Comparison of Ozone Production by Difference DBD Configuration Reactor and Dissolved Ozone in Waterwith Micro-Nano Bubble Technology

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Abstract:-Ozone is a technology that has been widely developed because it has many benefits in various fields. Therefore, the development of ozone generators continues to be carried out to find the most optimum setup. This paper will compare the effectiveness of ozone production from two reactors that have different geometries, namely a long reactor and a large diameter reactor with variations in flowrate and frequency. The ozone generator used is a dielectric barrier discharge (DBD). The increase in voltage applied to the reactor makes the current also greater. Apparently, the use of a small flowrate increases the concentration of ozone produced at each voltage variation. It was found that the large reactor had a better concentration than the long reactor at each variation of flowrate and frequency. Dissolved ozone was also observed, it was found that oxygen gas sources can dissolve ozone better than free gas sources in water.

Keywords:-Dielectric Barrier Discharge, ozone concentration nano-micro bubble, gas flowrate, dissolved ozone.

I. INTRODUCTION

Cold plasma that can operate at atmospheric pressure is widely used in various fields such as agriculture and the environment. This is because it can be generated in the surrounding air and by using a fairly simple tool such as a dielectric barrier discharge (DBD) [1]. In simple terms, two electrodes are given a high voltage and one of the electrodes is blocked by a dielectric material will produce an electric field that ionizes the gas around the electrode so as to produce energetic species such as reactive oxygen species (ROS) which include O2+, O2-, and O₃-[2]. DBD plasma reactor is also known as an effective ozone generator [3], [4].

Ozone is known as a strong oxidant which is widely used in killing bacteria, algae, spores, removing organic compounds, eliminating odors, to purifying and treating water [5],[6],[7]. The utilization of ozone dose used in each application requires a different characterization and shape of the DBD plasma reactor. This is because the use of the method affects the dose of ozone produced [8]. In general, voltage, type of electrode, reactor geometry, gas flow used, reactor configuration and gas source used are things that affect ozone production [9]-[15].

In this paper, we study the performance of two types of DBD reactors with different lengths and diameters. We also analyze the effect of flowrate and frequency variations on the ozone concentration and the capacity of the ozone produced. With micro-nano bubble technique, dissolved ozone in the water has been demonstrate.

II. RESEARCH METHODS

This research is an experimental research which carried out in the laboratory. Figure 1 show the experimental set up of this research. The equipment used includes: two Dielectric Barrier Discharge(DBD) Plasma Reactors with deference of diameters and longs. The DBD plasma reactor used in this study is in the form of a cylinder where the inside is a threaded cylindrical mesh aluminum and the outer electrode is also a cylinder made of aluminum foil.



Fig. 1: Experimental Set Up

Between one electrode to another there is a separator or barrier made of cylindrical pyrex material. The large diameter reactor has a diameter of 5 cm and a length of 30 cm. The long reactor has two steps, while the long reactor has a diameter of 3 cm and a length of each step 20 cm as shown in Figure 2. Both reactors have pyrex glass along the length of the reactor which functions as a barrier. For inside the reactor is flowed by dry air or oxygen gas which are made into plasma and finally it product. Source of High Voltage AC (High Voltage Alternating Current). The high voltage source serves to supply high voltage to the plasma generator (CPR, 0-20 kV).



Fig. 2. (a). Configuration of large DBD reactor (b) Configuration of long DBD reactor

The high voltage pulsed AC applied to the reactor varied from 0 to 10 kV and 9 kV and used a free gas source with variations of 5,10,15,20 L/min. The measurement of ozone concentration was carried out by the titration method and calculated using the equation below [15],

 $C = \frac{24 \, xV_t xMx \, 1000}{Flow xTime}$

Vt is the volume of titrant (sodium thiosulfate) in liters, M is the molar titrant in grams/L and t is the time the ozone is exposed to the KI solution.



Fig. 3: Experimental set up for

This study included three experiments, first experiment was explored on ozone productions under different air flow rate. The second experiment last was concern on the dissolved ozone in the water with air or oxygen as sources The ozone concentration (C_{0_3}) in the air was calculated by iodometry method [17]. The ozone

capacity was calculated by multiplying concentration with the flow rates [13]. The dissolved ozone was measured in the 10 L distilled water and obtained by *Spectroquant*[®] *Move DC* (173635) - *ozone test kit* (Merck KGaA, Germany).

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The micro-bubbles were continuously produced for 70 min with different ozone flow rate. The water temperature was between 25 to 27 °C during the experiments although without temperature controller. Fig. 1 shows a schematic representation of the experimental setup for ozone production and Fig.3 for experiment dissolved ozone in the water.

III. RESULT AND DISCUSION

A. Electrical Analysis of Reactor

This research uses a DBDP (Dielectric Barrier Discharge Plasma) reactor. Ozone in the reactor is generated by providing high AC voltage between 0 kV-10 kV. Figure 3 a and b shows the current characteristics as a function of voltage. Current and voltage characteristics are obtained by providing varying voltages at different flow rates between 5 L/min- 20 L/min. The graph in Figure 3 shows that the greater the applied voltage, the greater the current generated.



Fig.4. (a) Electric Current as voltage function at long DBD reactor with frequency of 400 Hz and variation of flowrates. (b) Electric Current as voltage function at long DBD reactor with flowrate of 20 lpmand variation of frequencies.

This shows that the AC voltage applied to the DBD reactor (Dielectric Barrier Discharge) will increase the amount of electric charge generated in the process of ionizing gas molecules and followed by the formation of plasma. The increase in voltage causes a change in charge because the change in time (capacitive current) generated according to the equation (I = C dV/dt) will be even greater.

This capacitive current is also an alternating current. From the graph, it can also be seen that the uncertainty current follows a second-order polynomial equation according to the formulation trend of modified Robinson [18]. The current in the dielectric barrier discharge plasma reactor is proportional to the square of the fitting voltage. This characteristic is as proposed, that the Robinson equation after being modified can apply more generally [18]. This equation can be used for DBD reactors with more than one dielectric material and takes into account the electrode area.

Ozone is usually formed as a result of a three-body collision reaction that occurs in a small gap between the dielectric surface of the DBD reactor which is fed with air.



Fig.5. (a) Electric Current as voltage function at the big DBD reactor with frequency of 400 Hz and variation of flowrates. (b) Electric Current as voltage function at big DBD reactor with flowrate of 81pm and variation of frequencies.

Usually M represents O, O_2 , N_2 and O_3 in air. Figure 4 shows a graph of the current characterization as a function of voltage in the DBD reactor in large diameter reactors and long reactors with free gas input. The increase in current is directly proportional to the supply of voltage, this applies to both types of reactors. The high voltage applied to the reactor causes the formation of a strong electric field, this strong electric field indicates the movement of electrons that accumulates very much in the electrodes. This movement of electrons increases the probability of the electron colliding with the oxygen molecule, resulting in ionization and dissociation.

B. The Influence of Flowrate on Ozone Concentration

The characterization of the ozone generator was charged using high voltage source of 10 kV. In the process when ozone has been produce, high energy needed to dissociate the Oxygen to produce O atoms. Then, the O atoms hit the O_2 and these O atoms recombined in a three-body reaction to produce ozone [12].

$$e^- + 0_2 \rightarrow 20 + e^- \tag{2}$$

$$0 + 0_2 + 0_2 \rightarrow 0_3 + 0_2$$
 (3)



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The effect of flowrate on ozone concentration is given by Fig.6 adn Fig.7. The maximum ozone concentration was obtained when gas flowrate was 5 L/minute and 400 Hz frequency at 172 ppm and the lowest value when the gas flowerate was 20 L/minute and 700 Hz frequencyat 43.1ppm. The increase in the gas flowrate results lower ozone concentration and on the contrary the decrease in the gas flow rate generate the higher ozone concentration. The graph of the ozone concentration as a function of stress is shown in Figure 6 and 7. The results show that the ozone concentration increases as the voltage increases. The increase in voltage makes the electrons have high energy to ionize and dissociate oxygen molecules into atoms which indirectly increases the possibility of ozone formation reactions.



Fig. 7: (a) Ozone Concentration as function of voltage at big DBD reactor with frequency of 400 Hz and variation of flowrates. (b) Ozone Concentration as function of voltage at big DBD reactor with flow rate of 8 lpmand variation of frequencies.

C. Disolved Ozone in the Water

The concentrations of the dissolved ozone in water were plotted against time as seen in Figure8. Solid lines are the Boltzmann fit results. We found that oxygen source produce better dissolved ozone than free gas. For a given constant gas flow rate, the dissolved ozone increase with aeration times goes by until reaching the equilibrium state. At equalibrium state, oxygen source can obtained 0.1 ppm meanwhile free gas 0.05 pppm. The gas flow rate also affects the size of the bubbles in the water. Higher gas flow rates produce larger bubbles. The larger the bubble size, the smaller the total surface area and bubble density, as a result the gas-liquid mass transfer decreases [19]. While [20] found that the average bubble size radius of the different air

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streams, between 1 L/min and 0.25 L/min, was the same. However, the number of microbubbles is greater at lower gas flow rates [20].

In addition to the input ozone concentration and bubble diameter, the solubility of ozone is also significantly affected by other factors such as temperature, pH, the electrical conductivity of the medium, and the presence of impurities [21].



Fig. 8: Dissolved ozone in the water as function of time for sources air and oxygen at long DBD reactor with frequency of 400 Hz, and 5 lpm

IV. CONCLUSION

The greater the voltage applied to the DBD reactor, the greater the current. At the same time, a small intake gas will produce a large concentration of ozone. This can happen because the contact time of the input gas in the reactor is getting longer so that the formation of ozone is getting better, this applies to both types of reactors. Large reactors have better ozone concentrations than long reactors at all flow and frequency variations. Also, oxygen source can produce better dissolved ozone than free gas in the equilibrium state in water.

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REFERENCES

- [1.] Judée, F., Simon, S., Bailly, C., &Dufour, T. (2018). Plasma-activation of tap water using DBD for agronomy applications: Identification and quantification of long lifetime chemical species and production/consumption mechanisms. Water research, 133, 47-59.
- [2.] Marotta, E., Ceriani, E., Schiorlin, M., Ceretta, C., Paradisi, C., 2012. Comparison of the rates of phenol advanced oxidation in deionized and tap water within a dielectric barrier discharge reactor. Water Res. 46, 6239-6246
- [3.] Fang, Z, Qiu Y, Sun,Y, Wang H, Edmund K 2008 Jurnal of Electrostatics 66.
- [4.] U. Kogelschatz, Dielectric Barrier Discharges: Their History, Discharge Physics, and Industrial Applications, *Plasma Chemistry and Plasma Processing*, vol.23 (2003

- [5.] Franken, L., The Application Of Ozone Technology For Public Health And Industry. 2005, Food Safety and Security at Kansas StateUniversity.
- [6.] Babikov, D., Kendrick, B.K., Walker, R. B., and Pack, R.T, Formation of Ozone: Metastable States And Anomalous Isotope Effect. Journal of Chemical Physics, 2003. 119(5).
- [7.] Agency, U.E.P., Wastewater Technology Fact Sheet Ozone Disinfection, O.O. Water, Editor. 1999, Epa 832-F-99-063: Washington, D.C
- [8.] Mastanaiah, N., Banerjee, P., Johnson, J. A., & Roy, S. (2013). Examining the role of ozone in surface plasma sterilization using dielectric barrier discharge (DBD) plasma. *Plasma Processes and Polymers*, 10(12), 1120-1133.
- [9.] Asgar A 2014 Ozonization Technology for Washing Vegetables, IptekHortikultura 10.
- [10.] Nur M Plasma Physics and Its Applications, Undip Press, Semarang.
- [11.] Boonduang S, Limsuwan S, Kongsri W, Limsuwan P 2011 Effect of Oxygen Pressure and Flow Rate on Electrical Characteristic and Ozone Concentration of a Cylinder-Cylinder DBD Ozone Generator, Procedia Engineering 32
- [12.] Suraidin, Nur M, Gunawan, Susan, AI 2016 Journal of Physics: Conference Series 776.
- [13.] Nur M, Susan AI, Muhlisin Z, Arianto F, Kinandana AW, Nurhasanah I, Sumariyah, Wibawa PJ, Gunawan, Usman A 2017 Bulletin of Chemical Reaction Engineering and Catalysis 12.
- [14.] Nur M, Restiwijaya M, Muhlisin Z, Susan AI, Arianto F, Widyanto SA 2016 Journal of Physics: Conference Series 776.
- [15.] Bourdet NM, Kirkpatrick MJ, Tuvache F, Frochot D, Odic E 2013 The European Physical Journal Applied Physics 57.
- [16.] Nur, M., Yulianto, E., Kinandana, A.W., Restiwijaya, M., and Arianto, F. 2019. Development of DDBD and plasma jet reactors for production reactive species plasma chemistry. IOP Conf. Series: Materials Science and Engineering, 509 (May) 8.

- [17.] Chasanah, U., Yulianto, E., Zain, A, Z., Sasmita, E., Restiwijaya, M., Kinandana, A. W., Arianto, F., and Nur, M. 2019. Evaluation of Titration Method on Determination of Ozone Concentration Produced by Dielectric BarrierDischarge Plasma (DBDP) Technology. Journal of physics: conference series, 1153
- [18.] Muhammad Nur, YovitaAsriAmeliaa, FajarAriantoa, AndiWibowoKinandana, IntanZahar, Ade Ika Susana, and JujurPratamaWibawa, 2017, Dielectric Barrier Discharge Plasma Analysis and Application for Processing Palm oil mill effluent (POME), *Procedia Engineering* 170 pp. 325 – 331
- [19.] Alam, H. S., Redhyka, G. G., Bahrudin, Sugiarto, A. T., Salim, T. I., and Mardhiya, I. R., 2018. Design dan Performance of swirl flow microbubble geenrator. International Journal of Engineering an Technology (UAE), 7(4), 66-60.
- [20.] Haris, S., Qiu, X., Klammler, H., and <ohamed, M. M. A., 2020. The use of micro=nano bubbles in groundwater remediation: A comprehensive review. Groundwater for Sustainable Debelopmet, 11 (March), 100463.
- [21.] Wei, C., Zhang, F., Hu, Y., Feng, C., and Wu, H., 2017. Ozonation in water treatment: the generation, bsic properties of ozone and its prctical application. Revoes in Chemical Engineering, 33 (1). 49-89.