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Ozonizer Design by Using Double Dielectric Barrier Discharge for Ozone Generation

G. P. Panta[1*], H. B. Baniya[1,2], S. Dhungana[1], D. P. Subedi[1] & A. P. Papadaki[3]

[1]*Department of Physics, School of Science, Kathmandu University, Dhulikhel, Kavre, PO Box No. 6250, Nepal.*
[2]*Department of Physics, Tri-Chandra College, Tribhuvan University, Nepal*
[3]*Department of Electrical Engineering, School of Engineering, Frederick University, Nicosia, Cyprus*
[*]*Correspondence email: gppanta@ku.edu.np*

We designed an ozone reactor made of co-axial cylindrical dielectric barrier discharge system with air as a working gas. The discharge was generated by using high voltage power supply (V = 0 -18 kV and f = 50 Hz). The flow rate of air was varied from 4 to 12 *l/min*. A comparison between O_3 generation using brass and copper as central electrodes and air as feed gas have been conducted and it was found that O_3 concentration was higher in the case of copper rod electrode than brass rod electrode for given flow rate and applied voltage. It was found that the concentration of ozone increases with increasing applied voltage for constant discharge time. Ozone concentration also increases with increasing discharge time at certain applied voltage. But this concentration decreases with increasing air flow rate. Smaller reactor and larger inner electrode will achieve better energy efficiency at the same specific energy density. But reactor with small diameter there exist an optimum inner electrode diameter. The glass tube reactor of internal diameter of 16 mm and brass and copper electrodes of diameters 4 and 8 mm have been used for this work. The ozone concentration was higher for copper electrode in air than brass electrode for fixed time, applied voltage and reactor diameter. It was also found that ozone concentration increases when the central electrodes diameter increases for fixed time, applied voltage and reactor diameter of double dielectric barrier discharge of glass tube.

1. Introduction

The world has witnessed a remarkable progress in the generation, diagnostics and application of atmospheric pressure non-thermal and low temperature plasma such as the generation of ozone [1]. Atmospheric pressure plasma technology offers an attractive perspective in today's industrial processes due to the elimination of expensive vacuum equipment, easier handling of the samples [2, 3].

A dielectric barrier discharge (DBD) plasma reactor enables the generation of plasma active species at atmospheric pressure without expensive vacuum systems. The emission of UV-light and generation of radicals and charged particles contribute to the destruction of microorganisms in plasmas [4]. DBD method is used for the generation of ozone. DBD is most commonly generated in the filamentary mode when operated at atmospheric pressure [5]. This method finds its extensive applications in waste gas treatment, surface activation & treatment, carbon dioxide laser, excimer lamps and plasma screens [4-6]. A low frequency (50 Hz) AC source consisting of a conventional step up transformer and a variac may be the best choice for generation of ambient air DBDs [7].

The typical DBD) reactor, in which a small gap is formed for gas flow by inserting at least one dielectric layer on the surface of one of the electrodes, has always been considered as the reliable way to generate ozone by researchers. Due to its strong oxidizing property and environmentally friendly nature, ozone sees an increasing demand for considerable industrial applications including in food chemistry, medicine, water treatment, plasma assisted combustion an flue gas treatment and successfully used in wide industrial application including bacteria, algae, spores killing and vanishing volatile organic compounds, odor treatment, enhancing fertilization purification of ambient air and potable water, disinfecting food products to increase shelf life; fumigation of operation theaters in hospitals, sterilization of operational tools and personnel [8- 9].

When air is passed through the DBD, the interaction of high energy electrons with the oxygen molecules within the inter electrode space may give rise to the dissociation of oxygen molecules [10]. Ozone formed from oxygen atoms with O_3 chemical symbols. Ozone is a relatively unstable molecule compared to oxygen, which is very relatively stable. Ozone naturally can be formed by UV radiation, through the method of sunlight, which can reduce oxygen gas in the air. O_3 is smell-irritating, colorless, gas capable of oxidizing organic compounds [11]. Ozone is a powerful oxidizing agent of oxidation potential of 2.07 V and has relatively higher disinfection potential than chlorine and other disinfectants [12].

Ozone finds its application mainly in water treatment and air purification [13, 14]. The oxygen molecule breaks down into two-radical oxygen then it reacts with oxygen to form ozone [15]. Ozone is far more efficient in killing bacteria, viruses, spores and cysts [16. 17]. The role of electrons generated in electrical discharge is to excite and dissociate oxygen molecules [8, 15]. The design of non-thermal plasma reactor for generation of ozone in different gases environment and its applications in different fields have been presented and discussed by many researchers in several research works as well [18-39].

The main aim and objective of this work is to design of ozonizer by double dielectric barrier discharge to produce ozone and its effect and applications.

2. Materials and methods

2.1 Mechanism of Ozone Generation

The role of electrons generated in electrical discharge is to excite and dissociate oxygen molecules [18-39].

$$e + O_2 \rightarrow O + O + e \quad (1)$$

$$O + O_2 \rightarrow O_3 \quad (2)$$

$$O + O_2 + O_2 \rightarrow O_3 + O_2 \quad (3)$$

$$O + O_3 \rightarrow 2O_2 \quad (4)$$

$$2O_3 \rightarrow 3O_2 \quad (5)$$

$$3O_2 \rightarrow 2O_3 \quad (6)$$

The ozone formation reaction is given by [18-39]:

$$M + O + O_2 \rightarrow O_3 + M \quad (7)$$

$$O + O + M \rightarrow M + O_2 \quad (8)$$

In air discharge, M represents molecular nitrogen or oxygen. The ozone yield of a process depends on both concentration and input electric energy density with efficiency η in g/kWh which can be calculated by [26]

$$\eta = C\,(O_3)\,F_r / P \quad (9)$$

Where, $C\,(O_3)$ is the ozone concentration (ppmv), F_r is the gas flow rate (liter/min) and P is the discharge power (kW).

2.2 Experimental Setup Details

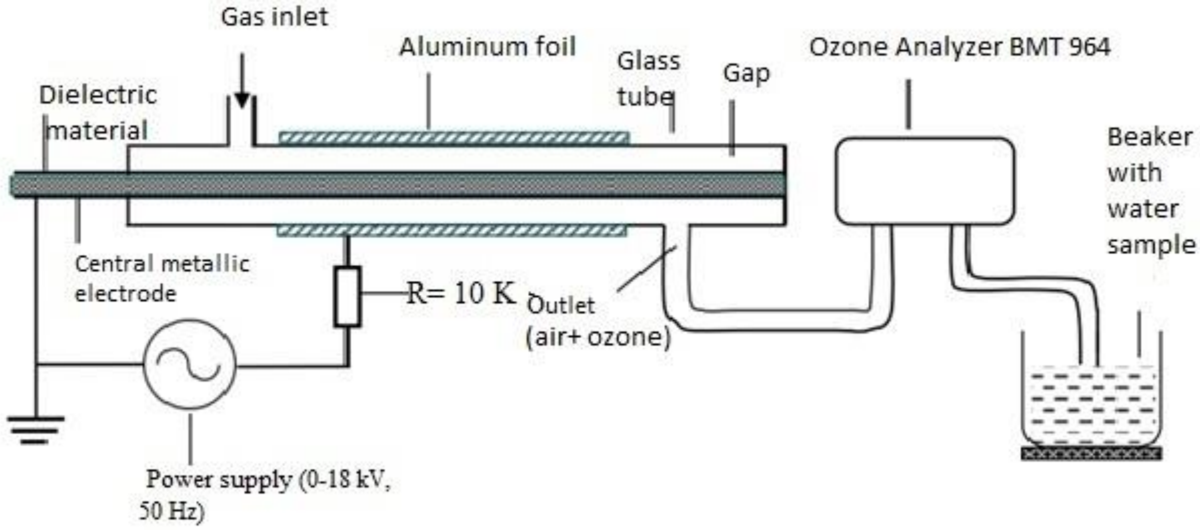


Fig.1: Schematic diagram of ozonizer for production of Ozone (0 – 18 kV, f = 50 Hz)

The production of O_3 by cylindrical DBD has been designed and an attempt to find the optimum condition for higher O_3 concentration in cylindrical glass tube reactor in air at atmospheric pressure is conducted. The schematic diagram of the experimental set up for production of O_3 in air and its measurement is shown in Fig. 1.

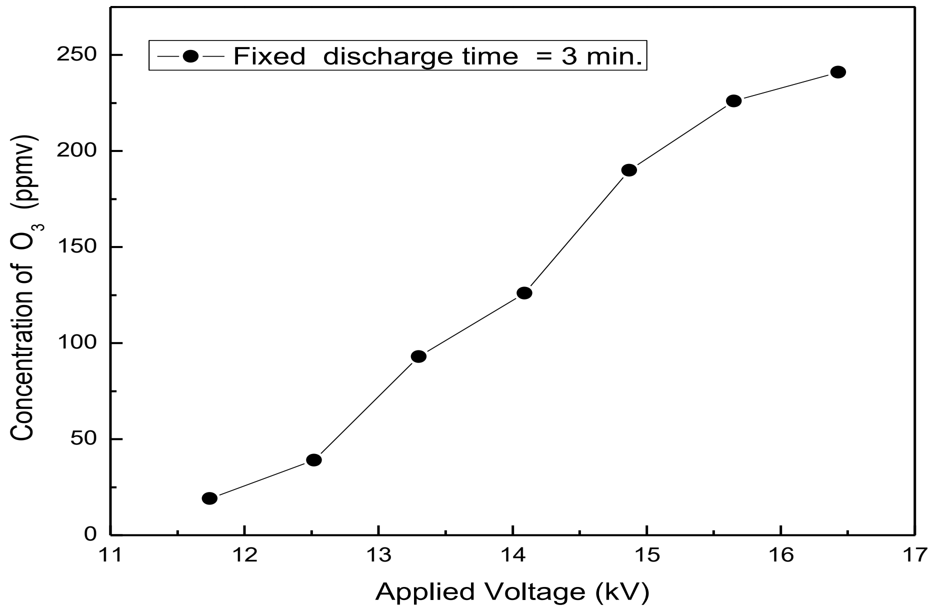
The discharge can be generated via line frequency (50 Hz) high voltage (maximum peak-to-peak value of 50 kV) power supply which is simply a step-up transformer with HT/LT ratio of 78.26. The central electrode is connected to the high voltage power supply through a ballast resistor of resistance 15 MΩ in series to limit the current. The outer electrode is grounded through a shunt resistor of resistance 10 kΩ across which the discharge current is measured. With this type of electrode configuration, as a discharge arrangement, at atmospheric pressure, electrical breakdown occurs in a very large number of randomly positioned, short-lived current filaments of a few nanoseconds duration referred to as micro-discharges. Thus, the filamentary DBD is an excellent source of micro-discharges containing energetic electrons. When the discharge occurred, ozone concentration was measured.

In this work, discharge from dielectric barrier was generated using high voltage AC source having voltage of 0-18 kV and operating at line frequency of f = 50 Hz was applied to the electrode system which was co-axial cylindrical geometry in type put inside the glass tube reactor of length 42 cm, internal diameter 16 mm, external diameter 18 mm and thickness 1 mm. The central electrodes were made up of copper and brass rods of length 45 cm, diameters 4 and 8 mm, fixed inside glass tube. The anode was connected to the central rod made of brass and copper placed inside the tube and cathode was connected by sheet of aluminum shielded outside of the tube. The central and outer electrodes which were separated by dielectric glass material. The air passed inside the tube through the gap between the anode and the glass tube. Aluminum foil of length 21 cm and thickness 0.5 mm wrapped outside the glass tube acted as outer electrode. Air was used as a working gas.

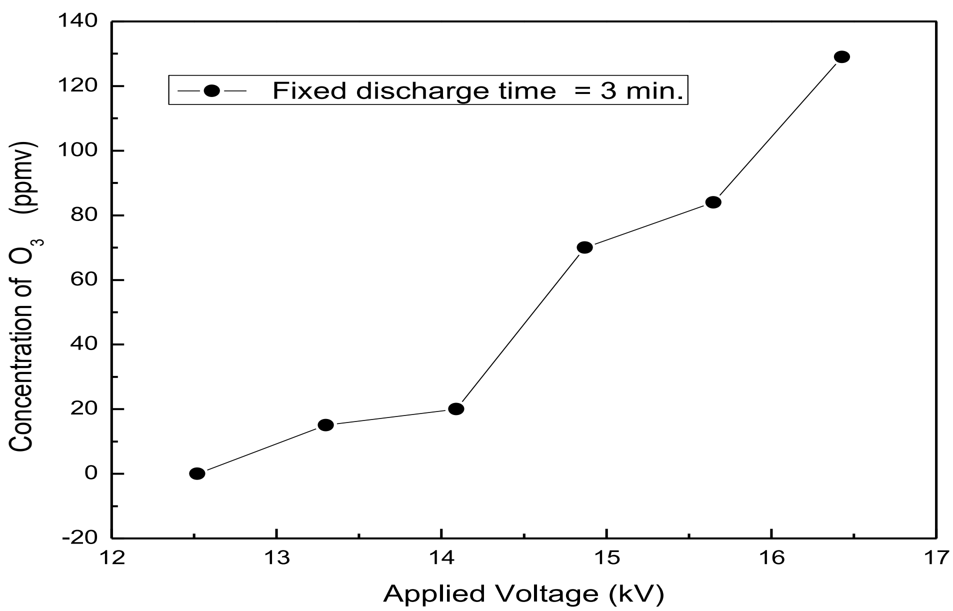
O_3 concentration was measured by using ozone analyzer BMT 964, Messtechnik GmbH, Germany after the discharge was developed. It is a microprocessor based dual beam photometer (UV 254 nm) for measuring the ozone content in air or oxygen. The ozone concentration is displayed in percent weight of ozone (%wt/wt), grams of ozone per normal cubic meter of sample gas (g/Nm^3) or ppmv. The concentration unit can be changed during operation.

The air passes inside the tube through the gap between the two glasses and it was supplied by air blower. The ozone analyzer can measure ozone concentration from 0 to 5000 ppmv after warm up pressure and time. Ozone was homogeneously bubbled into water with the help of spherical porous air stone to treat sample of water inside beaker to remove pungent smell or odor of ozone and prevent from its effect on human health.

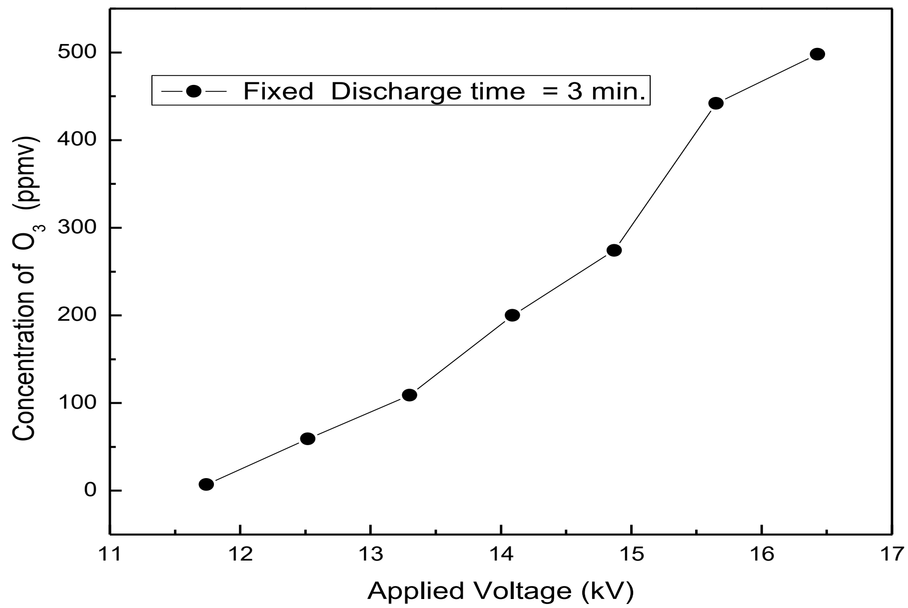
3. Results and Discussion



(a)



(b)



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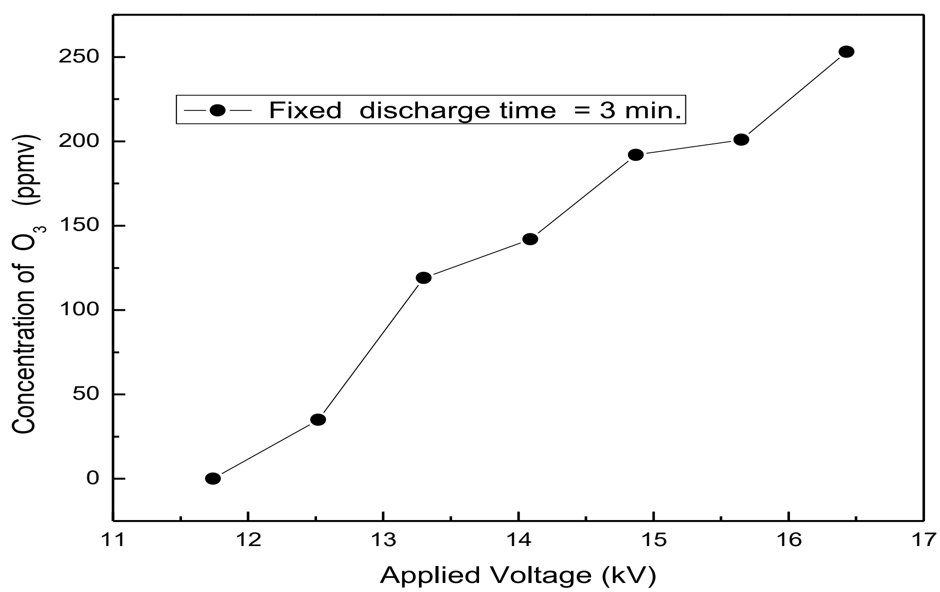
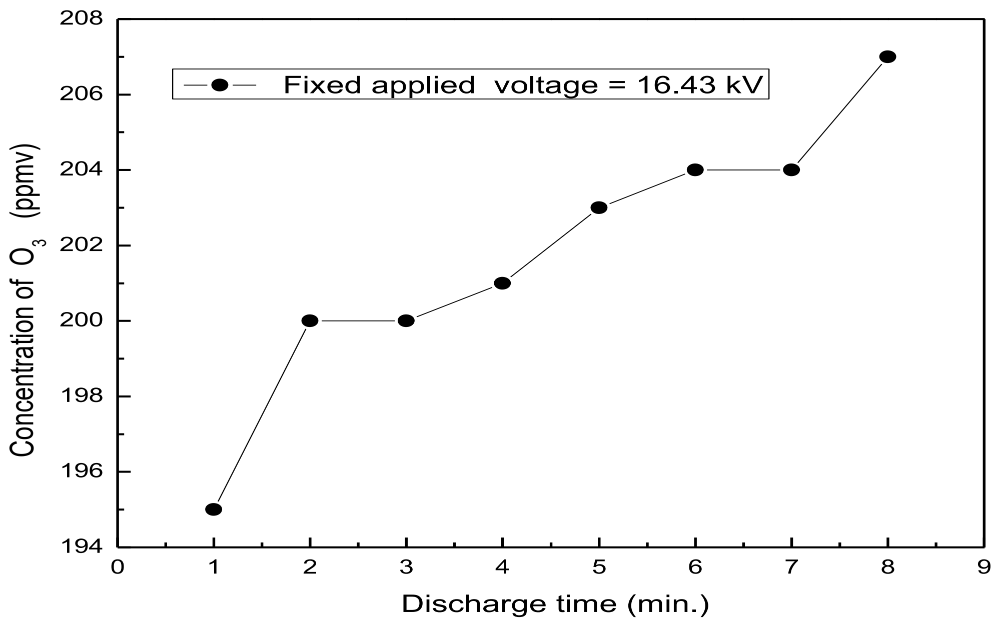
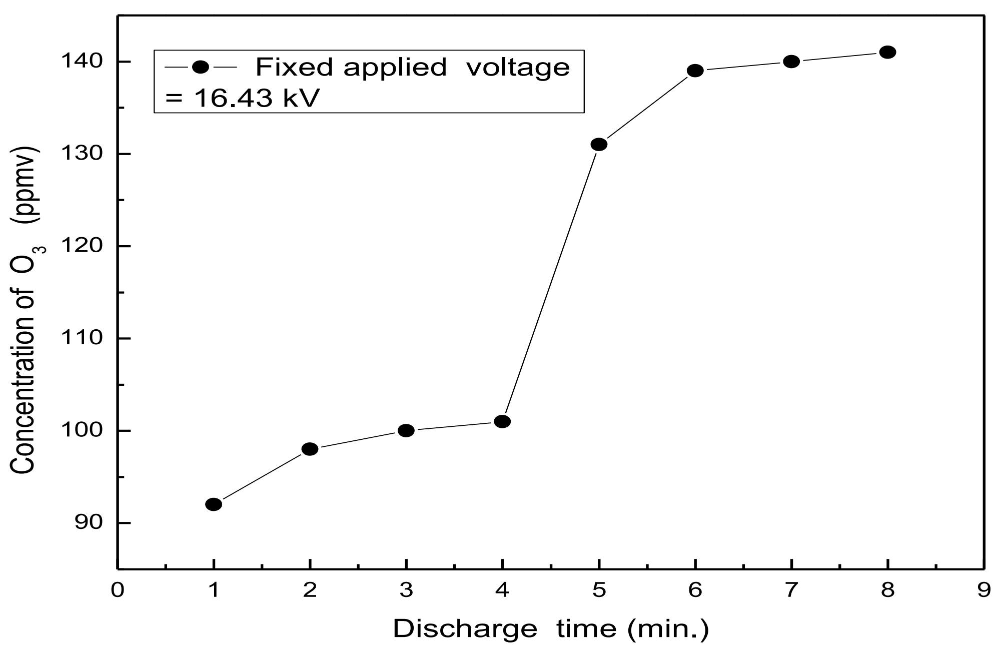


(d)

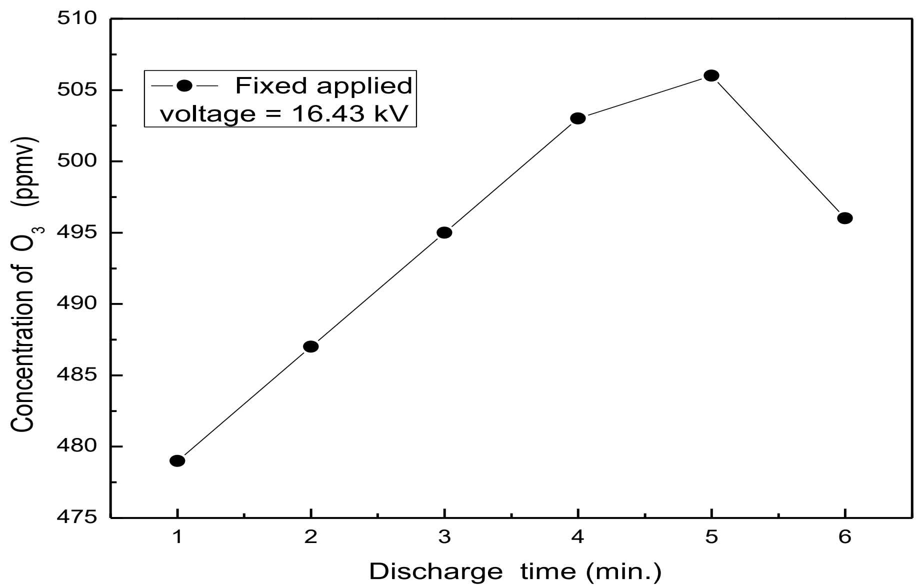
Fig. 2: O_3 concentration as a function of applied voltage for fixed discharge time and air flow rate for the brass electrode of diameter (a) d = 8 mm (b) and d = 4 mm, and the copper electrode of diameter (c) d = 8 mm and (d) d = 4 mm, and internal diameter of glass tube reactor 16 mm


Fixed applied voltage = 16.43 kV
Concentration of O_3 (ppmv)
Discharge time (min.)
208
206
204
202
200
198
196
194
0
1
2
3
4
5
6
7
8
9

(a)


Fixed applied voltage
= 16.43 kV
Concentration of O_3 (ppmv)
Discharge time (min.)
140
130
120
110
100
90
0
1
2
3
4
5
6
7
8
9

(b)



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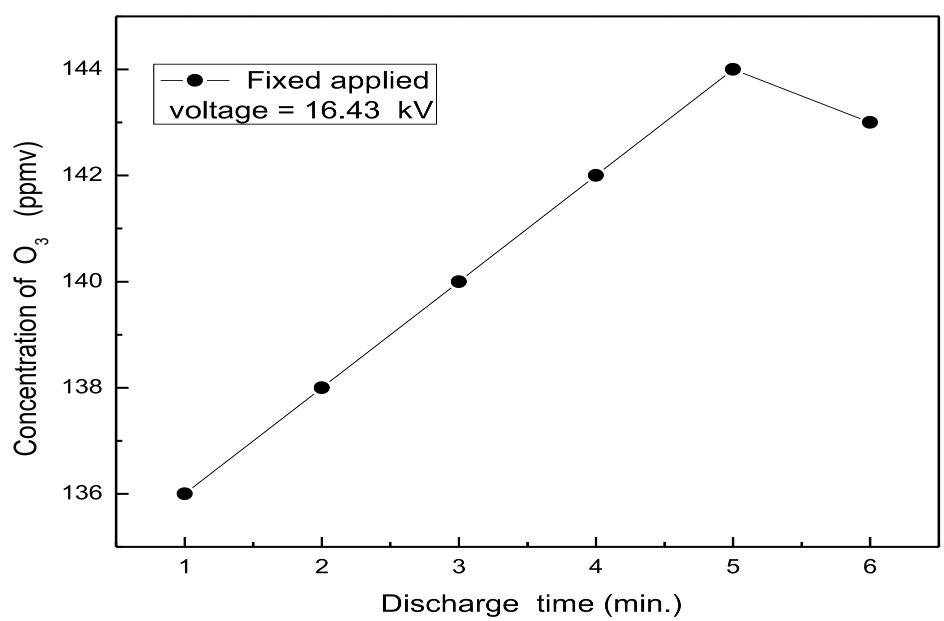
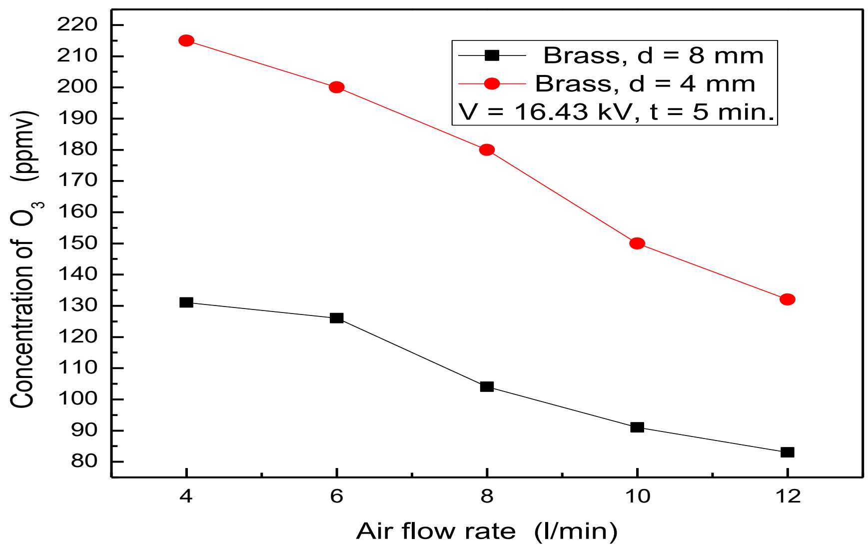
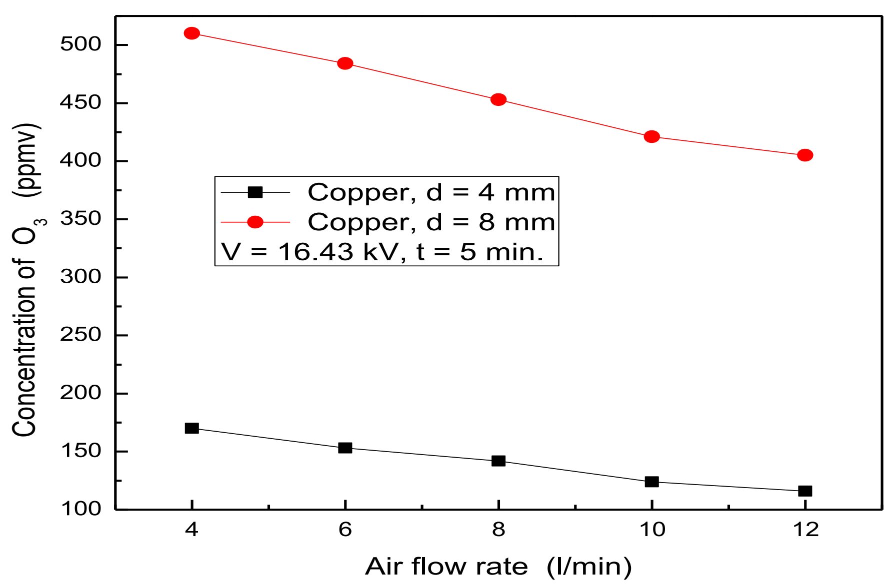


(d)

Fig. 3: O_3 concentration as a function of discharge time at fixed applied voltage and air flow rate for brass electrode of diameter (a) d = 8 mm (b) d = 4 mm and copper electrode of diameter (c) d = 8 mm and (d) d = 4mm and internal diameter of glass tube reactor 16 mm.



(a)



(b)

Fig. 4: O_3 concentration as a function of air flow rate at fixed discharge time and applied voltage for brass electrode of diameter (a) d = 8 mm and d = 4 mm and copper electrode of diameter (b) d = 8mm and d = 4mm and internal diameter of glass tube reactor 16 mm.

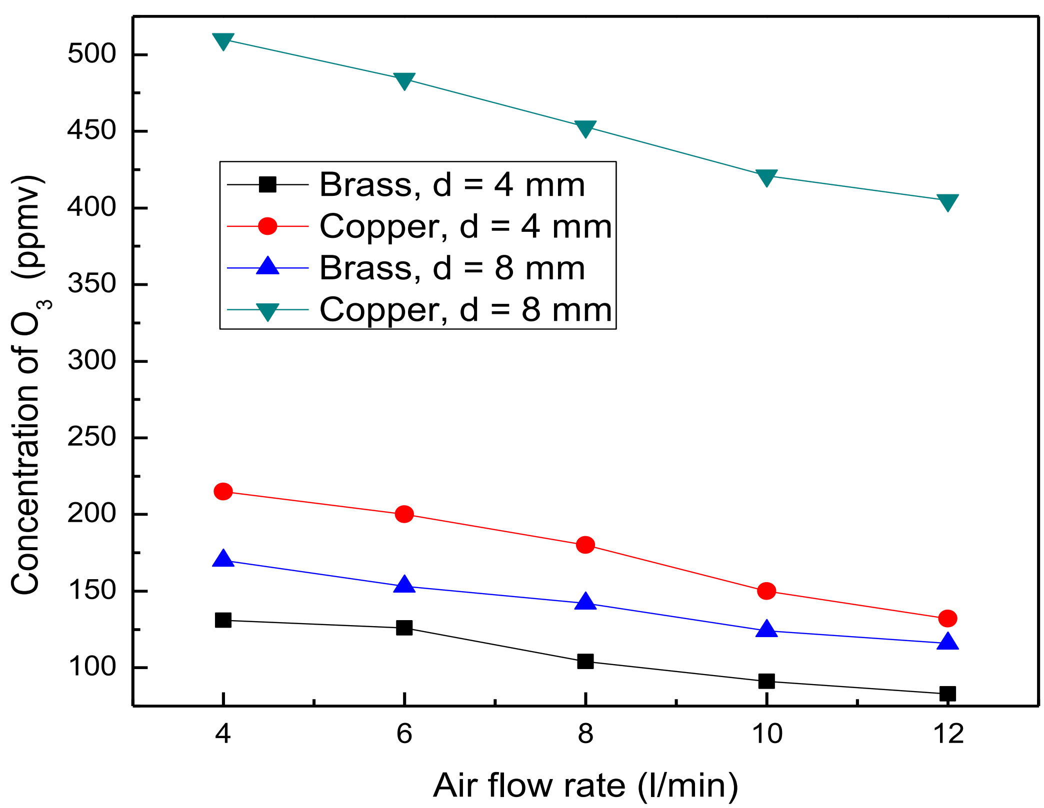


Fig. 5: O_3 concentration as a function of air flow rate at fixed discharge time (t) = 5 min. and applied voltage (V) = 16.43 kV for brass electrode of diameter d = 8mm and d = 4mm and copper electrode of diameter d = 8mm and d = 4mm and internal diameter of glass tube reactor 16mm.

Fig. 2 shows the O_3 concentration as a function of applied voltage at fixed discharge time 3 minutes for fixed flow rate 10 l/min of air for brass and copper central electrodes of diameters 8 and 4 mm using a double dielectric glass having thickness 1 mm and reactor of internal diameter 16 mm.

Fig. 3 shows the variation of O_3 concentration with discharge time at fixed applied voltage 16.43 kV for constant flow rate 10 l/min of air for brass and copper central electrodes of diameters 8 and 4 mm.

Fig. 4 and 5 show the variation of O_3 concentration with air flow rate at applied voltage 16.43 kV for constant discharge time of 5 minutes for brass and copper central electrodes of diameters 8 and 4 mm.

From Fig. 2, it was found that ozone concentration increases with increasing applied voltage. The reason is that the increase in applied voltage, the electrical energy density also increases i.e., more energy transferred to electrons which means increasing the possibility of collision of the air and oxygen in the reactor chamber. Voltage increase can produce higher electric field between two electrodes. High enough electric field can accelerate electron, molecules, or ions causing non-elastic collisions and finally augmenting the number of molecules ionized, radicalized, excited.

Consequently, under 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 ozone is produced. However, supplied energy may not always be sufficient for recombination of ions, radicals etc. This is because of the residence time of the gas on the ozone chamber.

From Fig. 3, it was found that ozone concentration increases with increasing discharge time and found more in copper electrode than brass electrode. Ozone concentrations increase with increasing discharge time. This is due to the increase of ozone concentration with increasing non-elastic collisions with oxygen molecules. The concentration of ozone was found more in copper than brass electrode. This is due to the reason that ozone concentration increases with increasing amount of non-elastic collisions with oxygen molecules. It is caused by reduced gas residence time with the increase of flow rate. Reaction to the formation of ozone in the discharge forms O_2 dissociation due to collision with electrons where at a constant flow rate, based on the number of the three body reaction rate, it is much slower compared to the dissociation of electron collision impact.

The residence time of gas on the reactor chamber is inversely related to the air flow rate. The increase residence time provides time for a reaction to occur and a correspondingly more ozone concentration to be generated. This is because of the residence time of the gas on the ozone chamber. From Fig. 4 and 5, the ozone concentration decreases with increasing air flow rate. The reason is the composition of air which is more complex and consists of different species such as Ar, N_2, O_2, He, H_2 etc.

The ozone production is gradually suppressed when flow rate increases because atom O generated runs out more quickly following the flow rate of the gas that comes out more quickly, without causing any ozone production so the concentration decreases with the increase of flow rate. The low concentration of ozone with feed gas air has low number of oxygen molecules per unit volume of air. When the applied voltage increases the ozone begins to be formed and its concentration increases rapidly with applied voltage for all values of the flow rate. The air flow rate itself also affects the ozone generation efficiency because of increasing cooling rate with air cooled zone reactor.

The ozone concentration is found to be more in copper electrode than brass electrode in air as a feed gas. This is because of higher electrical conductivity of copper than brass electrode. The effects of gap spacing on the electric field and power density in the discharge, larger discharge gap leads to the formation of a significantly different kind of discharge. Ozone concentration increases with increasing diameter of central electrodes in air as feed gas. This is because for fixed diameter of reactor, the diameter of central electrode increases, the gap space become smaller and less electric field or electric energy is required to break air and oxygen molecules. The ozone concentration is found to be more in copper electrode than brass electrode in air as feed gas. This is because of higher electrical conductivity of copper than brass electrode.

4. Conclusions

The concentration of ozone produced, increases with increasing voltage with the fixed discharge time and air flow rate. This concentration also increases with increasing discharge time at fixed applied voltage and air flow rate. The concentration of ozone increases with increasing gap space between the electrodes. But this concentration decreases with increasing the air flow rate.

For an efficient and cost effective ozone yield, smaller reactor and larger inner electrode will achieve better energy efficiency at the same specific energy density in that the mean electron energy is higher so that most of the energy can be used to produce atomic oxygen, the precursor for ozone formation. But for reactors with small diameters, there exists an optimum inner electrode diameter.

We have designed ozone reactor and used copper and brass as central electrode and air as a feed gas to produce ozone. The ozone concentration was higher when using copper electrode in air than brass electrode for fixed discharge time, applied voltage and reactor diameter. Ozone concentration increases with increasing diameter of central electrodes .and it was found higher in copper than brass electrode. This is because copper has high electrical conductivity and can produce high concentration of O_3, brass electrode. But basically brass electrode is used for the generation of O_3 due to its high mechanical strength and electrical resistance in the ozonizer. For the increase O_3 concentration, the gap distance is optimized in ozone generator.

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