Electron density and electron energy distribution functions in dual-mode microwave/radio frequency plasmas in argon and nitrogen

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Abstract

Dual-mode microwave/radiofrequency (MW/RF) plasmas were systematically investigated by means of Langmuir probe measurements and by mass spectrometry. Maxwellian-like electron distribution functions were found for both MW and MW/RF argon plasmas, while the distribution functions measured in nitrogen revealed two groups of electrons with different energies. Both in argon and nitrogen, the electron density, n_e, decreased with rising pressure, almost independent of the bias voltage on the RF electrode. The n_e value decays exponentially with distance from the MW applicator with a characteristic length comparable to the skin depth. Comparison of n_e values from the probe measurements with values calculated from the ion energy distribution functions and ion fluxes exhibit an excellent agreement.

Introduction

High-frequency discharges are frequently used in low-pressure plasma processing such as in plasma-enhanced chemical vapor deposition (PECVD), reactive ion etching (RIE) or surface modification [1,2]. Precise control of power delivered to the discharge, plasma uniformity as well as the flux and energy of ions impinging upon the substrate are the most important parameters for process optimization. In order to selectively control the chemistry in the plasma bulk and at the substrate surface, it is desirable to control independently the electron density, n_e, in the plasma and the ion flux, Φ_i, to the substrate. Dual-mode microwave/radiofrequency (MW/RF) plasma sources have been developed to fulfill these demands [2,3]. The MW/RF plasma has been used extensively to deposit functional coatings for barrier, optical and protective applications, and to modify materials surfaces for enhanced adhesion [1], but a complete characterization of the plasma bulk has so far not been performed. In the present work, microwave (MW), radiofrequency (RF), and MW/RF discharges are systematically investigated by means of Langmuir probe measurements and by mass spectrometry.

Experiments

The plasma system consisted of a RF-powered (13.56 MHz) stainless-steel electrode (150 mm diameter) facing a fused silica window, through which continuous MW (2.45 GHz) power was applied from a 25 cm long slow-wave applicator [4]. The MW power was supplied from a M1200 (Muegge) generator, with a 1.2 kW maximum output power. The RF power was coupled to the discharge by a tunable L-type matching unit (Advanced Energy) connected to the upper electrode (substrate holder). The distance between the upper electrode and the silica window was 100 mm.

The reactor chamber was pumped to a base pressure $<10^{-4}$ Torr, before Ar or N_2 were introduced through a mass flow controller at a flow rate of 20 sccm. The working pressure ranged between 20 and 200 mTorr.

The Langmuir probe (a tungsten wire, 50 µm in diameter and 6 mm in length) was attached to a translation/rotation feed-through, and it enabled spatially-resolved measurements along both the horizontal and vertical directions inside the plasma zone. The probe characteristics were recorded by a computer-controlled data acquisition system similar to that described in Ref. [5]. To enhance the signal-to-noise ratio, every point was averaged from 50 measurements. The probe was periodically cleaned by heating with high electron current.

The probe characteristics were evaluated by a computer program described in Ref. [6]. The obtained data were smoothed by digital filters and differentiated: The second derivative of the probe current, I, determines the electron energy distribution function, $F(E)$, according to the well-known Druyvestein formula $F(E) \sim E^{1/2} \cdot d^2I/dV^2$, where V is the probe potential and E is the electron energy. The plasma potential, V_p, and the electron temperature, T_e, were determined from the $d^2I/dV^2 = 0$ point, and from the slope of the semi-logarithmic plot of d^2I/dV^2 versus V, respectively. The n_e values were determined as $\int F(E)\,dE$.

Complementary measurements were performed using a Hiden EQP 1000 mass spectrometer, which sampled the discharge through a 50 µm diameter aperture in the RF electrode. The ions penetrating the orifice were accelerated by the electric field between the RF-electrode and the extractor into the drift region that precedes the ion energy analyzer (ESA). After crossing ESA the ions were separated in the quadrupole mass filter according to their mass-to-charge ratio. The flux of separated ions was detected by a channeltron secondary electron multiplier (SEM). As the SEM signal has not been calibrated, the Φ_i values will be quoted in arbitrary units and only relative changes will be discussed.

Results and discussion

Spatially-resolved measurements were performed in order to obtain information about plasma homogeneity in the reactor. The n_e values reached their maximum below the center of the substrate holder, with a small decay observed perpendicularly to the applicator. This n_e profile was the same for argon and nitrogen. For example, n_e measured 2 cm below the substrate holder at P_{MW}=300 W power varied from 8.2×10^{16}m^{-3} in the center to 5.4×10^{16}m^{-3} 6 cm from the center for argon, and from 4.1×10^{16}m^{-3} to 1.8×10^{16}m^{-3} for nitrogen, respectively. As expected, n_e decreased with decreasing power but the profile remained practically unchanged. In addition, n_e was found to be uniform within ± 10% along the whole length of the MW applicator.

The value of n_e *decreased exponentially with increasing distance from the MW-applicator. The vertical* n_e *profile could be approximated with an exponential function:*

$$n_e(y) = n_{e0}\exp(-y/d). \qquad (1)$$

The constants n_{e0} *and* d, *determined from the experimental data, correspond to* n_e *at the silica window surface, and to the characteristic decay length, respectively. The* n_{e0} *and* d *values calculated from vertical profiles of* n_e *measured in argon and nitrogen are summarized in Table 1.*

Table 1:

	Ar, 30 mTorr	Ar, 100 mTorr	Ar, 200 mTorr	N_2, 100 mTorr
n_{e0} *[m⁻³]*	2.56e18	1.075e18	6.9e17	3.88e17
d[cm]	3.088	3.016	2.899	3.948
δ_s *[cm]*	2.06	3.17	3.97	5.30

It is known that the fringing electric field intensity decreases exponentially with distance from the plane of the slow-wave structure [4] due to attenuation in the lossy (plasma) medium. The characteristic attenuation length, δ_s, *is inversely proportional to the plasma frequency, that is [7],*

$$\delta_s = \frac{2\pi c}{e}\left(\frac{\varepsilon_0 m}{n_0}\right)^{1/2} \qquad (2)$$

where m *and* e *are the electron mass and charge, respectively,* ε_0 *is the permittivity of free space, and* c *is the light velocity in vacuum. The values calculated using* n_{e0} *are also shown in Table 1. All* δ_s *values, both for argon and nitrogen, vary between about 2 and 5 cm. The observed good correlation between* δ_s *and* d *indicates that* n_e *in the plasma volume is controlled mainly by the intensity of the MW field rather than by free diffusion.*

The influence of the additional RF signal on the MW plasma in the MW/RF regime was investigated by a series of measurements in Ar and N_2 *at different pressures. The results for Ar are summarized in Fig. 1. One can see that* n_e *decreases with rising pressure from* 2.5×10^{17} m^{-3} *at 30 mTorr to* 4×10^{16} m^{-3} *at 200 mTorr. In* N_2 *the dependence of* n_e *versus pressure was similar to that in Ar:* n_e *decreased from* 1×10^{17} m^{-3} *at 30 mTorr to* 1.6×10^{16} m^{-3} *at 200 mTorr. Both in Ar and* N_2, n_e *was found to be almost independent of the* V_B *value on the RF electrode.*

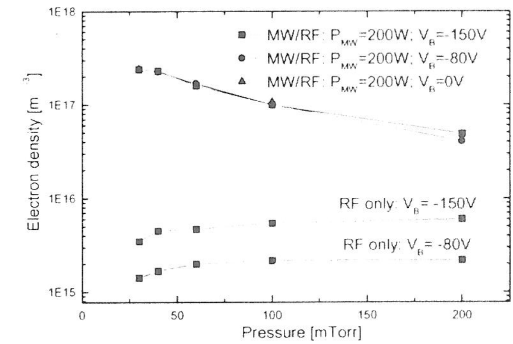

Fig. 1: *Electron density versus pressure in MW, RF and MW/RF plasma in argon, at a position 8 cm above the MW applicator.*

Examples of the measured d^2I/dV^2 behaviour are shown in Fig.2. Maxwellian-like electron distributions were found for both MW and MW/RF plasmas in Ar. T_e values varied between 1.3 and 1.5 eV, and were found to depend only little on other process parameters. *F(E)* measured in N_2 revealed two groups of electrons with different energies, namely: (i) $T_e \sim 0.8$ to 1.0 eV (low-energy group), and (ii) $T_e \sim 3$ eV (high-energy group). The difference between the high- and low-energy groups was less pronounced at higher pressures. The existence of these two groups can be attributed to energy losses resulting from inelastic collisions of electrons with nitrogen molecules.

A structured $F(E)$ was also found in RF discharges in Ar. Such a structure is well known in the RF discharges, as a result of a balance between the collisional frequency and the RF frequency [8], or as a result of stochastic heating in the so-called γ mode [9]. Unfortunately, it was impossible to calculate the second derivative from the probe characteristic measured in the RF discharge in N_2 because of an occurrence of plasma instabilities.

Assuming plasma quasi-neutrality and a collisionless Child-Langmuir sheath between the substrate holder and the plasma bulk, we developed a method to calculate n_e from measured ion energy distribution functions (IEDF) and ϕ_i data. This method was applied to determine n_e at the plasma-sheath edge for both MW/RF and RF plasmas [2,3]. In order to verify this model, we compared n_e obtained from the IEDF with the n_e values from our probe measurements.

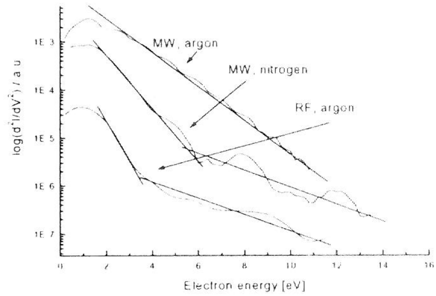

Fig. 2: *Examples of the d^2I/dV^2 plots.*

The n_e values calculated from the IEDFs relate to the plasma-sheath edge. The sheath thickness depends on the pressure, but except for a "pure" RF plasma, it never exceeds 2 mm in the pressure range investigated [3]. Since we were able to perform our probe measurements as close as 2 cm from the substrate (8 cm above the silica window), we extrapolated the exponential decay of n_e up to a distance of 10 cm from the silica window (substrate position), and calculated the corresponding $n_e(10)$ value. The n_e values at the plasma-sheath edge, n_s, correlate with the bulk n_e as $n_s = 0.61 n_e(10)$ [9]. The n_s values calculated from the IEDFs and from the probe data for MW/RF plasma at different pressures are summarized in Table 2.

Table 2:

Pressure [mTorr]	30	100	200
n_s – probe [m^{-3}]	6.1×10^{16}	2.5×10^{16}	1.4×10^{16}
n_s – IEDF [m^{-3}]	5.2×10^{16}	2.4×10^{16}	2.7×10^{16}

Good agreement between these values is evident, and it confirms the method of n_e calculation from the IEDF.

Beside the determination of bulk plasma properties (n_e, T_e,...), the probe measurements are also very attractive for the determination of ion fluxes towards the substrate holder, which otherwise requires another rather complex installation (differentially-pumped ion energy analyzer [10], mass spectrometry [2,3]).

The ion flux may be expressed by the following formula

$$J_i = 1.68\varepsilon_0 \left(\frac{2e}{M}\right)^{1/2} \frac{V_{dc}^{3/2}\lambda_i^{1/2}}{s_m^{5/2}} \tag{3}$$

where M is the ion mass, V_{dc} is the voltage across the plasma sheath, $\lambda_i = 1/330p$ is the mean free path, and p is the pressure. The sheath thickness, s_m, can be calculated according to [3] as

$$s_m = \frac{2\pi}{3}\left(\frac{2\varepsilon_0 V_{dc}}{n_i e}\right)^{1/2} \tag{4}$$

Fig. 3 shows the ϕ_i values calculated for different gas pressures from the probe data, together with the values determined from the MS measurements. Except for a discharge exited in "pure" MW plasma, the trend in ϕ_i deduced from probe measurements agrees with that calculated from the IEDFs. As s_m decreases with decreased V_{dc} and, simultaneously, λ_i increases with decreasing pressure, $s_m \approx \lambda_i$ at 30 mTorr. To estimate the influence of collisions, we also calculated ϕ_i assuming a collisionless sheath. As can be seen in Fig 3, only a small difference has been found; in fact, the difference between the probe and MS data cannot simply explain a decrease of ϕ_i by increased collisions. A possible explanation may be in terms of an angular distribution of ions, due to a lower voltage across the sheath, which reduces the total ion velocity perpendicular to the substrate. The combination of low ion energy with collisions in the sheath results in a broader angular distribution of ions, so that less ion flux is detected due to the limited acceptance angle of the EQP. A more detailed analysis is necessary to thoroughly clarify this effect.

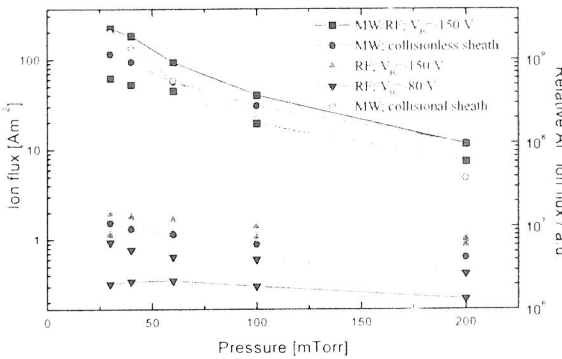

Fig. 3: *Relative flux of Ar⁺ ions calculated from probe and mass spectrometry measurements in MW, RF and MW/RF plasmas (solid line: calculated from probe measurements, dotted line: calculated from MS measurements).*

Conclusions

A systematic investigation of a dual-mode MW/RF plasma source has been performed using Langmuir probe and mass spectroscopy measurements. It was confirmed that the MW/RF excitation produces a large-volume, homogeneous plasma, with an independent control of plasma density and ion bombardment of the substrate. A structured electron energy distribution was found in nitrogen, while a Maxwellian distribution was observed in Ar. The plasma density was found to decay exponentially with increasing distance from the microwave applicator, with a characteristic length equal to the skin depth. The ion flux values calculated from the probe data for MW/RF and RF plasmas were found to be in good agreement with those calculated from the IEDFs.

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