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Research Article

Evaluation of Novel Integrated Dielectric Barrier Discharge Plasma as Ozone Generator

Muhammad Nur[1,2*], Ade Ika Susan[1], Zaenul Muhlisin[1,2], Fajar Arianto[1,2], Andi Wibowo Kinandana[1,2], Iis Nurhasanah[2], S. Sumariyah[2], Pratama Jujur Wibawa[3], G. Gunawan[3], Anwar Usman[4]

[1]Center for Plasma Research, Department of Physics, Faculty of Science and Mathematics, Diponegoro University, Jl. Prof. Soedarto, Kampus Undip Tembalang, Semarang 50239, Indonesia
[2]Department of Physics, Faculty of Science and Mathematics, Diponegoro University, Jl. Prof. Soedarto, Kampus Undip Tembalang, Semarang 50239, Indonesia
[3]Department of Chemistry, Faculty of Science and Mathematics, Diponegoro University, Jl. Prof. Soedarto, Kampus Undip Tembalang, Semarang 50239, Indonesia
[4]Department of Chemistry, Faculty of Science, Universiti Brunei Darussalam, Jl. Tungku Link, Gadong BE1410, Negara Brunei Darussalam

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Abstract

This paper presents a characterization of an integrated ozone generator constructed by seven of reactors of Dielectric Barrier Discharge Plasma (DBDP). DBDP a has spiral-cylindrical configuration. Silence plasma produced ozone inside the DBDP reactor was generated by AC-HV with voltage up to 25 kV and maximum frequency of 23 kHz. As a source of ozone, dry air was pumped into the generator and controlled by valves system and a flowmeter. We found ozone concentration increased with the applied voltage, but in contrary, the concentration decreased with the flow rate of dry air. It was also found that a maximum concentration was 20 mg/L and ozone capacity of 48 g/h with an input power of 1.4 kW. Moreover, in this generator, IP efficiency of 8.13 g/kWh was obtained at input power 0.45 kW and air flow rate of 9 L/min. Therefore, be the higher ozone capacity can be produced with higher input power; however, it provided lower IP efficiency. The effect of dry air flow rate and applied voltage on ozone concentrations have been studied. At last, spiral wire copper was very corrosive done to the interaction with ozone, and it is necessary to do a research for finding the best metals as an active electrode inside of the quartz dielectric.

Keywords: ozone generator; dielectric barrier discharge; ozone; corrosion; electrode

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1. Introduction

Ozone is a compound formed from oxygen atoms (O_3). Some techniques generate ozone that has been developed among others by DC corona discharge [1,2]. Generation of ozone is more widely used for the application purpose to obtain the desired concentration with dielectric barrier discharge plasma [3,4,5]. A dielectric barrier discharge (DBD) will appear in a gas gap when an AC or RF voltage is applied to an

* Corresponding Author.
E-mail: m.nur@undip.ac.id
Telp.: +62-24-7474754 Fax.: +62-24-76480690

electrode system with one or both electrodes covered by a dielectric layer [6]. Ozone can be produced by using electrical discharge in pure oxygen gas or air that was feed in the space between the active electrode with dielectric barrier. DBD is the most widely used in industrial pipeline applications since it produces the highest gas concentrations [8,9,10].

The factors that can affect ozone formation in general are voltage, dielectric materials, pressure, configuration system of the plasma reactor and a gas inserting in the plasma reactor [11,12]. The main mechanism of ozone formation are ionization, recombination, dissociation and association. The reaction of ozone formation can occur in a plasma reactor has been explained for example by Fridman in his book [13]. Experimental study on the current-voltage characteristics for atmospheric pressure DBD has been done by Pongsathon [14]. Nur *et al.* produced ozone by using DBD plasma and it was implemented in the rice storage for maintaining the rice quality [15].

2. Materials and Method

Figure 1 shows a series of experiments in this study. An ozone generator is made by combining seven Dielectric Barrier Discharge Plasma (DBDP) reactors. DBDP has a spiral-cylindrical configuration. Silence plasma produced ozone inside DBDP reactor was generated by AC-HV with voltage up to 25 kV and maximum frequency of 23 kHz. Input voltage was determined through a voltage divider (HV Probe AC Voltage max 28 kV; DC 40 kV, EC code number 1010, En G1010). Electrical signal for high voltage from the probe was detected by an Oscilloscope GOS-653, 50 MHz. The electric current was measured by using an Ammeter (AC/DC digital clamp meter, KYORITSU Kew Snap Model 2010). Input power (IP) can be determined by using the following eq. [16]: *Input Power (IP) = Applied Voltage (V) x Average Capacitive Current (I).*

The design of the generator that consists of seven DBDP reactors with spiral - cylinder configuration and pyrex tube serves as barrier is presented in the Figure 2a. An active electrode was made of spiral copper wire, and it is placed inside of a Pyrex tube. The pyrex tube has an outer diameter of 20 mm, 1.5 mm in thick and 20 cm in long. The outside of the pyrex tube was wrapped with cylindrical electrode of copper. Realization of the generator is showed in the Figure 2b.

In the Figure 2b, we can see that all of reactors are connected and fixed by Teflon with specific design, and the integrated DBDP reactors become an ozone generator. These reactors were connected together in parallel to AC-HV. Dry air was pumped into the generator with an air pump and controlled by valves system and a flowmeter (Kofloc Model RX1600A Kojima, scale 1-10 L/min). Finally, ozone concentrations were detected by the ozone monitor (Teledyne Instruments, API-454). Ozone Ca-

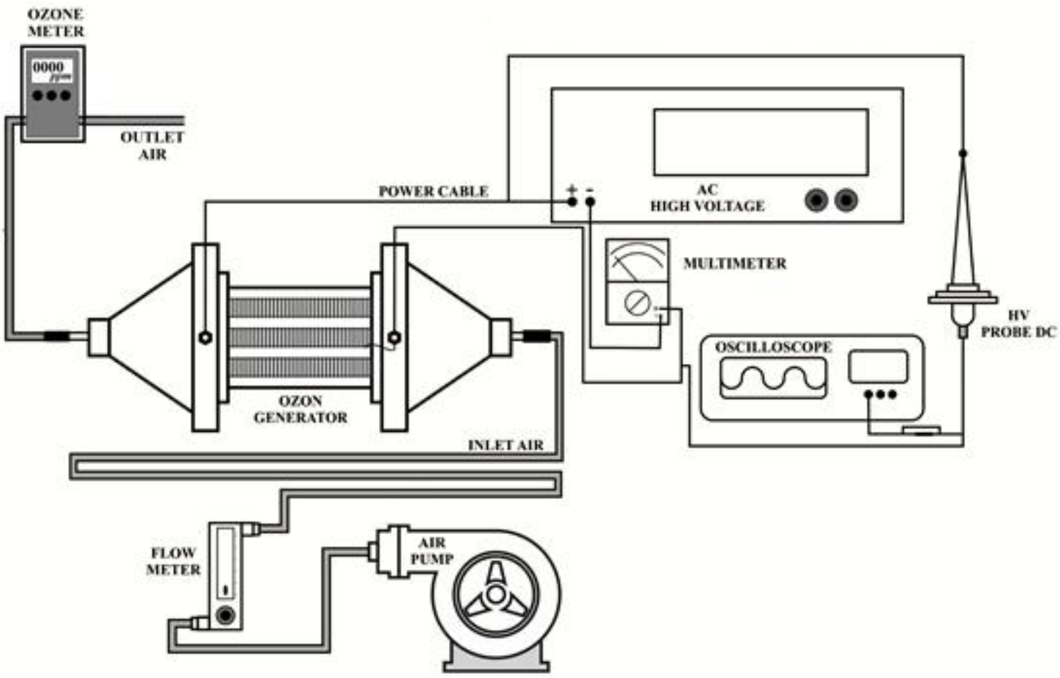


Figure 1. Experimental set up for an integrated ozone generator

pacity was calculated by a formula [17]: *Ozone capacity (g/h) = Ozone concentration (g/L) x Air Flow rate (L/h)*. Photograph of the experiments was taken using a CCD camera (Creative, DV Cam).

3. Results and Discussion

3.1. Characteristic of current - voltage

Figure 3 shows the average electrical current as a function of voltage and it was parabolic shapes. Similar results are recorded by [4] in the cylinder-cylinder and wire-cylinder configurations. This is due to the electric field intensification at the active electrode inside the reactor. The use of larger electric potential will generate strong electrical field around the active electrode. High intensity of the electric field further accelerates the motion of charged particles (ions and electrons) and these particles are involved in collisions among particles. Collisions among particles will produce ionization, dissociation and also electrical charges. The electrical charge changes with the change in time will generate an electric current.

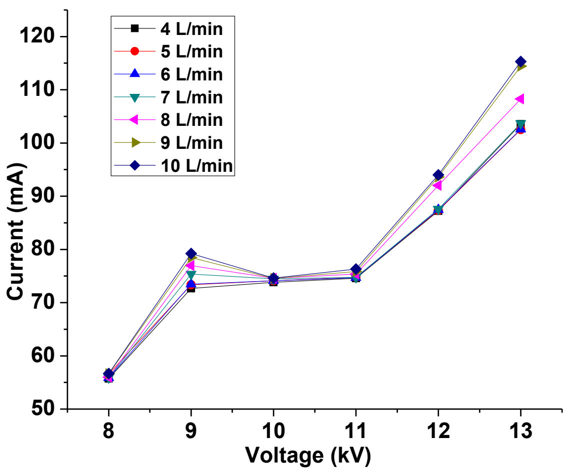


Figure 3. Current as function of voltage, for integrated reactor, for all variants air flow rate.

In the silence plasma or DBD plasma has two capacitors: dielectric barrier forming a capacitor connected in series with the gas capacitor [6]. Plasma itself is assumed to be a variable impedance. It is connected in parallel with gas capacitor. Total electrical current has two components: an electric current from the capacitive barrier and electric current of the capacitive gas. Therefore, the actual electrical current measured was capacitive current. In other world, there was presence of accumulated electrical charge changes due to the change of time.

3.2. Effect of voltage on ozone concentration

The influence of voltage on ozone concentrations are presented in Figure 4 for several flow rate of gas. In low flow rate, ozone concentration is more generated compare with high flow rate for the same of applied voltage. This trend can be explained that the rest time of gas in collision zone as well as ionization zone is longer for low flow rate. From the Figure 4 it can be seen that the concentration of ozone increased with the increase voltage, either for low and high flow rate. Fang *et al.* [4] suggested a similar result that the ozone concentration enlarged with the increase applied voltage for two types of DBD reactors with configuration cylinder-cylinder and wire-cylinder.

The graph presented in Figure 5 shows electrical current influenced by flow rate. From the graph can be observed that there is no significant change to the electrical current with the flow rate variation. The current did not affected by flow rate due to electric current is the cumulative capacitive electrical current of the gas discharge. Gas discharge can occur in case of collisions between energetic particles such as electrons, ions with atoms or molecules. The increasing in flow rate is not associated with an increasing the number of energetic plasma species. The increase in energy is obtained by increasing the applied voltage. It can be seen

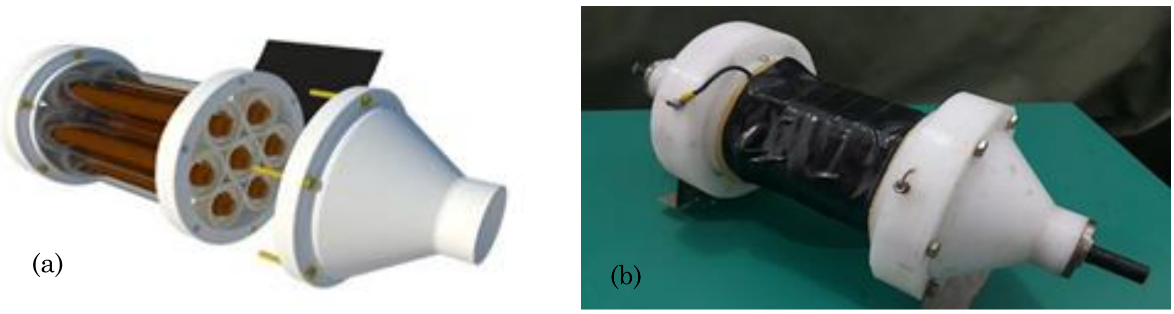


Figure 2. Integrated ozone generator with seven DBD reactors (a) Design (b) Realization

from the graph that the applied high voltage (13 kV) produced the largest electric current (116 mA).

To a certain voltage applied can be inferred to have a mean value of the current with small error bars. At the same graph, the effect of voltage applied to electrical current is very easy to find, the electrical current was greater with the increasing voltage applied. By using the average current value for each flow rate and applied voltage, the average input power can be determined for each flow rate. Input power (IP) can be determined by using the following equation [16]:

Input Power (IP) = Applied Voltage (V) × Average Capacitive Current (I)

The influence of input power to the ozone concentration in integrated reactor can be shown in Figure 6. In this research, ozone capacity has been determined using the following equation:

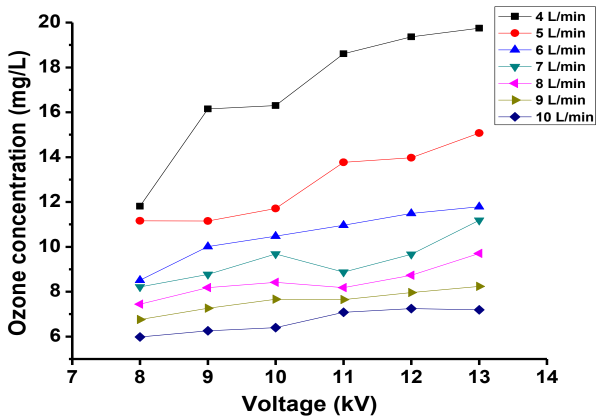


Figure 4. Ozone concentration as function of applied voltage for air flow rate variations.

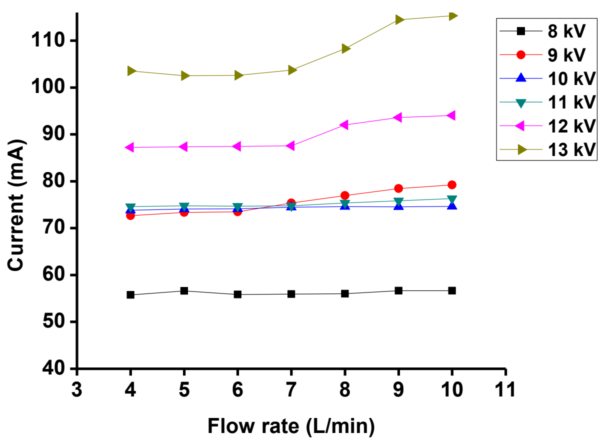


Figure 5. Electric current as a function of air flow rate for some voltage variation.

Ozone capacity (g/h) = ozone concentration (g/L) × air flow rate (L/h)

Figure 7 showed the ozone concentration in various air flow rates as function of input power. There are 7 variations of flow rates, from 4 to 10 L/min, with increment of every 1 L/min. It appears that the ozone capacity has significantly increased for all flow rates variation to the input power of 8 kW. It appears the presence of saturation or the tendency of constant value in ozone capacity with the increment of input power. We found that a maximum concentration is 20 mg/L and ozone capacity of 48 g/h with input power of 1.4 kW. The similar results were also obtained by Jodpimai *et al.* [17], but they used the higher input power up to 2.7 kW or two-fold greater than the input power used in this research. The tendency of constant value in ozone capacity with the increment of input power in this research [18] obtained at the O_2 flow rates of 5 L/min and 10 L/min. Figure 8 showed the input power (IP) efficiency, obtained by the following equation [17]:

IP efficiency (g/kWh) = Ozone capacity / Input power

3.3. Effect of flow rates on ozone concentration

Figure 9 shows the variation of ozone concentration with flow rate of air for several applied voltages. For all voltages, ozone concentration decreased with increasing flow rate. This decrease is due to partly reduced rest time of molecules of air or oxygen gas in the collision and ionization zones. As a result, the process of the ozone formation with tree body reactions cannot take place properly.

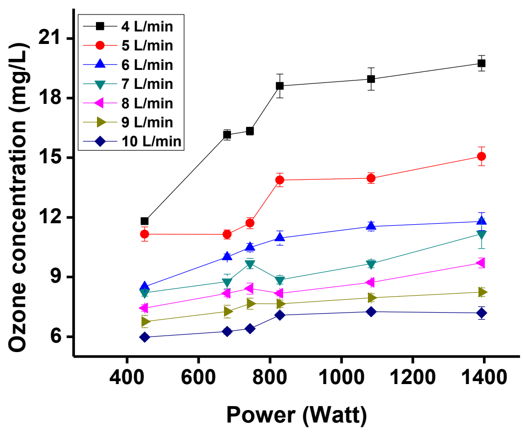


Figure 6. Ozone concentration as a function of input power.

Figure 9 shows that the ozone concentration decreases with the enlargement of the gas flow rate. Beside the mean free path for a gas molecule could be taken as the length of the path divided by the number of collisions. Thus, the greater the number of particles that pass through the ionization zone, the smaller the energy of the collision and the less oxygen dissociation may occur.

The complicated mechanism of ozone generation from air DBD has been studied by previous investigators [18,19,20]. The process of ozone formation in the high electric field as in DBD reactor generally follows several stages. High energy electrons (1-10 eV) collide with O_2 molecules. Then the oxygen molecules will be dissociated into oxygen atoms. Oxygen atoms may combine with oxygen molecules to form O_3. The formation of ozone can be explained by the reaction below:

$$e + O_2 \rightarrow O + O + e$$
$$O + O_2 + M \rightarrow O_3 + M$$

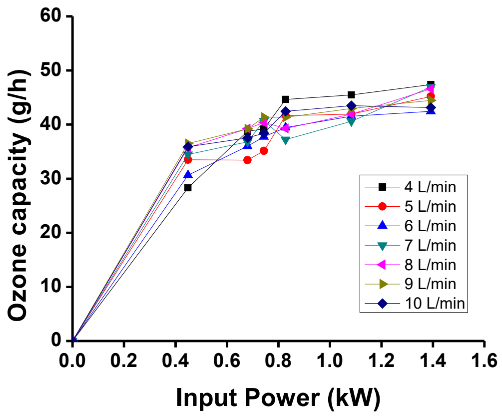


Figure 7. Ozone capacity as a function of input power at flow rates of 4-10 L/min with interval of 1 L/min

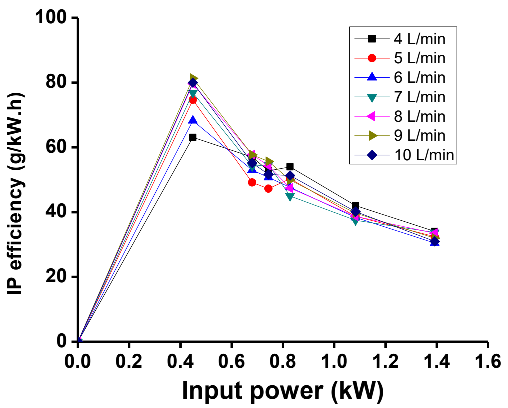


Figure 8. Input Power Efficiency as a function of input power at flow rates of 4-10 L/min with interval of 1 L/min

where, M is another molecule that functions to absorb part of energy in the process of formation of O_3. This molecule is not involved in chemical reactions. Therefore, the generation of ozone is based on silent discharge phenomena, electrolytic, opto-chemical reaction. The ozone gas is generated from oxygen or air by the electron bombardment of molecules in glass-barrier based discharge.

Figure 10 addressed average of ozone concentration as fuction of air flow rate. Effect of flow rate on the concentration of ozone follows the negative exponential function. In order to maintain high ozone productivity, we can conclude that the ozone concentration can be maintain on highly by using low flow rate and high voltage. The effect DBD reactor configuration and construction has been studied by Nur *et al.* [21]. This paper reported that the largest ozone concentration has been produced by using spiral-cylindrical configuration. The number of windings coil of spiral electrode is high, the concentration of ozone produced is also higher. Moreover, the closer the distance between the wire spiral with a dielectric (pyrex) the closer the distance to the electrode cylinder the greater the concentration of ozone generated [21].

3.4. Electrodes morphology analysis

The weakest component of the ozone reactor is the inner electrode. This electrode oxidizes continuously by the ozone produced in the reactor. Figure 11 shows the reactor with electrodes in the form of copper coils spiral before (A) and after used (B). Pure copper rod in an oxygenated environment will corrode to form copper oxide Cu_2O and/or CuO (if no compo-

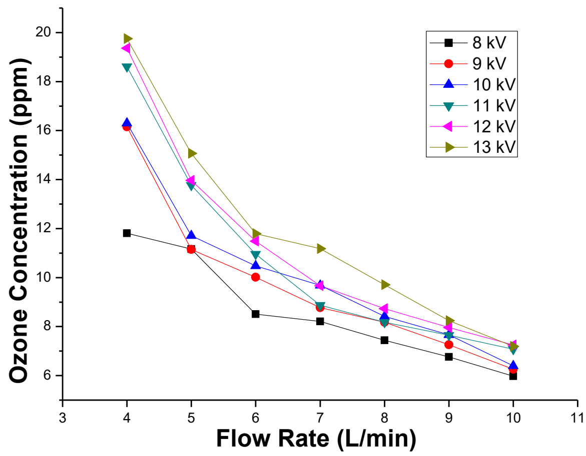


Figure 9. The concentration of ozone as a function of air flow rate in the integrated reactor, for some voltage variations.

nents of other atoms in the sample, this will need a data EDX). Cu_2O crystal structures are cubic, truncated octahedral, octahedral and polypoid (flower-like). Corrosion rate of copper will increase by increasing of the oxygen content and duration of exposure. The plasma reactor after certain time of usage should be clean using mineral acid to remove the remnant on the surface. This condition can be confirmed by XRD test. The images of SEM (Figure 12a and 12d) shows a solid surface and a relatively smooth, while the images (b and e) of the surface look rough and porous and there is a considerable hole (dark zone), this hole can be connected with the corrosion process where Cu eroded/sputter by oxygen radicals to form copper oxide. Furthermore, it appears tiny grains of nearly homogeneous and larger grains with diverse shapes (bright zone) on it. After washing (c and f) the grains on the top surface an invisible, black holes and the rod-shaped grains are randomly appearing more clearly, it shows the formation of Cu oxide (more clearly visible at magnification 10000×). Allegedly as a set of rod-shaped Cu_2O-like flower that is not yet fully formed.

Figure 13 shows the SEM's image of the surface morphology of the Cu electrode with d = 1.12 mm. Unlike the previous electrode, the electrode surface before use (a and d) is not too dense and coarse. Cu surfaces that are not washable (b and d) composed by casts (micro flake), also observed fragments that clustered to form flower-like structure with pores larger than the surface of the Cu (d = 0.52 mm). After washing, the Cu surface becomes denser composed by rod-shaped grains distributed randomly, although it appears the fracture extends. Black holes are thought to be due to cor-

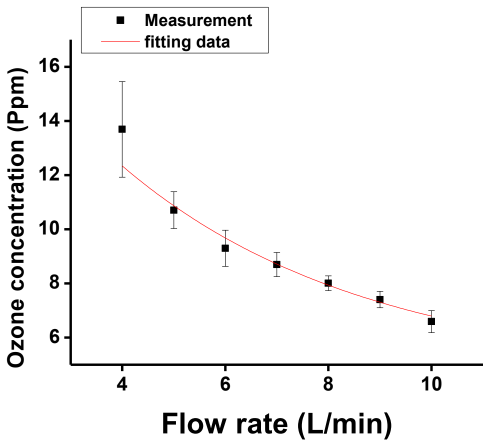
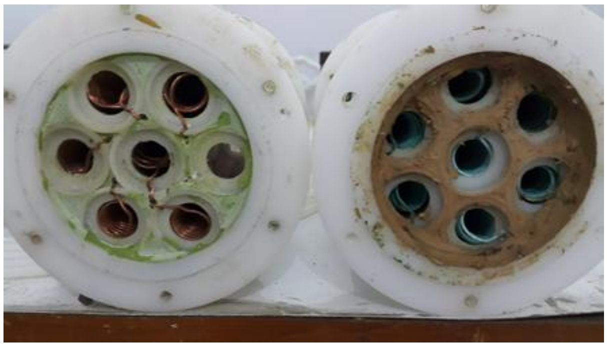


Figure 10. The average ozone concentration as a function of air flow rate



(A) (B)

Figure 11. Photographs of reactor with electrodes in the form of spiral coil copper before (A) and after used (B)

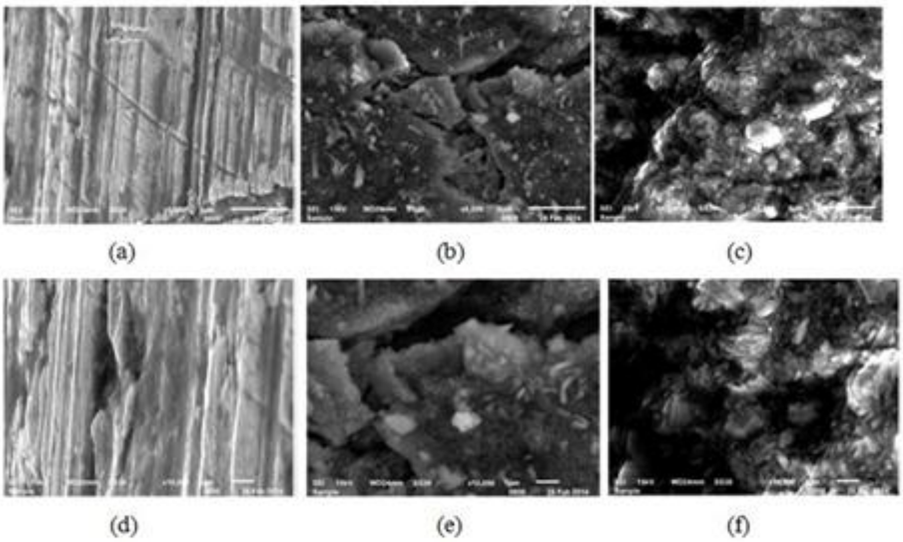


Figure 12. SEM images of the surface morphology of the Cu electrodes with diameter of 0.52 mm, top 5000×, 10000× bottom. (a & d) before use, (b & e) after use, (c & f) after use and washed.

rosion is more dominant on the electrodes with a diameter of 0.52 mm. Based on the SEM's images of Figures 12 and 13, stronger corrosion occurs at the electrodes with a diameter smaller morphological differences between the two electrodes which are summarized in Table 1.

4. Conclusions

An integrated ozone generator that constructed by seven of reactors of Dielectric Barrier Discharge Plasma (DBDP) has been developed. The characteristic of this ozone generator has been determined. We found that ozone concentration is increasing with the applied voltage but in contrary ozone concentration decreasing with flow rate of dry air. It is found also that a maximum concentration of 20 mg/L and ozone capacity of 48 g/h with input power of 1.4 kW. Moreover, in this generator, IP efficiency of 8.13 g/kWh was obtained at input power of 0.45 kW and an air flow rate of 9 L/min. Meanwhile higher ozone capacity was produced with the higher input power, however it provided the lower of IP efficiency. At last a spiral wire copper is very corrosive due to the interaction with ozone, and it is necessary to do a research to get the metal as an active electrode inside of the Pyrex dielectric.

Acknowledgement

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Table 1. Morphological identification of copper wire electrode inside of reactor

Electrode diameter (mm)	Before use	After use without washing	After use with washing
0.52	dense, relatively smooth	Spherical grains (~0.25 mm), rough, pores between grains, elongated black hole with a width of ~ 2 mm	Rod-shaped grains clumped, denser black hole shaped channel width
1.12	porous, more rugged	Composed of fragments, some of which appeared in groups, pore formed between flake	Composed by a rod-shaped grains (~ 0.5 mm length, diameter ~ 0.25 mm) more dense, elongated black hole

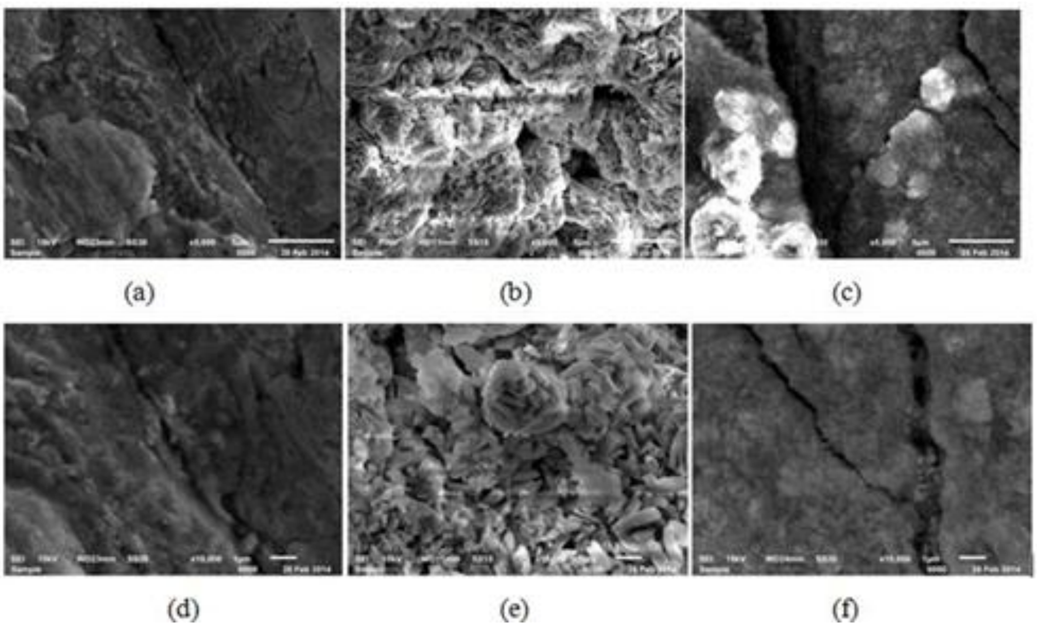

Figure 13. SEM image of the surface morphology of the Cu electrode with diameter of 1.12 mm, top 5000×, 10000× bottom (a & d) before use, (b & e) after use, (c & f) after use and washed

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