JOURNAL OF OPTOELECTRONICS AND ADVANCED MATERIALS Vol. 10, No. 8, August 2008, p. 2015 - 2019

Investigation of radiofrequency plasma jets at low and atmospheric pressure by optical emission spectroscopy

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Radiofrequency plasma jets generated in argon and nitrogen at low and atmospheric pressure have been investigated by means of Optical Emission Spectroscopy (OES) analysis. The local emissivity of the plasma along axial and radial direction of the plasma expansion has been determined by Abel inversion of the integral emission intensity. The gas temperature has been evaluated by simulating the roto-vibrational structure of the molecular bands.

(Received March 1, 2008; accepted June 30, 2008)

Keywords: Low pressure plasma, Atmospheric pressure plasma, Optical emission spectroscopy, Abel inversion, non-equilibrium plasma

1. Introduction

The potential of plasma application for surface processing (deposition or treatment) [1, 2], pollutants removal, or lighting has led to the development of various plasma sources and configurations. In order to control better the process parameters, the characterization of plasma properties is in demand.

Optical Emission Spectroscopy is a non-intrusive method, capable to provide detailed information on the plasma characteristics. From the optical spectra emitted by the plasma one can deduce all basic plasma parameters and its state: chemical composition (according to emitted lines), concentrations of excited atoms and molecules upon energetic levels (from the intensity of spectral lines), densities of charged and neutral particles (from the broadening of spectral lines), temperatures of excitation of atomic and molecular states (from the relative or absolute spectral line intensities) [3].

Among the most used plasma sources are the plasma jets, due to the possibility to control separately the parameters in the plasma generation region and in the processing region. However, because of the non-uniformity of plasma in its volume, a spatial plasma characterization is of great interest.

This paper reports on Optical Emission Spectroscopy (OES) investigation of radiofrequency plasma jets working in nitrogen at low pressure, and respectively in argon at atmospheric pressure.

The Abel inversion method was employed to determine the spatial distribution of emissivity of species responsible for the emission of the First and Second Positive Spectral Systems of N_2 (denoted as FPS and SPS), and of the spectral systems of NH and NO radicals.

The roto-vibrational band emission of OH radical was simulated and compared to the recorded one in order to determine the gas temperature.

2. Experimental set-up

2.1. Plasma jet sources

The plasma sources are working at low and respectively at atmospheric pressure, operating on similar principle: the RF discharge (13.56 MHz) is generated in flowing gas, in a small interelectrodic gap. The plasma jet is formed by the discharge expansion through a hole performed in the bottom (grounded) electrode, either at low pressure, in a vacuumed chamber, or at atmospheric pressure, in open atmosphere. Detailed description of the plasma sources can be found in [4, 5]. In Figure 1 are presented pictures of the plasma jets produced by the respective sources. The succession of luminous and dark regions in the image of the low pressure plasma jet (left image) is associated to the shock waves generated by the supersonic plasma expansion. The decrease of the emission at the middle of the atmospheric jet (in the right image) is unexpected and we do not have yet an explanation for it.

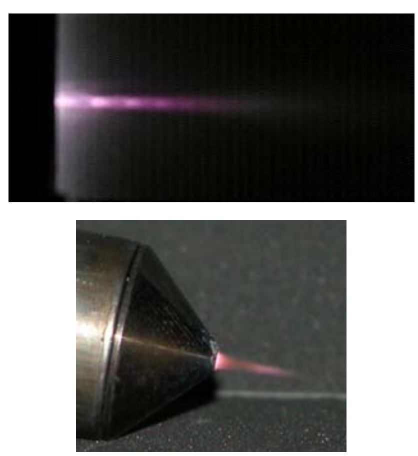

Fig. 1. Images of the plasma jets generated at low (left) and atmospheric (right) pressure

The parameters used during the OES investigations of the plasma sources and their values are presented in Table 1:

Table I. Operation parameters of the plasma jets studied in the paper

Parameter	Low pressure plasma	Atmospheric pressure plasma
Working gas	N_2	Ar
RF power	250 W	100 W
Interelectrodic distance	4 mm	2 mm
Electrode type	Steel	Cooper
Flow rate	1000 sccm	2300 sccm
R (plasma radius)	4 mm	1 mm

2.2 OES measurement geometry

The experimental setup used for optical emission spectroscopy measurements is presented in Figure 2. The plasma image is obtained by an imaging system formed by a lens L and an optical fiber OF mounted on a 2D translation stage. The other end of the fiber is connected to the entrance slit of the monochromator. The scanning was performed in the Ox direction, for different positions (z) from the plasma source nozzle. Every recorded spectra, corresponding to a given (x, z) position of the fiber, contains the integral emission of the axisymmetric plasma along a chord, as shown in Fig 2.

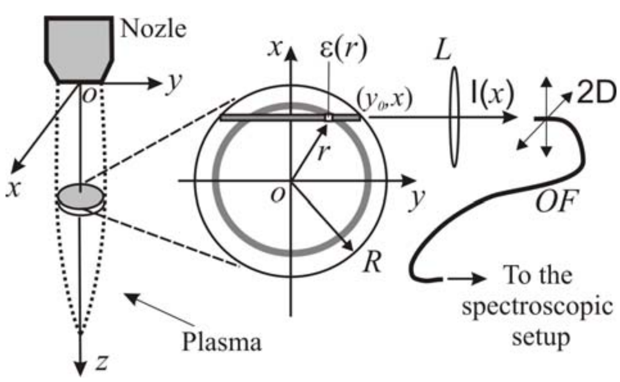

Fig. 2. Sketch of the OES measurements geometry

The collected light was analyzed in the spectral range of 200-1000 nm using an Ocean Optics SM-240 Optical Multichannel Analyzer for the low pressure plasma jet, while in the case of the atmospheric pressure plasma jet a monochromator (Carl Zeiss SPM-2) equipped with a 1200 mm^{-1} grating, 300 μm entrance slit and a QB 9958 photomultiplier was used.

3. Results and discussion

3.1. Emission spectra

In Fig. 3a is presented a typical OES spectrum of the low pressure nitrogen plasma, recorded in the nozzle vicinity (z=0, x=0). It is clearly dominated by the molecular nitrogen systems, namely the Second Positive System - SPS ($C^3\Pi_u$ - $B^3\Pi_g$) and First Positive System – FPS ($B^3\Pi_g$ - $A^3\Sigma_u$). The spectrum at atmospheric pressure, in argon, is presented in Figure 3b. Here, mainly the excitation of molecular species appearing due to the mixture of Ar plasma with the ambient atmosphere (air) is evidenced. Namely, the emission of γNO system ($A^2\Sigma^+$ - $X^2\Pi$), OH 3064 Å system ($A^2\Sigma$ - $X^2\Pi$) and NH 3360 Å ($A^3\Pi$ - $X^3\Pi$) system were observed.

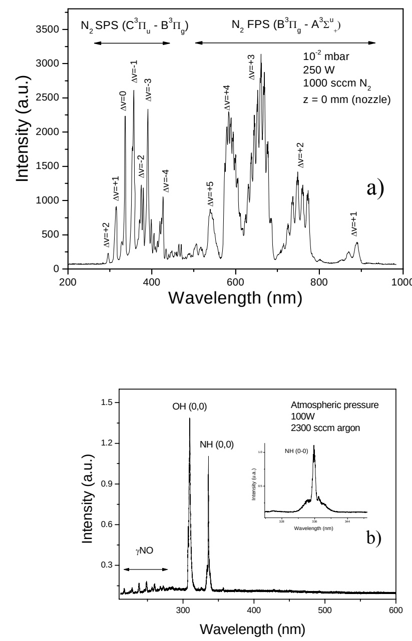

Fig. 3. Typical spectra of plasma jet operating: a) in nitrogen, at low pressure; b) in argon, at atmospheric Pressure.

In order to carry out systematic emission intensity measurements, we have selected the following wavelengths and spectral bands: λ = 337.13 nm (SPS, Δv=0), λ = 662.5 nm (FPS, Δv=0) in case of low pressure plasma and λ = 238 nm (γNO), λ = 336 nm (NH) for atmospheric pressure plasma.

3.2. Data processing and Abel inversion

Let's consider fixed the distance from the nozzle. When an axisymmetric radiation source is observed in the lateral direction, the measured spectral radiance I(x) represents the path integral of the plasma emissivity ε(r) along the chord delimited by the optical axis of the imaging optical system (Figure 2). If the source (the plasma jet in our case) is assumed to be cylindrically symmetric and optically thin, then the lateral observed signal can be written as:

$$I(x) = 2\int_{0}^{y_0} \varepsilon(r)dy \qquad (1)$$

where y_0 is the coordinate of the plasma edge for every x value where plasma scanning is performed. Using the Abel's transformation, this equation can be transformed to

$$\varepsilon(r) = -\frac{1}{\pi}\int_{r}^{R} \frac{dI}{dx}\cdot(x^2 - r^2)^{-\frac{1}{2}} \qquad (2)$$

where R represents the plasma slice radius. By Abel inversion (2), the integral observed signal I(x) is transformed into the radial profile of plasma emission coefficient (plasma emissivity) $\varepsilon(r)$.

From the spectra recorded at each (x, z) point the peak intensities of the bands of interest mentioned above were extracted. They represent the integral intensities data sets which are the input for data processing. Due to the insufficient resolution of the 2D scanning system, a limited number of experimental data points were available. Since the experimental curves I(x) fit satisfactory with Gaussian forms the raw data were fitted with Gaussians.

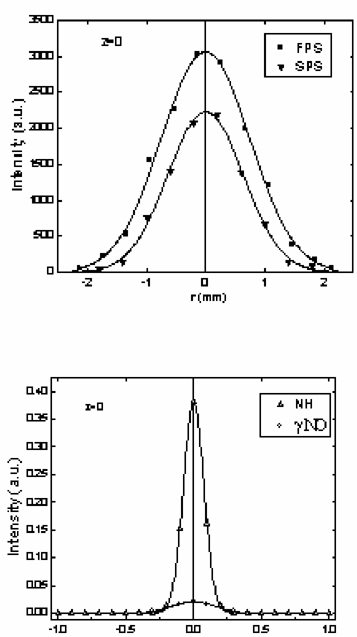

Fig. 4. The radial distribution of the emission intensity of the chosen wavelengths and their Gaussian fits for a) low pressure nitrogen plasma jet; b) atmospheric pressure argon plasma jet

Example of integral intensities profiles and their fitted curves are presented in Figure 4 for spectra recorded in the vicinity of the nozzle (z=0) for both investigated plasma jets. Further on the Gaussian curves were used as input data for the Abel inversion procedure. Several numerical methods were developed for performing the Abel inversion (deconvolution) [6-7]. For the data processing, we have used a LabVIEW program [8] based on Nestor-Olsen method [9].

3.3. Relative spatial distribution of emitting species

The Abel inversion has been performed for the above mentioned wavelengths (transitions).

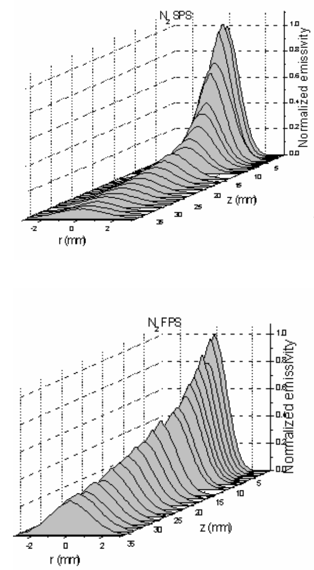

Fig. 5. The spatial distribution of emissivities of SPS and FPS systems of N_2 in low pressure plasma jet; values normalized to (0, 0) local emissivity

In Figure 5 are presented the nitrogen SPS and FPS space distribution of the emissivities for the low pressure plasma jet, by values normalized with respect to those at the nozzle position (coordinates z=0, r=0).

Because the emissivities are proportional with the concentration of the emitting species, Figure 5 gives also the map of $C^3\Pi_u$ and respectively $B^3\Pi_g$ state concentrations relative to those at nozzle level. One can see that the succession of luminous and dark zones observed in the image from Figure 1, related to the supersonic expansion of plasma, appears also in the distribution of emissivities (and therefore in the species concentration). Similar treatment of the OES data in the case of atmospheric pressure plasma jet led to the emissivities of γNO and NH systems (represented in Figure 6) and the respective emitting species (OH $A^2\Sigma$ and NH $A^2\Pi$).

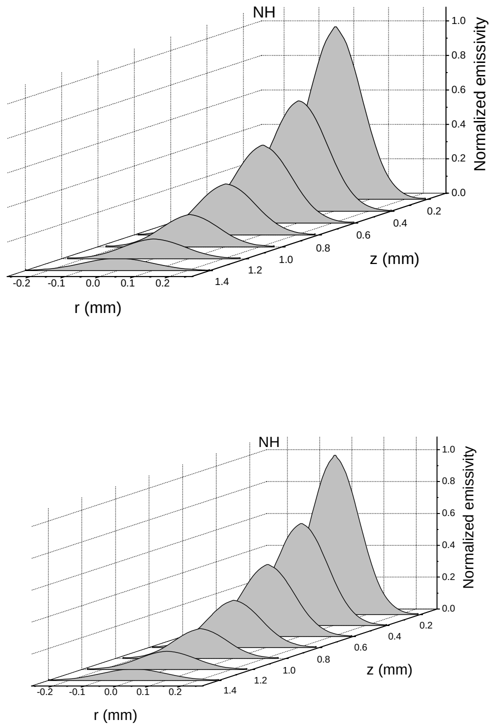

Fig. 6. The spatial distribution of emissivities of γNO and NH systems in atmospheric pressure plasma jet; values normalized to (0, 0) local emissivity

One must mention here that the Abel inversion procedure presents a good confidence interval over the investigated positions, except the places where the light intensities were low (at the plasma margins, r= r_{max}) and on the axis (r = 0) where the integral (2) presents a singularity. This can be observed in Figure 7, which presents the radial distribution of NO emissivity, at z=0 position, where unexpected behaviors are seen at these places.

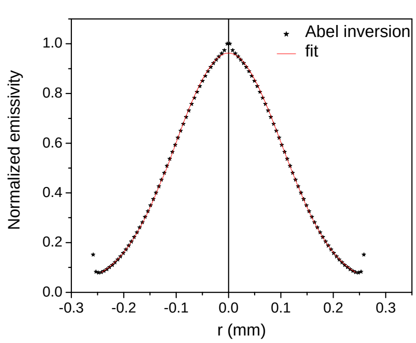

Fig. 7. Radial distribution of NO emissivity for atmospheric pressure argon plasma, for z = 0.

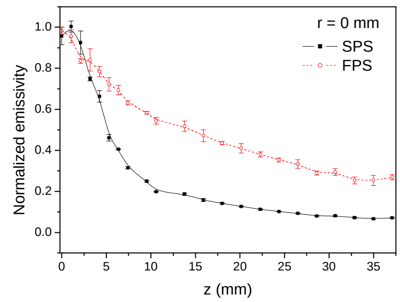

Fig. 8. Axial distribution of SPS and FPS emissivities (relative concentrations of $C^3\Pi_u$ and $B^3\Pi_g$ states) for low pressure nitrogen plasma, for r = 0.

In Figure 8 are presented the SPS and FPS normalized emissivities along the axial direction, in the center of the plasma jet. One may observe the different rate of the emissivity decay along the flowing axis for the two spectral systems. The much slow decreasing of the FPS emissivity (which describes the decrease of $B^3\Pi_g$ emitting state population) compared to the SPS (which describes the decrease of $C^3\Pi_u$ emitting state population) indicates an additional mechanism of population for the molecular state $B^3\Pi_g$. This suggests that N atoms are in large concentration in the plasma and the atomic nitrogen reassociation $N(^4S)+N(^4S)$ + M => short life intermediate state => $B^3\Pi_g$. + M (M is a third body), is populating the $B^3\Pi_g$ state, leading to the Lewis- Rayleigh afterglow [10].

3.4. Estimation of the gas temperature

In order to evaluate the plasma characteristics in the nozzle proximity for the atmospheric pressure plasma jet, simulation of the roto-vibrational structure of the OH 3064 Å system ($A^2\Sigma$ - $X^2\Pi$) has been performed. For this purpose, spectra with better resolution (0.15 nm line halfwidth) have been recorded. By comparison of the experimental data with the simulated one (see Figure 9), it was obtained that the rotational temperature is around 440 K. As this temperature corresponds to the gas temperature of the system, it proves the cold, non-equilibrium character of the generated plasma.

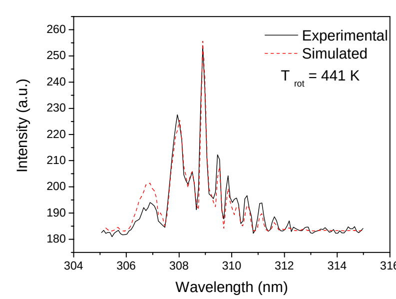

Fig. 9. Experimental and simulated spectra of OH band in atmospheric pressure argon plasma

Nevertheless, this value is obtained after the simulation of the integral emission of plasma. The spatial distribution of temperature can be determined only after Abel inversion of each point in the spectra, which would allow the OH-spectral emissivity reconstruction. This requires serious experimental effort and the handling of much larger data sets, being the subject of a future work.

4. Conclusions

The emission characteristics of two plasma jets, working at low pressure, in nitrogen, and at atmospheric pressure, in argon, have been determined by using Optical Emission Spectroscopy.

The spectra of low pressure nitrogen plasma jet are dominated by the emission of molecular nitrogen spectral systems. By using the Abel inversion procedure the spatial distributions of the SPS and FPS emissivities were obtained. They give the space distribution of the population of emitting states $C^3\Pi_u$ and respectively $B^3\Pi_g$ in the plasma volume. The slower decrease of the $B^3\Pi_g$ state along the flow axis indicates that plasma contains an important amount of dissociated atoms.

The spectral characteristics of atmospheric pressure plasma jet prove the excitation of impurities from ambient atmosphere. The low value of the gas temperature determined by fitting the experimental data corresponding to OH emission with simulated ones points out toward the cold character of that plasma jet.

Acknowledgements

The financial support of the Romanian Ministry of Education and Research under project PN 06 36 04 01 and CEEX project D11-13 is gratefully acknowledged.

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