Study of mode transition in low pressure capacitive RF discharges in nitrogen

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We observed two types of a capacitively coupled rf discharge in nitrogen – unconfined and confined mode. The modes differed significantly not only in a visual appearance but also in the dc self–bias voltage at the driven electrode, the plasma potential and the average electron energy. The ion energy distribution measured in the unconfined mode showed structure typical for the grounded electrode and lower pressure. The ion energy agreed relatively well with the results on the plasma potential.

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1 Introduction

Capacitively coupled rf discharges are used in numerous applications. Often the attention is focused on phenomena near the powered electrode where samples for nitridation or deposition are placed. However, for plasma treatment of polymers, there is a need for lower ion energies [1], e. g. obtained at the grounded electrode. We observed a transition of the discharge into the mode where the sheath at the grounded electrode expands by one order of magnitude and correspondingly the sheath voltage rises enormously. This effect can be very unpleasant in some treatment techniques. Therefore we decided to study this phenomenon in finer details.

2 Experimental

Rf capacitively coupled nitrogen discharges were studied in a stainless steel reactor of a spherical shape (i. d. 250 mm) with horizontally mounted two parallel–plate stainless steel electrodes separated by 57 mm. The bottom electrode (diameter 100 mm) was grounded. The upper electrode (diameter 80 mm) driven at 13.56 MHz was surrounded by a grounded shielding ring with the outer diameter of 100 mm. We used a constant nitrogen flow of 25.3 sccm which corresponded to the pressure of 2.5 Pa. The rf power delivered by the generator varied from 10 to 45 W. We measured rf voltage, rf current and a dc self–bias voltage on the driven electrode. For diagnostic purposes we used a Langmuir probe (Scientific Systems Ltd.) and the Plasma Processes Monitor PPM 421 (Balzers). The tip of the probe was placed at the discharge axis, 31 mm from the upper electrode. The horizontal distance of PPM's extraction hood was 5 mm from the electrode edges, the vertical distance was 9 mm from the bottom electrode. The extraction hood was grounded.

3 Results

We observed two discharge modes distinguished at the first glance from the visual point of view. The first mode was characterized by diffusive glow spread outside the interelectrode distance and a very thin dark sheath at the grounded electrode (see Fig. 1 left). We will refer to this case as to "unconfined mode". As we increased the power delivered by the rf generator up to maximum of 45 W the discharge abruptly changed its character. In this case, the discharge was evidently more confined between the electrodes and the dark sheath at the grounded electrode was comparable in the thickness with that at the powered one (see Fig. 1 right). We will further speak about this regime as about "confined mode". If the confined mode was once achieved the discharge stayed in this mode even after decreasing the power down to 10 W. Once ignited the confined discharge was sustained by rf voltage about 0.7 times lower than the unconfined mode. The difference between both modes can be seen also in the current–voltage characteristics of the discharges shown in Fig. 2. In both cases the current amplitude was a linear function of the voltage but the current for the same voltage was higher in the confined mode.

Two other features distinguished unambiguously the transition from the unconfined to the confined mode: (a) a significant decrease of a dc self–bias voltage measured on the powered electrode with respect to ground and (b) a significant increase of the plasma potential as well as the floating potential as measured by the Langmuir probe. As a result of these two opposite changes a sheath potential drop at the driven electrode, U_1, was similar in both modes for the same rf voltage as showed in Fig. 3. On the other hand, the potential drop at the grounded electrode, U_2, that is equal to the plasma potential U_p, increased by the factor of 5–10 due the mode transition (see Fig. 4). This fact resulted in the larger sheath thickness (compare two CCD camera images in Fig. 1). Theoretical considerations summarized by Lieberman [2] gives proportionality between the ratio of the sheath potentials and the ratio of their areas as $U_1/U_2 = (A_2/A_1)^\mathrm{q}$ where A_1 and A_2 are the areas of the

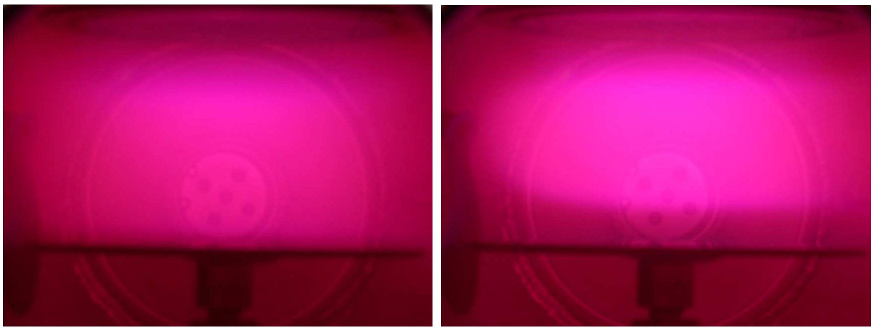

Fig. 1. CCD camera images of both modes of the discharge in nitrogen at similar applied rf voltage of 125 V: left side – "unconfined" mode, right side – "confined" mode.

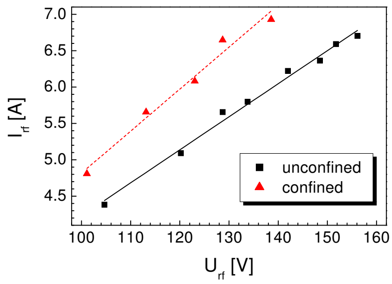

Fig. 2. Current voltage characteristics of rf nitrogen discharge in two modes.

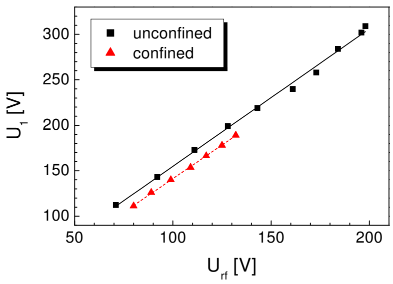

Fig. 3. Voltage drop in the sheath at the driven electrode for two discharge modes.

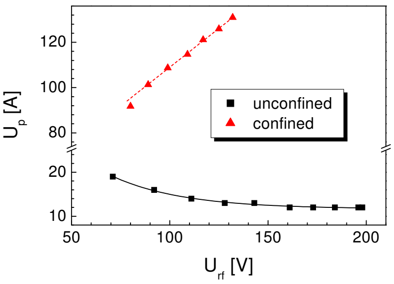

Fig. 4. Plasma potential as a function of applied rf voltage amplitude.

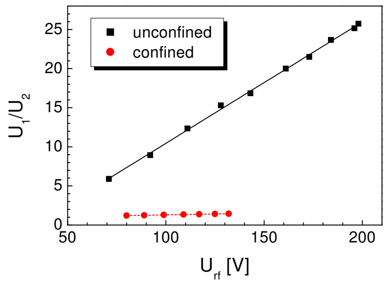

Fig. 5. The ratio of sheath voltages for two discharge modes.

driven and grounded electrodes, respectively and q depends on the model of the discharge, separately for the sheath and the glow. The sheath model depends on the pressure, i. e. on the assumption of the ion current density as a function of the sheath voltage and thickness. For the intermediate pressure used we could assume a collisional sheath with the constant mean free path for ions [2]. Then depending on the glow physics the exponent q varies as 1.25–2.5. In the Fig. 5 there is the ratio of the sheath voltages as a function of the applied rf voltage. In fact, this ratio should be constant. However, we have to take into account an effective area of the grounded surfaces in touch with the plasma that need not coincide with the area of the grounded electrode. Therefore the observed increase of the voltage ratio indicates a spread of the discharge, i. e. increasing the area of the grounded electrode. For the unconfined mode even at the lowest rf amplitude the effective area of the grounded electrode was higher than for the confined mode and depended linearly on the applied rf voltage (see Fig. 5). The variation of the area was about 12 % of the average value. In the case of the confined mode the variation was only 2 % although we can see again a linear dependence on the rf voltage.

The measured electron energy distribution function (EEDF) could not be fitted by one temperature distribution as was found by other authors too [3, 4]. Therefore

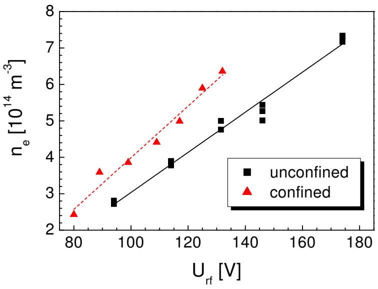

Fig. 6. The electron density calculated from the integral of the EEDF.

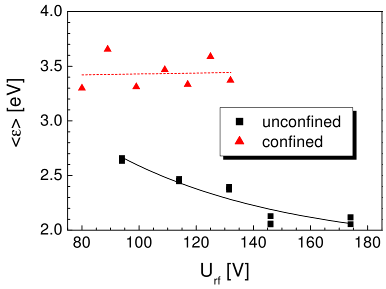

Fig. 7. The average electron energy calculated using integral of the EEDF.

we evaluated the electron density and the average electron energy directly integrating the EEDF obtained from the second derivative of the probe characteristics. The electron densities are given in Fig. 6. The density compared at the same rf voltages is higher in the case of the confined mode. We think that it is due to an effect of secondary electrons gained at the grounded electrode because of a higher potential drop in the case of the confined discharge (see Fig. 4). It is interesting to note that if we plot the density as a function of the rf current both the modes would behave similarly. The changes in the average electron energy are shown in Fig. 7. In the unconfined mode the average energy decreases from 2.7 to 2.1 eV with the rf voltage, i. e. with discharge spreading out. The energy in the confined mode is higher (approx. 3.4 eV) and does not change with the rf voltage.

The measurement of the ion energy distribution (IED) was in fact successful only in the unconfined mode. The reason for that is probably the position of the PPM. It was oriented horizontally as in Ref. [5] and not vertically through the electrode as usual in many other experiments. Olthoff *et al.* [6] showed that the horizontal position might not have a big influence on the measured IEDs. We hope this was the case for the unconfined mode in which the PPM head was in a contact with the glow and served like the grounded electrode. For the confined mode (Fig. 1 right) we can see a large dark space left from the grounded electrode near the PPM head. Moreover, due to the large sheath thickness at the grounded electrode the extraction hood was inside the sheath. These two facts probably caused a very low intensity of the ion current in the confined mode.

Shown in Fig. 8 are IEDs for N_2^+ ions sampled from the unconfined discharge as a function of the rf voltage. The ions had IEDs with relatively narrow asymmetric peak broadened at lower energies. The IEDs for N^+ ions were again peaked at the higher energies but the peak was even narrower and symmetric in the shape. The comparison of IEDs for both ionic species is made in Fig. 9 for the rf voltage of 144 V. Unlike at the driven electrode [8, 9] the IEDs did not contain any saddle shape or multiple peak structures. This is caused by a much lower sheath voltage modulation at the grounded electrode. The rf amplitude of the sheath voltages $U_{\mathrm{rf}1,2}$ can be estimated from the time average voltages $U_{1,2}$ given in Fig. 4 as

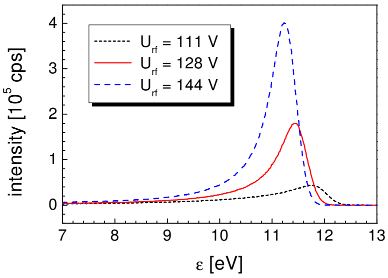

Fig. 8. IED of N_2^+ ions in the unconfined mode for different rf voltages.

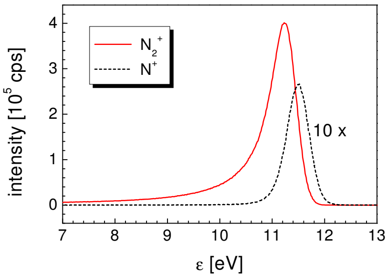

Fig. 9. Comparison of IED for N_2^+ and N^+ ions (unconfined mode, $U_{\mathrm{rf}} = 144$ V).

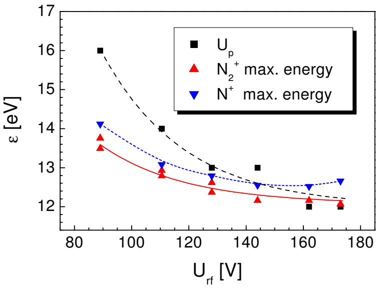

Fig. 10. Plasma potential compared with the maximum energy of N_2^+ and N^+ ions.

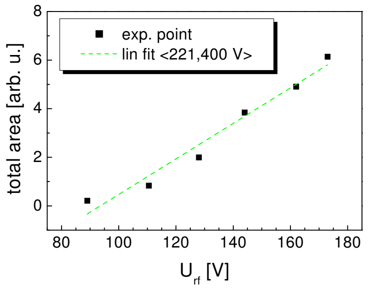

Fig. 11. Relative changes of ion density in the unconfined mode.

$U_{\mathrm{rf}1,2} = a\, U_{1,2}$ where a is 1.20 and 1.28 for collisionless and collisional sheaths, respectively [10]. The shape of the IEDs is in agreement with the results obtained by other authors at the low pressure [6, 9]. The peak at high energy is consistent with the expectation that the majority of ions are formed by electron impact in the bulk plasma, near the sheath–plasma interface, where the density of energetic electrons is highest [7]. Ions that experience energy loss by collisions contribute to the low–energy portion of the IEDs. The sheath at the grounded electrode is very thin for the unconfined mode – we estimated about 1 mm. The mean free paths for N_2^+ and N^+ ions with the energy of 13 eV calculated from the cross sections in Ref. [11] are 1.4 and 12.7 mm, respectively. It means that the sheath at the grounded electrode is collisionless for N^+ ions but there may be some effect of collisions for N_2^+. This is evidenced by different types of IEDs as shown in Fig. 9.

The transit time required for an ion to traverse the sheath region is at the frequency of 13.56 MHz much higher than the period of the rf voltage. This means that the maximum energy of the ions arrived at the surface is determined by the time average of U_{rf2}, i. e. by U_p. The ion maximum energy is compared for both ionic species, N_2^+ and N^+, with the plasma potential U_p in Fig. 10. Both, the maximum energy and the energy of the peak maximum exponentially decreases with the rf voltage similar as the plasma potential. For lower rf voltages the maximum energy of ions is about 2 eV lower than the plasma potential but then it coincides well, especially for N_2^+ ions. The error of the ion energy was estimated about 0.3 eV (0.1 eV of a standard measurement error and 0.2 eV of a systematic error in the energy scale calibration) and the error of plasma potential measurements was about 0.3 V. The relative ion density calculated as a sum of IEDs' integrals for both, N_2^+ and N^+, ions is given in Fig. 11 in dependence on the rf voltage. Except the first point at the lowest rf voltage it exhibits a linear dependence on the rf voltage similar to the electron density measured by the probe (see Fig. 6). In the range of the rf voltages the ion density increased by factor of seven although the electron density only by factor of three. It may be caused by the changes in the acceptance angle for spreading discharge but this fact has to be studied further in details.

4 Conclusions

Two observed discharge modes, confined and unconfined, differed significantly in the dc self–bias voltage and the plasma potential. As a result the sheath voltage at the driven electrode was similar in both modes but the sheath voltage at the grounded electrode raised from 13 V up to 130 V. The IEDs of N_2^+ and N^+ ions in the unconfined mode showed effect of different mean free path of both ions. The maximum ion energy agreed relatively well with the results on the plasma potential.

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