UV Dissociation and Calculation for O₃ Gas Generated from Nonthermal Atmospheric-Pressure Biocompatible Plasma Sterilizer

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ABSTRACT: Nonthermal atmospheric-pressure biocompatible plasma (NBP) sterilizers effectively deactivate microorganisms, such as viruses, bacteria, and fungi, using eco-friendly treatment methods without generating any waste or toxic by-products. The ozone density generated from the plasma source of surface microdischarge (SMD) in plasma sterilizers has been decreased by 70%, resulting in a density of less than 0.05 ppm, whereas the densities of OH and H₂O₂ have significantly increased due to UV-C dissociation with a high absorption cross section for ozone gas. The ozone density was calculated by estimating the oxygen atoms using the major reaction equations for ozone formation and dissociation, and measurement of the time constant of diffusion decrease (τ_{diff}). The ratio of the O density for the O₃ density generated from the plasma source decreased as the distance between the ozone measurement locations and the plasma source increased. In particular, the excited state density of atomic oxygen O(¹D) could be estimated at the downstream region of the air outlet in this study.

KEY WORDS: biocompatible plasma sterilizer, viruses, bacteria and fungi, ozone and oxygen reactive species, dissociation, disinfection, calculation

I. INTRODUCTION

People visiting hospitals, schools, concert halls, and the like, have a high risk of catching infections. People visiting medical institutions for diagnosis and treatment are highly exposed to many risks of infection transmission, and it is very important to protect patients, visitors, and staff from catching the infections. Microbial infections are spread by direct contact or indirect contact with aerosols in the air and splashes. Infections such as pneumonia, chickenpox, and measles are transmitted by aerosols in the air, whereas influenza, pertussis, and meningococcal infections are spread through splash. Multidrug-resistant bacteria are representative examples of infections caused by contact propagation.¹ Moreover, COVID-19 is transmitted directly and indirectly by aerosols in the air, causing people to become infected.^{2,3}

To prevent the spread of infection by aerosols in the air or splash, medical institutions usually rely on air circulation through air conditioning equipment with a high-efficiency particulate air (HEPA) filter that removes 99.97% particles larger than 0.3 μm in diameter. However, it takes a long time to purify the air this way in a large medical institution, and splashes are difficult to remove because they cover short distances. Bacteria

and viruses smaller than $0.3\ \mu\text{m}$ cannot be removed by a HEPA filter; therefore, they can be present even after air circulation. Moreover, chemicals such as chlorine, alcohols, aldehydes, and cresols are used periodically for sterilization and disinfection, which are expensive and environmentally damaging materials.⁴ UV sterilizers are used in hospitals and in many institutions to prevent infections. However, they can only sterilize a limited space where UV is irradiated.⁵

Nonthermal atmospheric-pressure biocompatible plasma (NBP) has been shown to effectively sterilize viruses and bacteria via reactive oxygen species (ROS) and reactive nitrogen species (RNS).^{6–10} NBP can be defined as low-temperature plasma enabled for use in human medical treatment and health care. ROS and RNS can oxidize and disassemble nucleic acids, proteins, and lipids, among bacteria.¹¹ NBP is used for the treatment of wounds¹² and cancers,¹³ making it a growth engine for the 21st century. Recently, permeation models for various reactions of plasma in skin and living cells have been proposed.¹⁴ NBP can be operated continuously for several months simply by turning on the power of a plasma sterilizer; it can also sterilize microorganisms smaller than $0.3\ \mu\text{m}$. Additionally, plasma sterilizers of different sizes can sterilize a wide area and spatial volume. Filipic et al.³ suggested that NBP can effectively deactivate microorganisms, such as viruses and bacteria, using eco-friendly methods, which generate no waste or toxic by-products. Hence, NBP is a novel, efficient, and clean solution to inactive viruses such as COVID-19. Inhalation of NO gas has been suggested as a therapy for COVID-19.¹⁵ Several studies on the sterilization of microorganisms using NBP have been published since 2000.¹⁶ However, only few studies have calculated the O_3 density¹⁷ and O density generated from plasma sterilizers in air.

We developed a plasma sterilizer which can be used in healthcare centers, and ambulances.⁶ Because O_3 gas has a relatively long life and harms the human body at high concentration, its release into the air from the plasma sterilizer should be adjusted not to exceed 0.05 ppm, according to environmental standards.¹⁸

In this study, a plasma sterilizer with six UV-C lamps was manufactured, and the dissociation rate of ozone gas was evaluated.¹⁹ O_3 densities were calculated by estimating oxygen atoms using the major reaction equations for ozone formation and dissociation, and measurement of the time constant of diffusion decrease (τ_{diff}); they were compared with the experimental results. The ratio of $R = [\text{O}]/[\text{O}_3]$ between the oxygen atom density $[\text{O}]$ and the ozone density $[\text{O}_3]$ was investigated for power density at measurement positions away from the plasma source. The atomic oxygen state was estimated, and $[\text{O}]$ was calculated at the exit of the plasma sterilizer.

II. MATERIALS AND METHODS

A. Experiment of O_3 Generated from Plasma Source

Figure 1 shows a plasma sterilizer with six UV-C lamps that increases reactive species by decomposing the ozone gas generated from the two plasma sources of surface microdischarge (SMD). The plasma sterilizer is composed of acrylic with a thickness

FIG. 1: (a) Plasma sterilizer with two plasma sources and six UV-C lamps; (b) driving module of plasma source of surface SMD

of 1 cm, a width of 40 cm, a length of 20 cm, and a height of 42 cm. By attaching an Al foil of 0.1-mm thickness to the inner and outer walls of the acrylic, external incident light is prohibited from entering the interior of the sterilizer and internal UV-C light is prevented from leaking to the outside environment. A plasma sterilizer is installed in the center of an acrylic box with a thickness of 1 cm and width, length and height of 1 m. The air sucked from the fan (air flow of 350 m³/h and diameter of 171 mm) at the bottom of the plasma sterilizer is mixed with reactive species generated from the plasma sources of the two SMDs. The air passes a TiO₂ filter with a thickness of 1 cm, UV-C lamps, and a TiO₂ filter. When air passes the top of the TiO₂ filter, its velocity is approximately 1.5 m/s. The surrounding gas velocity in the laboratory is 0, determined using a flow meter (KIMO, LV-50). The ozone density is measured using an ozone sensor (Test Auction, SKT-9300) installed on the air outlet at the top of the plasma sterilizer. Six UV-C lamps with 36-W power and sized 190 × 35 × 18 mm (length × width × thickness) are installed at 5-cm distance intervals, with two lamps at the same height. A TiO₂ filter is installed to adjust the air velocity and uniformity. The distance between the O₃ sensor and the plasma source of the SMD is approximately 30 cm. The density of reactive species, such as OH and O₂, generated by the TiO₂ filter owing to UV-C is important for sterilization, along with atomic O generated by O₃ dissociation generated by the UV-C lamps. We measured the dissociation of ozone gas using six UV-C lamps.

Figure 1(b) shows the driving modules with the plasma source of SMD. The area of the SMD is 0.12 cm². The power densities of 1.20, 2.37, and 3.53 w/cm² (4.0 kV, 27 mA) applied on the electrode of SMD are obtained by adjusting the duty ratio from 0.826% to 2.44%. The gap of modules equipped with SMD is 1 mm. The time constant of diffusion loss (τ_{dif}) for O₃ gas in an acryl box with a volume of 1 m³ is measured by turning off the power of the plasma sterilizer and measuring the O₃ density versus time. It should be noted that $\tau_{dif} = 400$ s is obtained via the equation $[O_3] = Ae^{-t/\tau}$,¹⁷ where A and t are the initial ozone density and time, respectively.

B. Calculation of O₃ Gas Generated from Plasma Source of SMD

Ozone generated and dissociated by atmospheric plasma in air is expressed using the three equations listed in Table 1. The ozone is generally produced by the dissociation of oxygen molecules due to electron impact and bonding of atomic oxygen with oxygen molecules in the atmosphere. Moreover, ozone is mainly quenched by NO and O; NO is produced by the reaction of N₂(AΣ_u⁺) + O, N(²D) + O₂, N(²D) + O₃, and N(²P) + O₂.²¹ Equations (1–2) were obtained using the three reactions listed in Table 1.

$$\frac{dn_{O_3}}{dt} = k_1 n_M n_O n_{O_2} - (k_2 n_{NO} + k_3 n_O + \tau_{dif}^{-1}) n_{O_3} \quad (1)$$

$$n_{O_3} = \frac{k_1 n_M n_O n_{O_2} (1 - e^{-(k_2 n_{NO} + k_3 n_O + \tau_{dif}^{-1})t})}{k_2 n_{NO} + k_3 n_O + \tau_{dif}^{-1}} \quad (2)$$

Here, n_i ($i = O_3, O, M, NO, O_2$) = density of each species; M = third body; and k_i ($i = 1-3$) is the rate constant. Equation (3) is obtained by $k_2 n_{NO} \cong k_3 n_O < 10^{-5} s^{-1} \ll \tau_{dif}^{-1} = 0.0025 s^{-1}$ from Equation (2) due to n_{NO} (or n_O)/ $n_{O_3} \cong 10^{-4}$ in the plasma source.²²

$$n_{O_3} = \frac{k_1 n_M n_O n_{O_2} (1 - e^{-\tau_{dif}^{-1}t})}{\tau_{dif}^{-1}} \quad (3)$$

TABLE 1: Rate constants for chemical reactions (rate constant: m³ s⁻¹ for k_1 and k_3 ; m⁶ s⁻¹ for k_2)

Index	Chemical reaction	Rate constant	Ref.
k_1	$O + O_2 + M \rightarrow O_3 + M$	3.4×10^{-46}	20
k_2	$O_3 + NO \rightarrow N O_2 + O_2$	1.9×10^{-20}	20
k_3	$O_3 + O \rightarrow 2 O_2$	8.0×10^{-21}	21

In Equation (3), the O₃ density is calculated by estimating the atomic oxygen density n_O using $k_1 = 3.4 \times 10^{-46} \text{ m}^{-6}\text{s}^{-1}$, $n_M = 2.6 \times 10^{25} \text{ m}^{-3}$, $n_{O_2} = 5.6 \times 10^{24} \text{ m}^{-3}$, and $\tau_{dif}^{-1} = 0.0025 \text{ s}^{-1}$.

III. RESULTS AND DISCUSSION

Figure 2 shows the results of the measurement and calculation of the O₃ density generated from a plasma sterilizer without using UV-C lamps. The graphs of the measurement and calculation of O₃ gas are represented by black and dotted red lines, respectively, and the time constant of the diffusion loss (τ_{dif}) of O₃ gas is 400 s. The O₃ density increases for elapsed time and is saturated at 0.14 ppm ($3.9 \times 10^{18} \text{ m}^{-3}$), 0.26 ppm ($7.1 \times 10^{18} \text{ m}^{-3}$) and 0.48 ppm ($1.3 \times 10^{19} \text{ m}^{-3}$) over 1,000 s, 1300 s, and 1,600 s at 1.2, 2.73, and 3.53 W/cm², respectively. The O₃ density was calculated by estimating [O] ($= n_O$) in the O₃ measurement graph in power densities of 1.2, 2.73, and 3.53 W/cm², respectively. [O] was estimated to be $1.9 \times 10^{11} \text{ m}^{-3}$, $3.4 \times 10^{11} \text{ m}^{-3}$, and $6.3 \times 10^{11} \text{ m}^{-3}$, respectively.

The calculation results for O₃ agree with the experimental results. The ratio of [O]/[O₃] was approximately 4.9×10^{-8} for a power density of 1.2–3.53 Wcm⁻². The saturated O₃ density was used as [O₃] for this ratio. Figure 3 shows the estimated O density for power density of 1.2–3.53 Wcm⁻². The O density is proportional to the power density. This result agrees well with Equation (3).

Figure 4 shows the results of the experiments and calculation for ozone density generated from the plasma sterilizer using six UV lamps. Approximately 70% of the O₃ density generated from the plasma source was dissociated by the lamps, which had a wavelength of 254 nm. Moreover, the graph of O₃ density was similar to that shown in Fig. 2. The dissociation of O₃ gas by UV-C lamp is expressed as follows:

FIG. 2: Experiments and calculation for ozone density generated from plasma sterilizer without UV lamps versus time at power densities of 1.2, 2.73, and 3.53 Wcm⁻²

FIG. 3: Measured O density at power densities of 1.2, 273, and 3.53 Wcm⁻²

FIG. 4: Experiments and calculation for ozone density generated from plasma sterilizer using six UV-C lamps with 254-nm wavelength.

The dissociation energy of O₃ gas was 1.1 eV and it was dissociated by a UV-C lamp that had a wavelength of 254 nm and energy of 4.9 eV; this was obtained using Equation (4).²³ The absorption cross section (σ) of 254-nm UV-C²⁴ was extremely high;

that is, σ (254 nm) = 1.1×10^{-17} cm². It should be noted that OH radicals and hydrogen peroxide were found to increase. This estimation was confirmed by the experiment with Ki,²⁵ where a UV-C lamp was installed in front of the plasma sterilizer. Here, the concentrations of OH radicals and hydrogen peroxide were found to increase by over 30%, whereas the O₃ gas density decreased by approximately 15%. However, there was no difference in the sterilization rate of *S. aureus* with or without UV-C lamps. H₂O₂²⁵ has an atmospheric lifetime²⁶ of 14–42 h. Moreover, it is reported to be effective in inactivating the respiratory viruses, such as adenovirus,²⁷ influenza A virus,²⁸ and COVID-19.²⁹

O₃ density ([O₃]) was saturated at 0.043 (1.2×10^{18} m⁻³), 0.079 (2.1×10^{18} m⁻³) and 0.14 ppm (3.9×10^{18} m⁻³) over 1,000 s, 1,300 s, and 1,600 s at 1.2, 2.73, and 3.53 W/cm², respectively. In contrast, [O₃] and [O] decreased by approximately 70% without UV-C lamps and [O] was calculated as 5.7×10^{10} , 1.0×10^{11} , 1.9×10^{11} , respectively, in 1.2, 2.73, and 3.53 Wcm⁻², and the ratio of [O]/[O₃] was similar to that without UV-C lamps, calculated as about 4.9×10^{-8} . The O₃ density decreased to 0.043 ppm (1.5×10^{18}) less than the 0.05 ppm at the exit of the plasma sterilizer using six UV-C lamps in a power density of 1.2 Wcm⁻².

Figure 5 shows the results for O₃ density calculated using measurement results for O₃ density generated from the plasma source and using $\tau_{diff} = 40$ s, according to experiments conducted by Shimizu.¹⁷ Powers of 15 kV and 5–10 kHz were applied to the stainless steel mesh installed on an Al₂O₃ dielectric with a thickness of 0.76 mm as a plasma source of SMD. The measurement position was 5 mm away from the SMD. The atomic oxygen [O] was 7.0×10^{15} , 1.2×10^{16} , 1.8×10^{16} m⁻³ at power densities of 9.5×10^{-4} , 4.1×10^{-3} and 9.5×10^{-3} Wcm⁻², respectively. [O]/[O₃] was approximately 6×10^{-7} in applied power density.

Figure 6 shows the value of $R = [O]/[O_3]$ versus distance (d) from the plasma source to the measurement location. The value of R determined in our study and by Shimizu¹⁴ was 4.8×10^{-8} ($d = 30$ cm) and 6.0×10^{-7} ($d = 5$ mm), respectively, as described

FIG. 5: Measurement and calculation for ozone gas generated from plasma sterilizer versus time. Measurement results provided by Shimizu et al.¹⁷ Measurement position = 5 mm from plasma source; $\tau_{diff} = 40$ s.

FIG. 6: Ratio ([O]/[O₃]) versus distance from plasma source to measurement

previously. The value of R (at $d = 0$ cm) obtained by Sakiyama et al.¹⁹ was 1×10^{-4} . Sakiyama and colleagues simulated the species density of O₃, O, OH, H₂O₂, and others, using 50 species and 600 chemical equations in the numerical model of SMD. A power of 10 kHz and a power density of 0.05 Wcm⁻² were applied to the SMD model. O₃ and O densities were simulated as about 10²⁴ and 10²⁰ m⁻³, respectively, in a steady state after discharge for 1,000 s, and $R = 1 \times 10^{-4}$ was obtained.

Because the life span of ozone is longer than 30 min¹⁸ and that of oxygen atoms is extremely short, R decreases as the measurement position is moved further away from the plasma source of SMD. The excited states of atomic oxygen of O(³P), O(¹D), and O(¹S) have life spans (ζ) of tens of nanoseconds, 110 s and 0.8 s, respectively.^{30,31} Therefore, the state of atomic oxygen at $d = 0$ and 30 cm can be estimated as O(³P), O(¹D), O(¹S), and O(¹D), respectively, owing to life span.

It is evident from this study that the ratio of atomic oxygen to ozone is about 10⁻⁴ inside the plasma generation region; however, the ozone concentration is controlled below 0.05 ppm, which is the international safety criterion. Hence, we emphasize that this biocompatible plasma sterilizer can be used in a hospital safely with high efficacy, mainly by using ozone at the downstream region of the air outlet.

IV. CONCLUSION

The O₃ density generated from a plasma sterilizer composed of two plasma sources of SMD and using six UV-C lamps of 254-nm wavelength decreased to 70% by O₃ dissociation in the generation of ozone density of 0.043 ppm and atomic oxygen of 5.7×10^{10} m⁻³ at the exit in 1.2 Wcm⁻². The O₃ density was calculated correctly by estimating the oxygen atoms using simple reaction equations for ozone formation and dissociation, and measurement of the decrease in the time constant of diffusion ($\tau_{dif} = 400$ s). The results for O₃ density were in good agreement with those for 30 cm away from the plasma source. The value of R ([O]/[O₃]) determined in our study was 4.8×10^{-8} . As the distance (d) between the measurement position from the plasma source increased, the value of R decreased due to the long life (ζ) of O₃ and the short life of O. Therefore,

the states of atomic oxygen at $d = 0$ and 30 cm can be estimated as O (³P, $\zeta =$ dozens of nanoseconds), O(¹D, $\zeta = 110$ s), O(¹S, $\zeta = 0.8$ s), and O(¹D), respectively. The plasma sterilizer can be used in human-occupied spaces as well as in human-free spaces, thus reducing viruses and bacteria.

It is evident from this study that the ratio of atomic oxygen to ozone remains about 10^{-4} inside the plasma generation region; however, the ozone concentration is controlled to be below 0.05 ppm, which is the international safety criterion. Hence we emphasize that this biocompatible plasma sterilizer can be used in hospitals safely and with high efficacy, mainly by using ozone at the downstream region the of air outlet.

ACKNOWLEDGMENTS

This research was supported by the Leading Foreign Research Institute Recruitment Program through the National Research Foundation of Korea (NRF) funded by Korean Government (MSIT), Grant NRF-2016K1A4A3914113 and partially by Grant NRF-2021R1A6A1A03038785 and by the ITRC (Information Technology Research Center) support program, Grant IITP-2020-0-01846. No conflict of interest of any kind exists in relation to this research.

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