Effect of Temperature on the Performance Characteristics of a Pin-Cylinder Discharge Type Ozonizer

Md. Fayzur Rahman, Byung-Joon Chun, Kwang-Sik Lee and Dong-In Lee

Abstract - A Pin-Cylinder discharge type ozonizer was designed and manufactured. The increase or decrease of temperature greatly influences on the characteristics of ozone generation of a discharge type ozonizer. The characteristics of ozone concentration (O_{3con}) , ozone generation (O_{3g}) and ozone yield rate $(O_{3\gamma})$ of the ozonizer were investigated by varying the gas flow rate (Q), the discharge power (W_d) and the temperature (T). At T = 20[°C], the values of O_{3con} were found as 7800, 5300, 3000 and 2300[ppm] at Q=1,2,4 and 6[l/min] respectively. The corresponding values of O_{3g} were found as 917, 1247, 1411 and 1623[mg/h] and those of O_{3Y} were 93, 126, 143 and 164[g/kWh] respectively. When the temperature is decreased to -50[°C], the values of O_{3con} became 12000, 8000, 5200 and 3600[ppm] at Q = 1, 2, 4 and 6[l/min] respectively. The corresponding values of O_{3g} were obtained as 1411, 1882, 2446 and 2600[mg/h] and those of O_{3Y} were 143, 190, 247 and 263[g/kWh] respectively. Hence as the temperature was decreased from 20 to -50[°C], the efficiencies of ozone generation were increased by 54, 51, 73 and 60[%] at Q = 1, 2, 4 and 6[1/min] respectively.

Keywords - pin-cylinder, ozone, discharge, temperature

1. Introduction

The main contributors to the environmental pollutions facing the world, such as acid rain, global warming, ozone depletion and smog are NOx, SOx, CO2 and volatile organic compounds [1-4]. It has been demonstrated that nonthermal plasma techniques offer an innovative approach to a cost effective solution to these problems [5-7]. Ozone plays a vital role in a number of different industrial applications including POZONE process used to bleach wood pulp, ozonated water for photoresist removal, sterilization of drinking water and treatment of industrial waste [8-10]. Since ozone is a pollution free oxidizing agent, it is attracting an increasing interest. In addition it has the second strongest oxidizing property next to Fluorine. In spite of these advantages, ozone is not used broadly in these days because of the high production cost. So, improvement of the efficiency of an ozonizer has become very essential for all research engineers. A number of ozonizers have been proposed in the literature [11-16]. The efficiency of a hybrid type ozonizer is higher in compared to that of a conventional ozonizer [17-18]. Considering all these points we tried to develop an ozonizer which uses the superposed operation of surface and corona discharge [19]. Although a number of ozonizers have been presented in the literature, the present ozonizer has a little different structure. By using this configuration it is possible to obtain higher values of O_{3con} , O_{3g} and O_{3Y} and in some cases these values may

be higher than those of other ozonizers. The present ozonizer consists of two glass tubes with three types of discharge electrodes: the central electrode (CE), the internal electrode (IE), and the external electrode (EE). When high voltage is applied between IE and CE electrodes, electric stress developed on the bottom parts of the discharging pins causes surface discharge. Also by applying high voltage between CE and EE electrodes corona discharge occurs between the discharging pins and the cylindrical electrode (EE). Since corona discharge is helpful for producing ozone, this design of the ozonizer is very suitable for the production of high ozone concentration and ozone yield. Also since pins are used for discharging purpose, there are enough space for the occurrence of surface discharge. These spaces are used for collection of the ozonized gas. Also since the discharge occurs between the tips of the pins and the EE electrode, the temperature of the region around the bottom parts of the pins do not rise too much. These are the main causes of obtaining higher ozone concentration (O_{3con}) and ozone yield rate (O_{3Y}) . O_2 was used as the source gas. By changing the gas flow rate (Q) and the discharge power (W_d), the effects of temperature (T) on the characteristics of ozone concentration (O_{3con}), the ozone generation (O_{3g}) and ozone yield rate (O_{3Y}) were investigated. As the temperature is reduced from 20 to -50[°C], the values of O_{3con} increase from 7800 to 12000[ppm] at Q = 1[1/min], from 5300 to 8000[ppm] at Q = 2[1/min], from 3000 to 5200[ppm] at Q = 4[1/min] and from 2300 to 3600[ppm] at Q = 6[l/min] respectively. With the same temperature decrease O_{3g} increases, from 917 to 1411[mg/h] at Q = 1[1/min], from 1247 to 1882[mg/h] at Q = 2[1/min], from 1411 to 2446[mg/h] at Q = 4[1/min] and

from 1623 to 2600[mg/h] at Q = 6[1/min] respectively. Un-

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der the same condition of temperature decrease O_{3Y} increases, from 93 to 143[g/kWh] at Q=1[l/min], from 126 to 190[g/kWh] at Q=2[l/min], from 143 to 247[g/kWh] at Q=4[l/min] and from 164 to 263[g/kWh] at Q=6[l/min] respectively. The maximum values of O_{3con} , O_{3g} and O_{3Y} were obtained as 12000[ppm], 2600[mg/h] and 320 [g/kWh] respectively.

2. Theory of Ozone Generation

The ozonizer is manufactured by using more than one electrode with glass or ceramic as insulator in such a way that a gap of 1~3[mm] is maintained between the electrodes. If ac voltage is applied, ozone is produced by chemical reaction in the oxygen portion of the supplied gas. Corona discharges are relatively low power electrical discharges that may take place at different pressures. The corona is invariably generated by strong electric fields associated with small diameter wires, sharp needles, or sharp edges on an electrode. Corona takes its name ("crown") from mariners observation of discharges during electrical storms. Corona appears as a faint filamentary discharge radiating outward from the discharge electrode. Corona discharge application process emphasizes one of the two aspects of the discharge: the ions produced or the energetic electrons producing the plasma. The two identities depend on the polarity of the discharge and the characteristics of the gas mixture especially on the electron attracting species. The electron energies depend on the gas characteristics and on the method of generating the corona.

Surface discharge occurs by narrow pulse type discharges and because electrons are generated from the different points on the surface of the electrodes, surface discharge is a useful one for the production of ozone caused by collisions of the electrons and oxygen molecules of the supplied gas. When high voltage is applied to a medium, interactions between the electrons and oxygen molecules take place to dissociate oxygen molecules into oxygen atoms. The rate of oxygen dissociation by the electrons depends on the energy distribution in the discharge region. In case of using atmospheric air as supplied gas existing Nitrogen affects the production of oxygen which plays an important role in ozone production.

 $O_2^- + High\ energy\ electron \rightarrow 2O + Low\ energy\ electron$

$$N_2 + e \to N_2^* \tag{2}$$

(1)

$$N_2^* + O_2 \to N_2 + 2O$$
 (3)

$$O_2 + e \rightarrow 2O + e \tag{4}$$

$$O + O_2 + M \rightarrow O_3 + M$$
 (5)
 $(K = 1.06 \times 10 -34 \exp(510/\text{T}) \text{ [cm3/s])}$

Here.

 N_2^{*} represents Nitrogen molecule in the excited state. N_2 represents Nitrogen molecule in the normal state. O_2 represents Oxygen molecule in the normal state. O represents Oxygen atom in the normal state. e represents electrons.

M represents third collision partner and may be O, O_2 or N_2 The decomposition of ozone are mostly represented by (6) - (13) as follows.

$$O_3 + N \to O_2 + NO \tag{6}$$

$$O_3 + NO \rightarrow O_2 + NO_2 \tag{7}$$

$$NO_2 + O_3 \rightarrow O_2 + NO_3$$
 (8)

$$M + O_3 + O \rightarrow O_2 + M \tag{9}$$

$$H_2O + O_3 \to O_2 + H_2O_2$$
 (10)

$$H_2O_2 \rightarrow HO_2 + H$$
 (11)

$$HO_2 + O_3 \rightarrow 2O_2 + OH \tag{12}$$

$$OH + O_3 \rightarrow O_2 + H O_2 \tag{13}$$

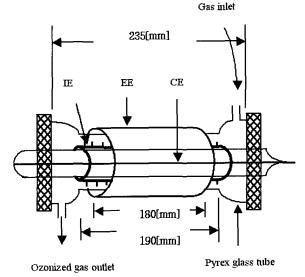


Fig. 1 Schematic diagram of the ozonizer.

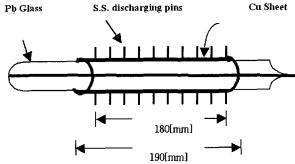


Fig. 2 Schematic Diagram of the inner tube.

Equations (10)-(13) show the reactions of ozone decomposition by moisture. The ozone concentration, ozone generation and ozone yield rate of the ozonizer can be improved by controlling the temperature and moisture content of the supplied gas. The proposed ozonizer has a configuration of improving ozone yield rate by controlling both the temperature and moisture content of the supplied gas.

3. Construction of Ozonizer

The ozonizer consists of two concentric glass tubes. The internal tube is made of Pb glass of 1[mm] thickness with the CE placed at the center. The outside diameter is 10[mm] and length is 250[mm]. After washing and drying the tube, it is sealed with the CE wire at the two ends and is evacuated by vacuum pump down to a pressure of 10-1 [torr]. The CE wire is 300[mm] long and one end is used for ground connection. A Cu sheet of about 0.2[mm] thickness was wrapped upon the surface and Stainless Steel (S.S.) pins of about 2[mm] length and 0.2[mm] diameter were inserted on the surface of the Cu sheet. A connection was brought out from the Cu sheet for using as IE electrode. The schematic diagram is shown in Fig. 2. The external tube is made of pyrex glass of 1.2[mm] thickness. The outer diameter is 19[mm] and the length is 235[mm]. A Cu sheet of about 0.2[mm] thickness was wrapped on the surface of the external tube. A wire was welded to the Cu sheet for using as EE electrode. The schematic diagram is shown in Fig. 1. The cross-sectional view is shown in Fig. 3. The measurements have been shown on the diagram.

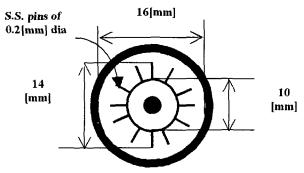


Fig. 3 Cross-section of the ozonizer.

4. Experimental Setup and Methods

4.1 Experimental Set Up

Fig. 4 shows the block diagram representation of the overall measurement process. The discharge voltage V_d and discharge current I_d were measured by means of a digital storage oscilloscope (500[MHz], 1[Gs/s]). The quantity of supplied gas was controlled by a flow meter (0-24[l/min]) . The ozone concentration of the ozonized gas was measured by an ozone monitor (0-110,000[ppm]). The

system is equipped with the combined discharge type ozonizer, cooler and dehumidifier. The gas flow line is shown with dotted line and this shows the O₂ supplier and flow of ozonized gas passing through the measuring apparatus. The cooling system was implemented by using liquid Nitrogen gas cylinder. One end of the cooling trap was connected to the Nitrogen gas cylinder and the other end was connected to the ozonizer. The dotted lines show the gas flow tube. The circuits of power supply source and measuring apparatus are shown with solid lines in the schematic diagram.

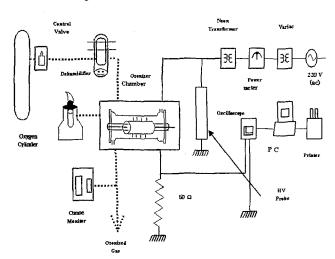


Fig. 4 Block diagram representation of the overall system.

4.2 Experimental Method

Experiments were carried out by applying high voltage between EE and IE with CE connected to ground. The amount of discharge power was varied by varying the supply voltage from 0 to 16[kV]. By varying the flow rate of the supplied gas (Q:1,2,4, and 6[l/min]) and the temperature of the supplied gas (T), the discharge voltage (Vd), discharge current (Id) and discharge power (Wd) were measured to investigate the characteristics of the proposed discharge type ozonizer. The different parameters like ozone concentration (O_{3con}) , the ozone generation (O_{3g}) and and ozone yield (O_{3Y}) were measured and calculated for variable values of Q, T and W_d .

5. Experimental Result and Discussion

Fig. 5 shows the O_{3con} - W_d characteristics for different values of Q at $T = -50[^{\circ}C]$. The temperature was kept constant at $T = -50[^{\circ}C]$ and the discharge power was increased from 0 to 10[W]. As seen from the figure for all values of Q ozone concentration increases continuously with W_d . But when W_d exceeds 8[W], the increasing rate becomes low. This means upto a value of 8[W] of W_d , the discharge is activated within the discharge chamber and depending on

the increase of the electrons produced in the discharge chamber, the number of collisions between the electrons and the oxygen atoms becomes very high. This leads the equations (1) - (5) to happen more. Hence the rate of ozone generation becomes very high. When the discharge power exceeds 8[W], the discharge becomes stronger and hence O_{3con} becomes high. Due to this high value of O_{3con} decomposition of ozone takes place at a higher rate resulting in the occurrence of (6) - (13) more and more. The oxygen atoms, molecules, electrons and excited atoms participate in decomposing ozone.

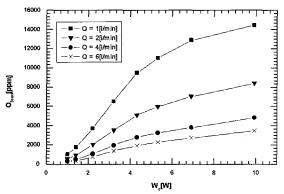


Fig. 5 Characteristics of O_{3con} against the variation of Q and W_d at $T = -50[^{\circ}C]$.

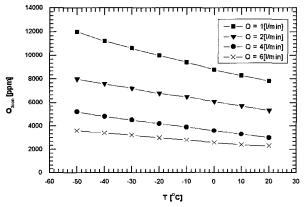


Fig. 6 Characteristics of O_{3con} against the variation of Q and T at $V_d = 16[kV]$.

Fig. 6 shows the ozone concentration (O_{3con}) characteristics against the variation of temperature (T). The discharge voltage (V_d) was kept at 16[kV] and the temperature was increased slowly from -50 to $20[^{\circ}C]$. It is seen from the figure that when the temperature increases from -50 to $20[^{\circ}C]$, the ozone concentration (O_{3con}) decreases from 12000 to 7800[ppm] at Q = 1[1/min], from 8000 to 5300[ppm] at Q = 2[1/min], from 5200 to 3000[ppm] at Q = 4[1/min] and from 3600 to 2300[ppm] at Q = 6[1/min]. This is due to the result of decreasing the moisture content of supplied gas in accordance with the decreasing temperature. So, the dissociation reaction of ozone was greatly decreased and the reactions of the equations (6) through (13)

happen to occur less. So ozone concentration decreases with the increase of temperature and vice versa.

According to the curves of Fig. 5 and 6 it is seen that O_{3con} increases as Q decreases and vice versa. When Q decreases, the duration of the oxygen molecules in the discharge chamber increases. This gives high value of O_{3con} . The maximum values of O_{3con} were found as 12000[ppm] at Q = 1[l/min], 8000[ppm] at Q = 2[l/min], 5200[ppm] at Q = 4[l/min] and 3600[ppm] at Q = 6[l/min] respectively.

Fig. 7 shows the O_{3g} characteristics against the variation of Q and W_d. Ozone generation increases with W_d for all values of Q. Hence it is considered that as long as W_d increases the electron density in the discharge chamber increases. So, both surface and corona discharges become stronger. The number of collisions among the electrons, oxygen atoms and molecules and excited oxygen atoms become larger, which lead O_{3g} to be proportional to O_{3con}. At a constant value of W_d the value of O_{3g} decreases with the decrease of Q. As Q increases, the ozone producing oxygen molecules also increase, but their duration of stay in the discharge chamber become less. This results in a few number of collisions among the generated electrons and oxygen molecules. The rate of increase of O_{3con} thus reduces. As a result O_{3g} is determined under such a condition that the reduction of O_{3con} is greater than the increase of the number of oxygen atoms. The maximum values of O_{3g} were found as 1700[mg/h] at Q = 1[1/min], 1980[mg/h] at Q = 2[1/min], 2277[mg/h] at Q = 4[1/min] and 2465[mg/h] at Q = 6[l/min] respectively.

Fig. 8 shows the characteristics of O_{3g} against the variation of Q and T. Here also O_{3g} decreases for all values of Q as T increases and vice versa. As T increases from -50[°C] to 20[°C], O_{3g} decreases from 1411 to 917[mg/h] at Q= 1[l/min], from 1881 to 1246[mg/h] at Q= 2[l/min], from 2446 to 1411[mg/h] at Q= 4[l/min] and from 2600 to 1623[mg/h] at Q= 6[l/min] respectively.

Fig. 9 shows O_{3Y} characteristics against the variation of W_d and Q at $T = -50[^{\circ}C]$. As seen from the figure for all values of Q ozone yield rate continuously rises until $W_d = 4.3[W]$. As W_d exceeds 4.3[W] the curves go downward.

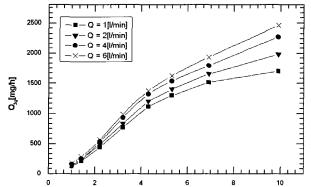


Fig. 7 Characteristics of O_{3g} against the variation of Q and W_d at $T = -50 [^{\circ}C]$.

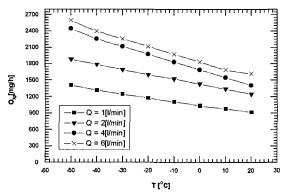


Fig. 8 Characteristics of O_{3g} against the variation of Q and T at $V_d = 16[kV]$.

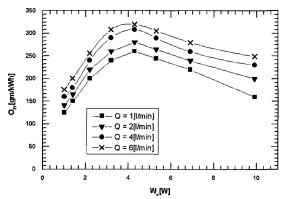


Fig. 9 Characteristics of O_{3Y} against the variation of W_d and Q at $T = -50[^{\circ}C]$.

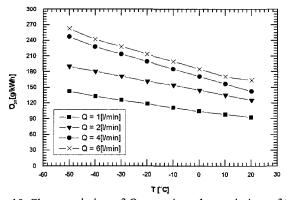


Fig. 10 Characteristics of O_{3Y} against the variation of W_d and T at $V_d = 16[kV]$.

As the discharge power starts increasing, the electron density increases. So, the number of collisions among the generated electrons, oxygen atoms and excited oxygen mole cules increase giving higher values of O_{3g} . After the time when O_{3Y} reaches peak value the temperature rises slightly. Due to this reason ozone decomposing reactions happen to occur more yielding equations (6) – (13). This reduces the value of O_{3g} . Since O_{3Y} is determined by O_{3g} and W_d , ozone yield rate after reaching a peak value decreases continuously for all values of Q.

At the same value of W_d the value of O_{3Y} is seen to be

higher at higher Q. At higher Q the number of oxygen atoms is larger. But the duration of their stay in the discharge chamber is less, which results in a low value of O_{3con} . The increase of oxygen atoms is dominant over the reduction of O_{3con} . So, O_{3Y} is higher with higher Q. For different values of Q the maximum values of O_{3Y} were found as 260[g/kWh] at Q = 1[l/min], 280[g/kWh] at Q = 2[l/min], 308[g/kWh] at Q = 4[l/min] and 320[g/kWh] at Q = 6[l/min] respectively.

Fig. 10 shows the ozone yield rate characteristics with the variation of both temperature and gas flow rate. The discharge power was kept constant at $W_d = 9.9 [W]$. The temperature was increased slowly from -50 to $20 [^{\circ}C]$ and the value of O_{3Y} was recorded each time. The cause of improving the ozone yield rate with the temperature reduction is exactly the same which was explained in Fig. 6. As the temperature increases from -50 to $20 [^{\circ}C]$, O_{3Y} reduces from 143 to 93 [g/kWh] at Q = 1 [1/min], from 190 to 126 [g/kWh] at Q = 2 [1/min], from 247 to 143 [g/kWh] at Q = 4 [1/min] and from 263 to 164 [g/kWh] at Q = 6 [1/min] respectively.

6. Conclusions and Discussion

The effect of temperature on the performance characteristics of the proposed Pin-Cylinder discharge type ozonizer has been studied in this paper. The ozone concentration was found nearly proportional to the gas flow rate. The maximum values of ozone yield rate were found at T = $20[^{\circ}C]$ as 93[g/kWh] at Q=1[1/min], 126[g/kWh] at Q=2[l/min], 143[g/kWh] at Q= 4[l/min] and 164[g/kWh] at Q = 6[1/min] respectively. When T was reduced to $-50[^{\circ}C]$, the corresponding values of ozone yield rate were recorded as 143, 190, 247 and 263[g/kWh] respectively. Hence the values of efficiency were improved by 53[%], 50[%], 72[%] and 60[%] at Q = 1, 2, 4 and 6[l/min] respectively. The maximum values of ozone concentration and ozone generation were recorded as 12000[ppm] and 2600[mg/h] respectively. Because of the simplicity in design the manufacturing cost is very less. The proposed ozonizer can be applied to the improvement of environmental pollution such as the treatment of industrial exhaust water.

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