LANGMUIR PROBE DIAGNOSTICS OF NEGATIVE-ION PLASMA

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Abstract

We present comprehensive Langmuir probe diagnostic, based on a MATLAB code determining from the current – voltage characteristic plasma potential, electron density, electron temperature and electron energy distribution function. Due to the implemented graphical user interface the program enables friendly and complex control of the computational process and its optional parameters. Moreover, a generalized method of determining negative-ion density from the current-voltage characteristic is proposed. This method is based on comparison of the characteristic measured in electronegative plasma (e.g. Ar/O_2) with a reference characteristic scanned in electropositive plasma (Ar).

The developed method was applied to the detailed diagnostic of a direct current discharge in Ar/O_2 gas burning in a planar magnetron operating in various regimes. Electron density, electron temperature, positive and negative ion densities as well as plasma potential are investigated in dependence upon various experimental conditions.

1. Introduction

Direct current (DC) magnetron sputtering is one of the methods widely used for deposition of thin layers. Magnetrons are mostly used in two basic configurations – cylindrical [1] or planar [2].

The common feature of both magnetron configurations is crossed electric and magnetic field that confines electrons in the vicinity of the cathode, connected with a target made from material to be deposited. The confined electrons form high-density plasma with positive ions accelerated towards the cathode (target) and sputtering the target material. The sputtered target material is then deposited on the substrate.

Planar magnetrons can be operated in balanced (BLM) and unbalanced (UNB) mode [3]. In the balanced mode the magnetic field lines are well confined around the cathode and the electron loss is reduced to minimum. The unbalanced magnetron allows electrons to release from the magnetic trap and ionize plasma away from the magnetron cathode.

The fundamental aspects of magnetron discharge used for practical applications were studied in [4]. These applications mostly work with mixtures of gases. As a carrier gas is used rare gas like Ar. Often nitride layers (N_2 added) or oxide layers (O_2 is added) are created because of their wide utilization. Mixtures Ar/O_2 plasma discharges were investigated spectroscopically [5] and by Langmuir probe method [6]. However, the presence of negative ions in plasma changes the current-voltage characteristic and therefore complicates the interpretation of Langmuir probe data. Moreover, Langmuir probe diagnostic in mixtures with O_2 is distorted by oxide layers deposited on the probe surface.

Methods determining negative ion density in electronegative plasma from Langmuir probe measurements were proposed, e.g., in Ref. [7-9]. The approach in papers [8,9] is based on comparison of two probe characteristics, the first taken in pure electropositive (Ar) plasma, the second measured in electronegative (Ar/O_2) plasma. Determination of negative ion density by the help of Langmuir probe investigation is a main goal of presented work.

Sec. 2 describes the experimental setup. In Sec. 3 we suggest a generalized method based on comparison of Langmuir probe measurements in electronegative plasma with reference measurements in electropositive plasma. Sec. 4 shortly describes the structure of developed MATLAB codes. Results and discussion are presented in Sec. 5.

2. Experiment

The experiments were carried out in spherical-shaped stainless-steel vacuum chamber. The magnetron operated either in BLM or UNB mode by mechanically changing permanent magnet configuration [10]. Water cooling protected the cathode from overheating and consequent destruction. The whole magnetron cathode was movable in vertical direction. The flow rate of working gas was controlled by mass flow controllers in the ranges up to 2 sccm (reactive gas) and 100 sccm (carrier gas).

The Langmuir probe was inserted into the chamber using radially movable feed-trough electrically isolated from the vacuum chamber. The probe was made of 100 μm tungsten wire of active length 6 mm. The probe wire was sleeved by a double-capillary glass tube with the outer capillary longer than the inner. This arrangement prevented the probe from short-circuiting of the probe to the (metal-coated) sleeve by the sputtered metal during the deposition process.

The probe was situated near the plane of the substrate. Different radial distances were achieved by inclining the probe holder. As follows from the symmetry, in this configuration the probe tip was perpendicular to the magnetic field lines and therefore the effect of magnetic field on the probe measurement was minimized [20]. In addition, as the magnetic field in the plane of measurement in both operating modes was of the order of 1 mT, the mean Larmor radius for electrons was much larger than the radius of probe wire.

For measurement of axial dependences the probe was kept at the axis of the target and the distance from the magnetron cathode was varied for distances z = 45, 65 and 85 mm. For measurements of radial dependences the probe radial position was changed with the step of about 10 mm.

As the probe data in Ar/O_2 plasma can be affected by a non-conductive oxide layer deposited on the probe tip, the probe was cleaned by ion bombardment during the pauses between each measurement. During the cleaning process the negative bias of about −180 V was applied to the probe for several seconds.

3. Evaluation of Langmuir probe data

The negative-ion density as well as density and temperature of electrons in electronegative plasmas significantly influence plasma etching or sputtering. Thus, determination of these parameters belongs to the most important subjects. We determined these parameters from Langmuir probe measurements by a method generalizing a procedure applied in [9]. In the following text the subscripts ‘e’, ‘p’ and ‘n’ denote quantities related to electrons, positive ions and negative ions, respectively. Quantities concerning electronegative plasma are labeled by dash.

Let us start our considerations with remembering some relations for electropositive plasma. For the sake of simplicity we suppose here the Maxwell electron energy distribution function. Thus, the electron part of the cylindrical characteristic in the unsaturated region is

$$
\begin{aligned}
I_\mathrm{e} &= eS\,n_\mathrm{e}\cdot\sqrt{\frac{eT_\mathrm{e}}{2\pi\,m_\mathrm{e}}}\cdot\exp\left(\frac{V}{T_\mathrm{e}}\right), \qquad V<0,\\
I_\mathrm{e} &= eS\,n_\mathrm{e}\cdot\sqrt{\frac{eT_\mathrm{e}}{2\pi\,m_\mathrm{e}}}\cdot\left(1+\frac{V}{T_\mathrm{e}}\right)^{1/2}, \qquad V>0,
\end{aligned}
\tag{1}
$$

where S is probe area, V is voltage bias between the probe and plasma, n_e is electron number density and T_e is electron temperature measured in volts ($= kT_\mathrm{e}/e$ with T_e measured in Kelvins).

Not a few theories have been proposed for description of the positive-ion current and ion current theories do not provide sufficiently reliable and undoubted results. We assume here a rather general form

$$I_{\mathrm{p}} = eS\,n_{\mathrm{p}}\sqrt{\frac{eT_{\mathrm{e}}}{m_{\mathrm{p}}}}\cdot F(V,T_{\mathrm{e}},\lambda),\quad V<0 \tag{2}$$

with function F specified by a particular ion-current model. Parameters λ are optional and can be determined empirically by fitting the formula (2) to the measured data in the region $V \ll 0$. Multiplicative factor $\sqrt{eT_{\mathrm{e}}/m_{\mathrm{p}}}$ denotes Bohm velocity v_{B} of positive ions at the sheath-presheath edge for the collisionless plasma sheath [11].

Let us now consider electronegative plasma. The presence of negative ions does not significantly influences the part of characteristics where electron current is dominant as the inequality $T'_{\mathrm{e}}/m_{\mathrm{e}} \gg T'_{\mathrm{n}}/m'_{\mathrm{n}}$ yields $I'_{\mathrm{e}} \gg I'_{\mathrm{n}}$. For instance, usual ratios $T'_{\mathrm{e}}/T'_{\mathrm{n}} \sim 10^2$, $m_{\mathrm{e}}/m'_{\mathrm{n}} \sim 10^{-3}$ and $n'_{\mathrm{e}}/n'_{\mathrm{n}} \sim 1$ give $I'_{\mathrm{e}}/I'_{\mathrm{n}} \sim 10^2$. Hence, the characteristics of electronegative plasma near the plasma potential can be treated as pure electronic and the plasma potential V'_{s}, electron number density n'_{e} and electron temperature T'_{e} can be determined by standard procedures.

On the contrary, more controversial is the assumption that the model (2) of positive-ion current holds true for electronegative plasma with the same parameters λ. There is also some discussion concerning validity of the Bohm criterion. Braithwaite and Allen [12] obtained for low pressures modified Bohm criterion

$$v_{\mathrm{B}}^2 = \frac{eT'_{\mathrm{e}}}{m'_{\mathrm{p}}}\cdot\frac{n'_{\mathrm{ps}}T'_{\mathrm{n}}}{n'_{\mathrm{es}}T'_{\mathrm{n}} + n'_{\mathrm{ns}}T'_{\mathrm{e}}}\,, \tag{3}$$

where the subscript ‘s’ denotes local values at the sheath edge. At low collisionality the negative ions are confined in the plasma bulk, thus $n'_{\mathrm{ns}} \approx 0$ and the normal Bohm criterion applies. We suppose here - with some uncertainty – that the model (2) remains valid for electronegative plasma, too.

A general method determining parameters of electronegative plasma from Langmuir probe measurements can be shortly described as follows. First, a comparative characteristic of electropositive plasma existing in similar conditions is measured and from its electron part the electron number density n_{e} and electron temperature T_{e} are determined. Then from its ion part and with the equality $n_{\mathrm{p}} = n_{\mathrm{e}}$ taken into account the optional parameters λ of the ion current model (2) are specified. Knowing these parameters, the characteristic of electronegative plasma can be processed. The electron part of the characteristic measured in electronegative plasma gives the electron number density n'_{e} and electron temperature T'_{e}. The model (2) with known λ applied to the positive-ion saturation current gives the positive-ion density, n'_{p}. Eventually, the negative-ion density n'_{n} is determined from the quasineutrality condition, $n'_{\mathrm{n}} = n'_{\mathrm{p}} - n'_{\mathrm{e}}$.

In our measurements the common procedure outlined above was specified as follows. In the electron regions of the characteristics we supposed Maxwellian (1) or double-Maxwellian characteristic. In both measurements in electropositive and electronegative plasmas the plasma potential was determined from the first or second derivative of the characteristics and the electron density and temperature were determined by fitting the single-Maxwell (1) or double-Maxwell formula to the measured data.

In the positive-ion regions we applied empirical ion current model (2) with

$$F(V) = C\left(1 - \frac{V}{T_{\mathrm{e}}}\right)^{\kappa} \tag{4}$$

or

$$I_{\mathrm{p}} = D\left(1-\frac{V}{T_{\mathrm{e}}}\right)^{\kappa}, \qquad D \equiv eS\,n_{\mathrm{p}}\sqrt{\frac{eT_{\mathrm{e}}}{m_{\mathrm{p}}}}\,C \tag{5}$$

Parameters $\lambda = (C,\kappa)$ were determined from the region $V << 0$ of characteristic measured in electropositive plasma. For DC discharge in Ar plasma under experimental conditions shortly described in Sec. 2 we obtained $\kappa \approx 1.4$, $C \approx 0.2$.

To express the ion densities n'_{p}, n'_{n} and degree of electronegativity $\alpha \equiv n'_{\mathrm{n}}/n'_{\mathrm{p}}$ in electronegative plasma without unnecessary multiplicative factors, we employ

$$\varepsilon \equiv \frac{n'_{\mathrm{e}}}{n_{\mathrm{e}}}, \qquad \gamma \equiv \frac{n'_{\mathrm{p}}}{n_{\mathrm{p}}} \tag{6}$$

Then

$$n'_{\mathrm{p}} = \gamma\, n_{\mathrm{e}}, \qquad n'_{\mathrm{n}} = (\gamma-\varepsilon)\, n_{\mathrm{e}}, \qquad \alpha = 1-\frac{\varepsilon}{\gamma} \tag{7}$$

In our method ε is evaluated directly as we explicitly determine the electron densities n'_{e} and n_{e}. The parameter γ is

$$\gamma = \sqrt{\frac{T_{\mathrm{e}}}{T'_{\mathrm{e}}}\frac{m'_{\mathrm{p}}}{m_{\mathrm{p}}}}\,\frac{D'}{D} \tag{8}$$

with factors D' and D determined by fit of the model current (5) to the ion parts of the characteristics measured in electronegative and electropositive plasma, respectively.

For comparison, the method described in ref. [9] is also as a special case included in the scheme (6,7), now with parameters

$$\varepsilon = \sqrt{\frac{T_{\mathrm{e}}}{T'_{\mathrm{e}}}}\,\frac{I'_{\mathrm{e}}(0)}{I_{\mathrm{e}}(0)}, \qquad \gamma = \sqrt{\frac{T_{\mathrm{e}}}{T'_{\mathrm{e}}}\frac{m'_{\mathrm{p}}}{m_{\mathrm{p}}}}\,\frac{I'_{\mathrm{p}}}{I_{\mathrm{p}}} \tag{9}$$

The first expression follows from the Maxwellian electron characteristic (1). The second one is a consequence of the simplest but not quite adequate model of constant positive-ion saturation current

$$I_{\mathrm{p}} = D \tag{10}$$

There are two error sources in determining the ion densities. The first one is connected with the assumption that parameters λ in (2) (or C,κ in (4)) are identical in both electropositive and electronegative plasmas. To minimize this error, the state of the reference electropositive plasma should be chosen to be as close to the state of electronegative plasma as possible.

The second error arises due to the positive-ion mass uncertainty. The effective mass m_{p} of a mixture of positive ions is defined by the formula (see Eq. 5)

$$\frac{1}{\sqrt{m_{\mathrm{p}}}} = \sum_X p(X)\frac{\nu(X)}{\sqrt{m(X)}} \tag{11}$$

where $p(X)$ is fraction of a positive-ion species X ($\Sigma\, p(X)=1$), $\nu(X)$ is its ionization degree ($\nu = 1,2,\ldots$) and $m(X)$ is its mass. The kind of positive ions and their density in Ar/O_2 plasmas were roughly estimated by plasma monitor measurements [13]. The obtained orders of magnitudes of ion concentrations are shown in the table. Similar values for Ar^+ and Ar^{++} were detected in pure Ar plasma.

X	n(X) [a.u.]
Ar^{+}	10^{0}-10^{1}
Ar^{++}	10^{0}
O_2^{+}	10^{-1}
O^{+}	10^{-2}
O^{++}	10^{-5}-10^{-4}

Neglecting fractions of positive oxygen ions one obtains from Eq. (11)

$$m_{\mathrm{p}} = \frac{m(\mathrm{Ar})}{[1+p(\mathrm{Ar}^{++})]^2} \tag{12}$$

On account of plasma monitor analysis we estimated $p(\mathrm{Ar}^{++})$ in the range (0.1, 0.3) for all measurements regardless the plasma was electropositive or electronegative. Hence, $m_{\mathrm{p}} = (0.7 \pm 0.1)\, m(\mathrm{Ar})$, $\sqrt{m'_{\mathrm{p}}/m_{\mathrm{p}}} = 1 \pm 0.2$. The error of the parameter γ in (8) is due to the positive-ion mass uncertainty estimated up to 20%.

4. Description of MATLAB codes

For comprehensive processing of Langmuir probe measurements we have developed two codes written in MATLAB.

The first program processes separately the electron and ion part of a characteristic and determines quantities common for both electropositive (e.g. Ar) and electronegative (e.g. Ar/O_2) plasma: plasma potential, floating potential, electron temperature, electron number density and some optional parameters of the models of characteristics determined by the least squares method. The procedure requires numerical computations of the first and second derivatives of the data overlapped by noise signal. To eliminate this noise the derivatives at each point are computed as the mean value of derivatives of several smoothing polynomials differing each from other by definition domain.

The program is equipped by sophisticated graphical user interface (GUI) involving dialog boxes, radio buttons, edit and static texts, popup menus, pushbuttons and various events and callbacks controlled by mouse clicks and motion (Fig.1). User by GUI specifies adequate models for electron and ionic part of characteristic including regions for fitting (Fig.2) and other parameters.

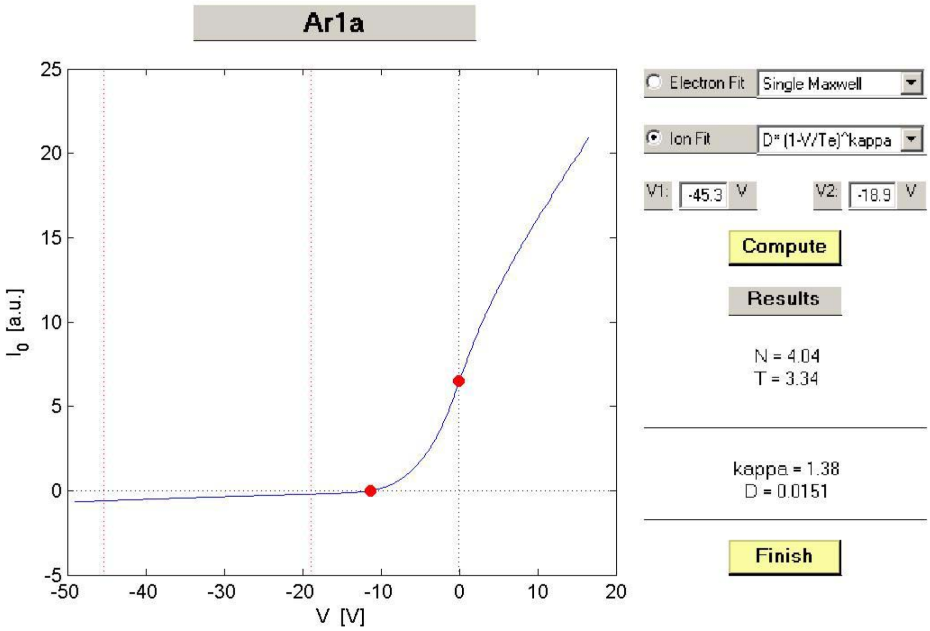

Fig. 1. Main window of GUI processing Langmuir probe characteristic

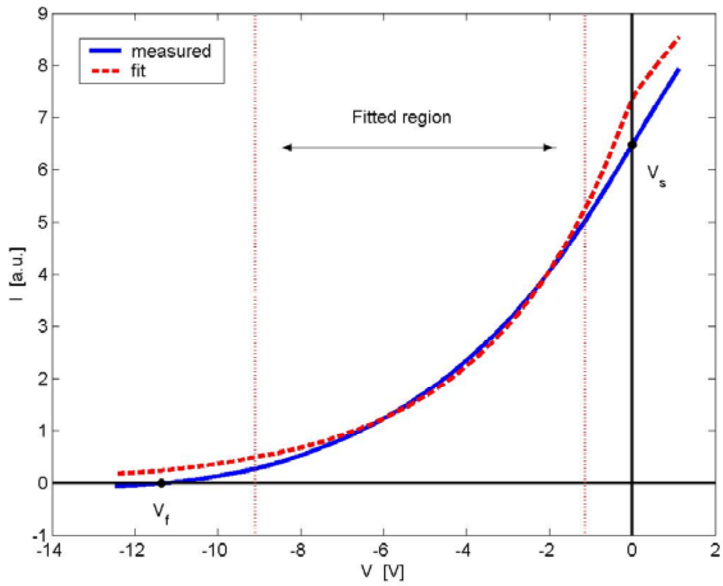

Fig. 2. Electron part of characteristic – measured and fitted

The second program compares a couple of characteristics – one for electropositive and the other for electronegative plasma. Then by the method described in detail in the previous section both the positive- and negative- ion densities in the electronegative plasma are determined. As input data serve results computed by the first program.

5. Experimental results and discussion

Direct current magnetron discharge was investigated with the main goal to diagnose the density of negative ions. Other plasma parameters as electron density, electron temperature and plasma potential were also evaluated with purpose to better understand negative ion creation. The negative-ion densities were calculated by the procedure described in the previous section.

In the following graphs we introduce experimentally determined plasma parameters depending on incoming power from 30 up to 110 W, on axial distances from the cathode (45, 65, 85 mm) and on radial distance from the axis of the target (cathode). The investigated plasma discharge was created from Ar as a carrier gas with flow rate 19.5 sccm. For Ar/O_2 mixtures oxygen was added as a reactive gas with two typical flow rates 0.75 and 1.5 sccm. Measurements were carried out for BLM as well as UNB mode. Power dependences were investigated when the probe was positioned at the axis of the target, i.e. at $r = 0$ mm. Radial and axial dependences are depicted for incoming power $P = 70$ W.

Power dependences of electron density for two different Ar/O_2 ratios are presented in Fig. 3. Similarly Fig. 4 depicts power dependences of negative-ion density. As plasma monitor data showed, relative concentration of atomic oxygen ions O^- is two orders of magnitude higher than concentration of molecular oxygen ions O_2^-. Hence, the evaluated negative-ion density concerns atomic oxygen ions O^-.

Figs. 3 and 4 also show that dependences of negative ion density on O_2 concentration or magnetron mode have opposite behavior compared to analogous dependences of electron density: if n_e is lower then n_n is higher and vice versa. More detailed analysis of this phenomenon has to be done.

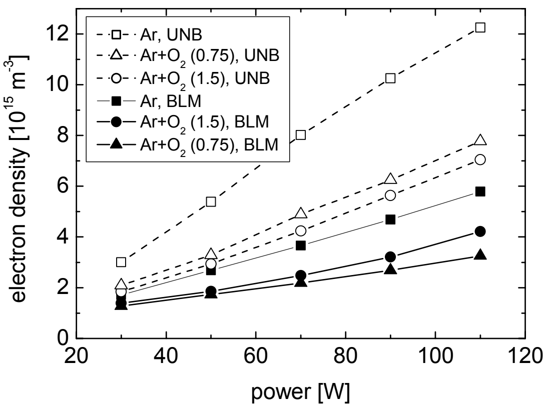

Fig. 3. Dependences of electron density on power, $z = 65$ mm, $r = 0$ mm, Ar (19.6 sccm), O_2 (0.75 and 1.5 sccm).

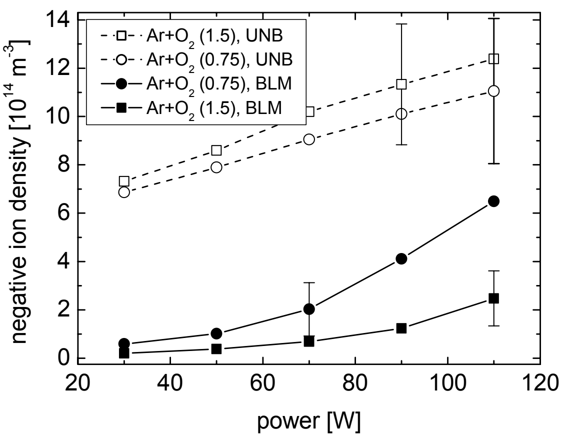

Fig. 4. Dependences of negative ion density on power, $z = 65$ mm, $r = 0$ mm, Ar (19.6 sccm), O_2 (0.75 and 1.5 sccm).

The radial profile of electron density is higher in UNB roughly by factor two compared to BLM. Stronger radial fall was observed only at the distance $z = 45$ mm, especially in UNB mode, see Fig. 5. The local electron density minimum is located at the distance $z = 65$ mm. This local density minimum is accompanied by the maximum of negative-ion density (Fig. 6).

As is shown in Fig. 6, the negative-ion density strongly depends on the radial position and – contrary to the electron density distribution - its maximum is shifted to the distance $r \approx 15$ mm. The pronounced inhomogeneous distribution of oxygen ions can be maintained due to their smaller diffusion coefficient. The fact that the radial distribution of negative-ion density in both BLM and UNB modes exhibits local maximum shifted apart from the symmetry axis is evident.

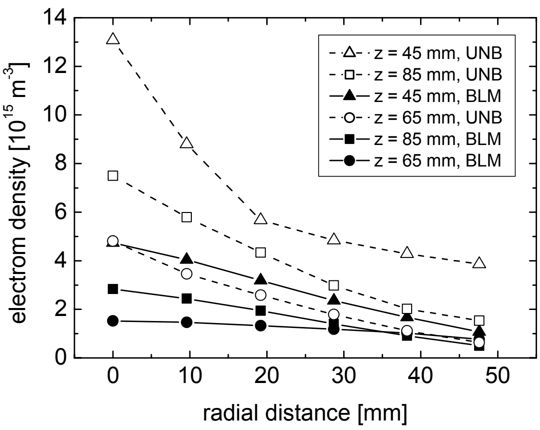

Fig. 5. Radial dependences of electron density, P = 70 W, Ar+O_2 (19.6+0.75 sccm), z = 45, 65, 85 mm.

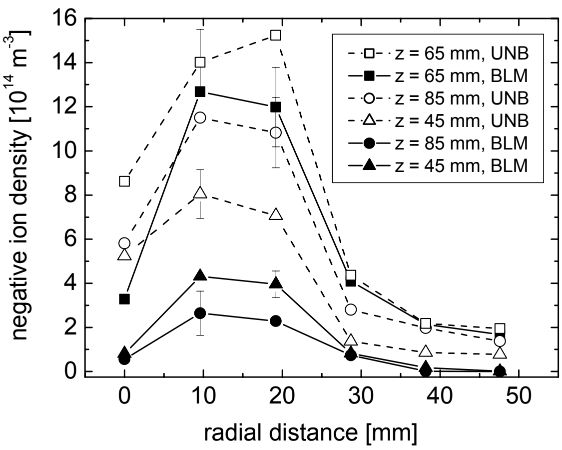

Fig. 6. Radial dependences of negative ion density, P = 70 W, Ar+O_2 (19.6+0.75 sccm), z = 45, 65, 85 mm.

6. Conclusion

DC planar magnetron discharge plasma in Ar and Ar+O_2 mixture was investigated by a new Langmuir probe method, determining the positive- and negative-ion density in electronegative plasma (e.g. in Ar/O_2 mixture) with negative ions (e.g. O^-). The proposed method is based on the comparison of the characteristic of the electronegative (Ar/O_2) plasma with the characteristic scanned in purely electropositive (Ar) plasma, where negative ions are absent. As is shown, the presence of negative ions does not significantly influence the electron part of the characteristic, therefore the electron number density and temperature as well as plasma potential for electronegative plasma can be determined by the same way as for electropositive plasma. The positive-ion density in electronegative plasma is estimated from the positive-ion saturation current with optional parameters of the current model determined from the characteristic measured in electropositive plasma, where the positive-ion density is known - it is equal to the electron density. This comparative procedure is valid under assumption that the presence of negative ions does not significantly influence the model of positive-ion current. After determining both the electron and positive-ion densities, the negative-ion density is evaluated as their difference.

Presented results show that Ar/O_2 plasma parameters are almost always higher in unbalanced than in balanced mode. In our experimental conditions the electron temperature and plasma potential have local maxima around 70 W. Electron density and negative ion density are related each to other. Negative ion density strongly depends on radial distance from the target (cathode) axis. Presented results suggest that negative ions originate at the distance around 65 mm from the target by dissociative electron attachment where the electron temperature reaches its maximum.

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