Study of ozone reactor with dielectric barrier discharge plasma (BDBP): variations of inner electrode based on Stainless steel, Galvalume, and Copper

Suraidin¹, M. Nur²,4*, Gunawan³,4, Ade Ika Susan¹

¹) Magister of Physics, Faculty of Science and Mathematics, Diponegoro University, Jl. Prof. H. Soedarto, SH, Tembalang. Semarang. INDONESIA,  
²) Department of Physics, Faculty of Science and Mathematics, Diponegoro University,  
³) Department of Chemistry, Faculty of Science and Mathematics, Diponegoro University,  
⁴) Center for Plasma Research, Science and Mathematics Faculty, Diponegoro University, Jl. Prof. H. Soedarto, SH, Tembalang. Semarang. INDONESIA.

E-mail: m.nur@undip.ac.id correspondence author

Abstract: The research of ozone reactor with dielectric barrier discharge plasma (BDBP) with variations of inner electrode based on stainless steel, galvalume, and copper has been done. The ozone was produced with oxygen as inputted gas. The ozone concentration has determined with variations of voltage (from 3 to 5 kV) and flow rate (i.e. 2, 3, 4, 5, 6, 8 and 10 L/min). Ozone reactor, has a frame with cylinder-cylinder configuration. Inner electrode (cylindrical) made of stainless steel, galvalume and copper. The three of materials was used for evaluates the inner electrode's corrosion resistance characteristics for DBDP reactor. The outer electrodes (cylindrical) made of zinc plate, while Pyrex used as dielectric material. AC High Voltage was used as power supply with frequency of 16 kHz. This research has been conducted with five variations of reactor’s length of 5, 10, 15, 20, and 25 cm. The ozone concentration has measured with KI solution and then titrated using sodium thiosulphate. After used as the inner materials of ozone reactor, the corrosion test has been conducted with SEM and EDX analysis. The results showed that ozone concentration was increasing by the increasing of voltage, and the greater gas flow rate gives the less ozone concentration. In addition, the most corrosion electrodes is one made of copper, which supported by SEM EDX result, that has CuO concentration of 60.77%. Otherwise, from the three of materials, stainless steel is obtained as the most stainless material.

1. Introduction
Ozone was first discovered by an European researcher, C.F. Schonbein, in 1839. He identified the smell, that arises from anode during the water electrolysis, as a new compound, named ‘ozone’. Ozone was first used commercially in 1907 to provide good water, and then used in 1910 in St. Petersburg [1] Ozone consists of oxygen atoms with the chemical symbol O₃. Ozone is a relatively unstable molecule when compared with oxygen O₂. Ozone can be generated using ultraviolet radiation, photochemical reaction and dielectric barrier discharge (DBD). Nowadays, DBD known as the most effective method for generating ozone [2]. DBD is a type of non-thermal plasma that generally produced using two electrodes which separated by a gap of several millimetres and covered by a dielectric layer. Both of
electrodes are connected with AC (alternative current) high voltage. Dielectric serves as a current limiter, preventing the formation of spark and distribute the discharge throughout the electrodes area[3]. DBD system is generally consists of a power supply, electrodes and dielectric barrier. According to the operating frequency of power supply, DBD is generated using alternative current (AC), radio frequency (RF), and pulse mode. Power supply and type of reactor provide a great effect on the characteristics of DBD. Reactor types can be classified by the electrodes configuration, electrode material, dielectric barrier material and the presence of catalysts between the electrodes. In addition, dielectric barrier materials used as insulation layer are glass, quartz, ceramic, and polymer coating. The type of material, thickness and surface structure of the dielectric material may affect the plasma discharge [4].

2. Research Methods
Figure 1 shows scheme of experiment set up of this study. The components and size of reactor that used are shown in table 1.

![Diagram of experiment set up](image)

Table 1. Component and size of each component of ozone reactor.

| No | name of parts   | material                  | length (mm) | Diameter (mm) | Thickness (mm) |
|----|----------------|---------------------------|-------------|---------------|----------------|
| 1  | Dielectric     | pyrex glass              | 225         | 50            | 1,1            |
| 2  | Inner Electrode| stainless stainless, galvalum, and copper, | 250         | 40            | 2              |
| 3  | Outer Electrode| zinc plate               | 250         | 52            | -              |
| 4  | Discharge gap  | -                         | 250         | -             | 2              |
| 5  | Cover          | flexion glass            | 22          | -             | 3              |
| 6  | Input line     | Cu                        | 20          | -             | -              |
| 7  | Output line    | Fe                        | 46          | -             | -              |
| 8  | Positive terminal| Fe                     | 35          | -             | 1              |
| 9  | Negative terminal| Cu                     | -           | -             | -              |

Inner electrode in the form of cylindrical framework made of stainless steel ring which connected by the 12 galvalume strings, and the diameter of each cylinder frame was 40 mm. The outer electrode in cylindrical form was made of zinc plate with diameter of 52 mm, and the barrier was made of Pyrex glass of 1.1 mm in thickness and 440 mm in length. AC high voltage used as power supply was varieties of 2, 3, 4 and 5 kV, while oxygen flow rate is variants of 2; 3; 4; 5; 6; 8, and 10 L/minutes.
Ozone’s concentration is equal with Na$_2$S$_2$O$_3$ solution’s volume that used in titration [7][8]. Ozone's concentration can be calculated by the following formula, where M is ozone’s mass, V is titration volume, N is sodium Thiosulfat’s concentration, $v_O$ is oxygen volume, and e is constant.

$$C_{O_3} = \frac{MVN}{v_O e}$$

3. Results and Discussions

Characteristic of Electric Current as Function of Voltage (I-V)

The characteristic of electric current as function of voltage has been conducted five variations of electrode's length of 5; 10; 15; 20; and 25 cm, as shown in Figure 2. Beside it, applied voltage has been varied of 3; 4; and 5 kV. Figure 2 shows that the increasing of voltage gives the increasing of electric current. It is caused by the charge accumulated in reactor. The electrical charge is formed by the multi-scattering when electric fields accelerate the electrons so that the electrons have enough energy to ionize the oxygen molecules inside of reactor.

![Figure 2. Electric current as function of voltage in some variations of reactor’s length.](image)

Effect of Voltage to Ozone Concentration.

The effect of voltage to ozone concentration in some variations of electrode's length can be seen in Figure 3. The greater the applied voltage provides the higher the potential between the electrodes, and then resulting the higher ozone concentration. These results are consistent with the results obtained by Syafrudin [9]. The results showed that ozone concentrations increase with smaller flow rate and higher voltage. The higher the voltage, the oxygen molecules inside the reactor will increase, so there are more oxygen molecules collide. These results are consistent with the results obtained by Nur [10].

On the reactor with electrode length of 5 cm, the smallest concentration of 67.2 ppm is obtained at voltage of 3 kV, while the highest ozone concentration of 384 ppm is obtained at voltage of 5 kV. On the reactor with electrode length of 10 cm, the smallest concentration of 76.8 ppm is obtained at voltage of 3 kV, while the highest ozone concentration of 384 ppm is obtained at voltage of 5 kV. On the reactor with an electrode length of 15 cm, the smallest concentration of 76.8 ppm is obtained at voltage of 3 kV, while the highest ozone concentration of 432 ppm is obtained at voltage of 5 kV. On the reactor with an electrode length of 20 cm, the smallest concentration of 96 ppm is obtained at voltage of 3 kV, while the highest ozone concentration of 480 ppm is obtained at voltage of 5 kV. While on the reactor with an electrode length of 25 cm, the smallest concentration of 96 ppm is obtained at voltage of 3 kV, while the highest ozone concentration of 624 ppm is obtained at voltage of 5 kV. Based on the results shown in figure 3, the applied voltage is affects the ozone concentration, the higher voltage provides the higher ozone concentration.
Figure 3. Ozone concentration as function of voltage in a reactor in length with (a) 5 cm, (b) 10 cm, (c) 15 cm, (d) 20 cm, and (e) 25 cm.

Morphology Analysis

After being used to generate ozone, the materials used as inner electrode in ozone reactor were evaluated using SEM EDX analysis. The components of inner electrodes is shown in table 2. Morphological analysis aims to determine the surface image on corrosion parts. These parts are magnified by 500×, 1000×, 3000×, and 5000×. The greater magnification of the image shows significant changes on the morphological surface. The increased of surface roughness indicates the defects on sample surface due to corrosion.

The test results of the three electrodes made of stainless steel, galvalume, and copper is shown in Figure 4 for a magnification of 3000x, In the image morphology for galvalume and copper electrodes already seen experience corrosion. The most corrosion electrode is an electrode made of copper. Results the supported by SEM EDX analysis in which electrodes are made of copper with CuO amounted to 60.77%. Figure 4.a, b, and c show the morphological image of stainless steel, galvalume, and copper, respectively, that used as inner electrodes, with magnification of 3000x. The morphology image of galvalume and copper already seen being rusty or corrosion. The most corrosive material is copper. These results are supported by EDX analysis, where the CuO concentration obtained was 60.77%. The higher magnification provides the better image of morphology.
Stainless Steel

Figure 4a. SEM image of Stainless steel sample 3000x magnification

Galvalum

Figure 4b. SEM image of galvalum sampleto 3000x magnification

Copper

Figure 4c. SEM image of copper sampleto 3000x magnification

For example at a magnification of 5000x seen that copper electrodes in the form of beam, Octoludral, and polypoid (such as flowers). Similar results were also found in galvalume electrode. From the aspect of corrosively, stainless steel was obtained as the most stainless material to used as the inner electrode.
Table 2 Components and composition of inner electrodes in the used ozone reactor.

| No | Sample  | Component          | Composition (% by weight) |
|----|---------|--------------------|---------------------------|
| 1  | Stainless steel          | Carbon, C       | 19,39                     |
|    |                      | Alumina, Al₂O₃  | 0,90                      |
|    |                      | silica Dioxide, SiO₂ | 2,26                    |
|    |                      | potassium Oxide, K₂O   | 2,38                     |
|    |                      | Mercury (II) Chloride, TiO₂ | 0,57                    |
|    |                      | chrome oxide, Cr₂O₃ | 9,57                      |
|    |                      | manganese Oxide, MnO | 5,18                     |
|    |                      | Iron (II) oxide, FeO | 34,70                    |
|    |                      | Copper (II) oxide, CuO Zink | 24,10                   |
|    |                      | Oxide, ZnO     | 0,95                      |
| 2  | Galvalume   | Carbon, C       | 19,58                     |
|    |                      | Alumina, Al₂O₃  | 0,89                      |
|    |                      | silica Dioxide, SiO₂ | 1,62                     |
|    |                      | potassium Oxide, K₂O | 0,60                    |
|    |                      | calcium Oxide, CaO | 0,39                     |
|    |                      | Iron (II) oxide, FeO | 62,06                   |
|    |                      | Zink Oxide, ZnO | 14,85                     |
| 3  | Copper       | Carbon, C       | 20,88                     |
|    |                      | Nitrogen, N   | 17,65                     |
|    |                      | Alumina, Al₂O₃  | 0,70                      |
|    |                      | Copper (II) oxide, CuO | 60,77                   |

4. Conclusion
The concentration of ozone is always higher with the smaller oxygen flow rate. The highest ozone concentration was obtained with flow rate of 2 L/min. The ozone concentration increases with the increasing of voltage. The length of the electrodes also give influence to the ozone concentration. The longer electrodes provide the higher ozone concentration. In this research, an electrode with the length of 25 cm produces the highest ozone concentration. Copper is the most corrosion materials as inner electrodes than the two others materials used in this research. From the aspect of corrosively, stainless steel was obtained as the best material for utilization as the inner electrode than galvalume and copper.

5. References
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