Generation and Decomposition of Ozone Gas by Ozone and Ozoneless Mercury Bulbs Excited by Microwave

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I. Introduction

Ozone gas having strong oxidizing power has been used in various fields such as environmental cleaning, sterilization [1)], inactivation of virus, decolorization, deodorization, removal of organic substances and so on [2)]. Ozone gas is often produced from oxygen molecules by silent discharge. Besides, it can be generated by ultraviolet light (UV light) emitted from a low-pressure mercury lamp. UV light at 184.9 nm reacts with oxygen molecules in the air to generate ozone as following [3)]:

$$O_2 + h\nu\ (184.9\ nm) \rightarrow O + O$$
$$O_2 + O \rightarrow O_3$$

On the other hand, UV light at 253.7 nm emitted simultaneously with UV light at 184.9 nm from a low-pressure mercury lamp decomposes ozone via active oxygen atoms:

$$O_3 + h\nu\ (253.7\ nm) \rightarrow O_2 + O$$
$$O + O_3 \rightarrow 2O_2$$

It is well known that ozone gas is toxic to humans. Health effect of ozone gas is summarized below. [4)]

0.01–0.02 ppm	Odour threshold
0.1 ppm	Nose and throat irritation
0.2–0.5 ppm	Reduced visual acuity, upper airway irritation
0.6–0.8 ppm	Chest pain, dry cough
1–2 ppm	Lassitude, headache, some reduction in lung function
5–10 ppm	Respiratory distress, pulse rate increase
> 50 ppm	Expected to be fatal

Moreover, the Occupational Exposure Limits of ozone gas, which is recommended by The Japan Society for Occupational Health (JSOH), is 0.1

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ppm (0.2 mg/m^3); the maximum allowable concentration of ozone gas established by the United States Food and Drug Administration (FDA) is 0.05 ppm (24 hours). According to the Air Pollution Control Law, the environmental standard of ozone is less than 0.06 ppm/h [5] In using ozone in safety, therefore, we need to control its concentration in the air.

Recently, ozone generators using UV light have been employed in general home for deodorization and disinfection. The ozone generator with corona discharge and glow discharge, instead of UV light [6], is also utilized, because the efficiency of its ozone generation is higher. Since these generators have no mechanism for decomposing ozone, however, there are safety concerns in using them. In addition, the ozone generator with the discharge also produces a large amount of nitrogen oxide, NOx [7], and involves a significant cost for equipment maintenance [7].

A low-pressure mercury lamp can be excited to emit UV light not only by electricity but also by microwave irradiation. In irradiation of microwave, gaseous mercury is exited and is changed to mercury plasma, and the low temperature plasma emits UV light [10]. The mercury lamp emitting UV light by microwave needs no electrode. We then call the lamp a mercury bulb. We made two types of mercury bulbs from different quartz glass materials [11, 12]; one emits UV light at 184.9 and 253.7 nm to produce ozone gas, and the other does the light at 253.7nm alone to decompose ozone gas. Here, we abbreviate the former and the latter as ozone mercury bulb and ozoneless mercury bulb, respectively.

We attempt to develop the ozone generator equipped each mercury bulb. The generator has a characteristic to produce and decompose ozone gas simultaneously. Moreover, the mercury bulb possesses no electrode, so it has no damage in electrode.

In the present study, we investigate time-dependence of change in ozone concentration by ozone and ozoneless mercury bulbs excited by microwave irradiation, and evaluate effect of these bulbs on generation and decomposition of ozone gas.

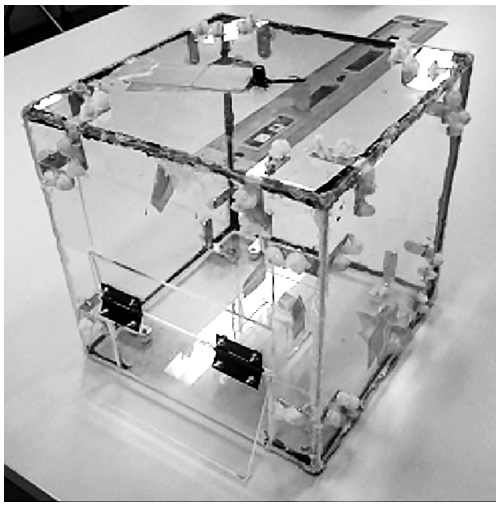

Fig. 1 Acrylic container used.

II. Experimental method

1. Ozone and ozoneless mercury bulbs.

Ozone and ozoneless mercury bulbs of which the diameter was 1.5 cm were prepared. The bulb including 1, 2, and 4 mg of mercury, and the vapor presser of mercury in the light-emitting bulb was controlled at less than 0.75 torr (100 Pa).

2. Generation and decomposition of ozone gas.

A container (30 cm x 30 cm x 30 cm) was produced with acrylic plates, shown in ***Figure 1***. The schematic diagram of the ozone generator used in this study is illustrated in ***Figure 2***. The mercury bulb socket (Hg socket) of the ozone generator was placed at the center of the bottom surface of the container, and a mercury bulb was settled in the socket. A 2.45 GHz of microwave was applied to the mercury bulb to emit UV light (***Figure 3***). The concentration of ozone gas in

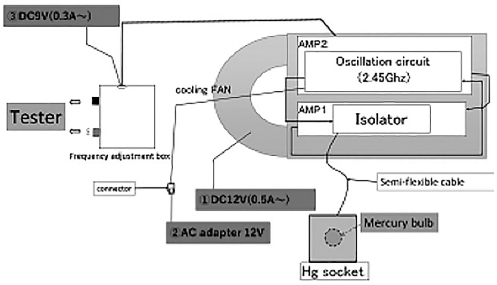

Fig. 2 Ozone generator used.

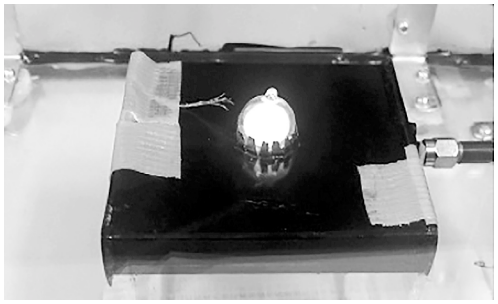

Fig. 3 Mercury bulb emitting UV light.

the container was measured using a gas sampling pump equipped with a short-term quick-measuring detector tube for ozone gas (18 L and 18 M, GASTEC Co., Ltd.).

III. Results

First of all, we measured emission spectra from ozone mercury bulbs including 1, 2, and 4 mg of mercury with a spectral radiometer. Here, since the lower wavelength limit of detection of the spectra radiometer is 200 nm, we measured the spectrum at the wavelength of more than 200 nm. The spectrum from the ozone mercury bulb with 4 mg of mercury is shown in ***Figure 4***. The large emission peak can be observed at 253.7 nm. The spectra of the other bulbs are almost the same. In addition, the fluence rate of the bulbs including 1, 2, and 4 mg of mercury are 0.564, 0.608, and 0.548 W/m^2, respectively.

Figure 5 depicts the time dependence of ozone concentration generated by the ozone mercury bulb with 1 mg of mercury. The ozone concentration increases with increasing emission time at any illuminance and becomes almost constant after about 50 min. The constant ozone concentration is 91, 120, and 162 ppm at illuminance of 610, 900, and 1200 Lx.

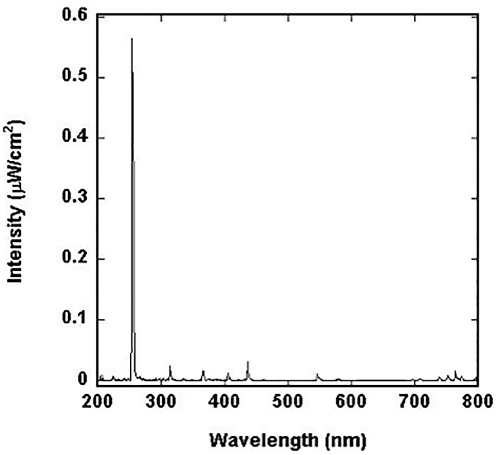

Fig. 4 Emission spectrum from ozone mercury bulb.

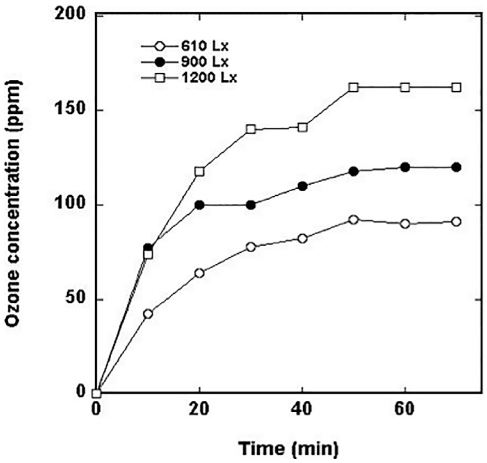

Fig. 5 Change in ozone concentration by ozone mercury bulb.

Next, the ozone decomposition by ozoneless mercury bulb with 1 mg of mercury was carried out. The result is illustrated in ***Figure 6***. Despite no emission of UV light, the ozone concentration in the container decreases from 20 to 6 ppm with increasing treatment time. It would be caused by leakage of ozone gas from the container and by

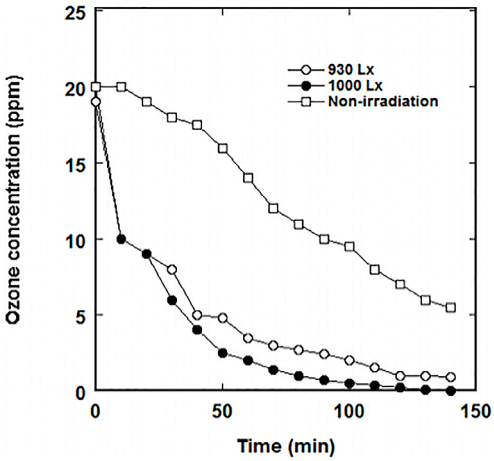

Fig. 6 Change in ozone concentration by ozoneless mercury bulb.

self-decomposition of ozone. In emission at 930 and 1000 Lx, moreover, the ozone concentration decreases immediately with an increase in irradiation time from 19 to 0.9 ppm and from 20 to 0 ppm.

IV. Discussion

The emission spectrum and fluence rate from the ozone mercury bulb including 1 to 4 mg of mercury is little significant difference. Furthermore, the maximum ozone concentration produced by ozone mercury bulb with 1 mg of mercury is about 160 ppm at 1000 Lx. This ozone concentration is comparable to the concentration inducing strong deodorization and sterilization for animal testing facilities, pharmaceutical manufacturing plants, and so on. In addition, about 20 ppm of ozone can be decomposed completely by the ozoneless mercury bulb including 1 mg of mercury for about 2 hr. Therefore, we consider that at least 1 mg of mercury in the mercury bulb is adequate to emit UV light for producing and decomposing ozone gas.

As can be seen from ***Fig. 5***, moreover, the constant ozone concentration in more than 50 min emission time is dependent on the illuminance of the mercury bulb. That is, the ozone concentration increases with an increase in illuminance. This result then indicates that we can control ozone concentration by the illuminance of the bulb.

According to the “Minamata Convention on Mercury” adopted in 2013, manufacturing, and importing and exporting of industrial products including mercury will be prohibited after 2021. Lighting equipment with mercury is no exception to the convention, and the equipment including more than 5 mg of mercury will be prohibited until the end of 2017. We suggest, however, the mercury bulbs prepared are ruled out in the convention as long as it includes 1 mg of mercury.

V. Conclusion

We demonstrate that the ozone and ozoneless mercury bulbs including at least 1 mg of mercury can readily generate and decompose ozone gas. We think that the ozone generator capable of effectively generating and decomposing ozone gas can be produced using the mercury bulb including 1 mg of mercury, not regulated by “Minamata Convention on Mercury”.

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