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Products of dielectric barrier discharge in screw and pyramid reactors as a soil processing environment

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Abstract

Dielectric barrier discharges (DBDs) generated in screw and pyramid type coaxial reactors with and without cooling and at different power supply and gas flow conditions, as a source of chemically active environment for soil processing are studied in this paper. The results show that in the proposed reactor both generation efficiency of DBDs products (O_3 and NO) and their concentrations can be controlled by the reactor geometry, voltage and frequency of power supply, gas flow rate and cooling conditions.

Keywords: Non-thermal plasma; Dielectric barrier discharges; Sterilization; Ozone; NO

1 Introduction

Application of plasma based technology in the agricultural and food industry results in improving sensory quality, nutritional value and storage life of agricultural products and eliminates from chemical compounds the process containing sulfur and chlorine that can persist in the environment [1].

Numerous technologies have been developed for the purpose of sterilization, including chemicals, moist and dry heat, microwaves, UV and γ radiation. At present, the worldwide need to develop environmentally friendly alternatives to

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chemical technologies for the agricultural and horticultural products sterilization has been observed [2]. As a source of ozone (O_3) and nitrogen monoxide (NO) gases a variety of non-equilibrium low-temperature discharges and reactors could be considered, like: dielectric barrier discharges (DBDs), atmospheric pressure glow discharges (APGD), RF and pulse discharges, ferroelectric bad-packed reactors or gliding arc discharges. The most of those non-thermal plasma discharges are already applied in many emergent technologies. The DBDs, applied for potable water treatment with ozone almost from the beginning of the last century, are still investigated to improve the efficiency of ozone generation from air and from oxygen.

Ozone, one of the main products of electrical discharges in air, is the most powerful oxidant that quickly kills insects, bacteria and fungi. The application of ozone for soil sterilization gives many benefits based on on-site generation, minimum human toxicity and lack of persistent chemicals in the soil [3]. The discharges in air are also the source of free radicals, energetic electrons, atoms and other excited species, among them nitric oxides (NO, NO_x) that could be used for soil sterilization. NO, which is also present in soil due to microbial and fertilization activities, is very reactive and unstable and has been expected to kill or eliminate microbial cells from soil and can strongly influence the soil quality.

Reported in the paper coaxial screw and pyramid dielectric barrier discharge reactors powered from high frequency voltage, generate useful environment for soil processing. Reactors can produce ozone and nitrogen oxides with both high concentration and efficiency and provide up to 40 g of ozone per cubic meter that can be controlled by the value of applied voltage, gas flow velocity and cooling conditions.

Results of soil processing with O_3 and NO, that show temporal and spatial changes of soil acidity (pH) and electrical conductivity (EC) are also presented in this paper.

2 Measuring system

The dielectric barrier discharge reactors (DBDs) with and without cooling system were investigated. The both geometries consisted of inner metal electrodes (100 mm length) in the form of screw and pyramid put in the outer glass quartz tube (ϕ 8 mm) that was wrapped around with aluminum or copper foil. Their construction allowed cooling both inner metal electrode and outer glass. In the first geometry (Fig. 1) the metal electrode was connected to high voltage and the glass tube with aluminum foil was grounded and can be cooled. In the second geometry (Fig. 2) the metal screw and pyramid electrodes with the hole inside was cooled by flowing water while the glass electrode covered with copper was connected to the high voltage source. The gap between the inner screw electrode and the glass tube was kept at 1 mm and the discharges were generated in dry air or oxygen.

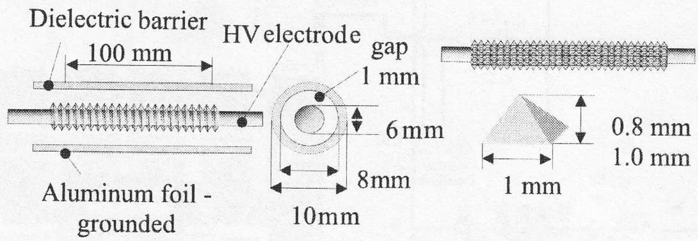


Figure 1. Geometry of DBD reactor with high voltage electrode in the form of screw and pyramid (without cooling).

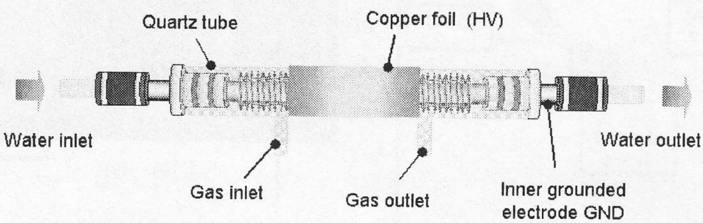


Figure 2. Water-cooled screw electrode reactor.

The measuring system, presented in Fig. 3, consisted of high frequency inverter, voltage divider, oscilloscope, flow meter and monitors of discharge products (ozone and nitrogen oxides). Measurements were made at three frequencies (10, 30 and 50 kHz) of supply voltage, that was changed from 1.7 to 7 kV and at varied process gas flow rates (1, 2 and 3 l/min).

Ozone and nitrogen monoxide were introduced into the soil through the drip-feed 1/4 inch Teflon pipe. Two sets of sensors were introduced into the soil container to measure its acidity pH and electrical conductivity EC (Fig. 4). Sensors arrangement in the ozone injection vessel is shown in Fig. 5. The soil temperature has also been measured. The treatment container was located in the incubator where temperature and light illumination could be automatically controlled. All measurements data can be transmitted to the PC through the wireless network system.

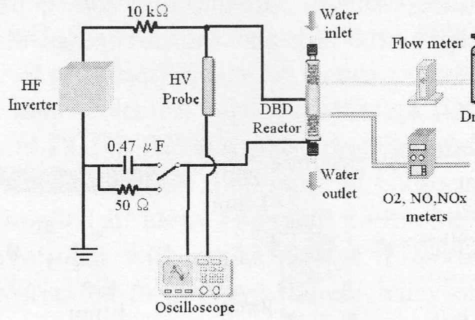
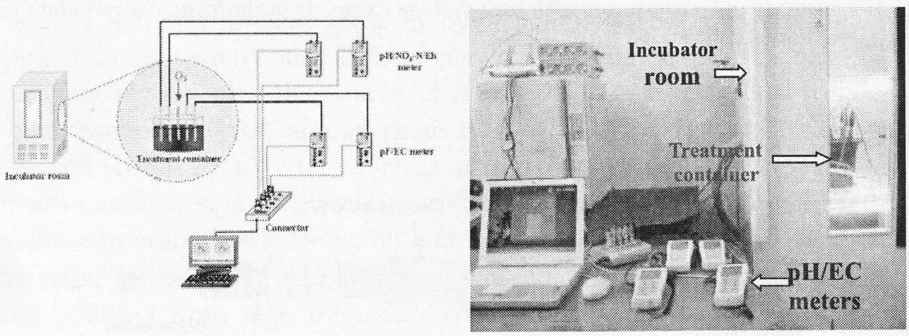


Figure 3. Experimental setup.



a) b)

Figure 4. Ozone and NO injection system into the soil with measuring sensors: a) scheme connection b) picture of the system.

3 Results and discussion

3.1 O_3, NO and NO_x generation

Concentrations of O_3, NO and NO_x in function of input power to the discharge were measured at three frequencies of supply voltage (10, 30 and 50 kHz) and at various O_2 flow rates. Results of measurements presented in Figs. 6, 7 and 8 show, that in the investigated reactor, the increase in the power system frequency is accompanied by decrease of the ozone concentration. At the 30 kHz and 50 kHz the content of O_3 is almost negligible. The ozone generation efficiency in function of ozone concentration at various O_2 flow rates is presented in Fig. 9. The highest ozone concentration of 80 g/m^3 is generated at the lowest gas flow rate of 0.3 l/min. On the other hand, the highest value of ozone generation efficiency (220 g/kWh) was obtained at 1–2 l/min at a low ozone concentration (10 g/m^2). It is shown that the maximum obtained concentration decreases with increase of

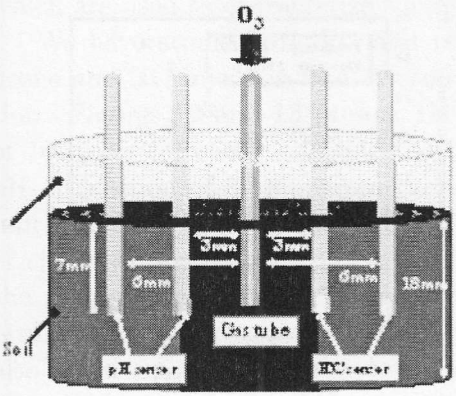


Figure 5. EC an pH sensors arrangement in the ozone injection vessel.

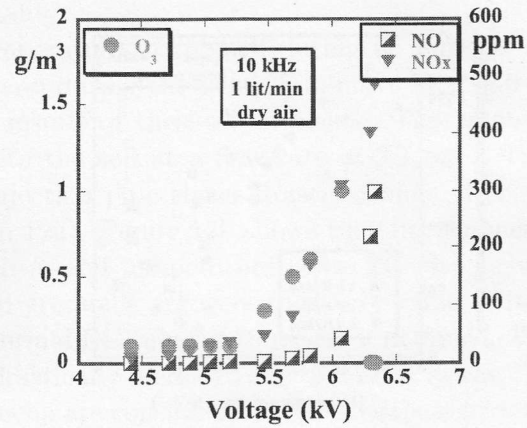


Figure 6. O_3, NO and NO_x concentration versus power at 10 kHz

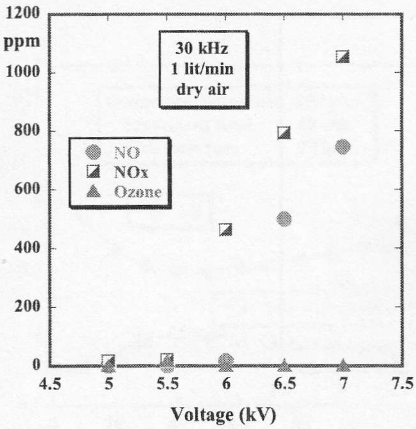


Figure 7. O_3, NO and NO_x concentration versus supply voltage at 30 kHz.

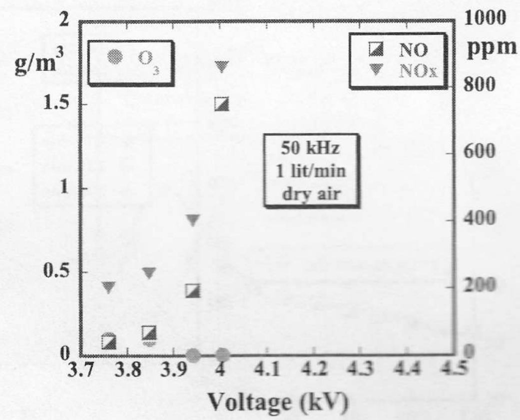


Figure 8. O_3, NO and NO_x concentration versus power at 50 kHz.

O_2 flow rate. Influence of cooling conditions on ozone generation is presented in Fig. 10. The maximum ozone concentration for the water-cooled electrode was 38 g/m^3 at 5.3 kV. In case of the non-cooling apparatus, rapid decrease of ozone concentration appears over 4.6 kV due to dielectric heating of the quartz tube.

Nitric monoxide (NO) is a very reactive and unstable free radical. In air, it quickly reacts with oxygen to form the poisonous nitrogen dioxide (NO_2). The NO has also been expected to kill or eliminate microbial cells. NO radicals were generated by using the dielectric barrier discharge. The nitrogen oxides concentration increases with both the frequency and the value of supply voltage. At 30 kHz

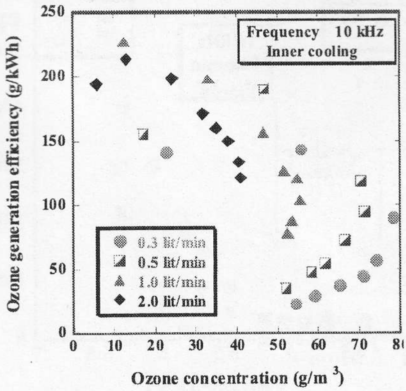


Figure 9. Ozone generation efficiency as a function of ozone generation for 4 different flow rates of oxygen.

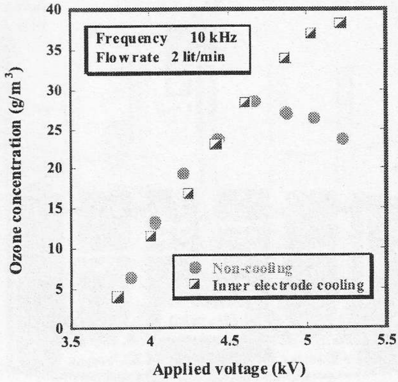


Figure 10. Influence of cooling on ozone concentration in function of applied voltage.

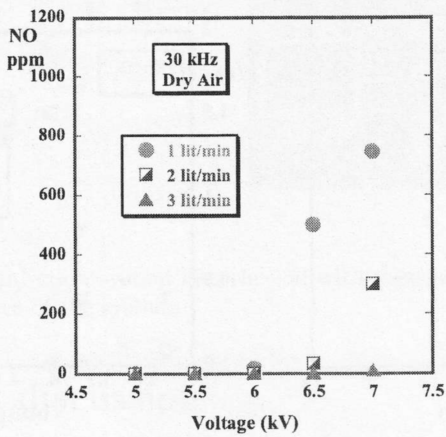


Figure 11. NO concentration versus supply voltage for different flow rates at 30 kHz.

NO began to be generated at 6 kV and it has attained its maximum concentration of 780 ppm at 7 kV (Fig. 7). Their concentration also substantially depends on gas flow rate. Figure 11 presents dependence of NO concentration in function of supply voltage for three values of gas flow rate at 30 kHz. NO concentration achieves lower values for higher flow rates and increases with supply voltage.

3.2 Soil properties after O_3 and NO injection

The soil acidity, pH, and electrical conductivity, EC, together with the content of moisture, pF, and nitrites NO_3NH, NH_3NO, are the most important indicators, which are used to characterize soil quality.

We have studied pH, EC and temperature of the soil during 60 minutes of ozone and NO injection into the soil and during the next 60 minutes after injection. Figures 12 and 13 present the results of these experiments. The amount of 100 g/m^3 of ozone was injected into the soil at a flow rate of 2 l/min. The pH at position of 30 mm from the injection pipe shows drastic change after 40 min and then gradually increases (Fig. 12a). Figure 12b shows time development of electrical conductivity (EC) as well as soil temperature. The EC also shows the rapid decrease in first 20 min and gradually recovers to steady value. After termination of the ozone treatment, normal EC value of 70 mS/m is attained. We also found out that soil temperature drastically increases and decreases as the EC and pH change rapidly. These phenomena are considered to be accompanied with chemical exothermic reaction of the soil with ozone. Figure 13 shows the change of the pH and the EC during and after NO treatment of soil. We notice only slow decrease of the EC. In this treatment, 500 ppm NO was injected into the soil at a flow rate of 2 l/min.

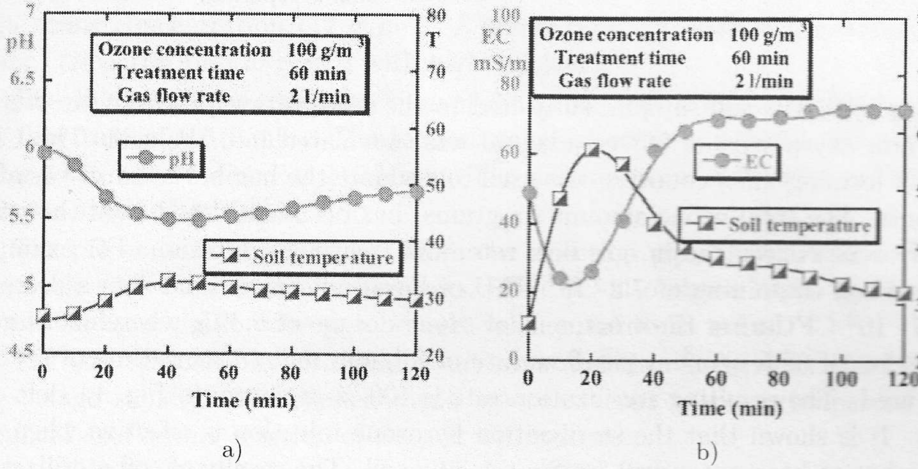


Figure 12. Time changes of pH, EC and soil temperature during 120 minutes (60 minutes of ozone injection and the next 60 minutes after injection).

3.3 Soil sterilization experiments

We have sterilized the agricultural soil by supplying ozone and nitrogen monoxide. Figure 14 presents previously published results [5]. The 50 g of agricultural soil was treated by varied ozone dosages (0–40 g O_3/m^3) and the duration (1–60 minutes). In this experiment the agricultural soil was filled in a chamber (ϕ 70 mm) and ozone gas was supplied from the top of the chamber and evacuated from the

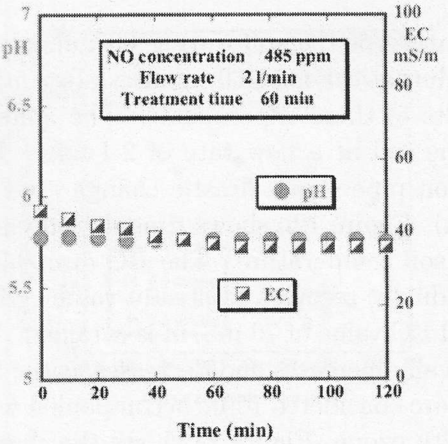


Figure 13. Changes of pH and EC of soil treated by NO.

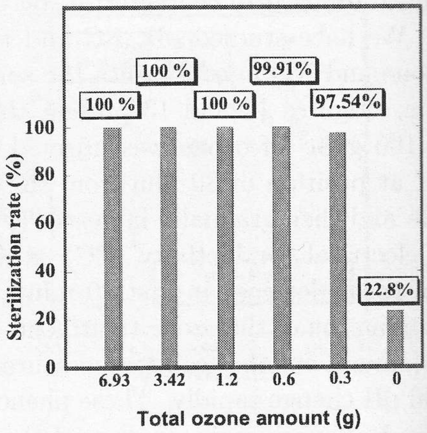


Figure 14. Sterilization as a function of ozone amount. The inoculated soil initially has $4\text{--}7{\cdot}10^6$ CFU/cc of fasarium oxysporum.

bottom exit. The soil samples subjected to the sterilization process were sterile. Fasarium oxysporum of 10^6 conidal/ml was inoculated into the soil. The CFU (colony forming unit) counting was used to evaluate the number of survivors after 72 hours. The total ozone amount (in grams) has been calculated from the product of ozone concentration, gas flow rate, and treatment duration. For example, the test soil containing of $7.2 \cdot 10^6$ CFU of Fusarium oxysporum shows decrease to $1.7 \cdot 10^2$ CFU after the treatment of ozone dosage of 6.39 g where ozone concentration of 38.5 g/m^3 at gas flow rate of 3 l/min and treating time of 60 min were used. The resulting sterilization rate is 100 % as shown in Fig. 14 (left end data). It is shown that the sterilization by ozone injection is effective when the ozone dosage becomes over 0.3 g for the 50 g soil. The results of soil sterilization are also presented in Table 1. Fusarium oxysporum in the soil was almost killed by ozone treatment of 20 g O_3/m^3 for 10 min. Over 80 % bacteria included in the soil was sterilized after 20 minutes treatment of 20 g O_3/m^3.

In another experiment for soil sterilization, the agricultural soil samples contaminated with root-knot nematode (M. incognita J2) were prepared. The ozone of the concentration of 45 g/m^3 was injected during 20 minutes at a flow rate of 3 l/min. In the treated soil any nematode in the depth range from the surface to 50 mm has been found. After 30 days, the root-knot index (the number of knots of root for planted melon) was measured. The measured index (2.0) for the ozone treated soil is decreased about half of the index (3.8) for the untreated soil.

Table 1. Soil sterilization by in-situ ozone treatment.

	Bacteria	Fusarium oxysporum	
Untreated [cfu/cc]	$1.8{\cdot}10^5$	$5.7{\cdot}10^6$	
Gas flow rate	1 l/min	3 l/min	
Concentration	20 g/m^3	10 g/m^3	20 g/m^3
Duration	20 min	10 min	10 min
Ozone treated [cfu/cc]	$2.7{\cdot}10^4$	$1.4{\cdot}10^5$	$1.7{\cdot}10^2$
Sterilization rate	86 %	97.5 %	99.9 %

4 Conclusions

We have studied the soil sterilization by using ozone and nitric monoxide, which are generated by the coaxial dielectric barrier discharge. A pyramid type electrode can control the ozone concentration from low ozone concentration (0.1 g/m^3) to high ozone concentration (12 g/m^3). A screw type electrode can generate high ozone concentration (20 g/m^3) with high efficiency.

The highest ozone concentration of 80 g/m^3 was generated at the lowest gas flow rate of 0.3 l/min. The highest value of ozone generation efficiency of 220 g/kWh was obtained at 1–2 l/min. The maximum concentration of 780 ppm NO was attained at 7 kV.

The pH at position of 30 mm from the injection pipe shows drastic change in 40 min and then gradually increases. The EC also shows rapid decrease till 20 min and gradually recovers to steady value equal to 70 mS/m.

The soil temperature drastically increases and decreases when the EC and pH change rapidly. These phenomena are considered to be accompanied with chemical exothermic reaction of the soil with ozone.

The sterilization of the agricultural soil was investigated using ozone injection into the soil. The results show that the sterilization rate could be equal to 100% for the ozone dosage over 0.6 g for the 50 g of treated soil.

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