone concentration of 72 ppm and AC HV is 1,8-1,9 kV. Those are calculated from the equation of 1,2, and 3 that have shown before [19, 20, 21]. Figure 7 shows the Olive oil control and ozonated olive oil during 3 hours. The color of the ozonated oil is more clear rather than control because ozone damage the chemical bond in oil so that form peroxide that is the rule in wound healing therapy [24].

![Figure 7. a. Control olive oil b. Ozonated olive oil during 3 hours](image)

The following is a table results of calculation of ozone dose in oil.

| Sample | Ozone doze [mg] | Ozone doze [mg/mL of oil] |
|--------|----------------|--------------------------|
| SO     | 0              | 0                        |
| SO15   | 1012.5         | 25.5                     |
| SO30   | 2025.0         | 50.6                     |
| SO45   | 3037.5         | 75.9                     |
| SO60   | 4050.0         | 101.3                    |
| SO75   | 5062.5         | 126.6                    |
| SO90   | 6075.0         | 151.9                    |
| SO105  | 7087.5         | 177.2                    |
| SO120  | 8100.0         | 202.5                    |

From the table, Sega, et al, 2010 result ozone dose of 202.5 mg/mL during 120 minutes ozonation and our result, the ozone dose is 421 mg/mL during 3 hours or 180 minutes. The suitable dose of ozone in oil can be applied for wound healing and chronic disease.

4. Conclusion
Double DBD plasma reactor can be generated to produce ozone and result hysteresis curve on characteristics of I-V. The electric current and ozone concentration will increase with the increase of voltage. By AC HV of 1.8 kV-1.9 kV and ozone concentration of 72 ppm, we can produce ozonated olive oil with ozone dose in oil is 421 mg/mL during 3 hours of ozonation. The treatment of ozonated oil for wound healing needs to be done by applying the variation of HV frequencies. The suitable dose of ozone in oil can be applied for wound healing and chronic disease.
5. References

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