Threshold ionization mass spectrometry and optical emission spectroscopy characterization of $Ar/O_2/N_2$ microwave discharge

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This study deals with the diagnostic of an $Ar_{1-x}/(O_{2\ 1-y}/N_{2\ y})_x$ ($0 \leq x \leq 0.1$ and $0 \leq y \leq 1$) plasma by means of threshold ionization mass spectromery and optical emission spectroscopy. The nature and density of neutral reactive species are determined as a function of the gas composition, working pressure and microwave power.

Oxygen and nitrogen mixture (or air) plasmas are an important topic of plasma research as they cover different application fields [1-4] such as atmospheric or low pressure discharges for sterilization, surface treatment of materials, active species production for plasma chemistry, plasma cleaning, blood coagulation, interaction with organic molecules of biological interest ... For all these plasma processes, several questions arise which are of crucial importance for the understanding and the control of such processes :

- what are the active species produced in the gas phase and what are their density ?
- how do these active species interact with surfaces ?

Different tools are commonly used for the diagnostics of charged or excited species (Langmuir probes, optical emission spectroscopy ...). For ground state, metastable and radical species, one must resort to more sophisticated techniques such as laser absorption, laser induced fluorescence, cavity ring down spectroscopy, Fourier transform infra-red spectroscopy which are either poorly sensitive or dedicated to the study of a particular species. Threshold ionization mass spectrometry [5] allows us to determine the nature and the density of the different radicals formed by the plasma.

In the present work, a combination of optical emission spectroscopy and mass spectrometry is used to characterize an $Ar_{1-x}/(O_{2\ 1-y}/N_{2\ y})_x$ ($0 \leq x \leq 0.1$ and $0 \leq y \leq 1$) microwave plasma and its post-dischargein the pressure range 1 - 10 mbar. Fig. 1 shows the signal intensity collected in RGA mode (for Residual Gas Analysis: collection of neutral species from the post-discharge and ionisation by electron impact inside the ionization chamber of the mass spectrometer) at m/z = 16 as a function of the electron energy. When the plasma is turned off, the signal comes only from O^+ ions created by the dissociative ionisation of O_2 molecules by electron impact ($O_2 + e \rightarrow O^+ + O + 2e$, $E_{th} = 18.8$ eV). When the plasma is turned on, a second component in the measured signal can be seen. This component can be attributed to O^+ ions created by the electron impact ionisation of O atoms ($O + e \rightarrow O^+ + 2e$, $E_{th} = 13.6$ eV). These O atoms are produced by the plasma and collected in the post-dischage zone.

Knowing the electron impact ionization cross sections for these processes and using the perfect gas law, the gas temperature, dissociation rate and absolute density of all species can be determined from these measurements and following equation:

$$I_{m/z=16}(\varepsilon) = K \times \left(\sigma_{O \rightarrow O^+}(\varepsilon) \times [O] + \sigma_{O_2 \rightarrow O^+}(\varepsilon) \times [O_2]\right) \tag{1}$$

$$\frac{[O]}{[O_2]} = \frac{\dfrac{\sigma_{O_2 \rightarrow O^+}(\varepsilon_2)}{\sigma_{O \rightarrow O^+}(\varepsilon_1)}}{\dfrac{I_{m/z=16}(\varepsilon_2)}{I_{m/z=16}(\varepsilon_1)} - \dfrac{\sigma_{O \rightarrow O^+}(\varepsilon_2)}{\sigma_{O \rightarrow O^+}(\varepsilon_1)}} \tag{2}$$

with 13.6eV $\leq \varepsilon_1 \leq$ 18.8 eV and $\varepsilon_2 \geq$ 18.8 eV.

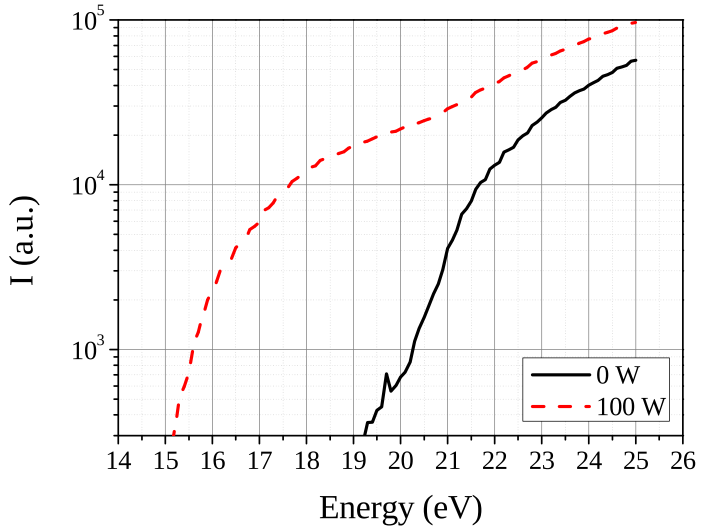

Fig. 1 : Mass spectrometry signal intensity in RGA mode at m/z = 16 as a function of electron energy for plasma on and plasma off conditions in Ar/O_2 gas mixture (70 sccm, 7.8 sccm, 100 W, 3.7 mbar)

In the experimental conditions given in Fig. 1, O atom density inside the post-discharge is determined to be 1.9x10^{14} cm^{-3} for a 1.3% dissociation rate of O_2. The same method is applied for the determination of N atoms densities when N_2 is added to Ar/O_2 plasma. Fig. 2 shows the evolution of densities in a Ar/O_2/N_2 plasma as a function of gas composition.

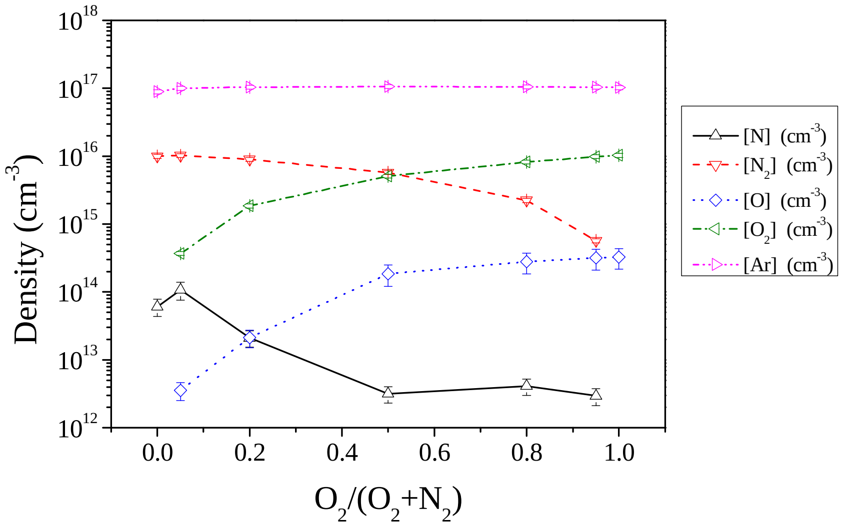

Fig. 2 : Species density vs gas composition in a 180 sccm Ar, 20 sccm O_2+N_2 for different O_2/(O_2+N_2) ratios at 100 W microwave power, 5.6 mbar pressure and 345 K gas temperature.

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References

[1] A. Ricard, V. Monna, *Plasma Sources Sci. Technol.*, 2002, 11 A150–3
[2] T. Czerwiec, J. Gavillet, T. Belmonte, H. Michel, A. Ricard, *Surf. Coat.Technol.* 1998, 98 1411–5
[3] M. Mrazkova, P. Vasina, V. Kudrle, A. Talsky, C. D. Pintassilgo, V. Guerra, *J. Phys. D: Appl. Phys.*, 2009, 42 075202.
[4] K. Kutasi, V. Guerra, P. A. Sa, *Plasma Sources Sci. Technol.*, 2011, 20 035006.
[5] E. Stoffels, Y. Aranda Gonzalvo, T. D. Whitmore, D. L. Seymour and J. A. Rees, *Plasma Sources Sci. Technol.*, 2007, 16 549