Suppression of the ozone generation in the positive and negative dc corona discharges

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Abstract— The effect of the electrode material on ozone generation in dc corona discharges has been reported, however, some of the reported results conflict. The aim of this paper is to investigate how the characteristics of the dc corona discharges are influenced by material of the discharge wire in both dry air and oxygen-fed reactors. The corona discharges were generated in a coaxial wire-cylinder reactor stressed by positive or negative dc voltage. A set of high purity wires made of different metals, having a constant radii, was tested as discharge electrodes in the reactor. The reactor was fed by either dry air or oxygen flowing with a constant rate at atmospheric pressure and room temperature. As the result, the effect of wire material on the ozone generation characteristics was the most significant with the positive corona in dry air, and not so significant with the negative corona in dry air, nor with both positive and negative coronas in oxygen. It was also confirmed that silver wire was effective to suppress the ozone production in dry air up to about 1/10 compared with that using tungsten wire.

Keywords— Corona discharge, DC corona discharge, Ozone, Ozone generation, Electrostatic precipitators.

I. Introduction

There are numerous applications of corona discharges. Typical examples are electrostatic precipitators in industry and in indoor air cleaning, spray coating and electro photography machines. The corona discharges are also used in dry-ore separation systems and in radiation detectors. Other important applications of the corona discharges include the surface treatment of polymers and the elimination of NO_x and SO_x from flue gases [1].

The importance of ozone generation in design of any equipment utilizing the corona discharges is well recognized. Ozone, which is one of the most powerful oxidants known, is very toxic to humans, animals and plants, and causes deterioration in many materials. Therefore, any equipment utilizing the corona discharge and is being used in real applications, where the air will be directly breathed by humans or animals, or will come into contact with sensitive plants and materials, should be designed to ensure that the ozone level does not exceed acceptable limits [2].

Among the parameters that influence the ozone generation by dc coronas is the material of the discharge electrode surrounded by the corona plasma. The previous studies, which concerned with studying effect of the wire material on characteristics of the corona discharges, are limited in the literatures. Two of these studies [3, 4] include some of the experimental results for effect of the wire material on the ozone generation by dc corona discharges in air. Moreover, there is a conflict between the results reported in these studies. In [3], it was found that the ozone generation by positive coronas depends on the wire material, while no significant change for the ozone generation was observed with negative coronas. On the contrary, it was found in [4] that the ozone generation depends on the wire material for both positive and negative corona discharges. With respect to the ozone generation by dc corona discharges in oxygen, some of the investigators [5] have only tested pure copper wire, while the other wires were composed of different metals (i.e., alloys).

In this paper, the current-voltage and the ozone generation characteristics of the dc corona discharges in both dry air and oxygen have been studied by using high purity wires made of different metals as discharge electrodes in the reactor. This has been aimed to clarify the conflicting results reported in the previous studies on the effect of wire material on ozone generation, and to investigate the effect on the voltage-current characteristics of corona discharge in both dry air and oxygen.

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II. Experimental setup and measuring technique

A. Experimental setup

Figure (1) shows a schematic diagram of the experimental setup used in this study. It was composed of the following:

(a) High-voltage dc power supply

This consisted of a step-up transformer (0.1/20 kV_{rms}, 60 Hz, 1 kW), a rectifying diode (100 kV, 20 mA), a

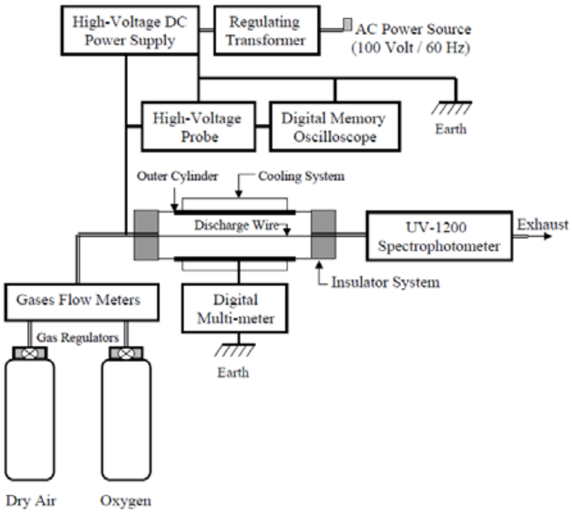

Fig. 1. Schematic diagram of the experimental setup.

charging capacitor (40 kV, 64 nF) and a load resistor (500 kΩ, 250 kV).

(b) Coaxial wire-cylinder reactor

It was made of aluminum cylinder having 15 cm length, 2 cm inner diameter, and 1.5 mm thickness. The edges of the cylinder were shaped to a semi-circle, and polished to avoid the local discharges. The discharge wire was tensioned coaxial with the cylinder by using insulator system, Figure (1). A set of high purity wires (99.9 %) made of different metals, and has standard radii of 0.2 mm, was used discharge electrodes in the reactor.

(c) Gas flow system

The reactor was fed by either dry air or oxygen from high pressure cylinders connected to it through gas flow meters.

B. Measuring technique

The reactor was connected to both the high voltage dc power supply and the gas flow system as shown in Figure (1). The ac voltage input to the power supply was adjusted with a regulating transformer (500 W). The net output positive (or negative) dc voltage was applied to the discharge wire of the reactor and measured by using a 1000: 1 high voltage probe (Tektronix P6015A, 75 MHz), and displayed on a two-channel digital real time memory oscilloscope (SONY-TDS360P, 200 MHz). The outer cylinder was earthed through a digital multi-meter (IWATSU-7411) for measuring the corona current. The flowing gas inside the reactor was controlled at a constant rate of one liter/minute. The ozone concentration generated in the gas flowing out the reactor was determined from absorption measurements with a quartz cell fixed in a UV-1200 spectrophotometer operated at a wavelength of 253.7 nm. The reactor was cooled by using water flow of two liters/minute at the room temperature through a rubber hose wound helically along the outer cylinder, Figure (1). The ambient temperature varied in the range of 8-12 ºC during the course of the measurements.

III. Results and discussion

A. The Current-Voltage Characteristics

Figures (2)-(5) show effect of the wire material on the current-voltage characteristics of positive and negative dc corona discharges in both dry air and oxygen. The curves in these figures summarize the following:

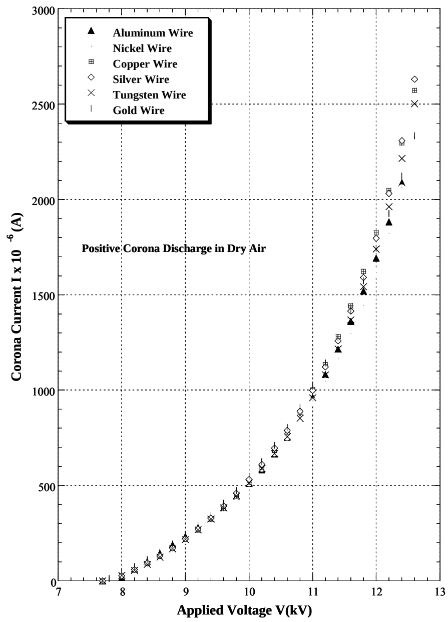

Fig. 2. Current-voltage characteristics of the positive corona discharge in dry air for different materials of the wire.

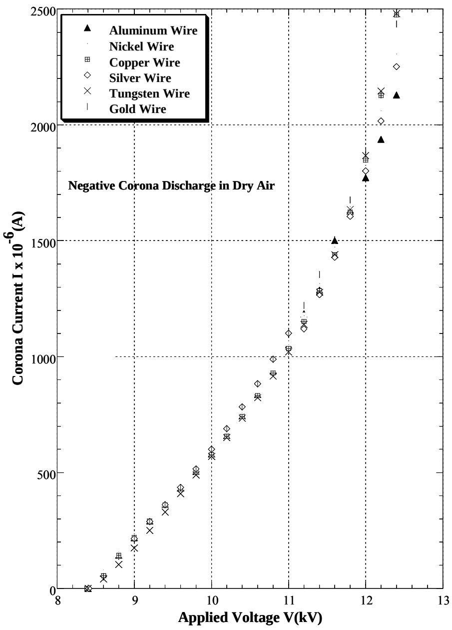

Fig. 3. Current-voltage characteristics of the negative corona discharge in dry air for different materials of the wire.

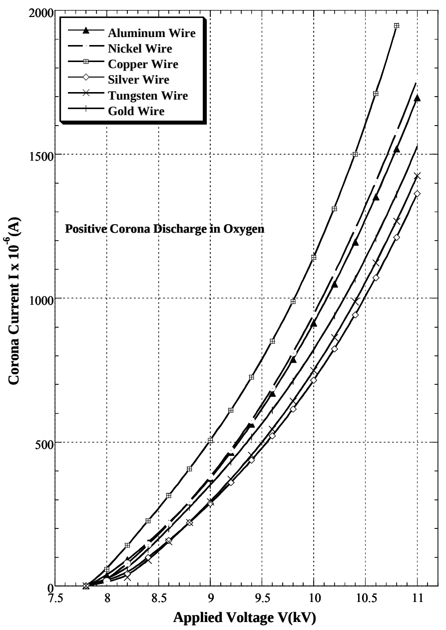

Fig. 4. Current-voltage characteristics of the positive corona discharge in oxygen for different materials of the wire.

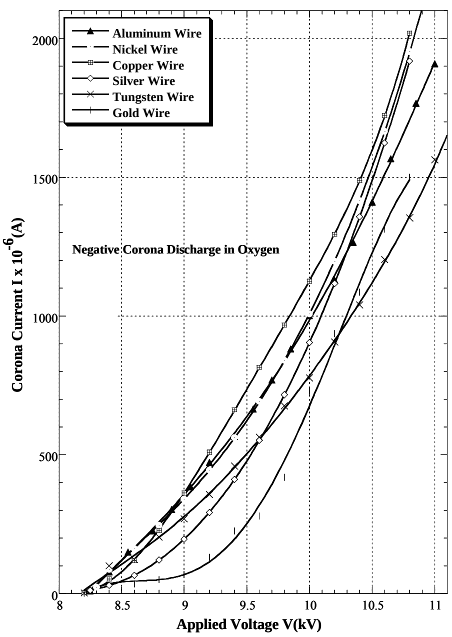

Fig. 5. Current-voltage characteristics of the negative corona discharge in oxygen for different materials of the wire.

The corona onset voltage is not affected by the wire material, whatever polarity of the corona or type of the flowing gas inside the reactor. The onset voltage of the positive coronas in either dry air or oxygen is slightly lower than that of the negative coronas.

The wire material does not affect the current-voltage characteristics of both positive and negative corona discharges in dry air. However, small differences in the corona currents were only recorded with the different wires in the intense discharge range. On the contrary, the wire material affects the current-voltage characteristics of both positive and negative corona discharges in oxygen. The current-voltage characteristics of the positive coronas have regular curves with the different wires, while those of the negative coronas have overlapping between the curves of the different wires. This may be due to the high stability of the positive coronas in comparison with the negative coronas [6].

The corona current is larger in oxygen than that in dry air when the discharge conditions are held constant (i. e. the wire material, polarity of the corona discharge and the voltage applied to the reactor). This difference in the corona current can be explained by the fact that the oxygen content in air is about 20% by volume in addition to the excitation and ionization energies of oxygen are lower than that of nitrogen [6]. Therefore, the more oxygen molecules are involved within the corona discharge plasma, the more positive or negative ions are created within the drift region inside the reactor, with a subsequent increase of the corona current at the same discharge conditions. This explains why the corona discharges yield larger current in oxygen than that in dry air at the same discharge conditions.

B. *The Ozone Generation Characteristics*

Figures (6)-(9) show how the material of the wire in the reactor influences the ozone generation characteristics of positive and negative dc corona discharges in both dry air and oxygen. The curves in these figures show the following:

In general, the ozone concentration generated in the reactor increases rapidly with the increase of the corona current in the weak discharge range whatever the operating conditions. This behavior is due to the absence of ozone destruction processes with the weak corona currents and low temperatures of the corona discharge plasma [7]. With further increase of the corona current, the ozone concentration mostly reaches saturation in the intense discharge range, where the corona discharge plasma destroys as much ozone as it creates [1, 8]. The saturated ozone concentration sometimes followed by a decrease in the ozone concentration with some of the investigated wires, especially with feeding the reactor by dry air. This is explained by the increase in temperature of the corona discharge plasma, where the rate of ozone decomposition increases while the rate of ozone formation decreases [4]. Also, the formation of nitrogen oxides (NO_x) led to an enhanced catalytic recombination of the oxygen atoms which are no longer available for ozone formation, with subsequent enhanced ozone destruction [8]. This explains why the saturated ozone concentration followed by a decrease in the ozone concentration with some of the discharge conditions.

The wire material generally affects the ozone generation characteristics of both positive and negative corona discharges in either dry air or oxygen. However, the effect of the wire material on these characteristics is the most significant with the positive corona discharges especially in dry air and not so significant with the negative corona discharges in either dry air or oxygen. The difference between the positive and negative corona discharges results from the direction of electron

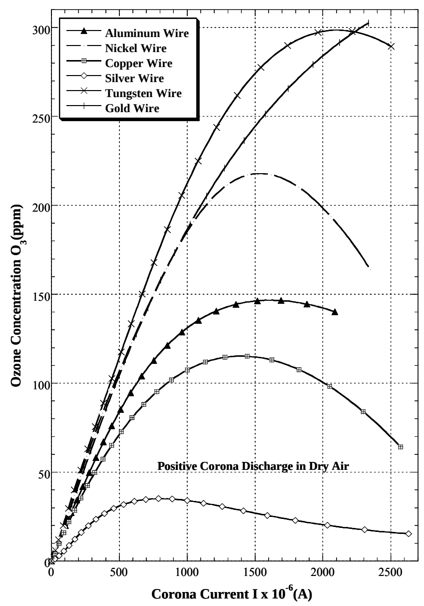

Fig. 6. The ozone concentration generated by the positive corona discharge in dry air as a function of the corona current for different materials of the wire.

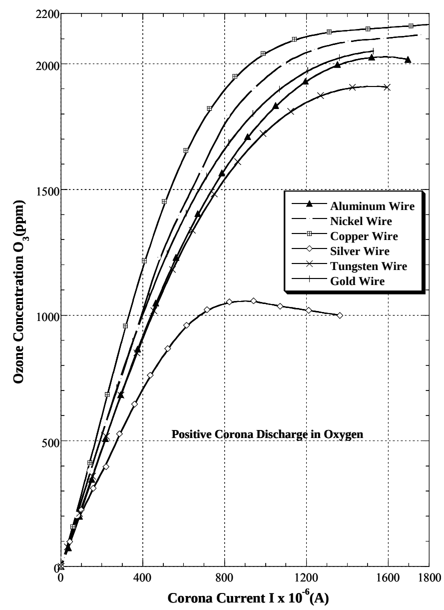

Fig. 8. The ozone concentration generated by the positive corona discharge in oxygen as a function of the corona current for different materials of the wire.

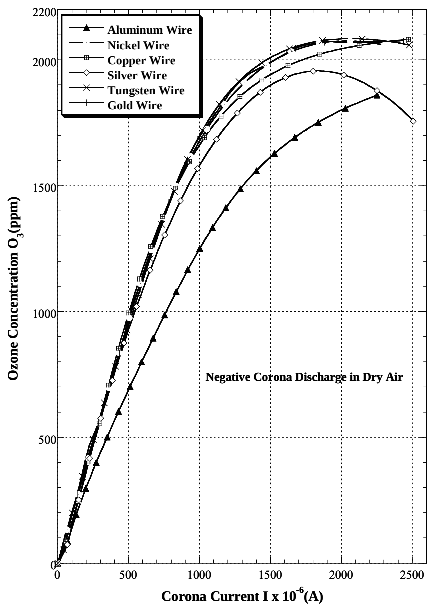

Fig. 7. The ozone concentration generated by the negative corona discharge in dry air as a function of the corona current for different materials of the wire.

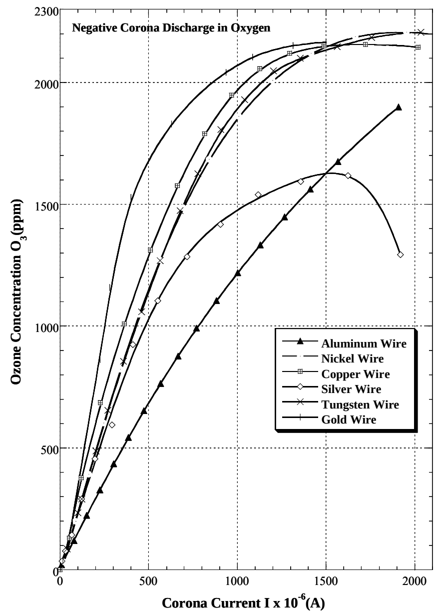

Fig. 9. The ozone concentration generated by the negative corona discharge in oxygen as a function of the corona current for different materials of the wire.

avalanches in the ionization-zone and the distribution of oxygen atoms surrounding the wire surface. In the positive corona, the electron avalanches progress towards the wire surface, while they move away from the wire in the negative corona [9]. The result is that the density of oxygen atoms created by electron avalanches around the wire surface is higher with the positive corona than that created with the negative corona. This in addition to the wire surface modifies the ozone formation and destruction processes according to the physical and chemical properties of the wire material with the reactions:

$$O + O_2 + M \rightarrow O_3 + M$$
$$O_3 + M \rightarrow O + O_2 + M$$
$$O + O + M \rightarrow O_2 + M$$

Where M represents the wire surface as a third body which is required to satisfy the condition of energy and momentum balance [10]. Therefore, the wire material must affects the ozone concentration generated by the positive corona more than that generated by the negative corona with the same discharge conditions. This explains why the wire material has a significant effect on the ozone generation characteristics of the positive corona discharges. Another important fact is that oxidation of the wire surface with the positive corona is more significant and occurs faster than with the negative corona. The decrease in the ozone generation with the rapid growth of the oxidation with the positive corona discharges mainly results from the following reduction reactions on the wire surface [3]:

$$O + MeO_x \rightarrow O_2 + MeO_{x-1}$$
$$O_3 + MeO_x \rightarrow 2O_2 + MeO_{x-1}$$

Where MeO_x means oxidized surface of the wire. This explains why the silver wire, which oxides easily, produces low concentrations of the generated ozone with the positive corona discharges in both dry air and oxygen over range of the corona current. On the contrary, both tungsten, copper and gold wires, which do not oxide easily, produce high concentrations of the ozone generated inside the reactor. The differing oxidation of the investigated wires with a reversal of the corona polarity indicates that chemical and electrical as well as thermal alterations take place in the gas in contact with the wire surface, and accordingly the generated ozone.

With dry air-fed reactor, the ozone concentration generated by the negative corona is several times higher than that generated by the positive corona at the same discharge conditions (i. e. the wire material and the corona current). The authors have already explained this phenomenon in [11, 12] by the fact that the non-thermal plasma generated by the negative corona is richer in energetic electrons than that generated by the positive corona [9]. Moreover, the thickness of the plasma layer generated by the negative corona around the discharge wire is thicker than that generated by the positive corona [13]. The larger the volume of the corona discharge plasma, the more air molecules are involved in the physico-chemical reaction processes for the ozone generation.

With oxygen-fed reactor, on the other hand, the ozone concentration generated by the negative corona is only higher than that generated by the positive corona at the same corona current with silver, tungsten and gold wires. With the other wires, the ozone concentration generated by the positive corona is higher than that generated by the negative corona at the same corona current. In a previous study [14], we have investigated modes of the corona discharge corresponding with the ozone generation in oxygen under conditions like that in the present study (i. e. the same reactor and the same oxygen flow rate inside it). Our investigations have shown that with stainless steel wires of radii 0.025, 0.05 and 0.1 mm, the ozone concentration generated by the negative corona was higher than that generated by the positive corona at the same discharge conditions. On the contrary, with the wire of 0.2 mm radius, the ozone concentration generated by the negative corona was exactly equal to that generated by the positive corona at the same discharge conditions. Moreover, both the positive and negative corona discharges appear in the form of streamers superimposed on the discharge current starting from the onset voltage up to the spark-over voltage. This behavior confirms that these streamers almost occupy the same discharge volume in the positive and negative coronas with the wire of 0.2 mm radius. This explains why the difference in the ozone concentration generated by both positive and negative corona discharges in oxygen is comparatively small at the same conditions with the investigated wires of 0.2 mm radius. In accordance with the above, the conflict between the previous results in [3] and [4] may be due to the difference in the wire diameter and the geometrical configuration of the discharge electrodes between the two studies.

The ozone concentration generated in oxygen-fed reactor is higher than that generated in dry air-fed reactor at the same discharge conditions (i. e., polarity of the corona discharge, the wire material and the corona current). As discussed before, the number of oxygen molecules involved within the corona discharge plasma with feeding the reactor by oxygen is much higher than that with feeding the reactor by dry air. The higher the numbers of oxygen molecules are exposed to the physico-chemical reaction processes of the corona discharge plasma, the more oxygen atoms are created for ozone generation. This explains why the ozone concentration generated in oxygen-fed reactor is higher than that generated in dry air-fed reactor at the same discharge conditions.

V. CONCLUSION

According to the present measurements, the following conclusions can be drawn:

(1) The wire material does not affect the current-voltage characteristics of both positive and negative corona discharges in dry air.
(2) The wire material affects the current-voltage characteristics of both positive and negative corona discharges in oxygen.
(3) Both positive and negative corona discharges yield larger current in oxygen than that in dry air at the same discharge conditions.
(4) The wire material affects the ozone generation characteristics whatever the discharge conditions. The ozone concentration increases with the corona current, and reaches saturation in the intense discharge range. The saturated ozone concentration sometimes followed by a decrease in the ozone concentration.
(5) Effect of the wire material on the ozone generation characteristics is the most significant with the positive corona in dry air, and not so significant with the negative corona in dry air, or with both positive and negative coronas in oxygen.
(6) Effect of the silver wire on the ozone generation characteristics is the most significant with the positive corona discharges in either dry air or oxygen. On the other hand, effect of the aluminum wire on the ozone generation characteristics is the most significant with the negative corona discharges in either dry air or oxygen.

(7) The ozone concentration generated by the negative corona is much higher than that generated by the positive corona at the same discharge conditions. This tendency is more clearly in dry air than in oxygen.
(8) The ozone concentration generated in oxygen-fed reactor is higher than that generated in dry air-fed reactor at the same discharge conditions.

REFERENCES

[1] Baldur Eliasson and Ulrich Kogelschatz “Nonequilibriun volume plasma chemical processing” IEEE Transactions on Plasma Science, Vol. 19, No. 6, page 1063-1077, 1991.

[2] G. S. Peter Castle, Ion I. Inculet and K. Irwin Burgess “Ozone generation in positive corona electrostatic precipitators” IEEE Transactions on Industry and General Applications, Vol. IGA-5, No. 4, page 489-496, 1969.

[3] Keiichi Nashimoto “The effect of electrode materials on O3 and NOx emissions by corona discharging” Journal of Imaging Science, Vol. 32, No. 5, page 205-210, 1988.

[4] Kimberly J. Boelter and Jane Davidson “Ozone generation by indoor, electrostatic air cleaners” Aerosol Science and Technology, Vol. 27, page 689-708, 1997.

[5] R. Peyrous and C. Lacaze “Technological parameters that influence the production of ozone in a dc corona discharge” Ozone Science and Engineering, Vol. 8, page 107-128, 1986.

[6] E. Nasser “Fundamentals of gaseous ionization and plasma electronics” New York: J. Wiley, 1971.

[7] R. Peyrous and R-M Millot “Ozone generation in oxygen by corona discharges in a point-to-plane gap subjected to a chopped DC positive voltage” J. Phys. D: Appl. Phys., Vol. 14, page 2237-2242, 1981.

[8] U. Kogelschatz “Silent discharges and their applications” Proceeding of the Tenth International Conference on Gas Discharges and Their Applications, Swansea, Wales, U. K., 13-18 September, Vol. II, page 972-980, 1992.

[9] M. P. Sarma and W. Janischewskyj “DC corona on smooth conductors in air” Proc. IEE, Vol. 116, page 161-166, 1969.

[10] M. B. Awad and G. S. P. Castle “Some parameters affecting the generation of ozone in positive and negative corona” Proc. IEEE-IAS Annual Meeting, page 373-380, 1973.

[11] A. Yehia, M..Abdel-Salam and A. Mizuno “On assessment of ozone generation in dc coronas” J. Phys. D: Appl. Phys., Vol. 33, page 831-835, 2000.

[12] A. Yehia, A. Mizuno and K. Takashima “On the characteristics of the corona discharge in a wire-duct reactor” J. Phys. D: Appl. Phys., Vol. 33, page 2807-2814, 2000.

[13] Y. Takahashi, M. Yoshida, Y. Anma, S. Kobayashi and M. Endo “Thickness and luminosity distribution of positive corona sheath in air” J. Phys. D: Appl. Phys., Vol. 15, page 639-653, 1982.

[14] A. Yehia “Analysis of silent and non-silent discharges in non-uniform field gaps” PhD Thesis, Faculty of Science, Assiut University, Assiut, Egypt, 2002.