New development of double dielectric barrier discharge (DBD) plasma reactor for medical

M Restiwijaya¹*, A R Hendrini¹, B Dayana³, E Yulianto¹, A W Kinandana¹,², F Arianto¹,², E Sasmita¹, M Azam² and M Nur¹,²

¹ Center for Plasma Research, Faculty of Science and Mathematics, Diponegoro University, Semarang
² Physics Department, Faculty of Science and Mathematics, Diponegoro University, Semarang
³ Medical Faculty of Diponegoro University, Semarang

*Corresponding author: resti_afid@yahoo.co.id

Abstract. Effect of gas source and flowrate to ozone concentration and capacity was investigated using double DBD plasma reactor with cylinder-cylinder configuration. The high AC voltage was applied in the range of 200-500 Volts and the frequency of 50 Hz. Gas sources; i.e. free air and pure oxygen; flowed into the reactor with several variations in flow rate, i.e. 2 to 24 L/min. The results showed that the ozone concentration decreased with increasing gas flow rate. Free air sources produced smaller ozone concentration and capacity compared to those produced using a pure oxygen source (O₂). While the ozone capacity increases with increasing gas flow rate. The pure oxygen (O₂) with a flow rate of 24 L/min had produced ozone concentration of 2.2 mg/liter and ozone capacity of 52.8 mg/min. A number of further studies need to be conducted related to the effect of voltage, diameter, and length of the electrode to obtain lower ozone concentration and capacities, for ozone applications in medical therapy. The suitable dose of ozone therapy for medical can be used for wound healing and chronic disease.

1. Introduction
Ozone is a form of allotropic oxygen (O₂) that have unique properties. The ozone molecule is very reactive and relatively unstable with very short lifetime (20-30 minutes) before going back into oxygen [1]. Ozone can be applied to medical therapy. As molecules that have the huge energy (3/2 O₂ + 143 KJ/mol), ozone inactivation of bacteria, viruses, fungi and some species of protozoa, so it can be used as a therapeutic agent for chronic wounds for clinical, as tropical ulcer, ischemic ulcer and diabetic wounds [2]. The use of topical ozonated oil is also used for the process of wound healing [2,3].

In Paol 2005 [4], use a method known as EBOO (Extracorporeal Blood Oxygenation and Ozonation) for therapy of wound healing of skin disease-specific patient PAD (peripheral artery disease). Bocci 2009, update its ozone-oxygen therapy for treatment [5]. The year 2011, Bocci analyze why medical ozone can be beneficial when it is dissolved in the blood or other biological fluids [6].
The utilization of ozone for medical should be in a safe and proper dosage. Excess ozone dose can be dangerous, but the oxidant in the proper concentration can be a drug [5].

Ozone can be generated from a plasma reactor dielectric barrier discharge (DBD) [7]. DBD reactor type is Single and Double DBD [8]. DBD is produced using high-voltage AC and the field of cylindrical electrodes or covered with a layer of dielectric barrier. A layer of dielectric barrier blocking the thermalisation of DBD i.e. extinguishes in short time duration discharges (a few dozen nanoseconds) [9]. In DBD flowed gas to free air or pure oxygen gas that passes through a gap between two electrodes. Under the influence of high energy electrons in the space between the electrodes then occurred the dissociation of oxygen molecules. Reaction to the formation of ozone begins with the formation of oxygen free radicals, oxygen radicals then react with oxygen to produce ozone [10].

Experimental studies of ozone production using DBD has already been done to improve the concentration of ozone, with a variety of conditions including: variations in voltage, polarity, gas flow rate, the width of the slit, dielectric material, arrangement of electrodes and gas who's running [11,7,12,13]. In this study using low voltage and frequency variation source flow and flow rate of the gas input is done to know its effects on ozone concentrations and get low concentrations. Expected with lower ozone concentrations can be utilized further to medical ozone therapy.

2. Methods
The reactor used is double DBD configuration with cylinder-cylinder (Figure 1). Consists of the inner electrode (positive) and the outer electrodes (negative) made of a copper plate, thickness (T) each 0.01 cm, length (L) each of 9 cm and 8.5 cm. Diameter pyrex Tubes ((Dp) and thickness (Tp) respectively is 2.7 cm and 0.63 cm; length (Lp) 15 cm) serves as a barrier is given on the outer electrode and inner electrode (double DBD).

![Figure 1. Double DBD reactor Scheme](image)

The AC voltage was applied in the range of 200-500 Volts and the frequency of 50 Hz. Gas sources; i.e. free air and pure oxygen; flowed into the reactor with several variations in flow rate, i.e. 2 to 24 L/min. The gas flow rate is measured using a flowmeter (WIEBROCK). Ozone concentration is measured using Iodometric titration method.

Measurement of ozone concentration begins by making a solution of KI (kalium iodide) 33 gr with a concentration of 0.2 M into aquadest 1 liter. Then prepare a solution of Na\textsubscript{2}S\textsubscript{2}O\textsubscript{3} (sodium thiosulphate) 6.32 gr with a concentration of 0.4 M to 100 ml of aquadest. Ozone streamed into the tube the Erlenmeyer flask containing 50 ml solution have KI. Ozonation time 2 minutes. Aqueous KI originally nodes because of the capture of ozone will change color to yellow. Then titrated with Na\textsubscript{2}S\textsubscript{2}O\textsubscript{3} using a micropipet (10-100 thoroughness) until the solution is clear coloured back. The calculation of the concentration of ozone is as follows [14,15]:
\[ O_3 \left( \frac{mg}{L} \right) = \frac{24000 \cdot V_t \cdot N_t}{V_g} \]  

(1)

\( O_3 \) is ozone concentration (mg/L), \( V_t \) is Na\(_2\)S\(_2\)O\(_3\) volume (ml), \( N_t \) is Normality Na\(_2\)S\(_2\)O\(_3\) (mol/L), and \( V_g \) is the volume of gas input (L).

3. Results and Discussion

3.1 Influence of resource flow and flow rate against the ozone concentration

The ozone concentration is measured by the variation of the source of the free flow of air and pure oxygen as a function of the flow rate shown in Figure 2. At a constant flow rate, the ozone concentration values on the source of free air flow are smaller compared to a source of pure oxygen flow [16]. This is because pure oxygen flow on the source, there are only molecules of oxygen as constituting. When the air is free, which included most of the reactors is resolved into the molecules of nitrogen (78%), oxygen (21%), argon (0.93%) and carbon dioxide (0.03%). If the high voltage is imposed on the reactor, then the electrons will be accelerated and early undergoes collisions with atoms or molecules of air constituents.

\[ O_2 \text{ molecules have ionization energy and dissociate lower than the molecules of } N_2. \text{ Therefore the molecule } O_2 \text{ more easily ionized and dissociate than the molecules of } N_2. \text{ The ionization energy of the molecule } O_2 \text{ is 12 eV while the ionization energy of molecular } N_2 \text{ is 15 eV [17].} \]

The ozone concentration decreases with growing with increasing flow rate provided (Figure 3). It is influenced by gas residence time decreases with increase in flow rate. Reaction to the formation of ozone in the discharge is from the \( O_2 \) dissociation due to collide with electrons (R1), followed by three body reaction (R2):

\[ O_2 + e \rightarrow 2O + e \]  

(R1)

\[ O + O_2 + O_2 \rightarrow O_3 + O_2 \]  

(R2)

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). When a given flow rate increases, the production of ozone gradually suppressed 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 increasing flow rate [11].
3.2 Influence of resource flow and flow rate against the capacity of the ozone

The capacity of the ozone production is the ability of a vessel expressed in the number of volumes of output per period of time. In this study, the value of the capacity of the ozone is determined by using the equation [18]:

\[
\text{Ozone capacity (g/h)} = \text{ozone concentration (g/L)} \times \text{air flow rate (L/h)}
\]  

Influence of flow rate against the capacity of the ozone with a variation source free air and the pure oxygen is shown in Figure 4. At a constant flow rate, the value of the ozone capacity on the source of the free air is smaller compared to a source of pure oxygen [16]. This is because the ozone concentration is generated on the source of the free air is smaller compared to a source of pure oxygen (Figure 2). The study rated ozone capacity with increased flow rate seems the presence of saturation or the tendency of constant value. Figure 5 shows that in the flow rate 2-6 L/min ozone capacity 48 mg/min and at a flow rate 8-24 L/min ozone capacity 52.8 mg/min.

![Figure 4. Ozone capacity with free air and pure oxygen as a function of flow rates](image)

![Figure 5. Ozone capacity measured with pure oxygen as a function of flow rates](image)

3.3 Application of ozone for medical

| Table 1. Empirically determined-ozone doses and antioxidant capacities of body fluids and tissues (modified from Bocci et al. [21]) |
|---|---|---|---|---|
| Body fluids and tissues | Antioxidant capacity | Total antioxidant status | Ozone concentration (µg/ml) | Gas (O₂-O₃) Volume (ml) | Response |
| Plasma | High | 1,28-1,83 mM | - | - | - |
| Total Blood | Very high | ++++ | 15-80 | 100-225 | Therapeutic |
| Bronchial associated lining fluid | Very low | + | None | - | Toxic |
| Skin | Low | ++ | 5-80 | Flux | Therapeutic |
| Subcutis | Moderate | ++ | 2-20 | 1-100 | Therapeutic |
| Colo-rectal mucosa | Low | ++ | 5-35 | 50-350 | Therapeutic |
| Tooth | Low | 4 | 100 in 10 seconds | Therapeutic |
| Nucleus pulposus | Moderate | ++ | 25-35 | 2-5 | Therapeutic |
| Epidurul space | Low | + | 10-20 | 20 | Therapeutic |
| Cerebro spinal fluid | Very low | + | none | - | Toxic |
| Muscular fluid | Moderate | ++ | 10-30 | 15-30 | Therapeutic |
| Synovial fluid | Moderate | ++ | 10-30 | 1-20 | Therapeutic |
In medicine, the use of ozone therapy is still controversial. Research in the form of clinical trials about the safety of the use of ozone in the treatment of various diseases and conditions that claimed to be treated with ozone apparently have not found [19]. Ozone has been used as a therapeutic agent for chronic wounds for clinical, as tropical ulcer, ischemic ulcers, and wounds of diabetes. The effects of ozone (O\(_3\)) on wound healing can be assumed due to the decrease in bacterial infection, dermal repair skin damage (small protrusions of the dermis into the epidermis layer), or an increase in oxygen levels resulting from exposure to ozone (O\(_3\)) in the wound [2]. Reference experience determines the ozone dose and a capacity of antioxidants in body fluids and tissues can be seen in table 1.

Bocci analyzes why medical ozone can be beneficial when it is dissolved in the blood or other body fluids. They expect that although toxic ozone on respiratory system are logged in inhalation, but can be recognized as useful agents in oxidative stress-related diseases, and join other medical gases that have recently considered important therapeutic [6]. The following is a table of the results of the calculation of the concentration of ozone and the capacity of our research results.

### Table 2. Comparison of ozone concentrations and ozone capacity

| Flowrate (L/min) | Gas source : Free Air | Gas source : Pure Oxygen O\(_2\) |
|-----------------|-----------------------|----------------------------------|
|                 | Ozone Concentration (ppm) | Ozone Capacity (mg/min) | Ozone Concentration (ppm) | Ozone Capacity (mg/min) |
| 2               | 13.44 | 26.88 | 2 | 24 | 48 |
| 4               | 7.68  | 30.72 | 4 | 12 | 48 |
| 6               | 5.12  | 30.72 | 6 | 8  | 48 |
| 8               | 4.32  | 34.56 | 8 | 6.6 | 52.8 |
| 10              | 3.456 | 34.56 | 10 | 5.28 | 52.8 |
| 12              | 2.88  | 34.56 | 12 | 4.4 | 52.8 |
| 14              | 2.469 | 34.56 | 14 | 3.77 | 52.8 |
|                 |       |       | 16 | 3.3 | 52.8 |
|                 |       |       | 18 | 2.933 | 52.8 |
|                 |       |       | 20 | 2.64 | 52.8 |
|                 |       |       | 24 | 2.2 | 52.8 |

The year 2010, Travagli review ozone and ozonated oils on skin diseases. The use of pure oxygen flow of resources for the application of medical ozone is more recommended instead of air, because the raw material air (78% Nitrogen), when used for ozonation of saturated substrate, can lead to production of NO (Nitric Oxide) that are potentially toxic, and a significant reduction of ozonation efficiency (Travagli, 2010).

In table 2 it looks that in pure oxygen flow rate 24 L/min obtained concentrations of 2.2 mg/L of ozone with ozone capacity 52.8 mg/minute. The capacity of the ozone is used to determine the dose of ozone with the equation:

\[
\text{The ozone dose (mg)} = \text{capacity of the ozone (mg/min)} \times \text{time (min)} \tag{3}
\]

The suitable dose of ozone therapy for medical can be used for wound healing and chronic disease. The development of the reactor for a lower concentration of ozone need to be done by applying a voltage variation, diameter and length of the electrode so that it can be used for the application of medical ozone therapy.
4. Conclusions
The concentration of ozone and the ozone reactor capacity Double DBD plasma is examined in terms of the influence of resource flow and flow rate of the gas. At a constant flow rate, the value of the concentration of ozone and the ozone capacity at the source of the free flow of air is smaller compared to the pure sources of oxygen flow. The ozone concentration decreases with growing with increasing flow rate. The development of the reactor for a lower concentration of ozone need to be done by applying a voltage variation, diameter and length of the electrode so that it can be used for the application of medical ozone therapy. The suitable dose of ozone therapy for medical can be used for wound healing and chronic disease.

References
[1] Miyake Y 2002 Generation of Ozonized Water by Surface Discharge (Japan: Musashi Institute of Technology)
[2] Campanati A, De Blasio S, Giuliano A, Ganzetti G, Giuliodori K, Pecora T, Consales V, Minnetti I and Offidani A 2013 Burns 39 (6) 1178
[3] Valacchi G, Zanardi I, Lim Y, Belmonte G, Miracco C, Sticozzi C, Bocci V and Travagli V 2013 Int. J. Pharm. 458 65
[4] Di Paolo N, Bocci V, Salvo D P, Palasciano F, Biagioli M, Meini S, Galli F, Ciari I, Maccari F, Cappelletti F, Di Paolo M and Gaggiotti E 2005 Int. J. Artif. Organs 28 1039
[5] Bocci V and Di Paolo N 2009 Blood Purif 28 373
[6] Bocci V A, Zanardi I and Travagli V 2011 J. Transl. Med. 9 66
[7] Fang Z, Qiu Y, Sun Y, Wang H and Edmund K 2008 J. Electrostat. 66 421
[8] Lopez J L 2008 Dielectric Barrier Discharge, Ozone Generation, and Their Applications (USA: Complex Plasmas Summer Institute New Jersey)
[9] Takaki K, Hatanaka Y, Arima K, Mukaigawa S and Fujiwara T 2009 Vacuum 83 128
[10] Nur M 2011 Plasma Physics and Applications (Semarang: Diponegoro University Publishing Agency, Indonesia)
[11] Sung T L, Teii S, Liu C M, Hsiao R C, Chen P C, Wu Y H, Yang C K, Teii K, Ono S and Ebihara K 2013 Vacuum 90 65
[12] Moon J D and Jung J S 2007 J. Electrostat. 65 660
[13] Sung Y M and Sakoda T 2005 J. Surf. Coat. Technol. 197 148
[14] Masschelein W J 1998 Ozone : Sci. & Eng. 20 489
[15] Rajbarath P 2005 Utilization of Double Dielectric Barrier Discharge (DBD) Plasma Reactor in The Destruction of Escherichia Coli and Bacillus Subtilis Thesis Oklahoma State University
[16] Nur M, Supriati A, Setyaningrum D H, Gunawan Munir M and Sumariyah 2009 Period. Phys. 12 69
[17] Svanberg, S 1990 Atomic and Molecular Spectroscopy (Berlin: Springer-Verlag)
[18] Nur M, Susan A I, Muhlisin Z, Arianto F, Kinanda A W, Sumariyah S, Wibawa P J, Gunawan G and Usman A 2017 Bull. Chem. React. Eng. Catal. 12 24
[19] Sastroasmoro S 2004 Terapi Ozon (Jakarta: HTA Indonesia)
[20] Travagli V, Zanardi I, Valacchi G and Bocci V 2010 Mediat. Inflamm 2010 1
[21] WFOT Scientific Advisory Committee 2015 WFOT’s Review on Evidence Based Ozone Therapy (London: WFOT UK)