

INVESTIGATION OF ACTIVE SPECIES IN ATMOSPHERIC-PRESSURE ARGON PLASMA-JET

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ABSTRACT

An atmospheric-pressure argon plasma-jet is generated, and optical emission spectroscopy of the argon plasma-jet is carried out. It is found that the emission lines of Ar, OH, N₂ and O are observed in the emission spectra, and that the emission line of NO is also found only when room-air is mixed into the plasma-jet. The emission intensities of Ar, OH and N₂ with mixing the room-air monotonously decrease toward the downstream, but those of OH and N₂ without mixing the room-air reach those maximum in the mid-stream of the plasma-jet and then decrease. Deionised-water is exposed to the argon plasma-jet, and dissolved ions in deionised-water after the exposure of the argon plasma-jet are investigated. It is found that H₂O₂, NO₂⁻ and NO₃⁻ are dissolved, and that those concentration increase with the increase of the exposure time.

1. INTRODUCTION

An atmospheric-pressure plasma-jet originated in a dielectric barrier discharge configuration has recently attracted attention, because plasma generated between electrodes can extend downstream into ambient air, allowing the plasma to reach to solid and liquid easily. Therefore, the plasma-jet can be used in the fields of plasma medicine, sterilization, etc.[1, 2] In those fields, reactive oxygen species and reactive nitrogen species (ROS/RNS) such as O₂⁻, H₂O₂, NO₂, etc. are regarded as important species, and those are produced by dissolving the active

species in the plasma-jet into liquid. For the effective use of the plasma-jet property and expanding its applications, it is essential that the active species in the plasma-jet and the ROS/RNS in an aqueous solution after the exposure of the plasma-jet to water are investigated.

In this paper, we generated an atmospheric-pressure argon plasma-jet, and carried out the spatially resolved optical emission spectroscopy of the argon plasma-jet. We also investigated dissolved ions in deionised-water after the exposure of the argon plasma-jet.

2. EXPERIMENTAL APPARATUS AND CONDITIONS

Figure 1 shows the schematic diagram of experimental apparatus. The reactor of the atmospheric-pressure argon plasma-jet consists of a T-shaped glass tube, a copper tube and an aluminium sheet. The main tube of the T-shaped glass tube has 3 mm in inner diameter and 90 mm in length, and the branched tube connected to the centre of the main tube has 3 mm in inner diameter and 45 mm in length. The copper tube of 3 mm in outer diameter, used as a high-voltage electrode, is inserted in an end of the main tube at a depth of about 10 mm, and the aluminium sheet of 15 mm in width, used as an earth electrode, is bound around the main tube. The distance between the electrodes is fixed at 10 mm. Deionised-water of 200 mL poured in a 500 mL beaker is located directly below the reactor, and the distance between the water

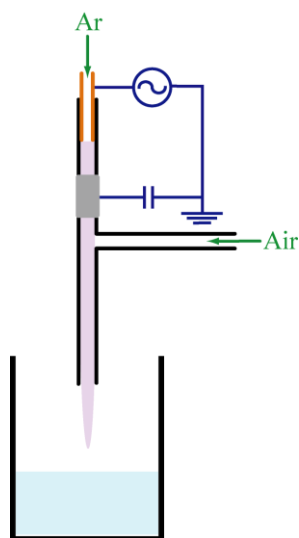


Fig. 1. Schematic diagram of experimental apparatus.

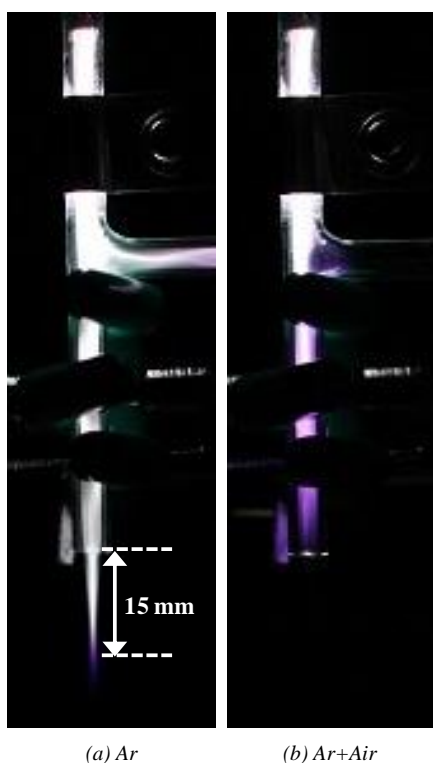


Fig. 2. Photographs of plasma-jet at input power of 4 W.

surface and the tip of the main tube is fixed at 65 mm.

Argon gas (99.99 %) is fed into the main tube through the copper tube at a constant flow rate of 10 L/min. A sinusoidal high-voltage of 13 kV_{p-p} with a frequency of 17 kHz generated by a neon-sign-transformer (Kodera, CR-N16) is applied to the copper tube, generating the plasma-jet. Compressed room-air is mixed into the plasma-jet from the branched tube at a flow rate of 0.1 L/min to produce active species such as excited nitrogen molecules, oxygen radicals, etc. Input

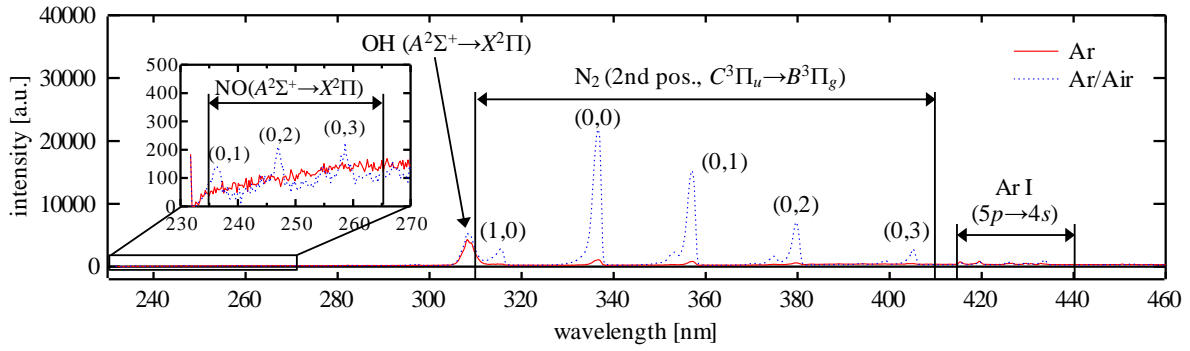
power is calculated by the Lissajous figure method[3]. The applied voltage is measured by a high-voltage probe (Iwatsu, HV-P30), and charge amount is calculated by a voltage drop across a 9.5 nF capacitor inserted between the aluminium sheet and earth. The voltage drop is measured by a high-voltage differential probe (GW Instek, GDP-100).

Optical emission spectra in a range of 230 to 980 nm are measured along the plasma-jet as a function of the distance from the end of the plasma-jet reactor, using High-Sensitivity Fibre Optic Spectrometer (Ocean Optics, Maya2000-pro) with a resolution of 0.31 and 0.33 nm, and the spatial resolution is 5 mm. Optical emission spectra on the central axis of the main tube are also measured. The deionised-water is exposed to the plasma-jet for 15 min, and the variations of ion concentrations in the water are measured using High-Performance Liquid Chromatograph (Shimadzu, Prominence, column: IC NI-424) by sampling the water of 1.5 mL every 3 min. The aqueous solution of acetic acid (3 mmol/L) and potassium hydroxide (2.25 mmol/L) is used as an elution, and the wavelength of an absorbance detector is fixed at 220 nm in HPLC.

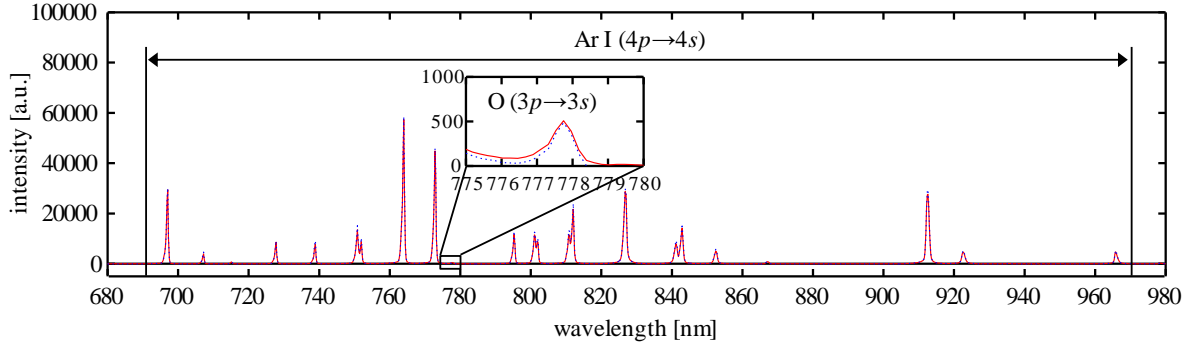
3. RESULTS AND DISCUSSION

Figure 2 shows the photographs of the plasma-jet with Ar and Ar/air mixture at the input power of 4W. The optical emission of the plasma-jet with Ar is observed up to 15 mm from the tip of the main tube, whereas that of the plasma-jet with Ar/air mixture is not observed beyond the inside of the main tube.

Figure 3 shows the emission spectra of the plasma-jet on the central axis of the main tube with Ar and Ar/air mixture in UV-VIS and VIS-IR region. The emission lines of Ar I ($4p \rightarrow 4s$)[4] in a range of 690 to 970 nm and Ar I ($5p \rightarrow 4s$)[4] in a range of 415 to 440 nm are observed with and without the room-air mixture. The emission line of OH ($A^2\Sigma^+ \rightarrow X^2\Pi$)[5], which is produced by dissociating H₂O in Ar or in ambient air of the plasma-jet, is found at 309 nm, and the weak emission of N₂ ($C^3\Pi_u \rightarrow B^3\Pi_g$, 2nd positive system)[5] in a range of 310 to 410 nm is also found. It is found that these intensity increase with mixing the room-air into the plasma-jet, and that the emission line of NO ($A^2\Sigma^+ \rightarrow X^2\Pi$)[5] in a range of 235 to 265 nm is newly observed at the



(a) UV-VIS



(b) VIS-IR

Fig. 3. Emission spectra of argon plasma-jet.

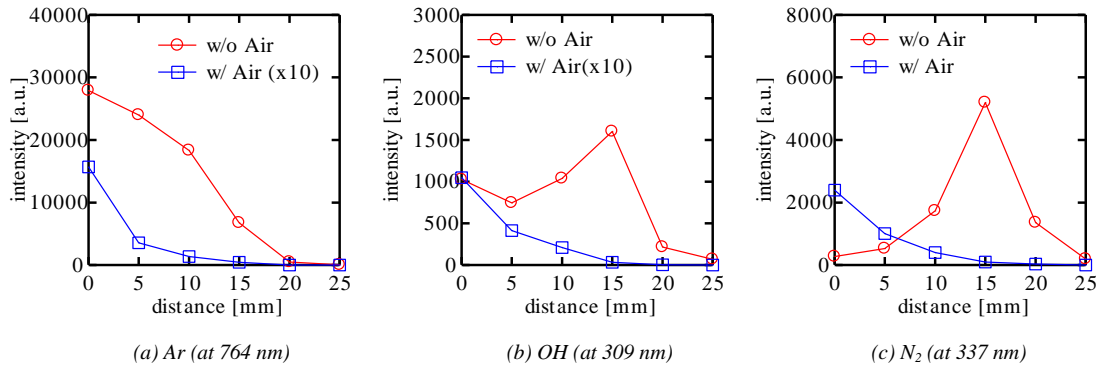


Fig. 4. Emission intensity of (a) Ar, (b) OH and (c) N₂ as functions of distance.

same time. In addition, the emission line of O ($3p \rightarrow 3s$)[4] at 777 nm is present in the emission spectra, and this intensity is almost the same with or without the room-air mixture.

Figure 4 shows the emission intensity of Ar I ($4p \rightarrow 4s$), OH ($A^2\Sigma^+ \rightarrow X^2\Pi$) and N₂ ($C^3\Pi_u \rightarrow B^3\Pi_g$) lines as functions of distance from the tip of the main tube. The emission intensity of Ar, OH and N₂ lines monotonously decreases with the increase of the distance when the room-air is mixed into the plasma-jet. However, when no room-air is mixed into the plasma-jet, the emission intensity of Ar line decreases with the increase of the distance, but that of OH and N₂ lines reaches its maximum at the distance of 15

mm and then decreases. This may be due to the engulfment of ambient air into the argon plasma-jet.

Figure 5 shows the chromatogram of the deionised-water after the exposure of the plasma-jet with Ar/air for 15 min. H₂O₂, NO₂⁻ and NO₃⁻ are detected at the retention time of 1.67, 6.00 and 9.55 min, respectively. These are probably produced from OH and NO in the plasma-jet. Figure 6 shows the ion concentrations in the deionised-water as a function of the exposure time. The concentrations of H₂O₂, NO₂⁻ and NO₃⁻ monotonously increase with the exposure time, and no saturation-tendency is observed within the exposure time of 15 min. The increasing rates

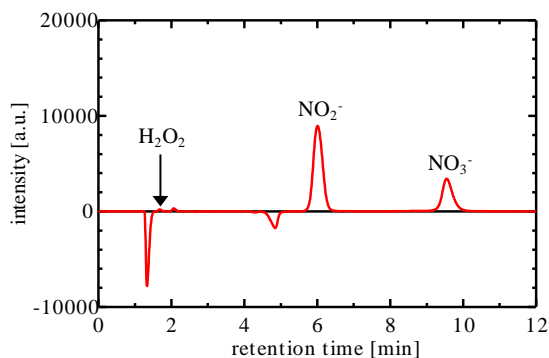


Fig. 5. Chromatogram of deionised water after 15 min of plasma-jet exposure.

of those are 0.063, 0.062 and 0.082 ppm/min, respectively.

4. CONCLUSIONS

An atmospheric-pressure argon plasma-jet is generated, and spatially resolved optical emission spectroscopy of the argon plasma-jet is carried out. It is found that the emission lines of Ar I ($4p \rightarrow 4s$), Ar I ($5p \rightarrow 4s$), OH ($A^2\Sigma^+ \rightarrow X^2\Pi$), N₂ ($C^3\Pi_u \rightarrow B^3\Pi_g$) and O ($3p \rightarrow 3s$) are observed, and that the emission line of NO ($A^2\Sigma^+ \rightarrow X^2\Pi$) is also found when the room-air is mixed into the plasma-jet. Although the emission intensity of Ar ($4p \rightarrow 4s$), OH and N₂ lines monotonously decreases with the increase of the distance from the tip of the main tube when the room-air is mixed, the emission intensity of OH and N₂ lines without the room-air mixture reaches its maximum at a distance of 15 mm and then decrease. Deionised-water is exposed to the argon plasma-jet, and dissolved ions in deionised-water after the exposure of the argon plasma-jet are also investigated. It is found that H₂O₂, NO₂⁻ and NO₃⁻ are dissolved in the deionised-water after the exposure of the plasma-jet, and that those concentrations monotonously increase.

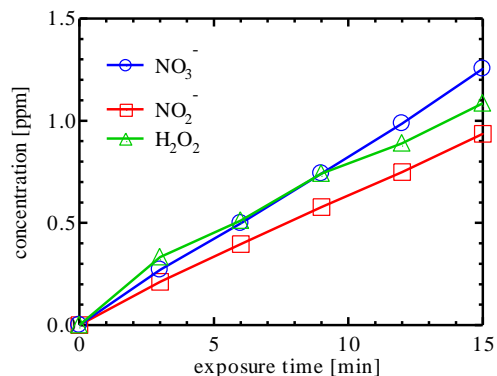


Fig. 6. Ion concentrations as a function of exposure time.

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