

Ozone Generator by Using Dielectric Barrier Discharge Plasma Technology With Spiral-Cylinder Configuration: Comparison Between Oxygen and Air As Sources

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Abstract

The ozone generator with Dielectric Barrier Discharge Plasma Technology (DBDPT) with Spiral-cylinder configuration has been developed. DBDP Reactor was constructed with spiral wire and cylinder. Plasma condition has been generated by using AC high voltage Ozone has been produced by this technology with oxygen and dry air as sources for ozone generating. In this research we found that the concentration of ozone produced increases with increasing voltage with a time constant. This concentration also increases with increasing time in certain applied voltage. Ozone concentration was higher than the concentration of dissolved ozone in water. Dissolved ozone in water was only 10 % of ozone produced and only 7 % for dry air as source. Oxygen as source of ozone was better than dry air; both for ozone produced and dissolved ozone in water.

Keyword : Ozone, Spiral-cylinder, DBDP, Oxygen, Dry air

Abstrak

Generator ozon dengan Teknologi lucutan plasma berpenghalang dielektrik (DBDPT) dengan konfigurasi Spiral-silinder telah dikembangkan. Reaktor DBDP dibuat dari kawat spiral dan silinder. Kondisi Plasma yang telah dihasilkan dengan menggunakan tegangan tinggi AC. Ozon telah diproduksi oleh teknologi ini dengan oksigen dan udara kering sebagai sumber untuk menghasilkan ozon. Dalam penelitian ini kami menemukan bahwa konsentrasi ozon yang diproduksi meningkat dengan meningkatnya tegangan dengan waktu konstan. Konsentrasi ini juga meningkat dengan meningkatnya waktu pada tegangan tertentu. Konsentrasi ozon lebih tinggi daripada konsentrasi ozon terlarut dalam air. Ozon terlarut dalam air hanya 10% dari ozon diproduksi jika ozon dibangkitkan dari oksigen murni dan hanya 7% untuk kering udara sebagai sumber ozon. Oksigen sebagai sumber ozon lebih baik daripada udara kering; baik untuk ozon diproduksi dan ozon terlarut dalam air.

Kata kunci : Ozon, Spiral-Silinder, DBDP, Oksigen, Udara kering

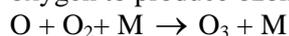
INTRODUCTION

Ozone (O₃) is a strong oxidizing agent and disinfectant that can be applied in various fields such as water treatment, food industries, medicine . In food industries research investigated the efficacy of gaseous ozone for the inactivation bacteria[1,2]. Ozone can be produced in three ways: electrical discharge; arc of ultraviolet [3] and

electrochemical [4]. Sources of ozone for practical applications are typically used in electrical discharge or always called corona discharge [4,5] as well as dielectric barrier discharges [6,7,8,9]. In dielectric barrier discharge (DBD) plasma with oxygen or air as resources, it will cause the electron energy will be transferred in the dominant gas molecules (N₂, O₂, H₂O) through the

collision process. Ion collisions produce primary radical (O^* , N^* , OH^* and others), positive ions, negative ions and excited molecules. One of the primary radical formation, electron-ion, ion-ion reaction and the release of electrons to produce more secondary radical (O_2^* , H_2O etc.)

Then oxygen radicals will react with oxygen to produce ozone.



$M = N_2$ or O_2 .

One of very important application of ozone is wastewater control. There have been many efforts for reducing pollution by wastewater, for example by the use of chlorine, the use of microbiology, activated carbon or membrane filtration. Water pollution degrades the quality of environment and the formation of oxygen in the water, that used for breathing fish and other aquatic animals. To overcome this problem can be done using non-thermal plasma technologies at atmospheric pressure. Ozone has been used for treating a municipal wastewater plant. The very important reasons for using ozone water treatment are it can destroy organic compounds and can kill bacteria. [9,10]. Ozone is a disinfectant that does not leave the rest of the reaction in the water because the ozone will decompose back in to oxygen and also the ability of ozone to dissolve in water thirteen times more easily than oxygen and dissolved ozone in water can be used as standard of water quality [11,12]. Ozone, used as a primary disinfectant, and it leaves no residual. Thus, ozone has been recommended by the World Health Organization (WHO) in the maintenance of drinking water [13]

In this research, the production of ozone and dissolved ozone in the water by using gas oxygen (O_2) pure and dry air as resources gasses. Production of ozone (O_3) has been done by generating non-thermal silent plasma

discharge or dielectric barrier discharge (DBD).

EXPERIMENTAL SETUP

The scheme of ozone generating system by using dielectric barrier discharge (DBD) plasma that was studied can be seen in Figure 1. The plasma reactor has been built with coaxial electrode geometry configuration (spiral-cylinder) [14,15] by adding a barrier layer dielectric (dielectric barrier) by providing pyrex glass and a gap of gas of 0.093 inches. The plasma was generated by a high voltage AC (alternating current) from 3.8 kV to 5 kV with variation time from 1 to 7 minutes. The electrical parameters ozone generator are determined using a high voltage divider. The positive polarity was connected to the positive electrode of copper wire spiral and positive polarity HV probe (HV Probe DC max Voltage DC 40 kV; AC 28 kV code number EC 1010, En G1010), negative polarity was connected to a negative polarity ammeters (Multimeter, Sunwa TRXn 360). Then the positive polarity of ammeters connected to the aluminum foil as negative electrode and the ground probe HV. While the negative polarity HV probe connected to a voltmeter (Multimeter, Sunwa TRXn 360).

After the ozone is formed and then inserted into the tube ozone erlenmeyer in which already contain a chemical solution consisting of KH_2PO_4 , and KNa_2PO_4 0,06 M that serves as a solution of ozone trappers. Ozone concentration has been characterized by UV-VIS spectrometer with a wavelength of 352 nm by obtaining its absorbance. The same method has been used to determine the concentration of dissolved ozone in the water. The concentration of ozone can be determined by using the following formulation:

$$C_{O_3} (mg / L) = 48 \times 10^{-3} \times V_1 \times \frac{M_{O_3}}{V} (mg / L)$$

Where the concentration of ozone in the absorbent solution (mol/liter), V_1 is the volume of absorbent solution of ozone (ml) and V is the sampling air volume (m³).

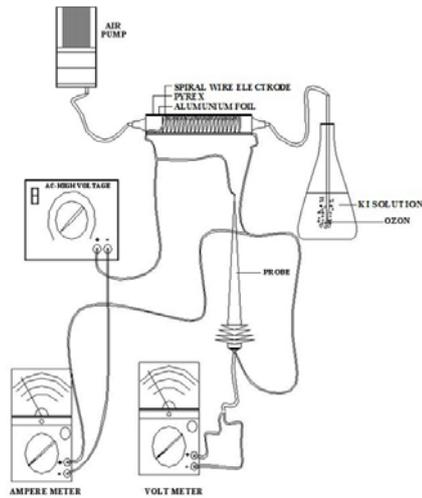


Figure 1. DBD Plasma generating system as ozone generator

RESULTS AND DISCUSSION

A. Influence of Voltage to Ozone Concentration

The influences of voltage to ozone concentration are presented in the figure 2. This figure shows that ozone concentration increases with increasing of voltages. In the range of applied voltage from 3.8 kV to 5 kV with some variation of time, we found that the

increasing of ozone concentration has same trends. We can perceive also in the figure 4 the influence of voltage for the oxygen flow rate of 0.5 liters/min, 1.5 liters/min and 2.5 liters/min. ozone concentration increases. By comparing of Figure 2a, 2b, and 2c, we can conclude that ozone generated decreases with increasing flow rate of oxygen. Figure 3 shows the same inclinations for air as source of ozone. This trend is cause of the higher voltage supplied to the system of dielectric barrier discharge plasma. Increasing of voltage can produce higher electric field between two electrodes. High enough of electric field can accelerate electron, molecules, or ions and its will entrance in the non-elastic collision, and finally augment the number of molecules ionized, radicalized, excited. Consequently, in this condition the ionization, excitation, dissociation process can be continued. The molecules of oxygen are excited as well as radicalized, thus the chemical reactions should be increased, and it produces ozone. In the figure 2a, 2b, and 2c, we found that ozone concentrations increase with increasing production times. This is due to the concentration of ozone increases with increasing the amount of non-elastic collisions with oxygen molecular.

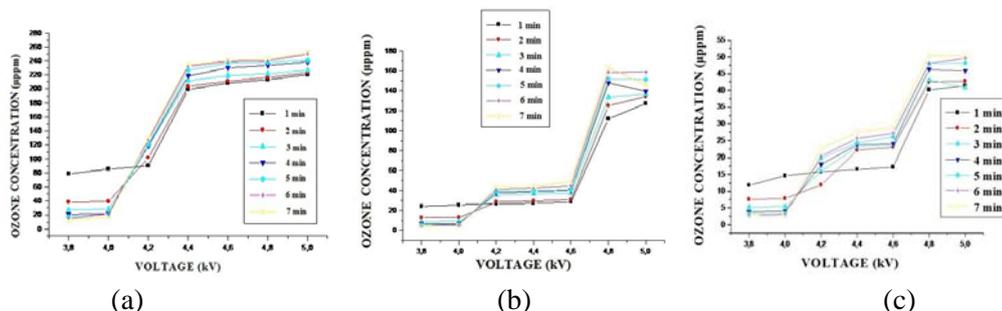


Figure 2: Ozone concentration as function of voltage with certain time of constant with flow rate of oxygen (a). 0.5 L/minutes, (b). 1.5 L/minutes , and (c). 2.5 L/minutes

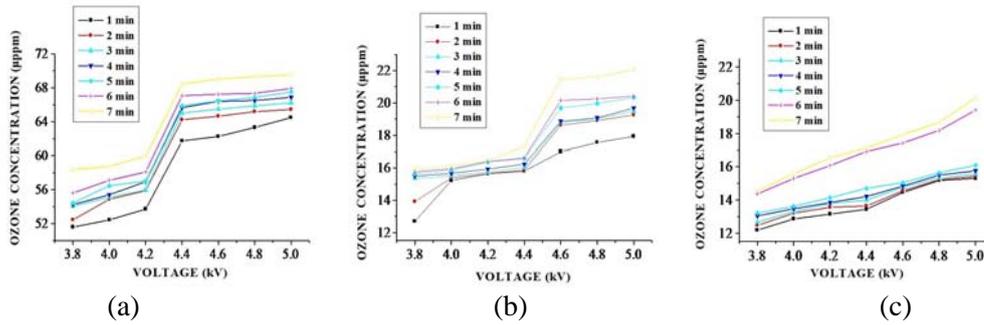


Figure 3: Ozone concentration as function of voltage with certain time of constant with dry air flow rate of (a). 1.5 liters/minutes, (b). 2.5 liters/minutes, and (c). 4.0 liters/minutes

Influence of oxygen or dry air flow rate to the production of ozone is shown in figure 4 and figure 5. The concentration of ozone produced with dry air as source is smaller than oxygen as source. This results is due to the composition of dry air more complex such as Ar, N₂, O₂, He, H₂.

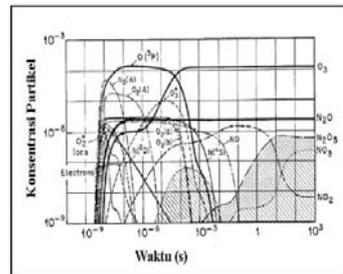


Figure 6: Micro discharge in dry air [16]

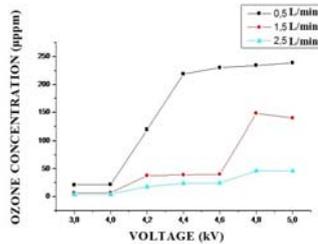


Figure 4. Concentration of ozone as function voltage with time constant of 4 minutes and rate flow of oxygen of 0.5 liters/minute, 1.5 liters/minute, 2.5 liters/minute

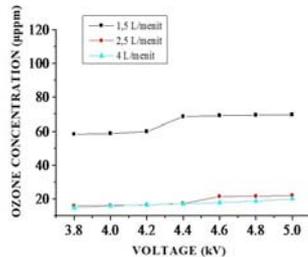


Figure 5. Concentration of ozone as function voltage with time constant of 4 minutes and rate flow of dry air of 1.5 liters/minute, 2.5 liters/minute 4.0 liters/minute

Moreover, from figure 4 and figure 5, we found that the concentration of ozone is always decreasing with increasing flow rate of oxygen or dry air.

Furthermore, according to Stanley [16], micro-discharge in dry air produced many compound of nitrogen oxides such as N₂O, N₂O₅, NO₃ and certainly O₃. Figure 6 shows the end of products after 10³ seconds production times of micro discharge in dry air. This is also reason why ozone concentration in oxygen as source is higher than dry air as source.

B. Dissolved Ozone in Water for Oxygen as Source

Figure 7 a,b,c and figure 8a, b, c shows the concentration of dissolved ozone in water, for oxygen and dry air as sources of ozone. Dissolved ozone in water is very important for wastewater treatment. According to Chen et al by using gas mixture of ozone and oxygen is introduced into the bottom of a column, and then transferred into the liquid. The pollutants in the wastewater are eliminated subsequently via oxidation by the dissolved ozone. There certainly exists a temporary and unsteady period before the ozonation system reaches steady state [17]. In this research, we found that the dissolved

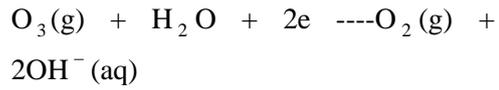
ozone increases with the increasing applied voltage on the system generating dielectric barrier discharge plasma. This increasing is due to increasing ozone concentration by augmenting applied voltage.

C. Ozone produced and Ozone dissolved in the water

Figure 9 and Figure 10 shows the comparing of ozone produced and dissolved ozone in water for oxygen as source and dry air as source. The concentration of ozone produced is higher than the concentration of

dissolved ozone in water by the applied voltages and times

This is caused of the ozone dissolved in water has a shorter life time during which reacts with molecules of water for 40 minutes and more easily break down into oxygen again as in the reaction [18]



According to our results, dissolved ozone in water are only 10 % of ozone produced and only 7 % for dry air as source.

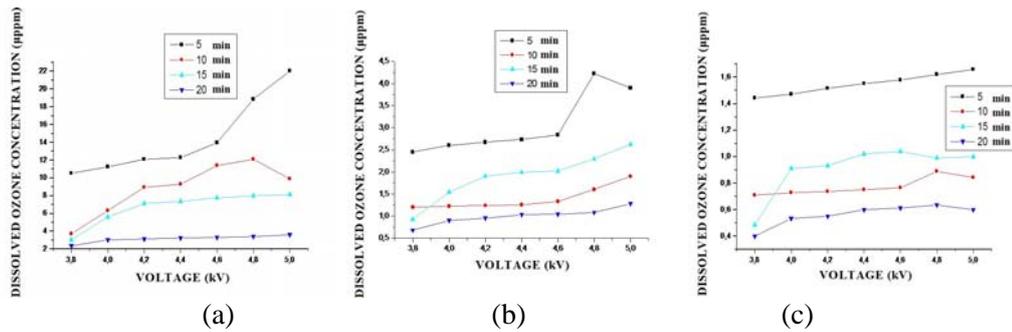


Figure 7. Concentration of dissolved ozone in water as function voltage with time constant of 4 minutes and flow rate of oxygen of 0.5 liters/minute, 1.5 liters/minute 2.5 liters/minute

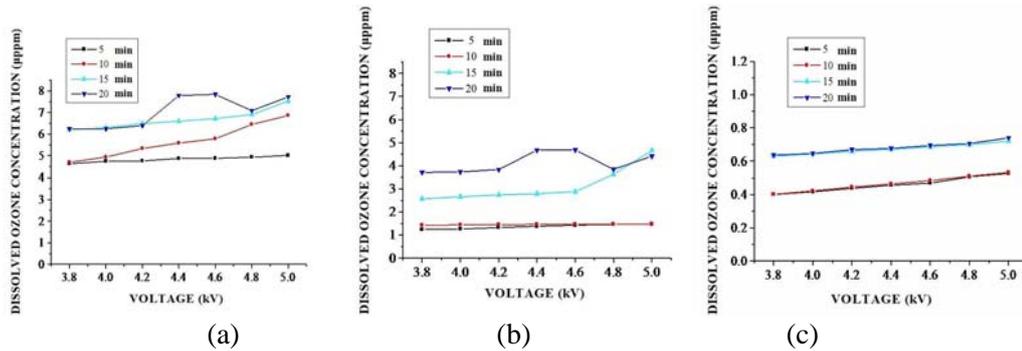


Figure 8. Concentration of dissolved ozone in water as function voltage with time constant of 4 minutes and flow rate of dry air of 1.5 liters/minute, 2.5 liters/minute 4.0 liters/minute

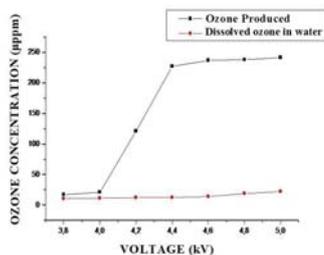


Figure 9. Comparison value concentration of Ozone produced and dissolved Ozone in water with contact time of 5 minutes for oxygen flow rate of 1.5 liters/minute

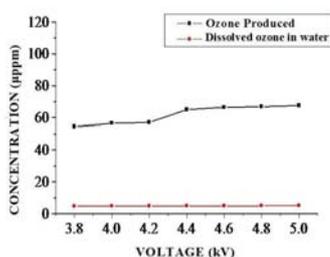


Figure 10. Comparison value concentration of Ozone produced and dissolved Ozone in water with contact time of 5 minutes for dry air flow rate of 1.5 liters/minute

CONCLUSION

The concentration of ozone produced increases with increasing voltage with a time constant. This concentration also increases with increasing time in certain applied voltage. Moreover, ozone concentration was higher than the concentration of dissolved ozone in water. Dissolved ozone in water are only 10 % of ozone produced and only 7 % for dry air as source. Oxygen as source of ozone is better than dry air, both for ozone produced and dissolved ozone in water.

ACKNOWLEDGMENT

Acknowledgments This work is supported by the Research Grand of Grand Competition Project Directorate Higher Education Republic Indonesia

REFERENCES

- [1] S. Patil, P. Bourke, J.M. Frias, B.K. Tiwari, and P.J. Cullen, *Inactivation of Escherichia coli in orange juice using ozone*, Innovative Food Science and Emerging Technologies 10 (2009) 551–557
- [2] C. Voidarou , A. Tzora, I. Skoufos , D. Vassos, G. Galogiannis, A. Alexopoulos and E. Bezirtzoglou *Experimental Effect of Ozone upon Some Indicator Bacteria for Preservation of an Ecologically Protected Watery System*, Water Air Soil Pollut (2007) 181:161–171
- [3] Bogaerts, A, *et al*, 2001, *Gas Discharge Plasmas and their Applications*, Elsevier, Spectrochimica Acta Part B 57 609-658, New York
- [4] Chen, J., dan Davidson, J.H., 2002, *Electron Density and Energy Distributions in the Positive DC Corona: Interpretation for Corona-Enhanced Chemical Reactions*, Plasma Chemistry and Plasma Processing, Vol. 22, pp 199-224.
- [5] Spyrou, N., *et al*, 1994, *New Result on a Point-to-Plane DC Plasma Reactor in Low-Pressure Dried Air*, Journal Phys. D: Appl. Phys., Vol. 27, pp 2329-2339.
- [6] Coogan, J.J., Jassal, A. S., *Silent Discharge Plasma for Point-of-Use Abatement of Volatile Organic Compound (VOC) Emission*, Sematech
- [7] DEL, 2002, *Ozone Water Purification Systems*, DEL Industries, United States of America.
- [8] Byoung Ho Lee, Won Chul Song, Biswaranjan Manna and Jong Kyu

- Ha, *Dissolved ozone flotation (DOF) — a promising technology in municipal wastewater treatment*, Desalination Volume 225, Issues 1-3, (2008), Pages 260-273
- [9] Javier Rivas, Olga Gimeno, and Fernando Beltrán "Wastewater recycling: Application of ozone based treatments to secondary effluents" *Chemosphere* Volume 74, Issue 6, (2009), Pages 854-859
- [10] Hiroshi Tsugura, Tetufumi Watanabe, Hiroshi Shimazaki and Shoichi Sameshima, *Development of a monitor to simultaneously measure dissolved ozone and organic matter in ozonated water* *Water Science and Technology* Volume 37, Issue 12, (1998), Pages 285-292
- [12] M. R. Viera, P. S. Guimet, M. F. L. de Mele and H. A. Videla, *Use of dissolved ozone for controlling planktonic and sessile bacteria in industrial cooling systems* *International Biodeterioration & Biodegradation* Volume 44, Issue 4, December 1999, Pages 201-207
- [13] WHO (2004). *Guidelines for drinking-water quality*, 3rd ed.,pp 55-60, World Health Organization, Geneva.
- [14] Kuraica, M. 1996, *Application of Coaxial Dielectric Barrier Discharge for Portable and Waste Water Treatment*, Faculty of Physics, Serbia and Montenegro.
- [15] McClellan, 2003, *The Silent Discharge Plasma Technology Nonequilibrium Nonthermal Plasma to Oxidize Organic Contaminants*, ENV America Incorporation, United States of America.
- [16] Stanley, B.T, *Feedgas for Modern High-Performance Ozone Generators*, ., (1999)Ozonias.Ltd., Switzerland
- [17] Y. H. Chen, C. Y. Chang, C. Y. Chiu, Y. H. Yu, P. C. Chiang, Y. Ku and J. N. Chen, *Dynamic behavior of ozonation with pollutant in a countercurrent bubble column with oxygen mass transfer*, *Water Research* Volume 37, Issue 11, (2003), Pages 2583-2594

