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SPECTRAL CHARACTERISTICS OF A RADIOFREQUENCY NITROGEN PLASMA JET CONTINUOUSLY PASSING FROM LOW TO ATMOSPHERIC PRESSURE

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We report and discuss the characteristics of the optical emission of a radiofrequency (13.56 MHz) expanding plasma generated in nitrogen in a double chamber configuration, when the discharge passes continuously from the low pressure (10^{-1} mbar) to the high pressure region (760 mbar), at constant gas mass flow. The pressure increase leads to a sudden change in plasma aspect, occurring at intermediate pressure of 200-400 torr. The plasma temperature estimation from spectral measurements indicates that the change in the aspect is accompanied by a sudden decrease of the plasma temperature.

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1. Introduction

Plasma sources operating at atmospheric pressure with the discharge covering a large volume in-between two electrodes are of large interest. Helium is mostly used as operating gas in order to create uniform plasma at high pressure because this gas prevents arcing phenomena. In the case of other gases, as example in nitrogen, it is difficult to obtain uniform plasmas at high and atmospheric pressure.

In a different way, plasmas suitable for processing materials in a large volume can be obtained by the fast flowing of the discharge out of the inter-electrodic zone. Expanding plasmas are realized by this technique.

The present work is focused on the investigation of the spectral behaviour of expanding radiofrequency plasma (rf plasma jet) upon pressure. The expanded plasma is first generated at low pressure. Afterwards, the pressure is increased at constant mass gas flow, up to atmospheric pressure. Previously we have shown that during the pressure increase, there is some pressure range (200-300 torr) where the aspect of the expanded plasma changes suddenly [1], suggesting a modification in the discharge regime when reaching the high pressure domain. The information obtained from the spectral measurements, shows that the change is accompanied by a decrease of the electrons temperature.

2. Experimental

The plasma jet is generated in a two-chamber system [2], the active chamber and the expansion chamber. The chambers are stacked one over the other, and communicate through an aperture. The discharge is mainly sustained in the active chamber (20 mm diameter) in a small gap

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(4-10 mm in the present experiments) delimited by the powered RF electrode and a planar grounded electrode (nozzle) which contains the aperture. The discharge may extend through the nozzle in the expansion chamber, which is a grounded stainless steel cylinder 120 cm long, and 20 cm diameter wide. The gas (nitrogen) is admitted in the active chamber via a mass flow controller, at values in the range 20-2000 sccm. Stationary pressure values are established in the expansion, by balancing the mass gas flow rate with the pumping speed via a valve with variable gas conductance. The pressure in the chamber was measured with mercury manometers in order to obtain reliable absolute pressure values in the range 1-760 torr.

The wall of the expansion chamber is provided with quartz windows that allow UV-Vis optical investigation along the plasma jet. The emission of plasma expansion was studied with a moderate resolution set-up consisting of quartz optics, optical fiber, monochromator (SPM-2, grating 1200 grooves/mm), photomultiplier (QB 9958), and a data acquisition system.

3. Results and discussion

The optical emission of the plasma jet in nitrogen, at low pressure, was described previously [3]. It consists mainly of the N_2(SPS) bands in the near UV and blue part of visible region, and the N_2(FPS) bands in the red part of the visible region. In the UV part of the spectra the γNO bands are observed, caused by the impurities from the gas or desorbed from the wall. Besides, in the early expansion, just downstream the nozzle, low intensity N_2^+(FNS) bands appear.

As reference, a spectrum recorded at the nozzle level, for a pressure of 10 torr is presented in Fig. 1, revealing the features of the N_2(SPS) bands. A spectrum recorded in similar conditions, but in the spectral range 230-280 nm, in which the γNO bands emission appears, is presented in Fig.2. These bands are of much lower intensity.

At increased pressure the spectra (not shown here) loose the definition (effect observed at atmospheric pressure by other authors, as well [4]). In addition, the pressure increase leads to an overall decrease of the emission. As such, the band height at 3371 Å at atmospheric pressure represents only 0.5 % from the height of the same band at 10 torr. The decrease can be assigned to the quenching of radiative levels by collisions and to the change in the distribution function of the electrons energy.

The behaviour of the relative intensities of bands, coming from species with emitting levels situated at different excitation energy can provide information, on the plasma temperature [5]. In Fig. 3 the dependence of the ratio of the bands intensities of γNO and N_2(SPS) intensities on the pressure is presented. The same ratio has been measured, for jet operation at atmospheric pressure along the plasma jet, an increasing trend being observed.

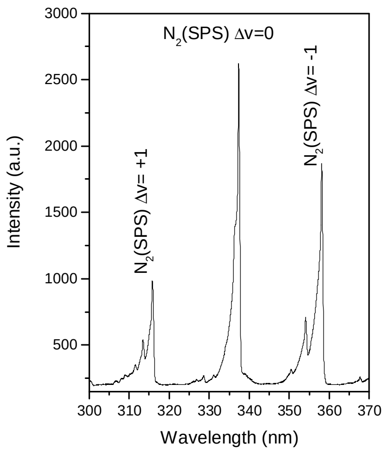

Fig. 1. Emission spectrum (p=10 torr, RF power 300 W, gas mass flow 1500 sccm, slit 100 μm).

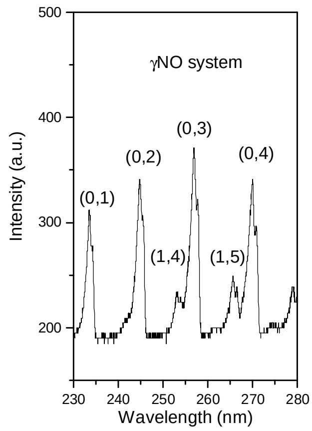

Fig. 2. Emission spectrum (p=760 torr, RF power 300 W, gas mass flow 1500 sccm, slit 400 μm).

The augmentation of the intensity ratio both with the pressure and with the position may be assigned to a decrease of the electronic temperature with the two parameters. Indeed, in agreement with an excitation scheme based on electronic collisions, the intensity ratio will be very much dependent on the electronic temperature and on the position of the energetic levels of species. Then, the excitation of species with lower excitation energies (the $A^2\Sigma^+$ electronic state of γNO bands has an energy of ~5.46 eV), against species with high electron energies (excitation energy of the nitrogen SPS system -$C^3\Pi_u$ state is ~11.06 eV) will be favored at low electronic temperature.

An estimation of the temperature behavior with the pressure can be done assuming roughly that a thermal equilibrium between the involved species exists (this circumvent a more complicated description in which one has to consider the collisional excitation transfer from the metastable $N_2(A)$ to NO(A) species, transfer favored by the small energy mismatch between the levels of the two states). The intensity of the N_2(SPS) system and of the **γ**NO system can be written, respectively:

$$I_{SPS\,(\gamma NO)}=C_{SPS\,(\gamma NO)}A_{SPS\,(\gamma NO)}N_{N2\,(NO)}\exp(-E_{SPS\,(\gamma NO)}/kT) \quad (1)$$

where $A_{SPS\,(\gamma NO)}$ represents the emission probability for the N_2(SPS) and respectively **γ**NO system, $N_{N2\,(NO)}$ the population of the fundamental states of the two molecules, while the $E_{SPS\,(\gamma NO)}$ are the energies of the upper states of the radiative transitions involved ($C^3\Pi_u$ for N_2 SPS and $A^2\Sigma^+$ for γNO). The intensity ratio $I_{SPS}/I_{\gamma NO}$ is function of the pressure through the subsystems temperature and the population of the fundamental states of the radicals involved. By knowing the electronic temperature at a given pressure (in our case at 10 torr), the temperature can be evaluated for all pressures. If one considers that the NO radical is coming as impurity from the feeding gas, the term related to the states population for two different pressures is leading to a constant and the equation for temperature dependence on pressure becomes:

$$T_e(p)=\frac{(\frac{E_{SPS}-E_{\gamma NO}}{k})*T_e(p_0)}{\left[\ln\frac{I_{SPS}}{I_{\gamma NO}}(p_0)-\ln\frac{I_{SPS}}{I_{\gamma NO}}(p)\right]*T_e(p_0)+(\frac{E_{SPS}-E_{\gamma NO}}{k})} \quad (2)$$

The electronic temperature in the low pressure regime of the discharge has been measured by Langmuir probes, obtaining $T_e(p_0)$~ 2 eV at 10 torr, in the nozzle proximity, in the expansion chamber. The temperature dependence upon the pressure, as estimated according to the Formula (2), leads to the behavior presented in Fig. 4. A striking aspect is the rapid decrease of temperature in the pressure range-200-300 torr, which corroborate with the change in the plasma aspect [1]. As was shown by complementary electrical measurements [1], a sudden change is observed in the discharge characteristics, too. Further measurements have to be performed in order to assign whether this behavior is caused by a change in the discharge regime, or by hydrodynamic instabilities created by the pressure increase

By using the same procedure the spectral emission of the plasma beam along the flow axis was studied at atmospheric pressure and the temperature behavior was obtained. The ratio of the $I_{\gamma(NO)}/I_{N2(SPS)}$ increases with the distance from the expansion nozzle, leading to a temperature decrease of almost 20% in the first 10 mm of expansion, in agreement with the fact that plasma cools out downstream. Nevertheless, these results are affected by large errors due to the influence of ambient air which will modify the ground state densities of N_2 and NO species along the flow.

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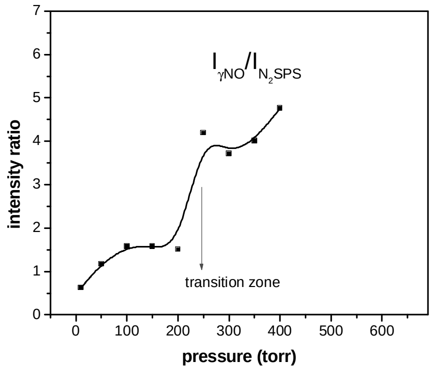

Fig. 3. Behaviour of the ratio of emission intensities of γNO and N_2(SPS) bands with pressure (RF power 300 W, gas mass flow 1500 sccm).

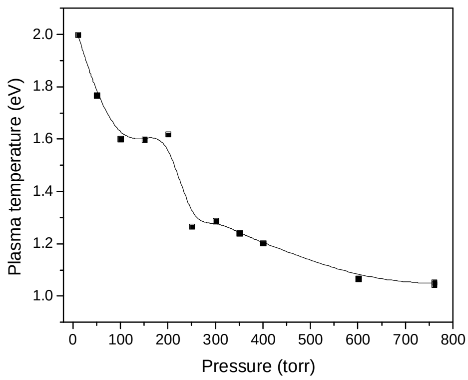

Fig. 4. Dependence of the plasma temperature on the pressure - thermal equilibrium assumption (RF power 300 W, gas mass flow 1500 sccm).

5. Conclusion

A nitrogen plasma jet, obtained by a radiofrequency discharge expansion in a double chamber configuration was continuously operated from low to atmospheric pressure. During the pressure increase, at constant mass flow, the expanded plasma exhibited – in the intermediate pressure range of 200-300 torr- a sudden change in its dimensions and aspect.

By Optical Emission Spectroscopy it was observed that during the pressure increase the emission of bands which belong to species having low excitation energy is favored against that of species with higher excitation energy. From the emission spectra the electronic temperature was estimated in the frame of the crude approximation of thermal equilibrium. The electron temperature decreases with the pressure increasing, according to an expected equilibration of the plasma subsystems due to collisional processes. A sudden decrease is observed in the specified intermediate pressure region, indicating a change in the discharge regime, or the intervention of a hydrodynamic instability.

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References

[1] G. Dinescu, B. Mitu, D. Crintea, S. Vizireanu, C. Petcu, Annals of University “A.I.Cuza”, Iasi, Fizica, XLIX, (2003) 25-35.
[2] G. Dinescu, B. Mitu, E. Aldea, M. Dinescu, Vacuum **56**, 83 (2000).
[3] G. Dinescu, O. Maris, G. Musa, Contrib. Plasma Phys. **31**(1), 49 (1991).
[4] J. M. Wiliamson, C. A. DeJoseph Jr., J. Appl. Phys. **93**(4), 1893 (2003).
[5] A. Ricard, Reactive plasmas, Published by SFV, Paris, France, 1996.