ESTIMATING PARTICLE TEMPERATURE FOR AN ARGON-OXYGEN DISCHARGE BY USING LANGMUIR PROBE AND OPTICAL EMISSION SPECTROSCOPY¹

M. Aflori^{2*}, G. Amarandei ^{*†}, L. M. Ivan ^{*}, D. G. Dimitriu ^{*†}, D. Dorohoi ^{*} ^{*}Department of Plasma Physics, "Al. I. Cuza" University, 11 Carol I Blvd., RO-700506 Iasi, Romania [†]Institute for Ion Physics, Leopold-Franzens University, Technikerstr. 25, A-6020 Innsbruck, Austria

Received 18 April 2005, in final form 28 June 2005, accepted 22 July 2005

The glow region of a argon-oxygen mixture rf asymmetrically capacitively coupled plasma was studied using a self-compensated Langmuir probe. Optical emission spectroscopy has been used to obtain information about different plasma parameters as well as the processes which take place in plasma in the range of the studied pressures and powers.

PACS: 52.27.Cm, 52.70.Ks, 52.70.Ds

1 Introduction

An experimental study of the electron energy distribution function (EEDF) has been carried out. Plasma parameters as plasma potential, electron temperatures, electron densities, are correlated with the elementary processes involved in the discharge.

Optical emission intensities of recorded spectral lines for atoms and ions increase with increasing both of power and pressure. Spectra have been recorded from 600 to 900 nm. Using the relationship between the relative intensities of the emission lines the electronic plasma temperature has been calculated for different pressure and powers. One method for characterizing ion and atom energies is to examine the Doppler profile of the emission lines spectra from these species. Ion energies derived from these emission profiles depend on the operating power and pressure. A good concordance between the temperatures estimated using optical emission spectroscopy and the ones calculated for the same device using Langmuir probe measurements was obtained. Those diagnostics are important for characterizing plasma parameters relevant to etching applications [1].

0323-0465/05 © Institute of Physics, SAS, Bratislava, Slovakia

¹Presented at Joint 15th Symposium on Applications of Plasma Processes (SAPP) and 3rd EU-Japan Symposium on Plasma Processing, Podbanské (Slovakia), 15 – 20 January 2005.

²E-mail address: maflori@plasma.uaic.ro



Fig. 1. Schematic of the experimental setup (the grounded chamber wall acts as anode).

2 Experimental results and discussion

The RF discharge investigated was confined in a plasma chamber of an asymmetrical industrial OTP Plasmalab 100 capacitively coupled system (Fig. 1). The top electrode diameter was 295 mm, inter-electrode spacing 50 mm and the driven electrode diameter was 205 mm. The argon flow rate was measured in cubic centimeters per minute at standard temperature and pressure and kept constant at 14.1 sccm, while the oxygen flow rate was 1.41 sccm. The pressure was controllable between 6 and 12 Pa and this was achieved automatically by measurement of the pressure via capacitance pressure gauge and a pumping throttle valve. The range of powers was 10-150 W. The apparatus was controlled by Oxford Instruments software from a computer. A Hiden Analitical RF-compensated Langmuir probe was inserted into the middle of the plasma, as shown in the Fig. 1. The quartz plate shown in the diagram was 12 mm thick and covered the cathode with the exception of a ring approximately 5 mm wide at the cathode's edge. Both cathode and anode are made from aluminium. The grounded dark space shield prevented the formation of any plasma around the sides of the cathode, and kept the equipotential lines closer and parallel to the surface of the cathode. A matching network was used to match the impedance in order to maximize the energy transfer from the power supply to the plasma. The distance between probe tip and quartz plate was 18.5 mm. The cylindrical probe, also used in this work, was made of tungsten (10 mm length, 0.35 mm diameter).

The existence of the RF structure on the plasma potential will cause an RF potential to appear across the sheath of the DC Langmuir probe. Such time-varying sheath potentials modify the current sampled by the probe, and, to overcome this problem, the probe described incorporates a subsidiary electrode, placed close to the measurement region (but not close enough to disturb



Fig. 2. Typical optical emission spectrum.

the measurement), which senses the plasma potential variation and feeds them to the probe tip. The probe assembly comprises three parts: the main body, the compensation electrode and the probe tip. A very common problem in probe diagnostics is the probe contamination [2]. In the RF discharges this problem becomes even more severe due to the sputtering of the RF electrode. Contamination of the probe tip with low-conductivity layers introduces additional resistance into the probe circuit that leads to severe distortion of the peak in the measured second derivative of the probe characteristic [3]. This distortion appears as a suppression, or even as an absence, of low-energy electrons in the measured EEDF An effective remedy of the probe contamination is continuous probe cleaning (by ion bombardment or by excessive electron current heating). In order to sputter off any impurities by energetic ion bombardment, prior to each measurement the probe was negatively biased down to -200 V for about 2 min.

Spectral scans from the discharge were obtained using a spectrophotometer Jobin-Yvon type coupled with a silica-silica high OH-PYROCOATTM fiber consisting of a pure silica core with a silica cladding for high damage threshold and high-performance optical proprieties. The chamber has a quartz window and the silica fiber was positioned outside the chamber, very close to this window. All data presented here were obtained by observing the emission at 90° from the central axis of the electrodes, parallel to the electrode surfaces (Fig. 1). A typical spectrum is shown in Fig. 2.

From the Langmuir probe characteristics, two populations of electrons, with temperatures $T_{e1} \simeq 2.8$ eV and $T_{e2} \simeq 0.8$ eV respectively, and densities $n_{e1} \simeq 10^{13}$ m⁻³ and $n_{e2} \simeq 2 \times 10^{14}$ m⁻³, are founded to be present in the discharge (Fig. 3).

The plasma potential is located by automatically examining $d^2I/dV^2 = 0$ (Fig. 4a). In Fig. 4b we can easy distinguish the maximum of the first derivative dI/dU and the plasma potential can be assumed to correspond to this maximum. In the Fig. 5 the mean values of the plasma



Fig. 3. Normalized EEDF at P = 130 W and p = 12 Pa.

potential (in volts) are presented as function of pressure and RF power. It shows that plasma potential is positive with respect to the anode (grounded electrode).

The simplest and most direct method of using spectral line intensities to determine the temperature of plasma is to use the relations existing between the intensities of the spectral lines, when the state densities of an atomic species are in equilibrium. In order to determine the electronic plasma temperature by using the relative intensities between two spectral lines, it is necessary to consider plasma in the partial Local Thermodynamic Equilibrium (LTE). The partial LTE basically demands that the excitation and ionization process have been produced only by electron impact. To satisfy this condition, it is necessary for the electronic plasma density and the electronic plasma temperature to be $n_e \geq 10^{14}$ m⁻³ and $T_e < 1$ eV, respectively. In our experiment these condition were satisfied for "cold" electrons. The electronic plasma temperature can be calculated using the relative intensity between two spectral lines, corresponds to the transition of the different lower levels, according to the following expression [4]:

$$T_e = \frac{1}{k} \frac{E_1 - E_2}{\ln(\frac{I_2}{I_1}) - \ln(\frac{(gf\nu^3)_2}{(gf\nu^3)_1})},\tag{1}$$

where E_1 and E_2 are the energies of levels, I_1 and I_2 are the intensities of spectral lines, ν_1 and ν_2 are the frequencies of the two spectral lines, g_1 and g_2 are the statistical weights, f_1 and f_2 are the oscillator strengths and k is the Boltzmann's constant. By using the equation (1), the electron temperature has been calculated. The atomic argon peak at 750.38 nm and atomic oxygen peak at 777.41 nm are considered. The values of E_1 and E_2 , g_1 and g_2 , f_1 and f_2 were obtained from the NIST Atomic Spectral Database [5]. The values obtained by using optical emission spectroscopy ($T_e \simeq 0.5 \text{ eV}$) are in concordance with those obtained using Langmuir probe measurements for the "cold" group ($T_e \simeq 0.8 \text{ eV}$).

The optical emission intensity of the atomic oxygen spectral line 777 nm is a superposition of 3 spectral lines [6] (777.41 nm, 777.63 nm and 777.75 nm). Each of these three components is Doppler broadened [7]. After removal of the instrument broadening, the signals were decon-



Fig. 4. Analysis of the Langmuir probe characteristic (P = 20 W, p = 8 Pa).

voluted (Fig. 6) and the resulting profiles were fit with a Gaussian line-shape function whose full width at half-maximum (FWHM) was used as a measure of the Doppler width [8,9]. The Stark effect is insignificant related to Doppler effect in the broadening of spectral lines in the domain of working RF powers. At lower pressures (<100 Pa) the effect of broadening of spectral lines due to atom collisions can be neglected, the thermal motions of ions are predominant and a Maxwellian velocity distribution can be suggested by equation (1). In this case the broadening of spectral lines is determined in principal by Doppler effect and the following equation can be used to determine the atom and ion energies [9]:

$$kT = \frac{mc^2}{8\ln 2} (\frac{\Delta\lambda_D}{\lambda_0})^2,\tag{2}$$

where $\Delta \lambda_{1/2} = 2 \ \Delta \lambda_D$ is the full-width of half-maximum intensity of the line determined from the deconvolution process, $\Delta \lambda_D = 2(\lambda_0/c)[\ln(2)(2kT/m)]^{1/2}$ is the Doppler width, λ_0 is the center wavelength of the emission line, *T* is the temperature of the emitting species, *k* is Boltzmann's constant, *m* is mass unit of emitting species, *c* is the speed of light in vacuum. The values of ionic temperatures are in order of hundred eV, while the values for neutral temperature are in order of tens eV.

Doppler linewidth measurements for atoms and ions illustrates that the plasma temperature depends on the power and pressure at which the system is operated. The emission profile for ions broadens as the pressure is reduced, while for the atoms it becomes narrower. The influence of the electrical properties of the plasma on ion energies is illustrated by the correlation between the ion emission linewidth and the plasma potential as function of power and pressure (Fig. 5 and Tables 1 and 2). With the decreasing of the pressures, the electron temperature increases. At lower pressures the ions suffer few collisions as they traverse the experimental device and



Fig. 5. Dependence of the plasma potential on the rf power and gas pressure.

the contribution of electric fields to ion motion is expected to be greatest at low pressures and high power since the magnitude of plasma potential increases under these conditions. It was observed (Tables 1 and 2) a drop in ion temperatures at higher pressure and a corresponding increase in linewidth for neutral emission (Tables 3 and 4). The linewidth for neutral species for argon-oxygen plasma is less then that for ions.

Pressure (Pa)	$\Delta\lambda_{1/2}$ (nm)	T (eV)
1.33	0.25 ± 0.005	98.16
3.99	0.23 ± 0.009	83.08
6.66	0.22 ± 0.006	76.02
9.33	0.21 ± 0.009	69.26
11.99	0.20 ± 0.007	62.82

Tab. 1. Temperatures of oxygen ions determined from the spectral line 656.52 nm using Doppler profiles

Fig. 7 shows the variation of spectral lines intensities for Ar 750.38 nm at different pressures and rf powers. At lower pressures the intensities of spectral lines are increasing with increasing of both power and pressure. An exponential dependence of optical emission intensities with pressure, when the power is maintained constant, can be observed. At higher pressures and



Fig. 6. Measured profile and profiles obtained by deconvolution process and fitting with a Gaussian function.

Pressure (Pa)	$\Delta\lambda_{1/2}$ (nm)	T (eV)
1.33	0.20 ± 0.006	177.14
3.99	0.19 ± 0.004	160.41
6.66	0.18 ± 0.009	143.96
9.33	0.17 ± 0.007	128.41
11.99	0.16 ± 0.008	113.75

Tab. 2. Temperatures of argon ions determined from the spectral line 617.22 nm using Doppler profiles

powers the intensity of spectral lines suffers saturation. At lower pressures, not all the electrons who have sufficient energy for excitations suffer collisions with the other particles. At higher pressures, the plasma density increases and the frequency of collisions became higher. In this case almost all electrons can transfer their energy by impact with the other particles and can excite them. By dezexcitation, the atoms and ions emit spectral lines.

3 Conclusion

Optical emission spectroscopy and Langmuir probe measurements were performed in a capacitively coupled 13.56 MHz RF discharge in argon-oxygen mixture, in order to determine ions, neutrals and electron temperatures. Two types of ions can be considered: those created in the sheath, respectively those that enter in the sheath from plasma and reach the anode surface without collisions. Both of the groups are related to the periodic extinction of the electric field leading



Fig. 7. Dependence of the Ar 750.38 nm intensity on the rf power and gas pressure.

Pressure (Pa)	$\Delta\lambda_{1/2}$ (nm)	T (eV)
1.33	0.12 ± 0.005	16.13
3.99	0.14 ± 0.009	21.95
6.66	0.15 ± 0.006	25.13
9.33	0.17 ± 0.009	32.28
11.99	0.19 ± 0.007	40.43

Tab. 3. Temperatures of oxygen atoms determined from the spectral line 777.41 nm using Doppler profiles

to a periodic penetration of electrons into the sheath [2]. High energetic ions came to the cathode surface because of high sheath voltage [10] and because of the asymmetry of the device. Similar results have been found by using a theoretical model [1, 11], which follows electrons and ions through time varying potentials. The high value of the atom temperatures are due to the charge-transfer collisions which transform a high-temperature ion into a high-temperature atom. Two groups of electrons are founded. The low-energy group is due to the electron-electron collisions (because of the high value of the electron density), while the high-energy group is a consequence of RF heating [2].

At low pressures, an increase in plasma potential values induces an increase in O^+ ion temperature. At high pressures and high rf powers, an increase in the number of ion-neutral collisions causes Doppler broadening of the emission from O atoms. Plasma parameters as plasma potential, electron and ion temperatures, electron densities, were measured in the glow region of an argon-oxygen mixture rf asymmetrically capacitively coupled plasma, by using electrical probes and spectroscopic methods. Doppler line width measurements for atoms and ions illustrates that the plasma temperature depends on the rf power and pressure at which the system is operated.

Pressure (Pa)	$\Delta\lambda_{1/2}$ (nm)	T (eV)
1.33	0.09 ± 0.009	24.35
3.99	0.11 ± 0.010	36.37
6.66	0.12 ± 0.007	43.29
9.33	0.14 ± 0.003	58.92
11.99	0.15 ± 0.006	67.64

Tab. 4. Temperatures of argon atoms determined from the spectral line 750.38 nm using Doppler profiles

The emission profile for the ions broadens as the pressure is reduced, while for the atoms it becomes narrower. The optical emission intensities saturates at high pressures and powers. It can be approximate by an exponential dependence of spectral lines intensities with pressure. The method of using spectral line intensities to determine the temperature of plasma was used. The temperature of cold electrons has been obtained in concordance with Langmuir probe measurements. The oxygen ion temperatures are in concordance with results obtained in a RF planar inductive oxygen discharge (25 eV at 4 Pa) [12] measured with a quadrupole mass spectrometer. By using a quadrupole mass spectrometer in our device, it was obtained the ion and atom energy distributions [2, 13–16], which provided similar results, taking into account that the low-energy ions obtained are in the order of tens eV.

References

- [1] C. C. Surdu-Bob, J. L. Sullivan, S. O. Saied, R. Layberry, M. Aflori: Appl. Surf. Sci. 202 (2002) 183
- [2] I. A. Rusu, G. Popa, J. L. Sullivan: J. Phys. D: Appl. Phys. 35 (2002) 2808
- [3] P. A. Chatterton, J. A. Rees, W. L. Wu, K. Al-Assadi: Vacuum 42 (1991) 489
- [4] G. Zambrano, H. Riascos, P. Prieto, E. Restrepo, A. Devia, C. Rincon: Surf. Coat. Tech. 172 (2003) 144
- [5] NIST Atomic Spectr Database, http://physics.nist.gov/egi-bin/AtData/main_asd
- [6] N. C. M. Fuller, M. V. Malyshev, V. M. Donnelly, I. P. Herman: Plasma Sources Sci. Technol. 9 (2000) 116
- [7] A. Rousseau, E. Teboul, N. Sadeghi: Plasma Sources Sci. Technol. 13 (2004) 166
- [8] J. A. O'Neill, M. S. Barnes, J. H. Keller: J. Appl. Phys. 73 (1993) 1621
- [9] M. H. Elghazaly, A. M. Abd Elbaky, A. H. Bassyouni, H. Tuczek: J. Quant. Spectrosc. Radiat. Transfer 61 (1999) 503
- [10] J. L. Sullivan, S. O. Saied, R. L. Layberry, M. J. Cardwell: J. Vac. Sci. Tec. A 16 (1998) 2567
- [11] R. L. Layberry: Ph.D. Thesis, Aston University, UK, 1999
- [12] J. T. Gudmundsson: J. Phys. D: Appl. Phys. 32 (1999) 798
- [13] M. Aflori, D. G. Dimitriu, J. L. Sullivan: 31st European Physical Society Conference on Plasma Physics, London, England, 2004
- [14] M. Aflori, D. G. Dimitriu, D. Dorohoi: 31st European Physical Society Conference on Plasma Physics, London, England, 2004
- [15] M. Aflori, D. O. Dorohoi, J. L. Sullivan: 15th International Conference on Gas Discharges and their Applications, Toulouse, France, 2004
- [16] M. Aflori, C. Gaman, L. M. Ivan, M. Mihai-Plugaru, D. G. Dimitriu: Bull. Am. Phys. Soc. 49 (2004) 32