

## Space-resolving spectroscopy of atmospheric plasma jet source

Y. Takemura<sup>1</sup>, N. Yamaguchi<sup>2</sup>, T. Hara<sup>1</sup>

<sup>1</sup> *Toyota Technological Institute, Hisakata, Tempaku, Nagoya 468-8511, JAPAN*

<sup>2</sup> *Graduate School of Medicine and Pharmaceutical Sciences, University of Toyama, 2630 Sugitani, Toyama 930-0194, JAPAN*

### 1. Introduction

Cold plasma processing is one of the most important industrial technologies for surface modification, etching, thin film deposition or formation, and so on [1]. Cold plasmas are non-equilibrium plasma, where electrons are at high temperature state ( $>10000$  K), on the other hand, ion and gas temperatures are low ( $\sim 300$  K). As a result, it causes little damages to materials and has high reaction rate. That is why cold plasma processing is the key industrial technology in electronics, mechanical engineering, manufacturing, and medical sciences etc, whereas it needs vacuum environments. Non-equilibrium plasmas at atmospheric pressure are studied for various applications, such as surface processing and light source generation. There are great advantages that reactions can be progressed with high speed and without vacuum systems [2].

In this work we have investigated an atmospheric-pressure plasma jet by means of optical emission spectroscopy. Characteristics of surface modification for the improvement in hydrophilicity of polymer materials have been studied and considered in relation to space-resolved spectroscopic data.

### 2. Experimental Apparatus

A schematic of the plasma jet is shown in Fig. 1. The plasma device is an atmospheric-pressure plasma jet (Openair PFW10, Plasmacreat), which consists of a conical inner electrode, a grounded outer electrode with a nozzle of 4 mm diameter. The inner electrode is coupled to a stepped high-frequency pulse current power supply, 180-270 V, 3-7 A and 13-19 kHz, through a high voltage transformer. Working gas is fed into the annular space between the two electrodes and flows spirally at 10-60 l/min. The gas used in this experiment is air,  $N_2$ ,  $O_2$  or Ar. A soft and white plasma jet is produced stably flowing from the nozzle with a length of 10-20 mm.

Spectroscopic diagnostic system consists of a space-resolving slit of 1 mm width, a collecting lens, an optical filter, a Czerny-Turner spectrometer with 250 mm focal length

(MC-25, Ritu Appl. Optics) and a CCD detector (SPH5, Apogee). The detector has  $1024 \times 1024$  pixels with  $24 \times 24 \mu\text{m}$  pixel size. The system has been calibrated by using a standard lamp through the wavelength range from 300 nm to 1000 nm. The schematic layout of the spectroscopic system is also shown in Fig. 1. Viewing direction of the observation is perpendicular to the axis of flowing plasma with a spatial resolution of about 1 mm.

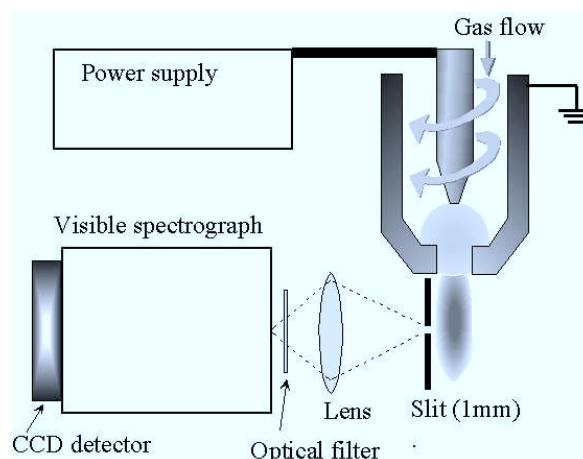


Fig. 1 Schematics of plasma jet device and optical emission diagnostics.

Surface-modification experiments have been performed by setting a piece of material on a translation stage, where the distance between the nozzle of plasma source and the sample is adjustable. The stage can be moved with a velocity of 10-40 cm/s. A contact angle meter is used to characterize improvement in hydrophilicity of sample surface.

### 3. Experimental Results and Discussions

In Fig. 2, typical emission spectra near the nozzle are shown in the wavelength range from 250 nm to 1000 nm where the working gas is air or nitrogen. Emission spectra of  $\text{N}_2$  molecule and N atom are observed for both cases, while spectral lines of O atom is significant in the air plasma jet. These spectra indicate that radical species like N and O atoms exist with high density in the plasma jet source.

Spatial distributions of UV spectral lines in the range 290 nm - 370 nm are shown in Fig. 3 as functions of distance from the nozzle outlet, (a) for  $\text{N}_2$  plasma and (b) for air plasma. The operation conditions were 270 V supply voltage and 30 l/min gas flow rate for both cases. Cluster of emission lines from  $\text{N}_2$  C-B (second positive system) transitions are

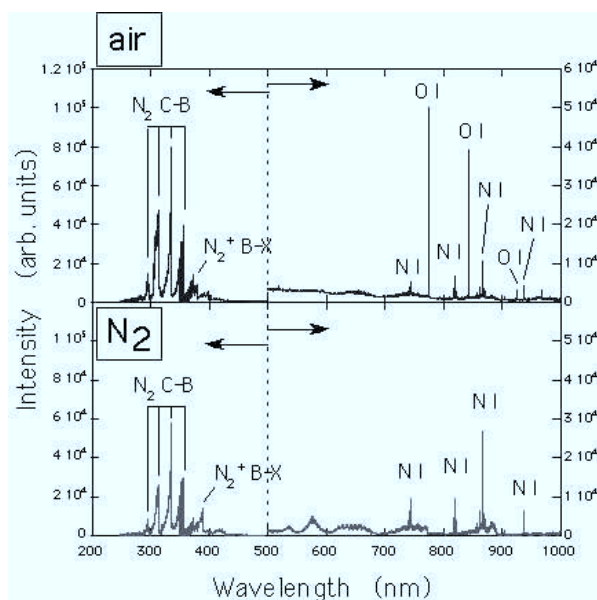


Fig. 2 UV/visible emission spectra of air plasma jet (top) and  $\text{N}_2$  plasma jet (bottom).

strong at the place within 2 mm from the nozzle in nitrogen and air plasmas. The spatial distribution of  $N_2$  molecular spectra in the  $N_2$  plasma has a different feature in the range farther than 3 mm away from the outlet compared with that in the air plasma. Area of weak emissions extends up to 15 mm in the  $N_2$  plasma, while in the air plasma the OH A-X transition lines around 310 nm appears at the place farther than 3 mm. This indicates that the air plasma contains a small amount of  $H_2O$  and consists of low temperature species along the jet [3]. It is thought that species with the higher temperature exist in the  $N_2$  plasma, though the very weak OH A-X emission lines are also observed at about 10 mm away from the nozzle. It should be noted that the temperature of the jet is as low as around 400 K as a whole.

Surface-modification experiments have been performed by spraying plasma jet onto a sample at a distance. Polyethylene terephthalate (PET) and polyimide (Kapton) films (25  $\mu\text{m}$  thick) were treated in this work. Significant augmentation in hydrophilicity was observed after the treatment where the distance from the nozzle was 1 cm and the exposure time was 0.01-0.04 s for both sample materials. The exposure time is defined as the time in which the sample passes through the nozzle diameter, and can be changed by controlling the transfer velocity of the sample stage. In Fig. 4, contact angles of water on treated Kapton film are shown as functions of the nozzle to sample distance for various working gases,  $N_2$ ,  $O_2$ , air and Ar. The experimental conditions were as follows; the driving voltage was 270 V, the gas flow rate 30 l/min and the exposure time 0.01 s. The augmentation in hydrophilicity can be seen for  $N_2$  or  $O_2$  plasma jet even when the sample is set about 20 mm distant from the nozzle. These results show that particles of radical species, *i.e.* N or O atoms, exist with enough density in the plasma jet at the above distance. On the other hand, it is thought that radical

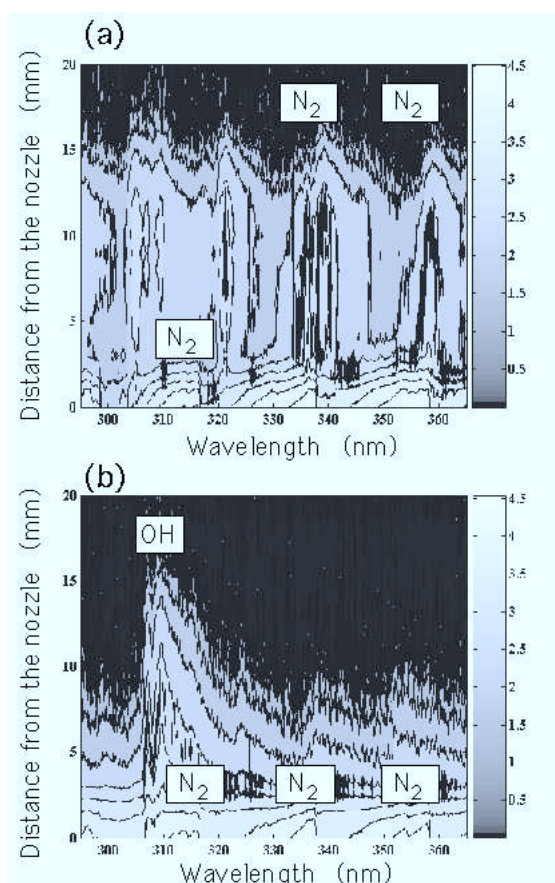


Fig. 3 Spatial distributions of UV spectral lines in the range 290 nm - 370 nm as functions of distance from the nozzle outlet, (a) for  $N_2$  plasma and (b) for air plasma.

neutral atoms cannot remain far from the nozzle output in the air plasma jet. The spatial distribution of UV spectral lines shown in Fig. 3 could support the above consideration.

The NO<sub>x</sub> compound analysis has been performed by controlled potential electrolysis (testo 350M) for produced gas in each plasma jet. Concentration of NO<sub>x</sub> compound was the highest in the air plasma jet, it was 22 times and 6 times as concentrated as that in the N<sub>2</sub> plasma jet and in the O<sub>2</sub> plasma jet, respectively. A set of NO  $\gamma$  (A-X) system lines (~260 nm)

was also observed in the air plasma jet. Therefore it is considered that the internal energy of plasma is spent to produce NO<sub>x</sub> compound particles in the air plasma jet and then the concentration of neutral N and O atoms is lower than in N<sub>2</sub> or O<sub>2</sub> plasma jet.

#### 4. Summary

In this paper we have investigated the atmospheric-pressure plasma jet. Spatially-resolved UV/visible emission spectra have been observed. It was found that the molecular spectral lines distributed far from the nozzle (>15 mm) in the N<sub>2</sub> plasma jet, while they decayed within a few mm in the air plasma jet. Characteristics of surface modification of polymer films have been studied and could be explained from the spectroscopic data.

#### References

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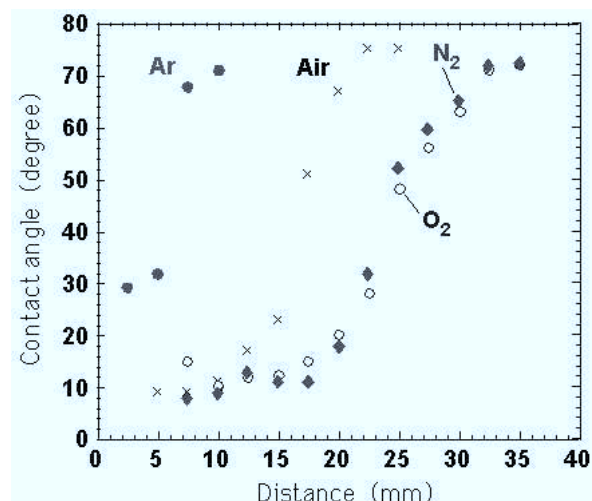


Fig. 4 Contact angles of water on treated Kapton film shown as functions of the nozzle to sample distance for various working gases, N<sub>2</sub>, O<sub>2</sub>, air and Ar. The contact angle of untreated sample is 75 degree.