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Ozone generation by dielectric barrier discharge for soil sterilization

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Abstract

We developed the ozone generation system for soil sterilization. The pyramid-type electrode was effective to reduce breakdown voltage and the ozone concentration can be widely controlled by changing the applied voltage. It is also shown that the screw-type electrode has the advantage of high ozone concentration with high efficiency. The model experiment of ozone diffusion in silica gel was carried out to estimate ozone diffusion phenomena in agricultural soil. The effectiveness of ozone treatment of agricultural soil is demonstrated by reducing soil pathogens.

Keywords: Ozone; Dielectric barrier discharge; Soil sterilization

1. Introduction

The global environmental destruction by the pollution of soil, air and water is a worldwide problem. Methyl bromide (CH_3Br) is frequently used as a fumigant to sterilize microbial activity in soil. But methyl bromide is being phased out rapidly due to claims of its deleterious long-term effects on the ozone layer and on human health and safety concerns.

It is necessary to develop environmentally benign alternatives to methyl bromide for soil fumigation. Ozone, possessing strong oxidative and germicidal properties, has a very short half-life of minutes or less in soil and decomposes in simple diatomic oxygen. It has also many benefits based on on-site generation, minimum human toxicity, low persistent chemicals in the soil. We have developed ozonation technology using the dielectric barrier discharge (DBD) [1,2].

DBD is a method to generate discharges by applying high voltage to dielectric between electrodes.

In this work, we have studied ozone generation by the DBD and ozone diffusion for soil sterilization [1–5].

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2. Experimental system and methods

2.1. Ozone generation experiment

The experimental setup for ozone generation using DBD is shown in Fig. 1. An electric power of high-frequency inverter was applied to the electrodes in the range of applied voltage (1.7–5.5 kV). Material gas (oxygen 99.9%) was supplied to the reactor of the dielectric quartz glass tube (ϕ 8 mm) with inner electrodes. The inner electrodes are structures of a screw-type electrode and a pyramid-type electrode (Fig. 2) of stainless steel rod (ϕ 6 mm). The ground electrode is an aluminum foil wrapped around the quartz tube. The gap separation between the inner electrode and the insulator tube was kept at 1 mm.

Time-resolved waveforms of discharge current and voltage were measured through a high-voltage probe and a resistor of 50 Ω and were recorded on a digitizing oscilloscope. Discharge power was estimated from the $Q-V$ Lissajous figure obtained by measuring the capacitor voltage of 0.47 μF.

2.2. Ozone diffusion experiment

Fig. 3 shows the experimental setup for ozone diffusion in soil. The 10 rings (ϕ 100 mm, height 10 mm) were piled up

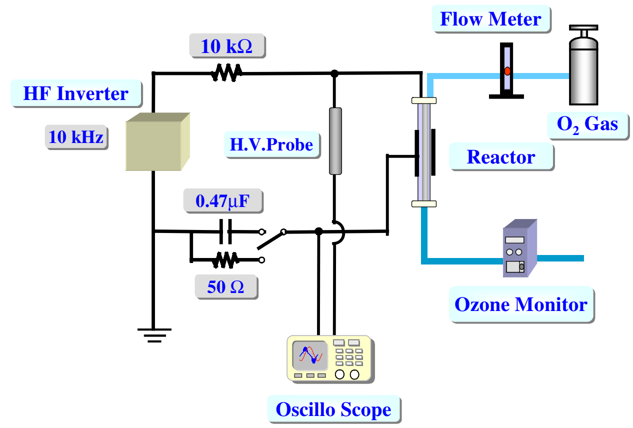

Fig. 1. Experimental setup for ozone generation.

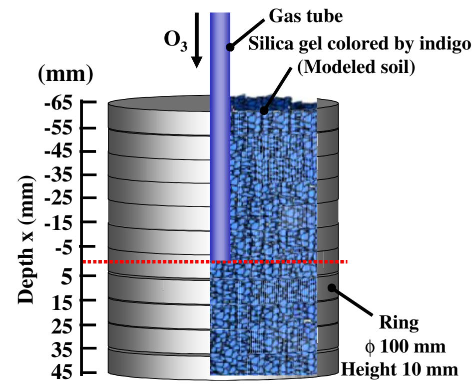

Fig. 3. Experimental setup for ozone diffusion.

and silica gel ($SiO_2 \cdot nH_2O$, 500 g) as modeled soil was filled. The ozone gas was supplied into the silica gel colored by indigo. The gas outlet of the drip tube (0.25 in. stainless) was located at the center of the piled container ($x=0$). After the ozone treatment at 22 °C, each pile was removed one by one and the decolorizing area was observed. The chemical decolorizing reaction during ozone treatment is

$$2O_3 + C_{16}H_{10}N_2O_2(\text{indigo}) \rightarrow 2C_8H_5NO_2(\text{isatin}) + 2O_2$$

The experimental conditions are given in Table 1.

3. Results and discussions

3.1. Ozone generation by various electrode types

Fig. 4 shows ozone concentration as a function of applied voltage. Fig. 5 shows the relationship between ozone generation efficiency and ozone concentration for various electrodes. The DBD discharges in the case of the pyramid electrode initiated at lower voltage of 1.7 kV comparing with 3.5 kV for the screw-type electrode under the same condition of the separation between the electrode surface and the quartz tube. The lowering of the voltage is due to the higher electric field strength of the pyramid electrode. The ozone concentration in the case of the pyramid type is higher at low applied voltage below 3.8 kV and can be controlled linearly by the applied voltage that is preferable to choose the optimum concentration for soil sterilization. A screw-type electrode can provide high ozone concentration (23 g/m^3) with high efficiency (100 g/kW h).

Fig. 6 shows voltage and current waveforms of the pyramid-type DBD at 4 kV. It was observed that the larger number of microdischarges appears in the case of a pyramid-type electrode comparing with the screw-type electrode at the same applied voltage. Although the increase of microdischarge promotes the generation of ozone through the oxygen decomposition, the rapid decomposition reaction of the generated ozone simultaneously occurs by contacting with the microdischarges. This is one of the reasons that the ozone concentration for the pyramid-type electrode is lower than that in the case of the screw-type electrode. Another is that the temperature rise of the quartz tube due to the high-frequency dielectric heating destroys the generated ozone. The quartz tube temperature was 90 °C for the pyramid-type electrode and 70 °C for the screw-type electrode at 4 kV and 2 l/min. These results show that the pyramid-type DBD is suitable to low ozone concentration for soil sterilization and that the screw type is useful for soil treatment requiring high ozone concentration.

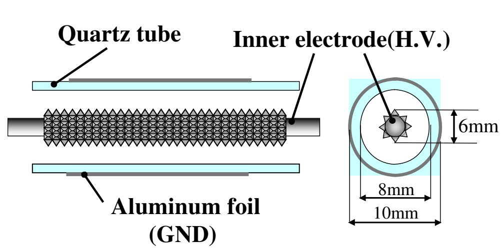

Fig. 2. Pyramid-type electrode.

3.2. Ozone diffusion in modeled soil

The agricultural soil consisting of mud, sand, organics, minerals and water shows a complicated change in physical and chemical properties after the injection of the ozone gas into the soil. Development of ozone injection system is necessary to make ozone treatment in practical use. We studied ozone diffusion dynamics in modeled soil to estimate the appropriate arrangement for the ozone gas drip tubes put in the soil. Because the in situ change of the agricultural soil after ozone treatment is difficult to observe, colored silica gel is used as the modeled soil.

Fig. 7 shows average diameter for the decolorizing areas as a function of the depth x at a flow rate of 1 l/min and ozone concentration of 1 g/m^3. It is shown that ozone can spread out to a diameter of 8 cm during 60 min irradiation.

Fig. 8 shows the effect of ozone concentration on the decolorizing area at a treatment time of 5 min. When ozone concentration is increased, the decolorizing area rapidly

Table 1
Experimental conditions for ozone diffusion

Ozone concentration	1–20 g/m^3	
Gas flow rate	1 l/min	
Treatment time	5–90 min	5 min
Modeled soil	Silica gel colored by indigo (500 g), volume 750 cm^3	

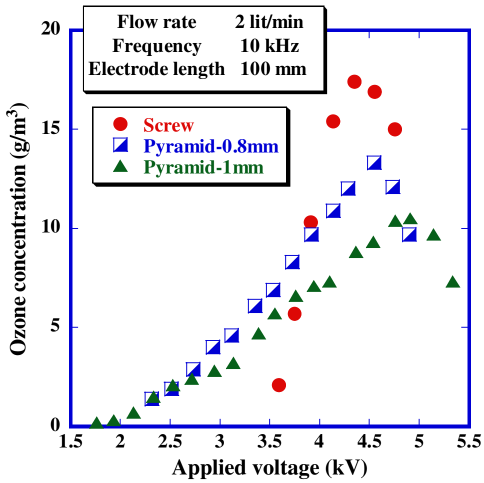

Fig. 4. Ozone concentration as a function of applied voltage at various electrodes.

increases. The ozone treatment rate, which is defined by the relation of the decolorizing volume divided by the injected ozone weight, was calculated. The rate during the first 5 min is 7.5 l/O_3 g for ozone concentration of 1 g/m^3 and 2.6 l/O_3 g for

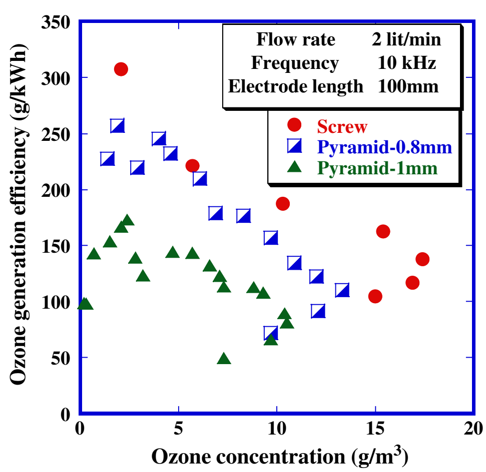

Fig. 5. Ozone generation efficiency as a function of ozone concentration at various electrode types.

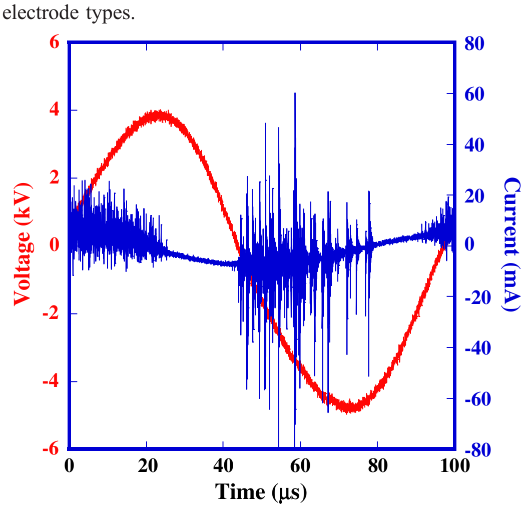

Fig. 6. Voltage and current waveform of DBD using pyramid-type electrode.

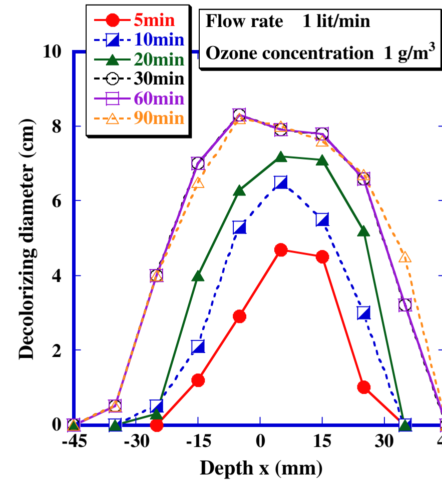

Fig. 7. Decolorizing diameter as a function of depth x at various treatment time.

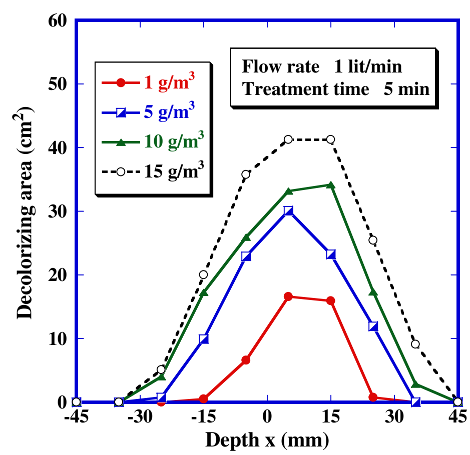

Fig. 8. Decolorizing area as a function of depth x at various ozone concentration.

10 g/m^3, respectively. Low decolorizing rate at high ozone concentration is due to excess absorption near the ozone outlet (depth $x = 0$).

3.3. Soil sterilization by ozone injection

Agricultural soil (Audisols) was sterilized by varying the application dosages and duration. The sample soil of 50 g placed in the chamber was treated by ozone injection [1]. The results are shown in Table 2. *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 min treatment of 20 g O_3/m^3.

Table 2
Soil sterilization by in situ ozone treatment

	Bacteria	*Fusarium oxysporum*	
Untreated [cfu/cm^3]	1.8×10^5	5.7×10^6	
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/cm^3]	2.7×10^4	1.4×10^5	1.7×10^2
Sterilization rate	86%	97.5%	99.9%

4. Conclusion

We performed ozone generation experiments and ozone diffusion experiment in the soil. 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 diffusion experiment using the modeled soil shows that the ozone can diffuse radically near 4 cm far from the gas outlet during 60 min. High ozone treatment rate is obtained at the conditions of long treatment time with low ozone concentration. High gas flow rate is effective to enlarge the treated volume in a short period.

Acknowledgement

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