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## 112

GDAŃSK 2003

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Financial support of publication of this journal is provided by the State Committee for Scientific Research, Warsaw, Poland

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ISSN 0079-3205

TRANSACTIONS OF THE INSTITUTE OF FLUID-FLOW MACHINERY

No. 112, 2002, 125-137

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# Plasma parameters in a surface wave discharge in nitrogen at atmospheric pressure†

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## Abstract

*Spatial distributions of plasma parameters are calculated for a surface wave discharge sustained in nitrogen at atmospheric pressure. A macroscopic model, applicable to an axially uniform two-temperature plasma, yields the radial distributions of the parameters. The axial distributions are determined from relations based on the approximation of local axial uniformity, which is valid provided the attenuation of the surface wave is weak. In the approximation adopted, the plasma parameters in any cross section of the discharge depend under given conditions solely on L, the power lost per unit length of the plasma column. Calculations made for a 2.45 GHz discharge in a capillary tube show an almost linear decrease of L and of the electron density towards the end of the discharge. The electron temperature for nitrogen is lower than for argon under similar discharge conditions. In nitrogen the electron density is about one order of magnitude lower and the gas temperature is much closer to the electron temperature than for argon. This may be explained by the presence of vibrational and rotational excitation and relaxation channels, which facilitate redistribution of the energy deposited in the discharge.*

**Keywords:** Surface wave; Microwave discharge; Plasma

# 1 Introduction

Microwave discharges have been finding ever wider use as plasma sources for diverse applications [1, 2]. When sustained at atmospheric pressure, such discharges are convenient and efficient sources of active particles for the plasma chemistry, elemental analysis, laser excitation or lighting.

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†This work was supported by the Institute of Fluid-Flow Machinery, Polish Academy of Sciences (IMP O3/Z1/T1)

In the last decades discharges sustained by the electromagnetic field of a wave propagating along the plasma, called traveling wave discharges (TWD) [3], have been developed and intensively investigated. TWDs that have been most widely studied and utilized are those sustained by surface waves. For low pressure surface wave discharges (SWD) the modelling is far advanced and the state of the art well documented [4]–[9]. Such is not the case for discharges sustained by surface waves at atmospheric pressure, although many experimental investigations as well as applications have been reported, see e.g. [10]–[13]. Both the radial and axial structures of discharge plasma components and their temperature are of interest when modelling such discharges. A systematic and complete work on the modelling of discharges of this kind is still to be done. A paper by Nowakowska *et al.* [14], concerning the SWD in argon, was a step in that direction.

The present paper concerns the SWD in nitrogen. A self-consistent model would require simultaneous solving of the Maxwell equations and the equations describing the discharge processes. A commonly used simplifying approximation is to assume that the influence of any axial gradient (in the $z$ direction) on the discharge maintenance processes and wave propagation is negligible. This is referred to as an *approximation of local axial uniformity* [5]. It is applicable if the condition $2\alpha(L)a \ll 1$ is satisfied, where $\alpha$ is the wave attenuation coefficient and $a$ is the discharge tube inner radius. It is customary to assume that the plasma column ends where $\alpha = \beta = 2\pi/\lambda_z$ (cf. [15]), $\beta$ being the phase coefficient and $\lambda_z$ – the wavelength. Therefore, at the end of the column $4\pi a/\lambda_z \ll 1$ must be fulfilled.

The assumption of weak attenuation of the wave (and, thus, of local axial uniformity) allows the analysis of the discharge to be performed in two separate stages. The first stage starts with energy removal from the wave, via plasma, to the wall. For any axial position the solution to the problem is the same as that for an axially uniform discharge. It gives the radial distributions of all the plasma parameters of interest, including its electromagnetic properties, for given discharge conditions (the gas composition and pressure, the wave mode and frequency, the tube transverse dimensions and the wall material) and given values of $L$. Consequently, the solution also yields $\alpha(L)$, the attenuation characteristic of the wave.

The second stage of the power balance analysis is directly connected with the specific nature of TWDs and concerns the gradual transfer of power from the wave field to plasma as the wave propagates along the column. It relates the local maintenance processes occurring in consecutive elementary slabs of the discharge to the overall power balance in the plasma column and provides the required axial distribution of the power loss $L(z)$ for given discharge conditions and for a given amount $P_0$ of the total power delivered by the wave launcher to plasma.

The distribution $L(z)$ of the power loss along the discharge may be found [5] from

$$\frac{1}{L(z)}\frac{\mathrm{d}L(z)}{\mathrm{d}z} = -2\alpha(L)\left[1-\frac{L(z)}{\alpha(L)}\frac{\mathrm{d}\alpha(L)}{\mathrm{d}L}\right]^{-1}, \tag{1}$$

with the initial condition $L(0) = 2\alpha(L_0)P_0$. This result, merged with the radial distribution of plasma parameters for a given $L$ as obtained from the analysis of an axially uniform discharge, provides full information about the spatial structure of the SWD.

## 2 Model of an axially uniform discharge in nitrogen at atmospheric pressure

At atmospheric pressure a macroscopic treatment is appropriate. We adopt a two-temperature model of the microwave discharge plasma [16, 17]. Maxwellian energy distributions characterized by temperatures $T_\mathrm{e}$ and $T$ are assumed for electrons and heavy particles (molecules, atoms and ions), respectively. It is further assumed that the electronic excited states have Boltzmann distributions characterized by $T_\mathrm{e}$. We also assume that owing to the strong coupling between the rotational and translational states of $N_2$ molecules the rotational temperature is equal to the gas temperature $T$. Similarly, the vibrational temperature is assumed to be equal to $T$ at low number densities of electrons in the discharge. Validity of that assumption was demonstrated in [18].

For discharges in capillary tubes the skin effect may be neglected in most cases and the rms value of the electric field intensity may be regarded as constant in the radial direction, $E(r) = E$. In this approximation the power deposited in the plasma per unit length of the column is $L = 2\pi\int_0^a \sigma E^2 r\mathrm{d}r \cong 2\pi E^2 \int_0^a \sigma r\mathrm{d}r$, where $\sigma$ is the electric conductivity. Neglecting energy losses due to radiation and diffusion we write the energy conservation equations for the electrons and heavy particles per unit volume, respectively, as (Section 7.8 in [16])

$$\frac{1}{r}\frac{\mathrm{d}}{\mathrm{d}r}\left(\lambda_\mathrm{e} r\frac{\mathrm{d}T_\mathrm{e}}{\mathrm{d}r}\right) + \sigma E^2 - \frac{3}{2}\delta_\mathrm{ef}\nu_\mathrm{ef}n_\mathrm{e}k\left(T_\mathrm{e}-T\right) = 0, \tag{2}$$

$$\frac{1}{r}\frac{\mathrm{d}}{\mathrm{d}r}\left[(\lambda+\lambda_\mathrm{D})r\frac{\mathrm{d}T}{\mathrm{d}r}\right] + \frac{3}{2}\delta_\mathrm{ef}\nu_\mathrm{ef}n_\mathrm{e}k\left(T_\mathrm{e}-T\right) = 0. \tag{3}$$

Here $\delta_\mathrm{ef}$ is the fraction of electron energy lost in an electron collision with a heavy particle, $\nu_\mathrm{ef}$ is the effective frequency of electron elastic collisions, $n_\mathrm{e}$ is the electron number density and $k$ is the Boltzmann constant. The coefficients $\lambda$ and $\lambda_\mathrm{e}$ designate the thermal conductivities for the molecules and electrons, respectively, and $\lambda_{\mathrm{D}}$ is a contribution to the thermal conductivity due to dissociation of N molecules.

Validity of those assumptions of a negligible influence of the skin effect, rad ation and diffusion has been checked *ex post* after having made calculations fo specific discharge conditions.

Both $\lambda$ and $\lambda_{\mathrm{D}}$ are functions of $T$. Their values were taken from [19]. Valu of the coefficient $\delta_{\mathrm{ef}}$, calculated with both elastic and inelastic collisions take into account, are used after [20]. Coefficient $\lambda_{\mathrm{e}}$ is calculated according to th formula (Section 7.4 in [16])

$$\lambda_{\mathrm{e}}(T_{\mathrm{e}}, T) = 2K_{\lambda}\,(k/e)^2\,\sigma(T_{\mathrm{e}}, T)\,T_{\mathrm{e}},$$

where the electrical conductivity is

$$\sigma(T_{\mathrm{e}}, T) = K_{\sigma}\frac{e^2 n_{\mathrm{e}}\nu_{\mathrm{ef}}}{m_{\mathrm{e}}(\omega^2 + \nu_{\mathrm{ef}}^2)}.$$

Here $e$ is the elementary charge, $m_{\mathrm{e}}$ is the electron mass and $\omega$ is the angu frequency of the electromagnetic field. Values of $K_{\lambda}$ and $K_{\sigma}$ were derived fro the literature [19, 21]. Their values were adjusted so that the conductivit calculated from (4) and (5) at $T_{\mathrm{e}} = T$ and $\omega = 0$ were equal to those give in [19] for a local thermodynamic equilibrium DC case. The effective collis frequency is calculated as $\nu_{\mathrm{ef}} = \overline{v}_{\mathrm{e}}(n_{\mathrm{e}}Q_{\mathrm{ei}} + n_{\mathrm{a}}Q_{\mathrm{ea}} + n_{\mathrm{m}}Q_{\mathrm{em}})$, where $Q$ deno the collision cross sections and $n$ are the number densities, the subscripts e, i and m refer to electrons, ions, atoms and molecules, respectively, and $\overline{v}_{\mathrm{e}}$ is th average thermal electron velocity $\overline{v}_{\mathrm{e}} = (8kT_{\mathrm{e}}/\pi m_{\mathrm{e}})^{1/2}$. The cross sections and $Q_{\mathrm{em}}$ were taken from [19], and $Q_{\mathrm{ei}} = 3.04\cdot 10^{-10}T_{\mathrm{e}}^{-2}\ln(2.8\cdot 10^{9}T_{\mathrm{e}}\,n_{\mathrm{e}}^{-1/2})$ wa adopted after [21]. The number densities of electrons, atoms, molecules and ion are determined based on the Dalton law $p = (n_{\mathrm{m}} + n_{\mathrm{a}} + n_{\mathrm{im}} + n_{\mathrm{ia}})kT + n_{\mathrm{e}}$ where $p$ denotes the pressure, the condition of quasi-neutrality $n_{\mathrm{e}} \cong n_{\mathrm{im}} +$ and the Saha equation for singly ionized molecules and atoms, respectively

$$\frac{n_{\mathrm{e}}n_{\mathrm{im}}}{n_{\mathrm{m}}} = \frac{2Z_{\mathrm{im}}}{Z_{\mathrm{m}}}\left(\frac{2\pi m_{\mathrm{e}}kT_{\mathrm{e}}}{h^2}\right)^{3/2}\exp\left(-\frac{U_{\mathrm{im}}}{kT_{\mathrm{e}}}\right),$$

$$\frac{n_{\mathrm{e}}n_{\mathrm{ia}}}{n_{\mathrm{a}}} = \frac{2Z_{\mathrm{ia}}}{Z_{\mathrm{a}}}\left(\frac{2\pi m_{\mathrm{e}}kT_{\mathrm{e}}}{h^2}\right)^{3/2}\exp\left(-\frac{U_{\mathrm{ia}}}{kT_{\mathrm{e}}}\right),$$

where $U_{\mathrm{im}}$ and $U_{\mathrm{ia}}$ are the respective ionization energies. Values of the parti functions $Z$ are calculated based on [21]. The degree of dissociation is calcula after [21] according to the formula

$$\frac{n_{\mathrm{a}}^2}{n_{\mathrm{m}}} = A(T)\,T^{-1/2}\exp\left(-\frac{U_{\mathrm{D}}}{kT}\right),$$

where $U_D$ denotes the dissociation energy. This completes the set of equations to be solved.

The two-temperature model used for the calculations represents an improvement over those assuming for a microwave discharge plasma a local thermal equilibrium as e.g. in [16, 17]. Use of the Saha equation, despite its shortcomings (see a discussion in [22]), seems to be at present the most convenient approach to the modelling of high pressure discharge plasmas.

The following boundary conditions are adopted: Owing to the axial symmetry there is at the tube axis ($r=0$)

$$\frac{\mathrm{d}T}{\mathrm{d}r}=0 \quad \text{and} \quad \frac{\mathrm{d}T_\mathrm{e}}{\mathrm{d}r}=0. \tag{9}$$

At the inner surface of the wall ($r=a$) the heavy particles lose their energy to the wall, therefore

$$\lambda\frac{\mathrm{d}T}{\mathrm{d}r}=\lambda_\mathrm{w}\frac{\mathrm{d}T_\mathrm{w}}{\mathrm{d}r}, \tag{10}$$

where $\lambda_\mathrm{w}$ and $T_\mathrm{w}$ are the thermal conductivity of the wall material and the temperature of the wall inner surface, respectively. The latter is related to the temperature $T_\infty$ of the outer surface of the tube wall by the expression

$$T_\mathrm{w}=T_\infty+\frac{L}{2\pi\lambda_\mathrm{w}}\ln\frac{b}{a}, \tag{11}$$

where $b$ is the outer diameter of the discharge tube. For the electron gas the wall surface at $r=a$ may be treated as an adiabatic envelope, as it can be assumed that electrons are reflected by the electric field within the sheath and do not reach the wall. Therefore no energy is transferred from the electron gas to the wall and we can set

$$\lambda_\mathrm{e}\frac{\mathrm{d}T_\mathrm{e}}{\mathrm{d}r}=0 \quad \text{at } r=a. \tag{12}$$

Equations (2), (3) are converted into four differential equations of the first order and solved, for a range of values of $L$ and given $\omega$, $a$, $b$, $\lambda_\mathrm{w}$, $p$ and $T_\infty$, using the relaxation method [23]. This yields for each $L$ radial distributions $T(r)$ and $T_\mathrm{e}(r)$, from which $n_\mathrm{e}(r)$ and $\nu_\mathrm{ef}(r)$ may be derived. These determine the electromagnetic characteristics of the plasma column.

## 3 Calculation of the wave propagation characteristics and the axial distribution of the power loss in the plasma

To calculate the wave propagation coefficient $\gamma=\alpha+j\beta$ as a function of $L$ we consider a cylindrical column of homogeneous plasma, characterized by the electric permittivity which is equal to the average over the column cross-section of t radially variable permittivity calculated for given $L$. Condition $n_e(r_p) = 0.1\, n_e$ defines the plasma radius $r_p$. We assume that along the column, which is in ge eral surrounded by three coaxial layers of dielectric (gas, tube wall materi gas), an azimuthally symmetric surface wave propagates. For determinatio the wave propagation characteristics, the *cold plasma linear dielectric appr mation* [20] is used. The local values of the complex relative permittivity $\varepsilon_p$ the plasma are calculated in a way similar to that shown in [16]. The real pa of the relative electric permittivity is given by

$$\mathrm{Re}(\varepsilon_p) = 1 - \frac{\omega_p^2(r)}{\omega^2 + \nu^2(r)},$$

where $\omega_p(r) = [n_e(r)e^2/(\varepsilon_0 m_e)]^{1/2}$ is the plasma frequency, $\varepsilon_0$ being the pe mittivity of vacuum. The imaginary part of the relative electric permittivi obtained from

$$\mathrm{Im}(\varepsilon_p) = -\frac{\sigma(r)}{\varepsilon_0 \omega}.$$

The cross-section average value of the complex relative permittivity is calcula as

$$\bar{\varepsilon}_p = \frac{1}{\pi r_p^2} \int_0^a 2\pi \varepsilon_p(r) r \mathrm{d}r.$$

To derive the dispersion equation, from which the propagation coefficie may be calculated, wave equations are written in the cylindrical coordinates the axial component of the electric field in the plasma column and the lay surrounding it

$$\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial E_z}{\partial r}\right) + \frac{1}{r^2}\frac{\partial^2 E_z}{\partial \phi^2} + (\gamma^2 + k_0^2 \varepsilon_i) E_z = 0,$$

where $k_0 = 2\pi/\lambda_0$ is the free-space phase coefficient of the wave and $\varepsilon_i$ de the relative electric permittivity of the i-th medium, $i$ =1 .. 4 for plasma, g dielectric tube and gas, respectively. For an azimuthally symmetric surface wa which propagates in a TM mode, this equation yields

$$E_{z,i} = [A_i I_0(\gamma_i r) + B_i K_0(\gamma_i r)] \exp(-\gamma z),$$

where $I_0$ and $K_0$ are the modified Bessel functions of the first and second ki respectively, of order zero; $\gamma_i^2 \equiv -(\gamma^2 + k_0^2 \varepsilon_i)$; $A_i$ and $B_i$ are coefficients to calculated from the boundary conditions.

The remaining field components may be obtained in terms of $E_{z,i}$ from the Maxwell equations

$$E_{r,i} = \frac{\gamma}{\gamma_i^2}\frac{\partial E_{z,i}}{\partial r} \tag{18}$$

and

$$H_{\phi,i} = \frac{j\omega\varepsilon_i}{\gamma_i^2}\frac{\partial E_{z,i}}{\partial r}. \tag{19}$$

The usual boundary conditions for the electromagnetic field require that $E_{z,i}$ and $H_{\phi,i}$ be continuous at all three media interfaces. This leads to a set of six homogeneous equations, which has a single non-zero solution provided that its determinant is equal to zero. That condition yields the dispersion equation, which solved for consecutive values of $L$, provides the functional dependence $\alpha(L)$.

The next step is to substitute $\alpha(L)$ into (1) and solve that equation for the axial distribution $L(z)$ of the power loss per unit length. Finally, complete spatial distributions of plasma parameters are determined by merging $L(z)$ with the radial distributions calculated for given $L$.

# 4 Numerical results

The method described in this paper has been applied to determination of plasma parameters in a surface wave discharge sustained at a frequency $f$ = 2.45 GHz and pressure $p = 1$ atm in nitrogen flowing through an alumina tube ($\lambda_{\mathrm{w}} = 30$ W/mK) with the inner diameter $2a = 4$ mm and the outer diameter $2b = 6$ mm. The temperature outside the tube was set to $T_\infty = 350$ K. The calculations were made for the values of $L$ between 1.75 kW/m and 15 kW/m. The lower limit was imposed by the condition $\alpha = \beta$ defining the end of the SWD. The upper limit corresponded to that value of $L$ above which the assumption of the skin effect having been negligible became invalid. Selected results of these calculations have already been presented in a review paper [24].

Figures 1 and 2 show radial distributions of $T_{\mathrm{e}}$, $T$ and $n_{\mathrm{e}}$ in the nitrogen discharge plasma. We compare them with the results of similar calculations for the SWD maintained in argon at the same frequency and pressure, but in a slightly narrower tube [25]. While in argon for $L = 1$ kW/m $T_{\mathrm{e}}$ was equal on the axis to 10000 K and its radial profile was almost flat, in nitrogen, with $L = 1.75$ kW/m, it drops from only 7000 K on the axis to 2000 K at the wall (Fig. 1). The difference in $n_{\mathrm{e}}$ is even larger. For nitrogen the electron density on the axis is about one order of magnitude lower than for argon and drops to negligible values at $r/a \cong 0.7$. For a discharge in argon $n_{\mathrm{e}}$ was at $r = a$ still equal to about 8.5% of its value on the axis. For the SWD in nitrogen the difference

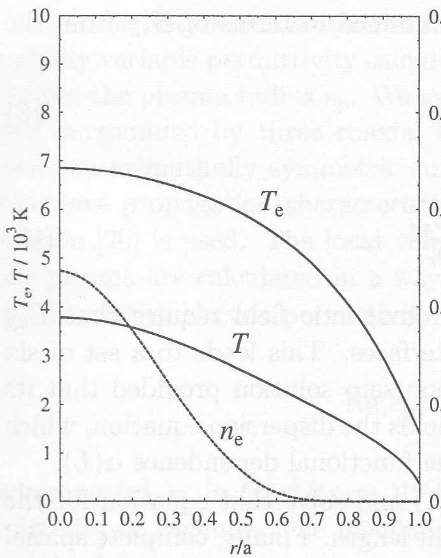


Figure 1. Radial distributions of $T_e, T$ and $n_e$ for $L = 1.75$ kW/m.

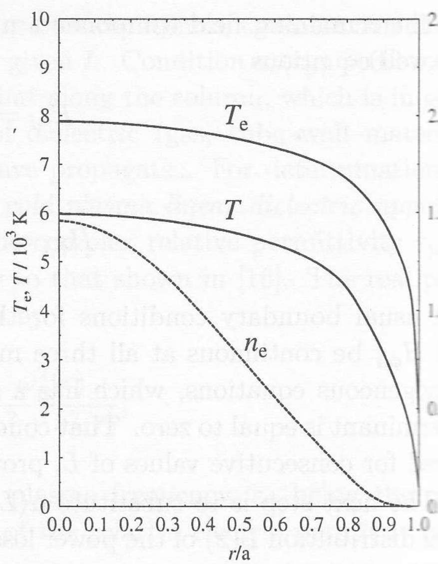


Figure 2. Radial distributions of $T_e, T$ and for $L = 15$ kW/m.

between $T_e$ and $T$ is smaller than that for argon. While for argon $T/T_e \cong$ 0. on the axis, for nitrogen this ratio exceeds 0.5.

About eightfold increase of $L$ results (Fig. 2) in a rise of $n_e$ of almost the same order, a slight increase of $T_e$ and a substantial rise of $T$, leading to $T/$ ratio on the axis equal to about 0.75. Thus, as expected, with an increase the local power loss the discharge plasma in nitrogen comes closer to the local thermodynamic equilibrium. The calculation results obtained for nitrogen are a qualitatively good agreement with results of calculations presented in [16] a microwave discharge in air. The calculated wave propagation characteristics $\alpha(L)$ and $\beta(L)$ are shown in Fig. 3. The axial variation of the power lost the plasma per unit length, $L(z)$, inferred from $\alpha(L)$, is shown in Fig. 4. The decrease of $L$ with decreasing distance to the end of the discharge is almost linear. Note that the length $l$ of the plasma column is determined by $P_0$, the total power delivered by the wave launcher to the plasma.

The variation of the plasma radius $r_p$ with $L$ is shown in Fig. 5. As it is seen the discharge does not fill the entire cross section of the tube even for large $L$.

Figure 6 shows the plots against $z$ of the electron density $n_{e,ax}$ on the axis and the average electron density $n_{e,av}$ calculated for the plasma column of radius $r_p$. Both those plots are almost linear. As the plasma radius also changes along the column, this result together with the linear change of $L$ with $z$ shows that the power needed to produce an electron in the discharge is not necessarily const

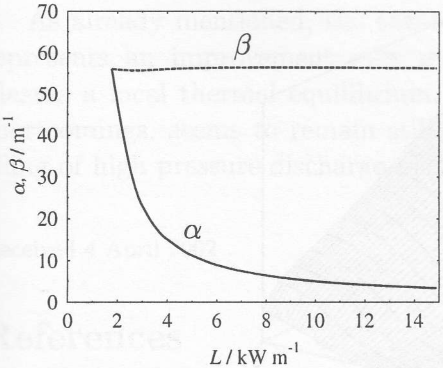


Figure 3. Calculated phase and attenuation coefficients for the wave.

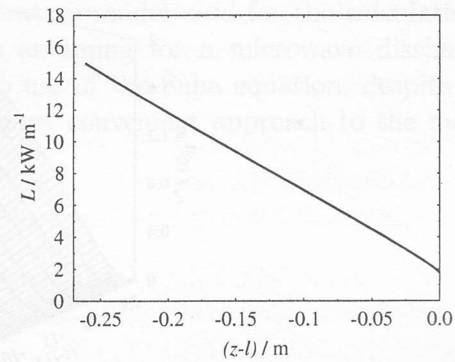


Figure 4. Variation of $L$ along the plasma column.

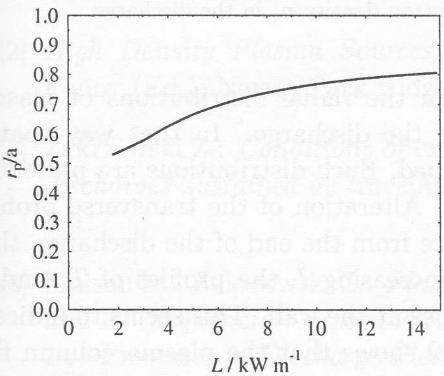


Figure 5. Variation of the plasma radius $r_p$ with $L$.

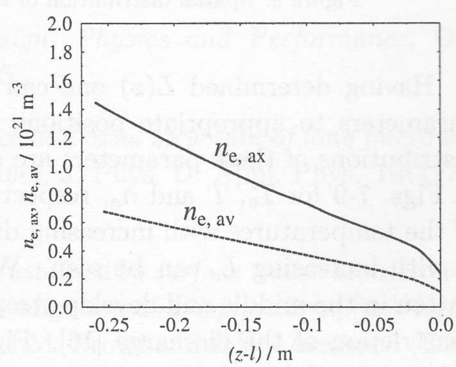


Figure 6. Variation of the electron density on the axis $n_{e,ax}$ and the average electron density $n_{e,av}$ along the plasma column.

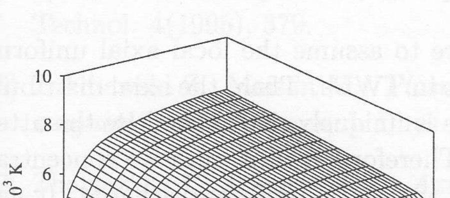


Figure 7. Spatial distribution of the electron temperature $T_e$ in the discharge.

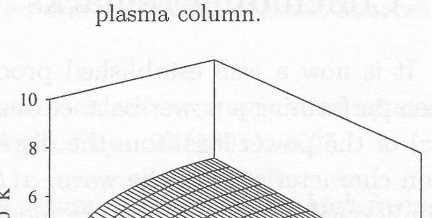


Figure 8. Spatial distribution of the gas temperature $T$ in the discharge.

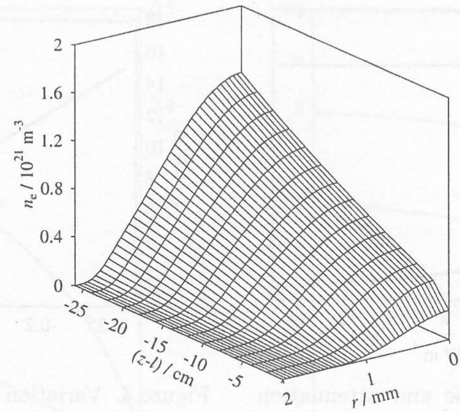


Figure 9. Spatial distribution of the electron density $n_e$ in the discharge.

Having determined $L(z)$ one can assign the radial distributions of pl
parameters to appropriate positions along the discharge. In that way sp
distributions of these parameters are obtained. Such distributions are pres
in Figs. 7-9 for $T_e$, $T$ and $n_e$, respectively. Alteration of the transverse pro
of the temperatures with increasing distance from the end of the discharge,
is with increasing $L$, can be seen. With increasing $L$ the profiles of $T_e$ an
flatten in the middle and develop steep slopes at the wall. This seems to ind
constriction of the discharge [16]. Figure 9 shows that the plasma column
only a part of the tube cross section even at the wave launcher, where $L$ is la

# 5 Concluding remarks

It is now a well established procedure to assume the local axial unifo
when performing a power balance analysis in TWDs. Then, the axial distri
$L(z)$ of the power loss from the discharge is uniquely determined by the at
ation characteristics of the wave, $\alpha(L)$. Therefore, in this work we concen
mainly on the two-temperature model of an axially uniform discharge. It
the radial distribution of the plasma parameters and the attenuation chara
istics of the wave. The latter provides $L(z)$ which, merged with the former,
the expected result of our modelling: the full spatial distribution of the pl
parameters of interest.

As expected, the transverse discharge structure at any position $z$ de
clearly on the local value of $L(z)$. However, $L$ affects various plasma para
differently. The dependence on L is strong for the charged particle densi
the gas temperature, and weaker for the electron temperature. The plasma
saturates with increasing $L$.

As already mentioned, the two-temperature model used for the calculations represents an improvement over models assuming for a microwave discharge plasma a local thermal equilibrium. Also use of the Saha equation, despite its shortcomings, seems to remain still the most convenient approach to the modelling of high pressure discharge plasmas.

Received 4 April 2002

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