

Simple Visualization of the Distribution of Ozone Concentration Generated by Atmospheric-pressure Plasma Jet Irradiation Using Test Strips

by

Hiroshi KUWAHATA*¹ and Takaaki YAMAGUCHI*²

(Received on Mar. 27, 2020 and accepted on Jul. 24, 2020)

Abstract

The distribution of ozone (O_3) concentration generated by atmospheric-pressure argon (Ar) plasma jet irradiation was visualized using test strips. The O_3 test strips become brown in the presence of O_3 , enabling the O_3 concentration to be easily determined on the basis of the darkness of the color. An Ar plasma jet generated with a frequency of 10 kHz, an applied voltage of 10 kV, and an Ar gas flow rate of 10 L/min was ejected into the air in the form of an inverse cone with a length of approximately 15 mm and a maximum diameter of 6 mm. After plasma jet ejection for 1 s, the color of the O_3 test strips turned brown ($>210 \mu\text{g}/\text{m}^3$) except in the region of the plasma jet (inverted triangle). This result indicates that O_3 was generated in the region where the plasma came into contact with the air.

The results of this study suggested that the distribution of the O_3 concentration generated by atmospheric-pressure Ar plasma jet irradiation is dependent on the plasma ejection/irradiation duration, irradiation distance, and applied voltage.

Keywords: Atmospheric-pressure plasma, Ozone, Visualization, Test strips

1. Introduction

Research is being carried out on the application of atmospheric-pressure plasma in the surface treatment of solid materials¹⁻⁶⁾, water purification⁷⁻¹⁰⁾, and the inactivation of bacteria and fungi¹¹⁻¹⁵⁾. Recently, active research on the plasma has been carried out on a wide variety of biomedical applications¹⁶⁻¹⁹⁾ including wound healing²⁰⁻²³⁾, cancer cell treatment²⁴⁻²⁶⁾, and the introduction of genes and drug molecules into cells²⁷⁻³¹⁾. It is known that ozone (O_3) is generated owing to the introduction of plasma in the air. O_3 is generated because high-energy electrons in the plasma collide with oxygen (O_2) molecules in the air and the oxygen (O) atoms produced by the dissociation of the O_2 molecule bind to another O_2 molecule^{32, 33)}. O_3 is one of the reactive oxygen species (ROS) and has a high oxidation-reduction potential of 2.07 V^{34, 35)}, thereby easily oxidizing objects. This oxidizing power is useful in various fields of research.

The concentration of O_3 generated when an atmospheric-pressure plasma jet is ejected into the air is measured by ultraviolet (UV) absorption³⁶⁻⁴⁰⁾, an O_3 monitor⁴¹⁾, and molecular beam mass spectrometry (MBMS)⁴²⁾. The distribution of the O_3 concentration was studied by Winter *et al.* by means of UV absorption using a deuterium (D_2) lamp, a monochromator, an imaging spectrometer, and an intensified charge-coupled device (iCCD) camera³⁸⁾. It was also studied by Zhang *et al.* by means of UV absorption using a mercury (Hg) lamp, a monochromator, and a photomultiplier³⁹⁾.

As explained above, there have been reports on the distribution of the O_3 concentration when an atmospheric-pressure plasma jet is ejected into the air. However, the suggested measurement methods require much equipment and time. In addition, the distribution of the concentration of O_3 existing on the surface of an object when it is irradiated with an atmospheric-pressure plasma jet has not been measured.

In our previous study, we measured the distribution of the concentration of hydrogen peroxide (H_2O_2) generated by atmospheric-pressure argon (Ar) plasma jet irradiation using H_2O_2

*1 Associate Professor, Department of Electrical and Electronic Engineering

*2 Undergraduate Student, Department of Electrical and Electronic Engineering

test strips⁴³⁾. H_2O_2 is also an ROS with an oxidation-reduction potential of 1.77 V³⁴⁾. The color of the H_2O_2 test strips turns blue in the presence of H_2O_2 , and the concentration of H_2O_2 can be simply determined on the basis of the darkness of the color. In that study, it was found that the distribution of the H_2O_2 concentration is dependent on irradiation duration, irradiation distance, and applied voltage. Test strips for the simple measurement of the O_3 concentration are commercially available. The color of the test strips turns brown in the presence of O_3 , and the O_3 concentration can be determined on the basis of the darkness of the color. The principle of the color reaction is due to the oxidation of the colorless iodide on O_3 test strips to brown iodine induced by O_3 . There have been no reports on visualizing the distribution of O_3 concentration obtained using O_3 test strips to the best of our knowledge.

In this study, using O_3 test strips, we tried to visualize the dependences of the O_3 concentration on plasma ejection/irradiation duration, irradiation distance, and applied voltage when an atmospheric-pressure Ar plasma jet is ejected into the air or when an object is irradiated with an atmospheric-pressure Ar plasma jet.

2. Experimental Methods

Figure 1 shows a schematic of the experimental setup used in this study. In the unit used to generate a plasma jet, a copper tube (inner diameter, 4 mm; outer diameter, 6 mm) as the discharge electrode was inserted into a dielectric quartz tube (length, 50 mm; inner diameter, 6 mm; outer diameter, 8 mm), around which a copper foil (thickness, 0.05 mm; width, 10 mm) was wrapped as the ground electrode⁴⁴⁾. A high AC voltage was applied to the electrodes to induce a dielectric barrier discharge inside the quartz tube between the electrodes and to convert the Ar gas into plasma, which was then ejected into the air in the form of a jet. A high-voltage power supply (LHV-10AC, Logy Electric Co., Ltd.) was used to generate the Ar plasma jet (frequency, 10 kHz; applied voltage, 3–10 kV; Ar gas flow rate, 10 L/min). The gas flow rate was adjusted using a gas flow meter (RK-1250, Kofloc Kyoto, Kojima Instruments Inc.). The voltage and current were measured using a high-voltage probe (P6015A, Tektronix, Inc.) and a current probe (A621, Tektronix, Inc.), respectively, which were both connected to a digital oscilloscope (TDS1001B, Tektronix, Inc.).

The plasma jet irradiated in the container made of transparent acrylic resin. The dimensions of the container are 120 (W) × 100

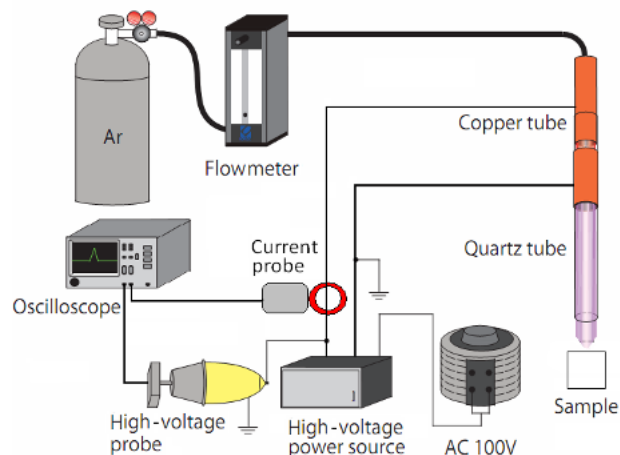


Fig. 1 Schematic of experimental setup.

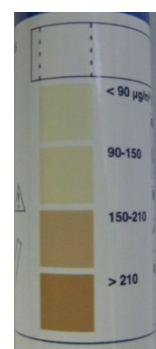


Fig. 2 Color chart of O_3 test strip.

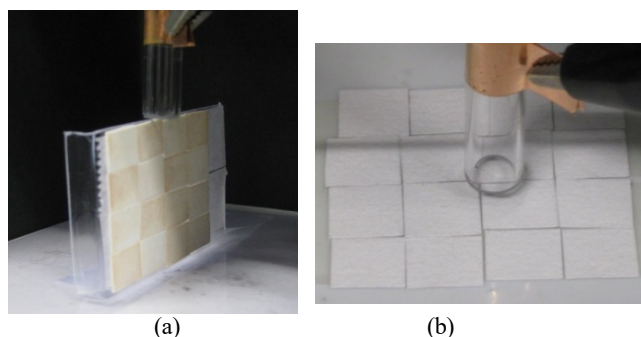


Fig. 3 Photographs of O_3 test strips during measurement of distribution of O_3 concentration

(a) Ejection into air and (b) Irradiation onto object.

(D) × 180 (H) mm³. The front side of the container is open. The height of the sample stage is variable in the range of 60–110 mm.

O_3 test strips (Macherey-Nagel) were used to determine the O_3 concentration. Figure 2 shows the color chart of the O_3 test strips. The color of the O_3 test strips turns brown in the presence of O_3 , and the O_3 concentration can be determined in four levels (<90, 90–150, 150–210, and >210 µg/m³) on the basis of the darkness of the color.

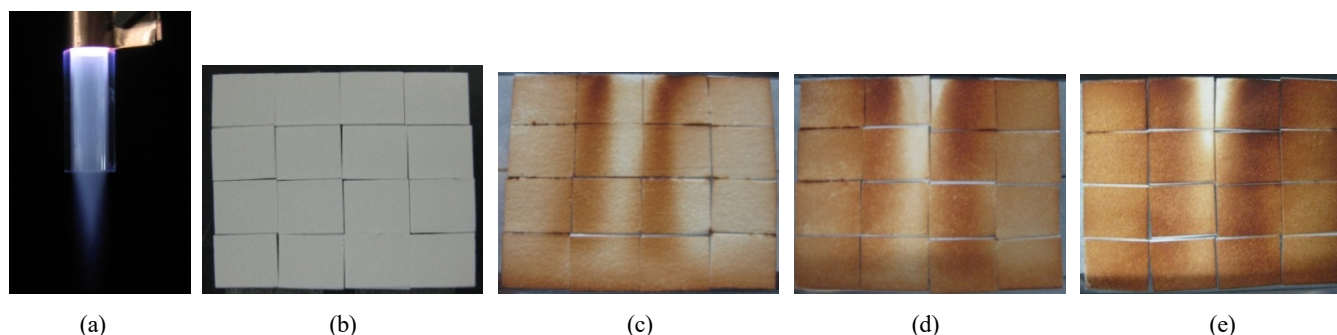


Fig. 4 Photographs of atmospheric-pressure Ar plasma jet ejected into air and O₃ test strips for different irradiation durations (applied voltage, 10 kV; Ar gas flow rate, 10 L/min) (a) Plasma jet, (b) Before ejection, (c) Ejection for 1 s, (d) Ejection for 10 s, and (e) Ejection for 30 s.

Figure 3 shows photographs of the O₃ test strips during the measurement of the distribution of the O₃ concentration. When the plasma jet was ejected into the air [Fig. 3(a)], a total of 16 (4 × 4) O₃ test strips (dimensions of one test strip, 8 × 10 mm²; dimensions of 16 test strips, 32 × 40 mm²) were arranged and set up vertically immediately below the center of the quartz tube. When the plasma jet was irradiated onto an object [Fig. 3(b)], a total of 16 (4 × 4) O₃ test strips were placed immediately below the quartz tube.

3. Results and Discussion

3.1 Ejection of atmospheric-pressure Ar plasma jet into air

3.1.1 Dependence of distribution of O₃ concentration on plasma ejection duration

Figure 4 shows photographs of the atmospheric-pressure Ar plasma jet ejected into the air and O₃ test strips for different plasma ejection durations. When an Ar plasma jet was generated with an applied voltage of 10 kV and an Ar gas flow rate of 10 L/min, an atmospheric-pressure Ar plasma jet with an inverse cone shape having a length of ~15 mm and a maximum diameter of 6 mm was ejected into the air [Fig. 4(a)]. As shown in Fig. 4(b), the color of the O₃ test strips remained white before the plasma ejection. After ejecting for 1 s [Fig. 4(c)], the color of the test strips that came into contact with the plasma jet remained white but that in the vicinity of that area turned brown. From this result, it was found that O₃ was generated simultaneously with the ejection of the plasma jet in the region where the plasma jet came into contact with the air, but it was not generated in the plasma. The nearer the region was from the plasma jet, the greater the darkness of brown became. With further distance from the plasma jet, the color became lighter. This finding indicates that the O₃ generated through contact between the plasma jet and the air diffused in its vicinity. By comparing the darkness of the color of the O₃ test

strips with the color chart, the O₃ concentration was found to be >210 µg/m³ at the edge of the plasma jet (within a radius of ~4–8 mm from the center of the plasma) and was 150–210 µg/m³ in its vicinity (within a radius of 8–20 mm from the center of the plasma). The shape of the white area was almost identical to that of the plasma jet. The length of the white area was ~16 mm and the width was ~10 mm at the top of the O₃ test strips (immediately below the quartz tube), which gradually decreased with distance from the top. After 10-s ejection [Fig. 4(d)], the color of the O₃ test strips in the vicinity became darker than that in the case of 1-s ejection. This finding indicates that the O₃ concentration increased; the O₃ concentration was found to be >210 µg/m³ even in the vicinity. The size of the white area decreased slightly [~16 (L) mm × ~8 (W) mm]. After 30-s ejection [Fig. 4(e)], the color of the O₃ test strips in the vicinity became much darker. This finding indicates that the O₃ concentration further increased; the O₃ concentration was found to be >210 µg/m³ even in the vicinity. The size of the white area was further decreased [~13 (L) mm × ~6 (W) mm]. With increasing plasma ejection duration, the white area decreased. This is because the O₃, which was generated at the site where the plasma jet came into contact with air, came into continuous contact with the O₃ test strips and changed the color of the test strips at the edge of the plasma jet. From the above findings, with increasing plasma ejection duration, the concentration of O₃ generated in the vicinity of plasma was found to increase. It is considered that the concentration of generated O₃ does not change with time and remains constant when measured in a completely open space. In our experiment, however, the O₃ concentration increased with the plasma ejection duration. This is because the O₃ test strips were placed in an acrylic container, which was only open at the front side and thus was not completely open. In this case, some of the generated O₃ remained in the container and the O₃ concentration increased with time.

In the study of Winter *et al.*, a plasma jet of ~4 mm length was

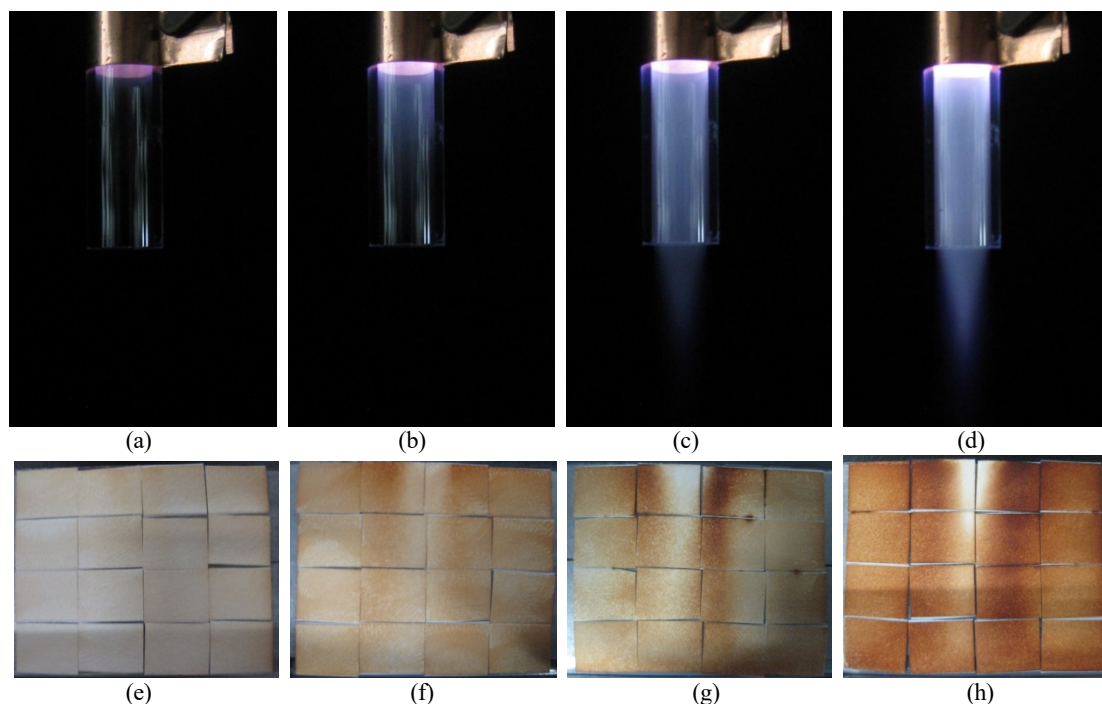


Fig. 5 Photographs of atmospheric-pressure Ar plasma jet ejected into air and O₃ test strips for different applied voltages (Ar gas flow rate, 10 L/min; plasma ejection duration, 30 s) (a, e) 3 kV, (b, f) 6 kV, (c, g) 8 kV, and (d, h) 10 kV.

generated by introducing a flow of an Ar gas [3 standard L/min (slm)] mixed with O₂ into a quartz capillary (inner radius, 0.8 mm) with a high-frequency applied voltage (~ 1 MHz, 2–6 kV_{pp})³⁸⁾. The distribution of the O₃ concentration was studied by means of UV absorption at a wavelength $\lambda = 254$ nm using a D₂ lamp, a monochromator, an imaging spectrometer, and an iCCD camera. They reported that the O₃ concentration of $1.5 \times 10^{16} \text{ cm}^{-3}$ at the end of the plasma jet (position in the axial direction along the capillary, $z = 4$ mm) was the highest when the gas mixture of Ar +1.0% O₂ was used. With increasing distance in the ejection direction, the O₃ concentration gradually decreased to $2.5 \times 10^{15} \text{ cm}^{-3}$ at $z = 19$ mm. Regarding the spread of O₃ in the horizontal direction, the radius r of the plasma jet was 0.75 mm at $z = 4$ mm; it reached the maximum ($r = 2.0$ mm) at $z = 15$ mm and decreased to $r = 1.75$ mm at $z = 19$ mm. In the study of Zhang *et al.*, a plasma jet of ~ 9 mm length was generated with a flow of Ar gas (2slm) mixed with 2% O₂ in a quartz tube (inner diameter, 1.8 mm) with a plasma power of 13.56 MHz and 6.5 W³⁹⁾. The distribution of the O₃ concentration was studied by means of UV absorption at a wavelength $\lambda = 253.7$ nm using a Hg lamp, a monochromator, and a photomultiplier. They reported that the O₃ concentration of $1.9 \times 10^{15} \text{ cm}^{-3}$ was the highest at 4 mm from the end of the plasma jet ($z = 13$ mm). With increasing distance in the ejection direction, the O₃ concentration gradually decreased to $6 \times 10^{14} \text{ cm}^{-3}$ at $z = 25$ mm. Regarding the spread of O₃ in the

horizontal direction, the radius r of the plasma jet was 1.5 mm at the end of the plasma jet ($z = 9$ mm), reached the maximum ($r = 2.5$ mm) at $z = 15$ mm, and decreased to $r = 1.5$ mm at $z = 19$ mm. In the study of Deng *et al.*, a plasma jet was generated with a flow of nitrogen (N₂) gas (8 slm) in the quartz tube (inner diameter, 5 mm) with the application of direct current (5–30 mA)⁴¹⁾. By ejecting the plasma jet through a metal mesh, an afterglow was generated. They examined the distribution of the O₃ concentration from the afterglow using an O₃ monitor. In the case of the current of 25 mA, they reported that the O₃ concentration was ~ 22 ppm at 2 mm from the metal mesh ($z = 2$ mm). With increasing distance in the ejection direction, the O₃ concentration gradually increased, peaked (90 ppm) at $z = 15$ mm, and then gradually decreased to ~ 30 ppm at $z = 30$ mm.

3.1.2 Dependence of distribution of O₃ concentration on applied voltage

Figure 5 shows photographs of the atmospheric-pressure Ar plasma jet ejected into the air and O₃ test strips for different applied voltages. At an applied voltage of 3 kV [Fig. 5(a)], a plasma jet was observed only in the vicinity of the electrode and it seemed that no plasma jet was ejected from the quartz tube. The color of the entire O₃ test strips turned slightly brown [Fig. 5(e)] and no white area indicating plasma ejection was observed. The O₃ concentration over the entirety of the O₃ test strips was 90–150

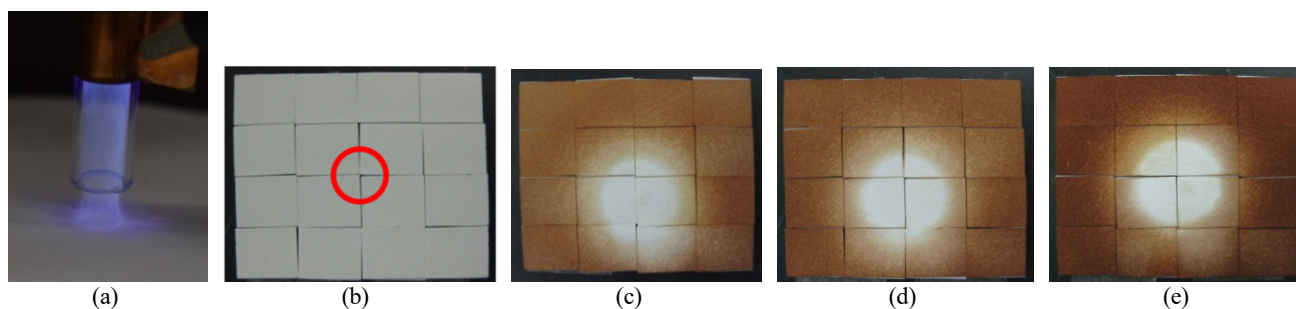


Fig. 6 Photographs of atmospheric-pressure Ar plasma jet irradiation and O₃ test strips irradiated with plasma jet for different irradiation durations (applied voltage, 10 kV; Ar gas flow rate, 10 L/min; irradiation distance, 5 mm)

(a) Plasma jet, (b) Before irradiation, (c) Ejection for 1 s, (d) Ejection for 5 s, and (e) Ejection for 10 s.

$\mu\text{g}/\text{m}^3$. This finding indicates that O₃ was generated in the quartz tube even when the plasma jet was not ejected from the quartz tube but generated in the quartz tube. At an applied voltage of 6 kV [Fig. 5(b)], no plasma was again ejected from the quartz tube. A small white area indicating plasma jet ejection was observed on the O₃ test strips [Fig. 5(f)]. The dimensions of the white area were ~ 16 (L) mm $\times$ ~ 7 (W) mm. The O₃ concentration was >210 $\mu\text{g}/\text{m}^3$ at the edge of the plasma jet and $150\text{--}210$ $\mu\text{g}/\text{m}^3$ in its vicinity. At an applied voltage of 8 kV [Fig. 5(c)], a plasma jet appeared to be ejected from the quartz tube to a distance of ~ 10 mm. The white area indicating plasma jet ejection was observed on the O₃ test strips [Fig. 5(g)]. The dimensions of the white area were ~ 16 (L) mm $\times$ ~ 8 (W) mm. The O₃ concentrations were >210 $\mu\text{g}/\text{m}^3$ at the edge of the plasma jet and $150\text{--}210$ $\mu\text{g}/\text{m}^3$ in its vicinity. At an applied voltage of 10 kV [Fig. 5(d)], the plasma jet was apparently ejected from the quartz tube to a distance of ~ 15 mm. A white area indicating plasma jet ejection was observed on the O₃ test strips [Fig. 5(h)]. The dimensions of the white area were ~ 13 (L) mm $\times$ ~ 6 (W) mm. The O₃ concentration was >210 $\mu\text{g}/\text{m}^3$ at the edge of the plasma jet and in its vicinity. Comparing the results in Figs. 5(f)–5(h), we can see that the color of the O₃ test strips became darker with increasing applied voltage. From this result, it was concluded that the O₃ concentration increased with increasing applied voltage, suggesting that the number of high-energy electrons is larger in a plasma jet generated at a higher applied voltage.

3.2 Irradiation of atmospheric-pressure Ar plasma jet onto object

3.2.1 Dependence of distribution of O₃ concentration on irradiation duration

Figure 6 shows photographs of plasma jet irradiation and O₃ test strips irradiated with the plasma jet for different irradiation durations. Filter paper, instead of O₃ test strips, was placed under

the quartz tube to take a photograph shown in Fig. 6(a). The plasma jet reached the filter paper and extended horizontally on the filter paper. The diameter of the region over which the plasma extended was ~ 17 mm. The color of the O₃ test strips before plasma irradiation was white [Fig. 6(b)]. The plasma jet was ejected onto the center of the O₃ test strips. After plasma jet irradiation for 1 s [Fig. 6(c)], the color of the O₃ test strips turned brown except in the area irradiated with the plasma jet. This finding indicates that O₃ was generated immediately on the O₃ test strips irradiated with the plasma jet but was not generated in the region irradiated with the plasma jet. The diameter of the white area was ~ 12 mm, which was smaller than that of the region over which the plasma extended (~ 17 mm). This result indicates that O₃ was generated inside the edge of the region over which the plasma extended horizontally on the O₃ test strips. The area of the O₃ test strips that turned brown extended to a radius of at least 26 mm from the center of the region irradiated with the plasma and the O₃ concentration was >210 $\mu\text{g}/\text{m}^3$. Namely, when an object was irradiated with atmospheric-pressure Ar plasma jet for 1 s, O₃ with a concentration of >210 $\mu\text{g}/\text{m}^3$ was generated in the vicinity of the object. The region with a O₃ concentration >210 $\mu\text{g}/\text{m}^3$ was considered to extend to a radius of at least 26 mm from the center of the region irradiated with the plasma. As seen from the results in Figs. 6(c)–6(e), the brown color of the O₃ test strips became darker with increasing irradiation duration. This finding indicates that the O₃ concentration in the vicinity of the object increased with increasing irradiation duration. The diameter of the white area changed negligibly and was in the range of $\sim 12\text{--}14$ mm regardless of the irradiation duration. On the basis of the above findings, we concluded that O₃ was not generated in the region of the object irradiated with the plasma and no effect of O₃ was expected in this region.

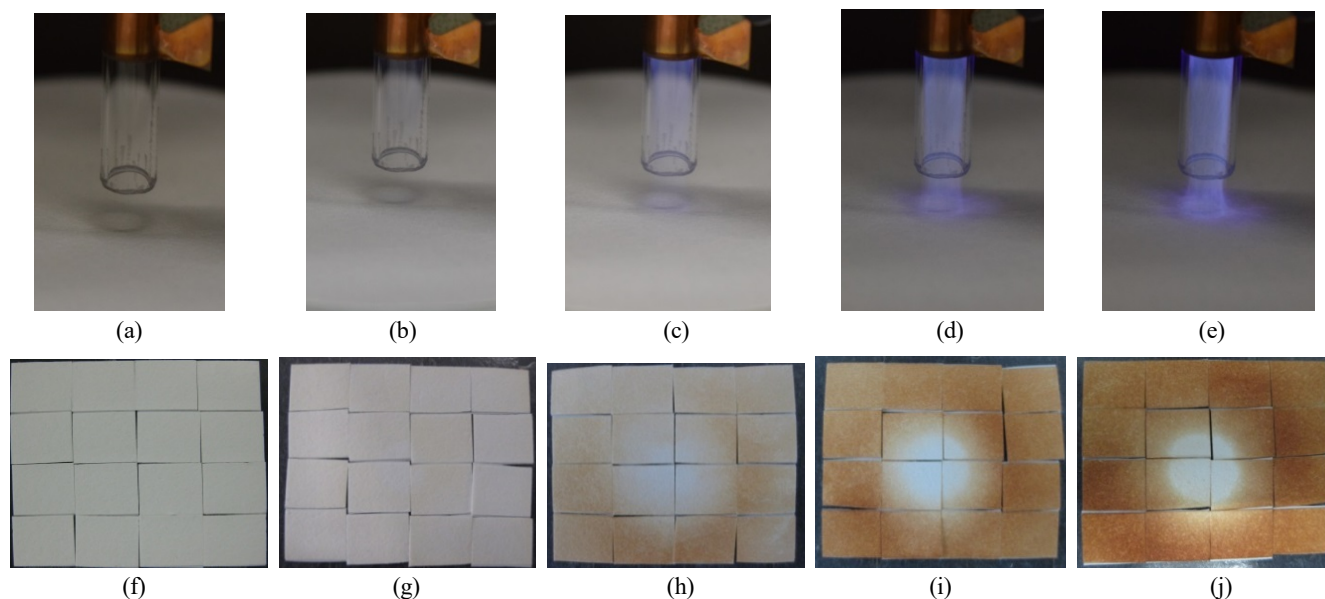


Fig. 7 Photographs of atmospheric-pressure Ar plasma jet irradiation and O_3 test strips irradiated with plasma jet for different applied voltages (Ar gas flow rate, 10 L/min; irradiation distance, 5 mm; irradiation duration, 10 s) (a, f) 3 kV, (b, g) 4 kV, (c, h) 6 kV, (d, i) 8 kV, and (e, j) 10 kV.

3.2.2 Dependence of distribution of O_3 concentration on applied voltage

Figure 7 shows photographs of plasma jet irradiation and O_3 test strips irradiated with the plasma jet for different applied voltages. At an applied voltage of 3 kV [Fig. 7(a)], no plasma was generated. As shown in Fig. 5(a), at the same voltage, a plasma jet was generated in the vicinity of the electrode. From this finding, an applied voltage of 3 kV is deemed the minimum voltage required for the generation of a plasma jet. At an applied voltage of 3 kV, the color of the O_3 test strips remained white, as shown in Fig. 7(f), because no plasma jet was generated. At an applied voltage of 4 kV [Fig. 7(b)], a plasma jet was generated only in the vicinity of the electrode. The color of the entirety of the O_3 test strips became slightly brown, as shown in Fig. 7(g). The O_3 concentration was $<90 \mu\text{g}/\text{m}^3$. This finding indicates that O_3 was generated and extended horizontally on the O_3 test strips to a radius of at least ~ 26 mm from the center of the region irradiated with the plasma jet, even when no plasma jet was ejected from the quartz tube. At an applied voltage of 6 kV [Fig. 7(c)], a plasma jet was ejected from the quartz tube and came into weak contact with the O_3 test strips. As shown in Fig. 7(h), the color of the area irradiated with the plasma jet remained white but the color of the O_3 test strips in the vicinity of that area turned slightly brown, and the O_3 concentration was estimated to be $90\text{--}150 \mu\text{g}/\text{m}^3$. At an applied voltage of 8 kV [Fig. 7(d)], plasma jet was ejected from the quartz tube, came into contact with the O_3 test strips, and then

extended horizontally over the O_3 test strips. The diameter of the region over which the plasma extended was ~ 13 mm. As shown in Fig. 7(i), the brown color of the area on the O_3 test strips in the vicinity of the region irradiated with the plasma jet became much darker. The O_3 concentration was $150\text{--}210 \mu\text{g}/\text{m}^3$. The diameter of the white area was ~ 12 mm, which is almost the same as that of the region over which the plasma jet extended. At an applied voltage of 10 kV [Fig. 7(e)], a plasma jet was ejected from the quartz tube, came into contact with the O_3 test strips, and then extended horizontally over the O_3 test strips. The diameter of the region over which the plasma extended was ~ 17 mm. As shown in Fig. 7(j), the brown color of the area on the O_3 test strips in the vicinity of the region irradiated with the plasma jet became much darker. The O_3 concentration was $>210 \mu\text{g}/\text{m}^3$. The diameter of the white area was ~ 12 mm. From the above findings, the diameter of the region over which the plasma extended was found to increase with increasing applied voltage. However, the diameter of the region where no O_3 was generated (white area) remained the same because the diameter of the area on the O_3 test strips that came into contact with the plasma jet remained the same. From these results, it was found that the O_3 concentration in the vicinity of an object increased with increasing applied voltage.

3.2.3 Dependence of distribution of O_3 concentration on irradiation distance

Figure 8 shows photographs of plasma jet irradiation and O_3

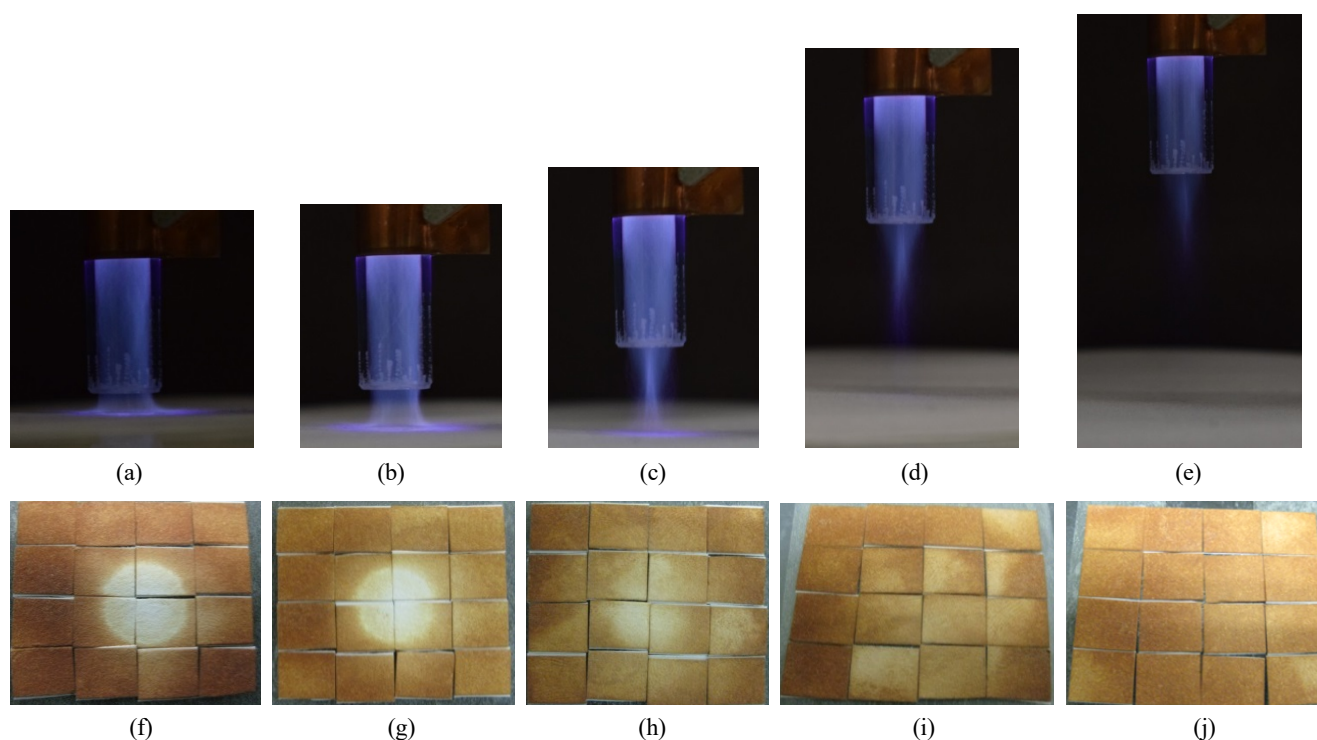


Fig. 8 Photographs of atmospheric-pressure Ar plasma jet irradiation and O_3 test strips irradiated with plasma jet for different irradiation distances (applied voltage, 10 kV; gas flow rate, 10 L/min; irradiation duration, 10 s) (a, f) 2 mm, (b, g) 5 mm, (c, h) 10 mm, (d, i) 20 mm, and (e, j) 30 mm.

test strips irradiated with the plasma jet for different irradiation distances. At irradiation distances of 2–10 mm [Figs. 8(a)–8(c)], a plasma jet was ejected from the quartz tube, came into contact with the filter paper, and then extended horizontally over the filter paper. However, at an irradiation distance of 20 mm [Fig. 8(d)] or 30 mm [Fig. 8(e)], a plasma jet ejected from the quartz tube did not come into contact with the filter paper. The diameters of the region over which the plasma extended were ~ 18 , ~ 16 , and ~ 13 mm at irradiation distances of 2, 5, and 10 mm, respectively. At irradiation distances of 2–10 mm [Figs. 8(f)–8(h)], the color of the region irradiated with the plasma jet remained white and the color of the O_3 test strips in the vicinity of this region turned brown. The O_3 concentration was $>210 \mu\text{g}/\text{m}^3$ regardless of the irradiation distance. The diameters of the white area were ~ 14 , ~ 13 , and ~ 11 mm at irradiation distances of 2, 5, and 10 mm, respectively. However, there were no white areas on the O_3 test strips at an irradiation distance of 20 or 30 mm, indicating that the diameter of the white area decreases with increasing irradiation distance. From these findings, when the irradiation distance was small (2–10 mm), no O_3 was generated in the region irradiated with the plasma jet and the effect of O_3 was not expected in this

region. In contrast, when the irradiation distance was large (≥ 20 mm), the plasma jet did not come into contact with the object but the O_3 generated at the edge of the plasma was diffused over the vicinity of the target. Therefore, the effect of O_3 is expected when the irradiation distance is large.

4. Conclusions

The distributions of O_3 concentration generated by atmospheric-pressure Ar plasma jet ejection in air and irradiation on an object were visualized using O_3 test strips. The dependences of the distribution of O_3 concentration on plasma ejection/irradiation duration, applied voltage, and irradiation distance were clarified.

When the plasma jet was ejected into air, the color of the O_3 test strips that came into contact with the plasma jet remained white, but the color of the O_3 test strips in the vicinity of that region turned brown. The region of the shape of plasma jet ejection (inverted triangle) was white on the O_3 test strips. The O_3 concentration in the vicinity of the region was found to increase with increasing plasma ejection duration, and applied voltage.

When an object was irradiated with the plasma jet, the

color of the O₃ test strips irradiated with the plasma jet remained white but that in the vicinity of that region turned brown. The shape of the white area was circular. The O₃ concentration in the vicinity of the object was found to increase with increasing irradiation duration and applied voltage. The manner of contact between the plasma jet and the O₃ test strips differed depending on the irradiation distance. When the irradiation distance was large (≥ 20 mm), the plasma did not come into contact with the O₃ test strips and the entirety of the O₃ test strips turned brown. In contrast, when the irradiation distance was small (2–10 mm), the color of the O₃ test strips irradiated with the plasma jet remained white but the color in the vicinity of that region turned brown. The shape of the white area was circular. In addition, when the irradiation distance was smaller, no O₃ was generated at the region where the plasma came into contact with the O₃ test strips and the effect of O₃ is not expected in this region. Therefore, to induce the effect of O₃ on an object, it is necessary to eject the plasma jet from above the object so that the plasma jet does not come into contact with the object.

The results of this study demonstrated that the visualization of the distribution of the O₃ concentration generated by atmospheric-pressure plasma jet ejection into air and irradiation onto an object was possible using O₃ test strips.

References

- 1) S. Kanazawa, M. Kogama, T. Moriwaki, and S. Okazaki, Stable glow plasma at atmospheric pressure, *J. Phys. D: Appl. Phys.* **21** p.838 (1988).
- 2) M. C. Kim, S. H. Yang, J. -H. Boo, and J. H. Han, Surface treatment of metals using an atmospheric pressure plasma jet and their surface characteristics, *Surf. Coatings Technol.* **174-175** p.839 (2003).
- 3) Z. Fang, Y. Qiu, and E. Kuffel, Formation of hydrophobic coating on glass surface using atmospheric pressure non-thermal plasma in ambient air, *J. Phys. D: Appl. Phys.* **37** p.2261 (2004).
- 4) H. Kuwahata, T. Haraki, and I. Mikami, Superhydrophilization of Si Surface by Atmospheric-Pressure Plasma Jet Irradiation: Analysis by Time-of-Flight Secondary Ion Mass Spectrometry (TOF-SIMS), *e-J. Surf. Sci. Nanotechnol.* **11** p.36 (2013).
- 5) H. Kuwahata and T. Yamaguchi, Removal of Aluminum on Glass Substrate by Atmospheric-Pressure Plasma Jet Irradiation, *e-J. Surf. Sci. Nanotechnol.* **14** p.231 (2016).
- 6) H. Kuwahata, Y. Murata, N. Hashimoto, and R. Segawa, Superhydrophilization of Surface of Aluminum Thin Film by Atmospheric-Pressure Plasma Jet Irradiation, *e-J. Surf. Sci. Nanotechnol.* **16** p.27 (2018).
- 7) W. F. L. M. Hoebe, E. M. van Veldhuizen, W. R. Rutgers, and G. M. W. Kroesen, Gas phase corona discharges for oxidation of phenol in an aqueous solution, *J. Phys. D: Appl. Phys.* **32** L133 (1999).
- 8) T. Kuroki, K. Yoshida, H. Watanabe, M. Okubo, and T. Yamamoto, Decomposition of Trace Phenol in Solution Using Gas-Liquid Interface Discharge, *Jpn. J. Appl. Phys.* **45** p.4296 (2006).
- 9) H. Kuwahata, K. Kimura, and R. Ohyama, Decolorization of Methylene Blue Aqueous Solution by Atmospheric-Pressure Plasma Jet, *e-J. Surf. Sci. Nanotechnol.* **8** p.381 (2010).
- 10) M. Uchida, S. Kezuka, Y. Oda, M. Isomura, and H. Kuwahata, Decolorization of Methylene Blue Aqueous Solution by Atmospheric-Pressure Plasma Jet Irradiation: NMR and MS Analyses, *Proc. Sch. Eng. Tokai Univ., Ser. J*, **56** p.9 (2016) [in Japanese].
- 11) M. Laroussi and X. Lu, Room-temperature atmospheric pressure plasma plume for biomedical applications, *Appl. Phys. Lett.* **87**, p.113902 (2005).
- 12) S. Ikawa, K. Kitano, and S. Hamaguchi, Effects of pH on Bacterial Inactivation in Aqueous Solutions due to Low-Temperature Atmospheric Pressure Plasma Application, *Plasma Process. Polym.* **7** p.33 (2010).
- 13) H. Kuwahata, E. Kumazawa, R. Ohyama, and A. Ito, Scanning Electron Microscopy Image of Escherichia Coli Exposed with Atmospheric-Pressure Plasma Jet, *e-J. Surf. Sci. Nanotechnol.* **8** p.74 (2010).
- 14) K. Oehmigen, J. Winter, M. Hähnel, C. Wilke, R. Brandenburg, K. D. Weltmann, and T. von Woedtke, Estimation of Possible Mechanisms of Escherichia coli Inactivation by Plasma Treated Sodium Chloride Solution, *Plasma Process. Polym.* **8** p.904 (2011).
- 15) H. Kuwahata, T. Yamaguchi, R. Ohyama, and A. Ito, Inactivation of Escherichia coli using atmospheric-pressure plasma jet, *Jpn. J. Appl. Phys.* **54** 01AG08 (2015).
- 16) G. Fridman, G. Friedman, A. Gutsol, A. B. Shekhter, V. N. Vasilets, and A. Fridman, Applied Plasma Medicine, *Plasma Process. Polym.* **5** p.503 (2008).

- 17) M. G. Kong, G. Kroesen, G. Morfill, T. Nosenko, T. Shimizu, J. van Dijk, and J. L. Zimmermann, Plasma medicine: an introductory review, *New J. Phys.* **11** p.115012 (2009).
- 18) G. Y. Park, S. J. Park, M. Y. Choi, I. G. Koo, J. H. Byun, J. W. Hong, G. J. Sim, J. Y. Collins, and J. K. Lee, Atmospheric-pressure plasma sources for biomedical applications, *Plasma Sources Sci. Technol.* **21** 043001 (2012).
- 19) T. von Woedtke, S. Reuter, K. Masur, and K. -D. Weltmann, Plasmas for medicine, *Phys. Rep.* **530** p.291 (2013).
- 20) M. Laroussi, Low-Temperature Plasmas for Medicine?, *IEEE Trans. Plasma Sci.* **37** p. 714 (2009).
- 21) S. U. Kalghatgi, G. Fridman, M. Cooper, M. Nagaraj, M. Peddinghaus, M. Balasubramanian, V. N. Vasilets, A. F. Gutsol, A. Fridman, and G. Friedman, Mechanism of Blood Coagulation by Nonthermal Atmospheric Pressure Dielectric Barrier Discharge Plasma, *IEEE Trans. Plasma Sci.* **35** p.1559 (2007).
- 22) G. Isbary, G. Morfill, H. U. Schmidt, M. Georgi, K. Ramrath, J. Heinlin, S. Karrer, M. Landthaler, T. Shimizu, B. Steffes, W. Bunk, R. Monetti, J. L. Zimmermann, R. Pompl, and W. Stolz, A first prospective randomized controlled trial to decrease bacterial load using cold atmospheric argon plasma on chronic wounds in patients, *Br. J. Dermatol.* **163** p.78 (2010).
- 23) S. Kalghatgi, G. Friedman, A. Fridman, and A. M. Clyne, Endothelial Cell Proliferation is Enhanced by Low Dose Non-Thermal Plasma Through Fibroblast Growth Factor-2 Release, *Ann. Biomed. Eng.* **38** p.748 (2010).
- 24) G. Fridman, A. Shereshevsky, M. M. Jost, A. D. Brooks, A. Fridman, A. Gutsol, V. Vasilets, and G. Friedman, Floating Electrode Dielectric Barrier Discharge Plasma in Air Promoting Apoptotic Behavior in Melanoma Skin Cancer Cell Lines, *Plasma Chem. Plasma Process.* **27** p.163 (2007).
- 25) S. Iseki, K. Nakamura, M. Hayashi, H. Tanaka, H. Kondo, H. Kajiyama, H. Kano, F. Kikkawa, and M. Hori, Selective killing of ovarian cancer cells through induction of apoptosis by nonequilibrium atmospheric pressure plasma, *Appl. Phys. Lett.* **100** p.113702 (2012).
- 26) A. R. Gibson, H. O. McCarthy, A. A. Ali, D. O'Connell, and W. G. Graham, Interactions of a Non-Thermal Atmospheric Pressure Plasma Effluent with PC-3 Prostate Cancer Cells, *Plasma Process. Polym.* **11** p. 1142 (2014).
- 27) Y. Ogawa, N. Morikawa, A. Ohkubo-Suzuki, S. Miyoshi, H. Awakawa, Y. Kita, and S. Nishimura, An epoch-making application of discharge plasma phenomenon to gene-transfer, *Biotechnol. Bioeng.* **92** p.865 (2005).
- 28) Y. Sakai, V. Khajoe, Y. Ogawa, K. Kusuhara, Y. Katayama, T. Hara, A novel transfection method for mammalian cells using gas plasma, *J. Biotechnol.* **121** p. 299 (2006).
- 29) M. Leduc, D. Guay, R. L. Leask, and S. Coulombe, Cell permeabilization using a non-thermal plasma, *New J. Phys.* **11** p. 115021 (2009).
- 30) T. Nakajima, H. Yasuda, H. Kurita, K. Takashima, and A. Mizuno, Generation of Bactericidal Factors in the Liquid Phase and Approach to New Gene Transfer Technology by Low Temperature Plasma Jet Treatment, *Int. J. Plasma Environ. Soc. Technol.* **5** p.42 (2011).
- 31) S. Sasaki, M. Kanzaki, and T. Kaneko, Highly efficient and minimally invasive transfection using time-controlled irradiation of atmospheric-pressure plasma, *Appl. Phys. Express* **7** 026202 (2014).
- 32) J. C. Devins, Mechanism of Ozone Formation in the Silent Electric Discharge, *J. Electrochem. Soc.* **103** p. 460 (1956).
- 33) J. S. Chang, P. A. Lawless, and T. Yamamoto, Corona Discharge Processes, *IEEE Trans. Plasma Sci.* **19** p. 1152 (1991).
- 34) I. Somiya, The Ozone: I. Introduction on Ozonation, *T. IEE Japan* **114-D** p. 353 (1994) [in Japanese].
- 35) T. Ishijima, H. Hotta, H. Sugai, and M. Sato, Multibubble plasma production and solvent decomposition in water by slot-excited microwave discharge, *Appl. Phys. Lett.* **91** p. 121501 (2007).
- 36) S. Kühn, N. Bibinov, R. Gesche, and P. Awakowicz, Non-thermal atmospheric pressure HF plasma source: generation of nitric oxide and ozone for bio-medical applications, *Plasma Sources Sci. Technol.* **19** 015013 (2010).
- 37) S. Reuter, J. Winter, S. Iseni, S. Peters, A. Schmidt-Bleker, M. Dünnebier, J. Schäfer, R. Foest, and K. -D. Weltmann, Detection of ozone in a MHz argon plasma bullet jet, *Plasma Sources Sci. Technol.* **21** 034015 (2012).
- 38) J. Winter, M. Dünnebier, A. Schmidt-Bleker, A. Meshchanov, S. Reuter, and K. D. Weltmann, Aspects of

- UV-absorption spectroscopy on ozone in effluents of plasma jets operated in air, *J. Phys. D: Appl. Phys.* **45** p.385201 (2012).
- 39) S. Zhang, W. van Gaens, B. van Gessel, S. Hofmann, E. van Veldhuizen, A. Bogaerts, and P. Bruggeman, Spatially resolved ozone densities and gas temperatures in a time modulated RF driven atmospheric pressure plasma jet: an analysis of the production and destruction mechanisms, *J. Phys. D: Appl. Phys.* **46** p. 205202 (2013).
- 40) J. Winter, K. Wende, K. Masur, S. Iseni, M. Dünnbier, M. U. Hammer, H. Tresp, K. -D .Weltmann, and S. Reuter, Feed gas humidity: a vital parameter affecting a cold atmospheric-pressure plasma jet and plasma-treated human skin cells, *J. Phys. D: Appl. Phys.* p. 295401**46** (2013).
- 41) X. L. Deng, A. Yu. Nikiforov, P. Vanraes, and Ch. Leys, Direct current plasma jet at atmospheric pressure operating in nitrogen and air, *J. Appl. Phys.* **113** 023305 (2013).
- 42) D. Ellerweg, A. von Keudell, and J. Benedikt, Unexpected O and O₃ production in the effluent of He/O₂ microplasma jets emanating into ambient air, *Plasma Sources Sci. Technol.* **21** 034019 (2012).
- 43) H. Kuwahata and T. Yamaguchi, Two-Dimensional Concentration Distribution of Hydrogen Peroxide Generated by Atmospheric-Pressure Plasma Jet Irradiation, *e-J. Surf. Sci. Nanotechnol.* **13** p.474 (2015).
- 44) R. Ohyama and A. Nagai, Japan Patent 244938 (2006).