Characteristic Research on the Generation of Ozone Using Dielectric Barrier Discharge

Zhehua Du¹*, Xin Lin²

¹Wuhan Second Ship Design and Research Institute, Wuhan, Hubei, 430205, China
²Hubei Province Engineering Consulting Co., LTD., Wuhan, Hubei, 430071, China

*Corresponding author’s e-mail: Jackydzh@163.com

Abstract. Ozone has been widely used in many fields due to its strong oxidation ability. Under the conditions of the experiments, the results are as follows: With an increasing applied voltage, both ozone concentration and ozone generation rate increase, while ozone generation efficiency increases at first, reaching peak value, then decreases slowly; With an increasing gas flow rate, ozone concentration decreases, while both ozone generation rate and ozone generation efficiency increase at first, reaching peak value, then decrease; When power supply frequency is changed in the range from 5.0kHz to 9.9kHz, no apparent laws of ozone concentration, generation rate and generation efficiency are found; When gap spacing is changed in the range from 0.3mm to 0.5mm and dielectric thickness is unchanged, shorter gap doesn’t show better effect.

1. Introduction
Ozone, as a strong oxidant, will not produce carcinogenic secondary pollutants, and is widely used in the treatment of drinking water, industrial wastewater and other fields. Dielectric barrier discharge (DBD) is mainly used to produce ozone in industry. However, the existing ozone equipment energy consumption, high cost, greatly limited the application of ozone. In order to improve the ozone production rate, this paper studies the rule of ozone generation in high frequency dielectric barrier discharger with thin gap on the basis of experiments.

2. Overview
Dielectric barrier discharge is a typical non-thermal equilibrium gas discharge occurring at high atmospheric pressure (0.1-10bar) with dielectric inserted into the discharge space [1]. When a sufficiently high AC voltage is applied between the electrodes, a discharge occurs between them. Because of the existence of dielectric, the discharge is different from the general gas discharge. Macroscopically, it is uniform and stable blue-purple, distributed in the whole air gap space. It can be considered that dielectric barrier discharge is composed of numerous micro discharges that occur and disappear from time to time[1].

Oxygen in dielectric barrier discharge will produce a large number of active particles, including: electron, positive ion O⁺, O₂⁺, negative ionsO⁻, O₂; O₃; the ground state atoms O(^1P), O₂(X^3Σg⁻), O₁(^1A), excited atoms O(^1D), O₂(a'Σg⁺), O₂(b'3Σg⁺), O₂(A^3Σu⁺), O₂(B^3Σu⁻), O₂(V), O₁⁺, etc[2]. The reactions in the process of oxygen plasma are very complicated[3].
3. Experimental equipment and measurement method

3.1. Experimental equipment
The experimental circuit principle is shown in Figure 1. W is a power meter used to measure the total power consumption including the dielectric barrier discharger and its high-frequency power supply. HF is a high frequency generator used to provide an adjustable high frequency square wave signal of 5.0-9.9 kHz. T is a high-frequency transformer for raising the voltage of the output of the front stage to achieve the discharge condition of the dielectric barrier discharger. O.G, which is the dielectric barrier discharger, is the object of experimental research. P is a high-voltage probe that can attenuate 1000 times, providing measurement signal for oscilloscope. OSC is an oscilloscope, which is used to measure and analyze the electrical parameters in the experiment.

![Figure 1. Circuit principle diagram](image1)

3.2. Dielectric barrier discharger
The design of the experimental device is based on the idea of studying thin gap discharge. Figure 2 shows the dielectric barrier discharger in the experiment. In order to facilitate the observation of the internal discharge, the outer wall of the device uses a transparent plexiglass cylinder with a diameter of 400 mm and a height of 500 mm. The electrode is made of stainless steel and is designed as a disc-shaped cavity with a hole in the middle. The hole in the middle is used to allow airflow, and the cavity is used to pass the cooling medium. The dielectric is a 2.0 mm thick quartz glass plate. The discharge gap is a padded polytetrafluoroethylene gasket, so that the gap thickness can be adjusted.

![Figure 2. Dielectric barrier discharger Schematic diagram](image2)

3.3. Measurement method
The gas flow during the experiment is shown in Figure 3. In the Figure, 1 is valve, 2 is gas flowmeter, 3 is ozone generator, 4 is Na₂S₂O₃ solution, 5 is KI solution and 6 is atmospheric sampler. The concentration and yield of ozone in the experiment are determined by the iodometric method [4].

![Figure 3. Ozone collection and measurement schematic](image3)
4. Experimental results and analysis
In order to study the rule of ozone generated by dielectric barrier discharge, this experiment changes the applied voltage, power frequency and gas flow respectively under different gap thickness, and studies the influence of these factors on ozone generation by measuring the ozone concentration, production rate and power consumption under different conditions.

4.1. Voltage effect
The experiment changes the applied voltage of the dielectric barrier discharger under the premise of keeping the power frequency and gas flow constant. The curves of ozone concentration and ozone yield with applied voltage are shown in Figure 4 and Figure 5. Ozone production rate can be obtained by directly multiplying the ozone concentration by the gas flow rate.

As can be seen from Figure 5, when other factors remain unchanged, the ozone concentration generated by the dielectric barrier discharger increases with the increase of the applied voltage regardless of the gap thickness. This shows that when the applied voltage increases, the field intensity of the discharge gap increases, and the energy injected into it increases, which is conducive to the dissociation of oxygen molecules, that is, more oxygen atoms are produced, thus more ozone molecules are formed, which leads to the increase of ozone concentration.

It can be seen from Fig. 6 that as the applied voltage increases, the yield of the dielectric barrier discharger also increases. However, when the applied voltage is increased to a certain value, the growth of the yield is slowed down, and after reaching the maximum value, it starts to decrease slightly. This shows that with the increase of applied voltage, although the output of ozone is also increasing, with the increase of oxygen atom concentration and ozone concentration, the formation of ozone is gradually inhibited, thus slowing down the formation rate of ozone[5].

4.2. Power frequency effect
Under the premise of keeping the applied voltage and gas flow constant, the measured curve of ozone concentration and ozone yield changing with the power supply frequency is shown in Figure 6 and Figure 7.
As can be seen from Figure 7 and Figure 8, when other factors remain unchanged and the power supply frequency changes, there is no obvious change rule in the ozone concentration and ozone yield of the dielectric barrier discharger. This may be because, when the power supply frequency changes within the range of 5.0kHz ~ 9.9kHz, and the applied voltage rating remains unchanged, the field intensity of discharge gap basically changes little, and the energy injected into it is subject to the power supply performance, so there is no obvious change rule of ozone concentration and ozone yield. Abnormal runout at 7kHz may be caused by resonance in the loop, which results in abnormal power supply output.

4.3. Gas flow effect
Under the premise of keeping the applied voltage and the power frequency constant, the gas flow rate is changed, and the curves of ozone concentration and ozone yield as a function of gas flow rate are shown in Figure 8 and Figure 10. The ozone production rate can be obtained by directly multiplying the ozone concentration by the gas flow rate, and the curve of its change with the gas flow rate is shown in Figure 9.
As can be seen from Figure 8, when other factors are constant, regardless of the thickness of the gap, as the gas flow rate increases, the concentration of ozone generated by the dielectric barrier discharger is continuously reduced. This shows that when the gas flow rate is small, oxygen can pass through the discharge area at a slower speed, that is to say, oxygen molecules can stay in the discharge area for a long time, so the chances of oxygen molecules dissociation is greater, and thus more ozone is generated per unit volume of oxygen, that is, ozone concentration is higher. As the gas flow rate increases, the time for oxygen to pass through the discharge region is shortened, eventually leading to a decrease in ozone concentration.

As can be seen from Figure 10, with the increase of gas flow rate, the production speed of dielectric barrier discharger will also increase, but when the gas flow rate increases to a certain value, the production speed reaches the maximum, and then begins to decline. This shows that when the gas flow is small, although the ozone concentration is very high, the product of the two, namely, the production rate of ozone, is still small, which also reflects that the high ozone concentration has an inhibitory effect on the formation of ozone. With the increase of gas flow, the total amount of gas plays a major role, so the production rate of ozone is also increasing. However, when the gas flow increases to a certain extent, the advantage of the total amount of gas can no longer offset the reduction of ozone concentration, so the production rate of ozone will start to decline. For the electrode structure, size and power supply conditions in this experiment, the ozone production rate reaches the maximum when the gas flow rate is 0.8m³/h.

It can be seen from the comparison between figure 10 and figure 9 that the production rate of dielectric barrier discharger changes with the gas flow rate, which is similar to the rule that the ozone production rate changes with the gas flow rate. On the one hand, this is because of the variation law of ozone production rate. On the other hand, under certain output conditions (such as the same frequency and voltage), the total power output of the power supply basically does not change greatly due to the change of gas flow. For the electrode structure, size and power supply conditions in this experiment, the ozone production rate reaches the maximum when the gas flow rate is 0.8m³/h.

5. Conclusion

- With the increase of applied voltage, the ozone concentration and production rate of dielectric barrier discharger increase. With the increase of the applied voltage, the yield of ozone also shows an increasing trend. However, when the applied voltage increases to a certain value, the yield growth slows down and starts to decline slightly after reaching the maximum value.
- Under the condition of square wave power supply in this experiment, when the power frequency changes, the ozone concentration, production rate and production rate of dielectric barrier discharger have no obvious change rule.
With the increase of gas flow, the concentration of ozone produced by dielectric barrier discharger decreases. With the increase of gas flow rate, the production rate and rate of ozone also increase, but when the gas flow rate increases to a certain value, the production rate and rate reach the maximum, and then start to decline. Under the conditions of this experiment, the ozone production rate and yield reach a maximum at a gas flow rate of 0.8 m$^3$/h.

Due to the comprehensive influence of gap thickness on gap field intensity and oxygen residence time in the discharge area, under the conditions of this experiment, when the dielectric thickness remains unchanged, the ozone effect at the gap thickness of 0.5mm is slightly better than that at 0.4mm and 0.3mm.

Acknowledgments
This paper is funded by the National Key R&D Program（Item Number:2017YFC0307800）.

References
[1] Xu, X. (2001) Dielectric barrier discharge properties and applications. Thin Solid Films, 390(1-2): 237-242.
[2] Eliasson, B. (2006) Electron impact dissociation in oxygen. Journal of Physics B, 19:1241-1247.
[3] Eliasson, B. (2007) Ozone synthesis from oxygen in dielectric barrier discharges. Journal of Physics D: Applied Physics, 20:1421-1437.
[4] Sun, L.L. (2018) Characteristic analysis of dielectric barrier discharge with different electrode structures. Electrotechnical Application, 8:60-64.
[5] Wang, B.W. (2018) Progress in the preparation of ozone by dielectric barrier plasma. Modern Chemical Industry, 7:31-35.