

Ozone Production in Cylindrical Co-axial Double Dielectric Barrier Discharge Ozone Generator

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Abstract

This study developed an ozone generator of a double co-axial cylindrical dielectric barrier discharge system with air, argon, and oxygen as the working gases. The discharge was produced by using a high voltage power supply of 0 - 18 kV and a line frequency of 50 Hz. The flow rate of air, argon, and oxygen was varied from 1 to 6 L/min. A comparison of O₃ generation in air, argon, and oxygen using brass as a central electrode was conducted and it was found that O₃ concentration was higher in the case of oxygen than in the air and in argon gases environment for given fixed discharge time, applied voltage, and diameter of the brass electrode. This study revealed that the concentration of ozone increased along with the increase in the applied voltage for constant discharge time and gas flow rate. The O₃ concentration also increased with the increase in the discharge time at fixed applied voltage and gas flow rate; however, the concentration decreased with the increase in the gas flow rate at fixed discharge time, applied voltage, and diameter of the electrode. A small reactor with a large inner electrode generated a high concentration of O₃. Yet, a reactor with a small diameter, there seemed to have an optimum inner electrode diameter. The glass tube reactor of the internal diameter of 18 mm and the brass electrode of diameter 8 mm were utilized in this study. The ozone concentration was higher for oxygen as feed gas than both in the air and in argon and the O₃ concentration was also higher in the air than in argon at fixed discharge time, applied voltage, and diameter of ozone generator.

Keywords: Applied voltage, Brass electrode, Double dielectric barrier discharge, Ozone, Ozone analyzer

Introduction

Dielectric Barrier Discharge (DBD) non-thermal or cold plasma reactor enables the production of plasma active species at atmospheric pressure without expensive vacuum systems. The emission of UV light and the generation of radicals and charged particles contribute to the destruction of microorganisms in cold plasmas [1,2]. DBD technique is used for the generation of ozone and is most commonly produced in the filamentary mode when operated at atmospheric pressure [3]. DBD technique finds its extensive applications in waste gas treatment, surface activation and treatment, carbon dioxide laser, excimer lamps, and plasma screens [2-4]. A low line frequency (50 Hz) AC source consisting of a conventional step-up transformer and a variac may be the best choice for the formation of ambient air DBDs [5].

DBD generator, in which a small gap is formed for gas flow by inserting at least 1 dielectric layer on the surface of one of the electrodes, has always been considered as the reliable way to generate ozone by researchers and 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 and flue gas treatment and successfully used in the 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 [6,7].

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 [8]. Ozone is formed from oxygen atoms with O₃ chemical symbol. 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. O₃ is smell or odor irritating, colorless, capable of oxidizing organic compounds [9]. Ozone is a powerful oxidizing agent of the oxidation potential of 2.07 V. It has relatively higher disinfection potential than chlorine and other disinfectants [10]. Ozone finds its application mainly in water treatment and air purification [11,12].

The oxygen molecule breaks down into 2 radicals oxygen then reacts with oxygen to form ozone [13]. Ozone is far more efficient in killing bacteria, viruses, spores, and cysts [14,15]. This study aims to design of ozone generator by double dielectric barrier discharge of glass tube to generate ozone in air, argon, and oxygen gases environment at atmospheric pressure.

Materials and methods

Ozone production mechanism in double DBD reactor

The role of electrons generated in electrical discharge is to excite and dissociate oxygen molecules [16-24].



where e is the charge of the electron, O is atomic oxygen, O₂ is the oxygen molecule and O₃ is the ozone molecule.



The ozone formation reaction is given by [18-26]:



In air discharge, M represents molecular N₂ or O₂. The ozone yield of a process depends on both concentration and input electric energy density with efficiency η in g/kWh which can be estimated by the following equation [24].

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

where $C(O_3)$ is the ozone concentration in ppmv, F_r is the gas flow rate (L/min) and P is the discharge power (kW).

Experimental set-up and procedures description

The production of O_3 by cylindrical DBD was designed to find the optimum condition for higher O_3 concentration in cylindrical glass tube reactor in the air, in argon, and in oxygen at atmospheric pressure. The schematic diagram of the experimental set-up for the production of O_3 and its measurement is shown in **Figure 1**. A high voltage AC source having a voltage of 0 - 18 kV and frequency of 50 Hz was applied to co-axial cylindrical geometry of glass tube reactor of internal diameter 18 mm and thickness 1 mm. The discharge was generated via line frequency of 50 Hz high voltage (maximum peak to the peak value of 50 kV) power supply which was simply a step-up transformer with an HT/LT ratio of 78.26. The central electrode was connected to the high voltage power supply through a ballast resistor of resistance 15 M Ω in series to limit the current and to avoid overheating of the DBD reactor was the total effective impedance of the DBD reactor. The outer electrode was grounded through a shunt resistor of resistance 10 k Ω across which the discharge current and voltage were measured. The central electrode was made up of a brass rod of diameter 8 mm, fixed inside the glass tube. The brass electrode was used for its resistance to oxidation and corrosion when exposed to ozone. The dielectric layers were glass tubes. Although the quartz window is better in spectral transmittance than glass, glass has been conveniently obtained, as they are economically available. Two ports on the top and bottom cover of the reactor were used as the gas inlet and outlet. Aluminum foil wrapped outside the glass tube acted as an outer electrode. Air, argon, and oxygen were used as the working gases. The O_3 concentration was measured by using ozone analyzer BMT 964 after the discharge was formed. The air passed inside the tube through the gap between the 2 glasses and it was supplied by air blower and argon and oxygen were supplied through gas cylinders. The ozone analyzer can measure ozone concentration from 0 to 5000 ppmv after warm-up pressure and time.

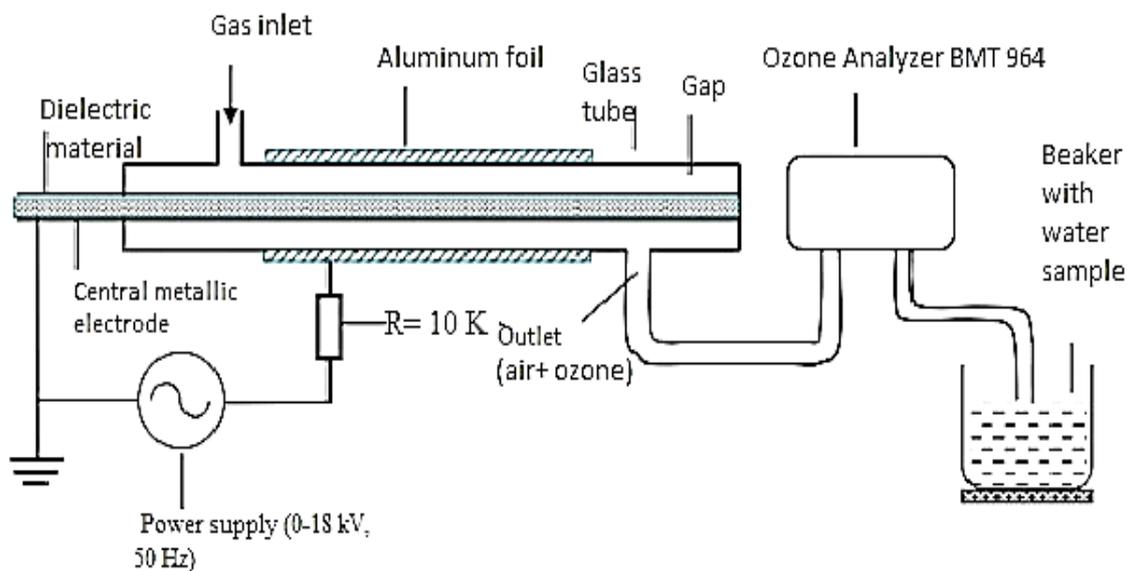
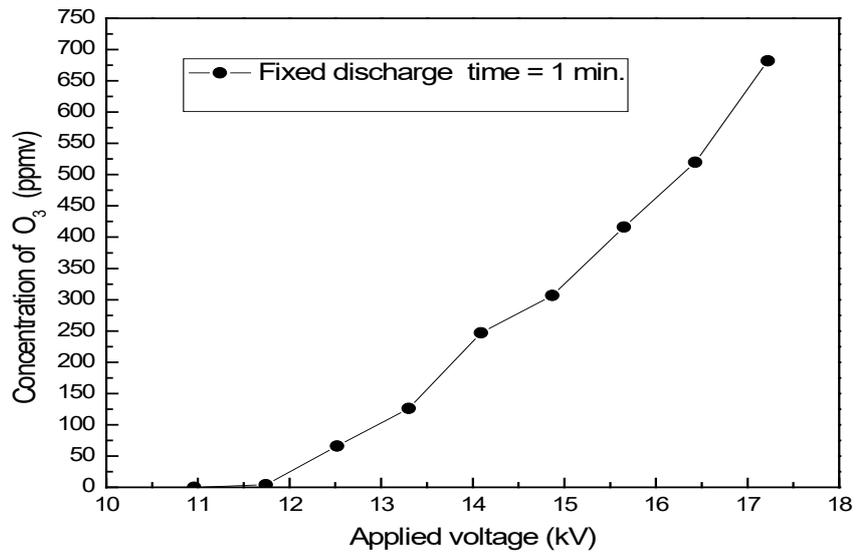


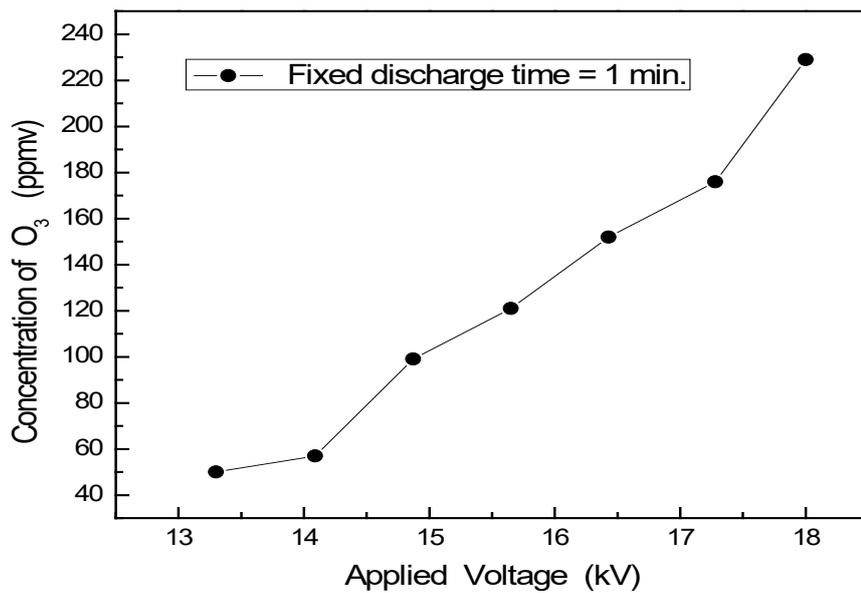
Figure 1 Schematic diagram of ozonizer for production of ozone.

Results and discussion

Figure 2 shows the plot of O₃ concentration with applied voltage at fixed discharge time 1 min for a fixed flow rate of air and argon 2 and 1 L/min, respectively for the brass central electrode of diameters 8 mm using a double dielectric glass having thickness 1 mm and reactor of internal diameter 18 mm. **Figure 3** shows the plot of O₃ concentration with discharge time at fixed applied voltage of 17.28 kV for a constant flow rate of air 2 and argon 1 L/min, respectively for brass central electrode of diameters 8 mm. **Figure 4** shows the plot of O₃ concentration with air, argon, and oxygen flow rate at applied voltage 17.28 kV for constant discharge time of 1 min for the brass central electrode of diameters 8 mm. **Figure 5** shows the plot of O₃ concentration versus gas flow rate for the brass electrode of diameter 8 mm and reactor of internal diameter 18 mm at fixed applied voltage of 17.28 kV and discharge time 1 min. From **Figures 2(a) - 2(b)**, it was found that ozone concentration increases with increasing applied voltage at fixed discharge time and fixed gas flow rate for both air and argon environments. This is because of 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. A voltage increase can produce a higher electric field between 2 electrodes. A high enough electric field can accelerate electrons, molecules, or ions causing non-elastic collisions and 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 in the ozone chamber. Ozone concentration was high in the case of air as feed gas than argon due to inert gas nature of argon which need more applied voltage for a long discharge time to generate ozone and argon reduces the dissociation of oxygen molecules to form nascent oxygen to combine with an oxygen molecule to produce ozone. From **Figure 3(a) - 3(b)**, it was found that ozone concentration increases with increasing discharge time and found more in the air than in argon. Ozone concentrations increase with increasing discharge time. This is due to the increase of ozone concentration with increasing non-elastic collisions with oxygen molecules. This is due to the reason that ozone concentration increases with an 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 O₂ dissociation due to collision with electrons where at a constant flow rate, based on the number of the 3 body reaction rates, 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 airflow rate. Increasing residence time providing time for a reactor to occur and a correspondingly more ozone concentration to be formed. This is because of the residence time of the gas in the ozone chamber. From **Figures 4(a) - 4(c)** and **5**, the ozone concentration decreases with increasing gas flow rate. The reason is the composition of air which is more complex and consists of different species such as Ar, N₂, O₂, He, H₂, etc. The ozone production is gradually suppressed when the 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 a 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 gas flow rate itself also affects the ozone generation efficiency because of the increasing cooling rate with air-cooled ozone reactor. The ozone concentration is found to be more in oxygen than in both air and argon. This is because oxygen easily combines with atomic oxygen to generate ozone in the generator than air which is more complex and argon is an inert gas which produce low ozone concentration than air. 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.

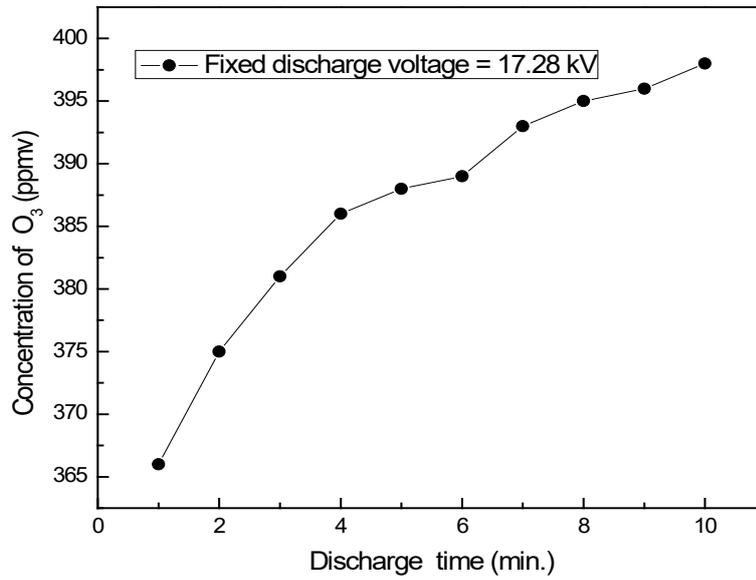


(a)

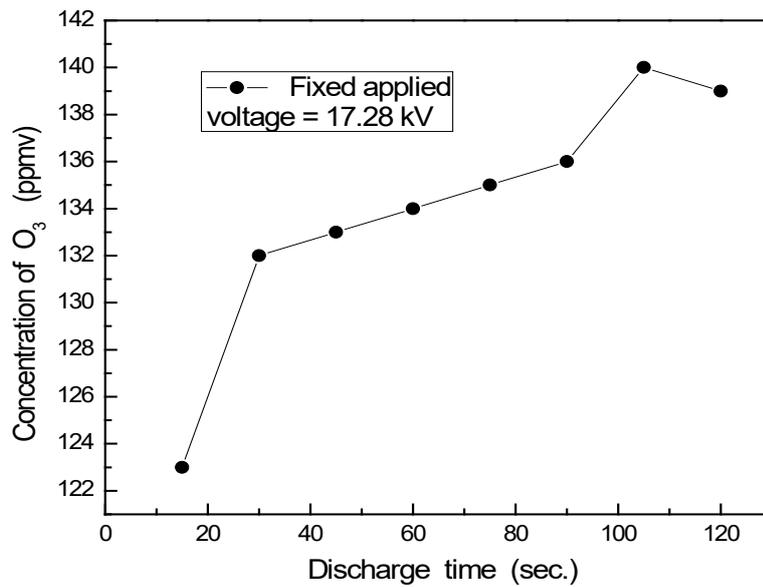


(b)

Figure 2 Plot of O₃ concentration versus applied voltage for the brass electrode of diameter 8 mm and reactor of internal diameter 18 mm in (a) Air (b) Air + Ar (fixed airflow rate 2 L/min and Ar flow rate 1 L/min).

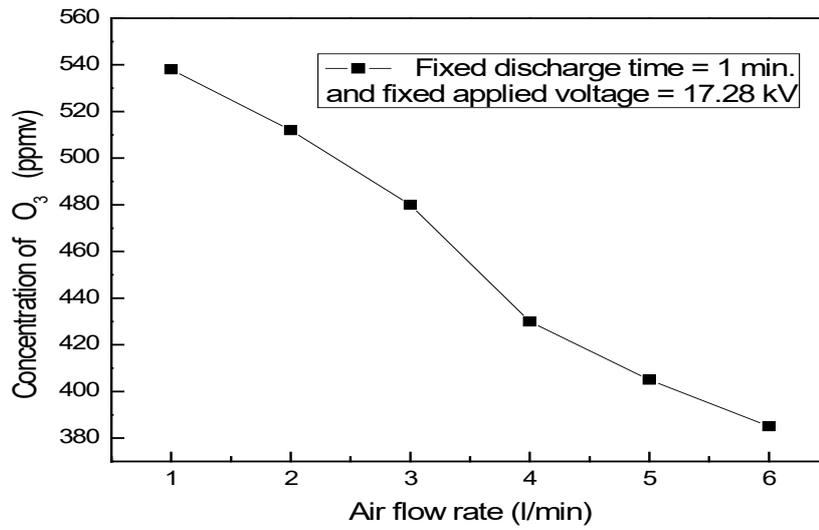


(a)

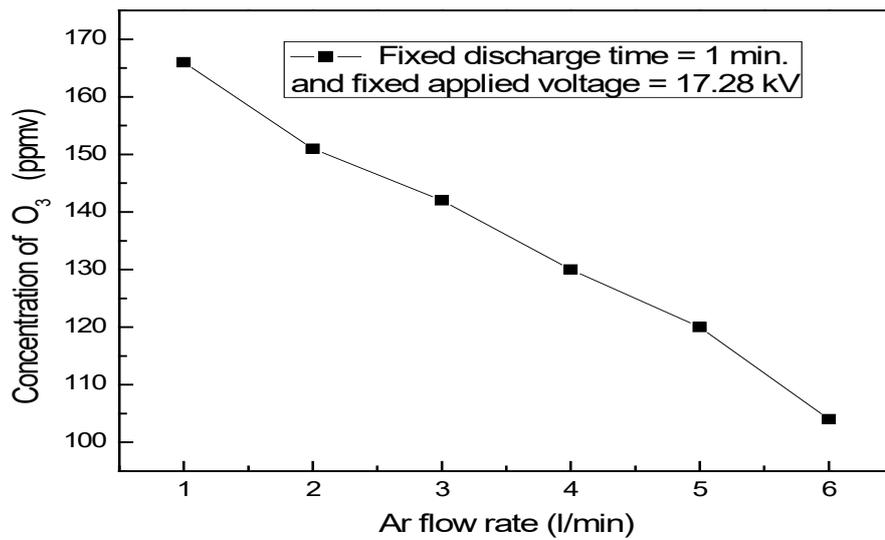


(b)

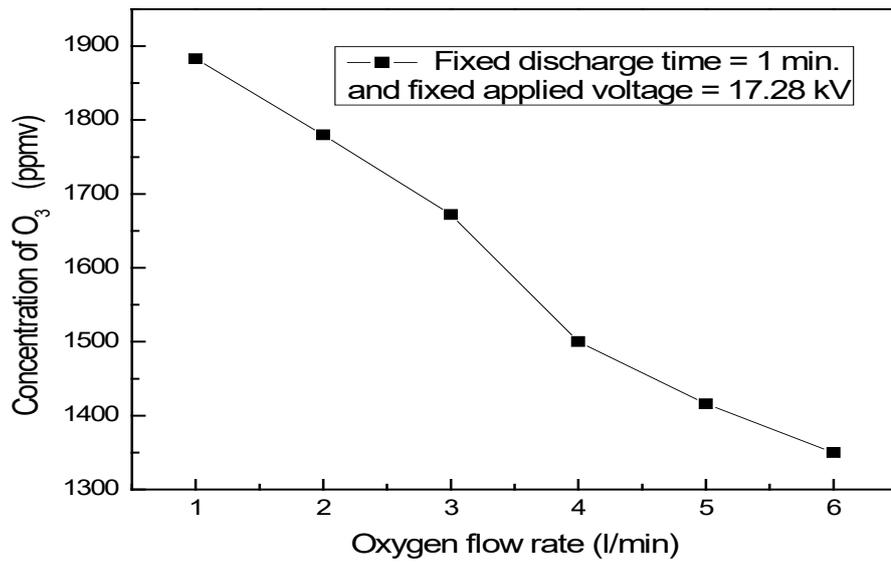
Figure 3 Plot of O₃ concentration versus discharge time for the brass electrode of diameter 8 mm and reactor of internal diameter 18 mm in (a) Air (b) Air + Ar (fixed airflow rate 2 L/min and Ar flow rate 1 L/min).



(a)



(b)



(c)

Figure 4 Plot of O₃ concentration versus flow rate for the brass electrode of diameter 8 mm and reactor of internal diameter 18 mm at a fixed applied voltage of 17.28 kV and discharge time 1 min in (a) Air (b) Air + Ar (c) Air+O₂.

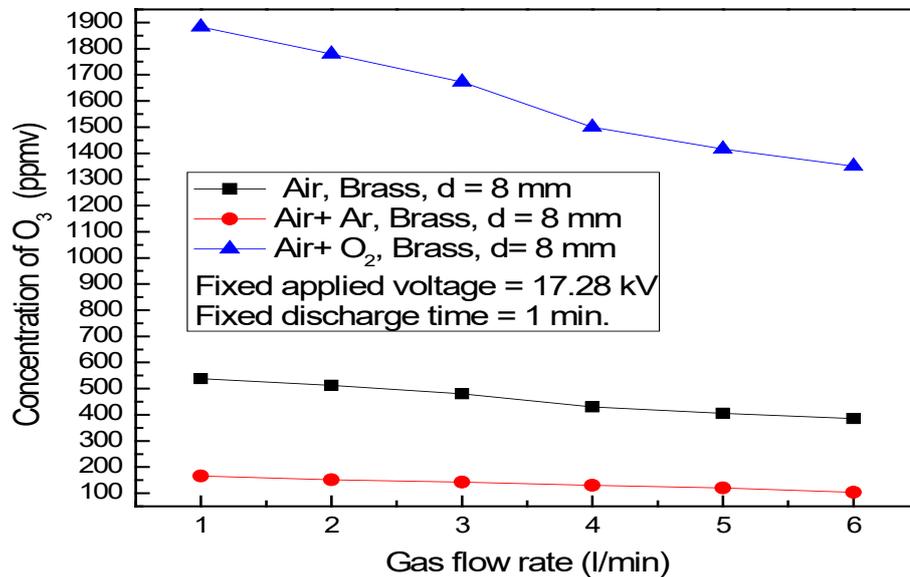


Figure 5 Plot of O₃ concentration versus gas flow rate for the brass electrode of diameter 8 mm and reactor of internal diameter 18 mm at a fixed applied voltage of 17.28 kV and discharge time 1 min.

Conclusions

In this work, we have found that ozone concentration increases with increasing applied voltage at fixed discharge time and gas flow rate. This concentration also increases with increasing discharge time at fixed applied voltage and gas flow rate. But this concentration decreases with increasing the gas 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 developed an ozone generator and used brass as a central electrode and air, argon, and oxygen as the feed gases to produce ozone. The ozone concentration was higher when using oxygen as feed gas than air and argon for fixed discharge time, applied voltage, and reactor diameter. The gap space is optimized in the ozone generator to increase O₃ concentration, yield, and efficiency.

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