Influence of antenna dielectric cover on the parameters of the electrode microwave discharge

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Abstract. One possibility of reducing the influence of contamination of the plasma volume, when initiating the microwave discharges using antennas, is to cover the antenna with a dielectric. The results of 1-D and 2-D modeling showing the impact of such dielectric cover on the parameters of a non-equilibrium plasma of an electrode microwave discharge in hydrogen at reduced pressure (the change of the electrodynamics of the discharge, charge deposition on the dielectric surface, changing the catalytic properties of the surface of the antenna) are presented in the article.

1. Introduction

One of the advantages of microwave discharges is the absence of electrodes in the discharge chamber that can lead to contamination of the plasma by the products of erosion. Because of that reason it was the electrodeless microwave systems like the waveguide, resonator and surface wave, etc systems [1] that have been widely used for plasma generation until recently. However there are conditions, when it is necessary to introduce the electrode for microwave plasma generation. This applies primarily to the initiated discharges when the microwave field from an external source is insufficient to initiate and maintain the discharge; creation of a discharge requires additional electrodes where an increase in the local field values exists [2]. In addition, there is a whole class of microwave devices where the plasma is initiated by electromagnetic vibrators of different types [3-6]. Such devices are used to obtain low pressure [7-9] and atmospheric pressure [4-6] plasma. These include widespread coaxial microwave plasma torch [10-12]. The microwave antenna systems are also applied for generating plasma inside liquids [13-16].

In these cases, there is the problem of contamination of the volume by the products of erosion of the metal electrodes. It seems that the possibility of contamination of the plasma with electrode material in microwave devices is exaggerated at least at low power. In [17] in the electrode microwave discharge in conditions typical for diamond deposition (a mixture of hydrogen with a small addition of methane, at pressure of about 15 Torr, the substrate, located at a distance of 1.5 cm from the electrode, is heated up to ~1000^{o}C) the ESCA and Auger spectroscopy methods have not revealed even trace quantities of the electrode material in the deposited material. This is due to the fact that in the microwave plasma the electrode surface is not probed by high energy ions. In RF discharge, for example, such ions appear as a result of their acceleration in large DC fields that exist in the electric double layer at the electrode due to the rectification of the applied RF voltage (~100-300V) on the nonlinearity of double layer. In the microwave plasma the electron concentration is high enough ($\geq 10^{11}$ cm^{-3}) and thickness of a near-electrode layer is small. Under these conditions, the layer impedance is capacitive, the rectification of the microwave signal, though exists, but is not a factor leading to the occurrence of high energy ions. Therefore the microwave discharge, as a rule, is the discharge with predominant volume ionization (α-form level) and the secondary ionization (γ-processes) can be neglected. However in high energy regimes, when the temperature of the electrode becomes large, melting and even explosion of metallic electrodes are possible [8, 9], which leads to the mass transfer of the material of electrodes into a gas media. Thermal erosion of the electrode also occurs in the microwave discharges in liquids [18, 19].

One method of eliminating the influence of electrode material is coating the electrode with a heat-resistant dielectric, as it was done, e.g., in [19]. In this regard the problem of influence of such coatings on the plasma parameters arises. This influence may be related to: the deposition of charges on the dielectric surface in the plasma, changing of the electrodynamic properties of the discharge system and also change of the catalytic properties of the surface (changing of the metal to dielectric) and an associated change of the surface impact on concentration of heavy plasma components. All of these factors are analyzed in the present work, based on modeling of the electrode microwave discharge in hydrogen at pressure of 1 Torr.

2. The model

We used one- and two-dimensional models for analyzing the effect of dielectric coating at the antenna on properties of the microwave plasma.

In the present paper a two-dimensional model is applied for studying steady-state microwave field which is set inside the empty discharge chamber with the inner electrode covered with dielectric. The discharge chamber is described in detail in [7-9]. The discharge chamber is a stainless steel cylinder with a diameter of 15 cm. An electromagnetic wave with a frequency of 2.45 GHz is fed into the discharge chamber through a waveguide-coaxial adapter and is introduced into the chamber along the electrode-antenna (a copper cylindrical electrode with a diameter of 5mm). Full self-consistent two-dimensional model for the description of the discharge in hydrogen is described by the authors [20, 21] in detail. It includes the Maxwell equations, balance equations for the plasma particles and the Poisson equation, taking into account the field of charge separation.

Dielectrics with ε = 1, 2, 3, 4 and thicknesses d = 1, 2, 3 mm have been used for coating of the inner electrode.

A one-dimensional self-consistent spherically symmetric model includes the equation for the electromagnetic field in the quasi-static approximation. The *rms* electric component of the microwave field in the electrode system with spherical symmetry coincides with the field of a spherical capacitor. The inner electrode of such a capacitor is covered with a dielectric layer ε and the remaining discharge region is filled with plasma of dielectric permeability $\varepsilon_{pl} = \left\{(1 - n_e/n_{norm})^2 + \left(\frac{\nu}{\omega}\frac{n_e}{n_{norm}}\right)^2\right\}^{1/2}$, where $n_{norm} = m(\omega^2 + \nu^2)/4\pi e^2$, ω is a circular frequency of the microwave signal, ν is the frequency of collisions of electrons with neutral heavy particles and n_e is the electron concentration. The microwave field in the capacitor is determined from the Poisson equation. The field amplitude at the internal electrode boundary is set.

The model also includes: the Boltzmann equation for the electron energy distribution function (EEDF), recorded in the two-term approximation of the expansion of the EEDF into spherical harmonics; non-stationary balance equations for the concentrations of charged and neutral particles, and the equation for the field calculation of charge separation. The solution of the Boltzmann equation considering both the microwave and DC fields showed that the influence of the DC field on the EEDF was negligible. Therefore, when calculating the EEDF only the microwave field has been taken into account. The effect of the vibrationally excited hydrogen molecules on the EEDF and rates of different plasma chemical processes was taken into account by using the vibrational distribution function of hydrogen molecules obtained in the diffusion approximation described in [22, 23]. All processes in hydrogen plasma which are used in the model are described in [20, 21] in detail.

Boundary conditions for charged plasma particles, described by the balanced equations, are set by the expression for fluxes to the surface of the insulator covering the inner electrode and to the outer wall of the discharge camera:

$$\boldsymbol{\Gamma_e}\cdot\boldsymbol{n} = -\frac{1}{4}n_e\upsilon_{T_e} - (a-1)\cdot\mu_e n_e\,\mathbf{E}\cdot\mathbf{n}\cdot$$

$$-\boldsymbol{\Gamma}_{\mathbf{p}}^{\mathbf{i}}\cdot\boldsymbol{n} = -\frac{1}{4}n_p^i\upsilon_{T_p}^i - a\cdot\mu_p^i n_p^i\,\mathbf{E}\cdot\mathbf{n}\cdot$$

Flows for electrons and ions of the i-th type to the wall consist of two components: diffusion and drift. Here υ_{Te} and υ^i_{Tp} are thermal velocities of electrons and ions on the wall, $\boldsymbol{n}$ is a unit vector normal to the electrode surface (normal vector). The corresponding electron temperature at the wall was computed using the Boltzmann equation, and the temperature of ions on the walls was taken equal to the gas temperature. The coefficient $a = 1$, if the DC field vector $\boldsymbol{E}$ is directed towards the wall, i.e. $\boldsymbol{E}\cdot\boldsymbol{n} > 0$, and $a = 0$, if $\boldsymbol{E}\cdot\boldsymbol{n} < 0$.

The equation for the accumulation of surface charge σ, deposited from the discharge, is solved at the dielectric-plasma boundary: $\frac{\partial\sigma}{\partial t} = -e\left(\sum_i \boldsymbol{\Gamma}_{\mathbf{p}}^{\mathbf{i}} - \boldsymbol{\Gamma}_{\mathbf{e}}\right)$. The boundary conditions for the potential φ, are the following: on the surface of the dielectric $\varepsilon_0\varepsilon\nabla\varphi = \sigma$; the outer wall of the working chamber is grounded.

The concentration of neutral excited particles of plasma on the wall was supposed to be zero. The flux of unexcited hydrogen atoms incident from plasma to the wall is equal to the heat flux (diffusion) multiplied by the probability of their losses due to recombination γ_{rec}: $-\boldsymbol{\Gamma_n}\cdot\boldsymbol{n} = -\gamma_{rec}\frac{1}{4}n_e\upsilon_{T_e}$.

The probability of the recombination loss for hydrogen atoms on the copper electrode was taken 0.05. According to [24] this value lies in the range 0.02-0.1. The probability of the recombination loss for hydrogen atoms on the dielectric surface covering the electrode (to be specific quartz was chosen as an insulator), was 0.01 [25]. The probability of the recombination loss of the hydrogen atoms on the chamber walls of stainless steel was taken equal to 0.1 [26].

The one- and two-dimensional simulations have been carried out numerically, using finite element methods implemented in the commercial package Comsol 3.5a [27] on 12-core 2.3GHz Xeon of 64 bit system with 32 GB of RAM.

The calculations were carried out at hydrogen pressure of 1 Torr and gas temperature of 600 K. In accordance with experimental results [28] the temperature was considered constant

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throughout the volume. In the following we present the stationary distributions of plasma parameters.

3. Results and discussions

3.1. . The change of the electrodynamic properties of the discharge system.

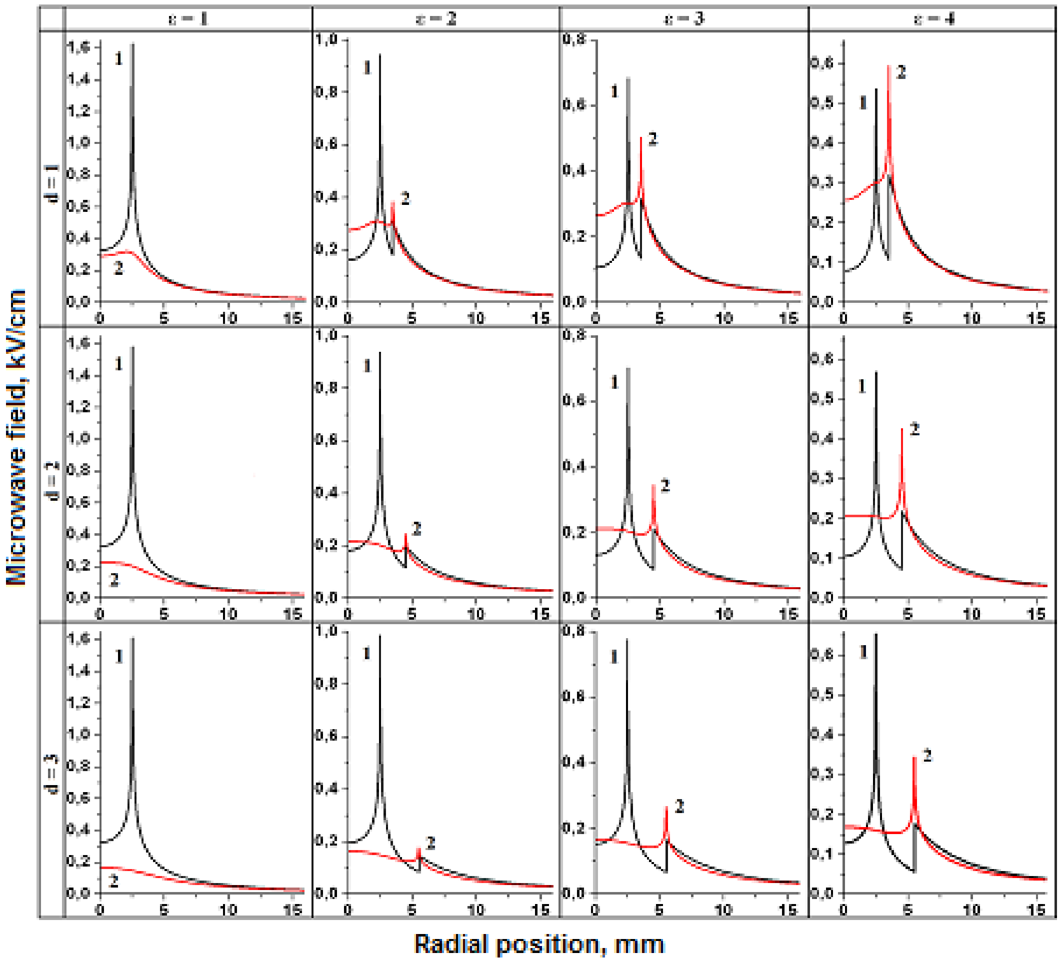

Figure 1. Radial distributions of microwave field at distance 10 microns below the metal electrode (black curves - 1) and 10 microns below the dielectric cover (red curves - 2) for different thickness of the cover and different dielectric constants (results of 2-D modeling).

The microwave energy is delivered into the discharge chamber along the antenna with the discharge ignited at its tip. A dielectric cover should lead to a change in the distribution of electromagnetic fields around the antenna. This problem has been studied in [19] by modeling of creating a microwave plasma in water. The antenna, coated with aluminum oxide, served as a central conductor of the coaxial line with a teflon insulator. The end of antenna is surrounded by water. It is shown that at the triple point, where the aluminum oxide, teflon and water are in contact, there has been a sharp increase in the intensity of the microwave field, which may lead to a discharge ignition. Note that in this case the discharge exists near the point of introduction of the antenna into the chamber. The problem on finding a place where the discharge is sustained at the body of antenna is investigated in [29] and it is shown that the in the media with high tangent loss the discharge always burns at the entrance of the antenna into the discharge chamber. In the media with low losses the discharge always burns at the end of the antenna.

In this paper we have studied the coaxial electrodynamic system (Fig. 1), where the discharge was excited by means of the antenna in hydrogen at a pressure of 1 Torr. Our previous experiments and calculations (see reviews [7-9]) have shown that the discharge at low power is sustained at the end of the antenna. Since plasma absorbs the microwave energy and represents a region with large losses, the plasma area shifts to the place of insertion of the antenna into the chamber at higher power. In this section we present a distribution of the microwave field in the discharge chamber without plasma (ε =1) with a cylindrical antenna covered by a lossless dielectric of different thickness and dielectric permeability. Radial distribution of the electric field at the border of the antenna and dielectric is shown in Fig.1.

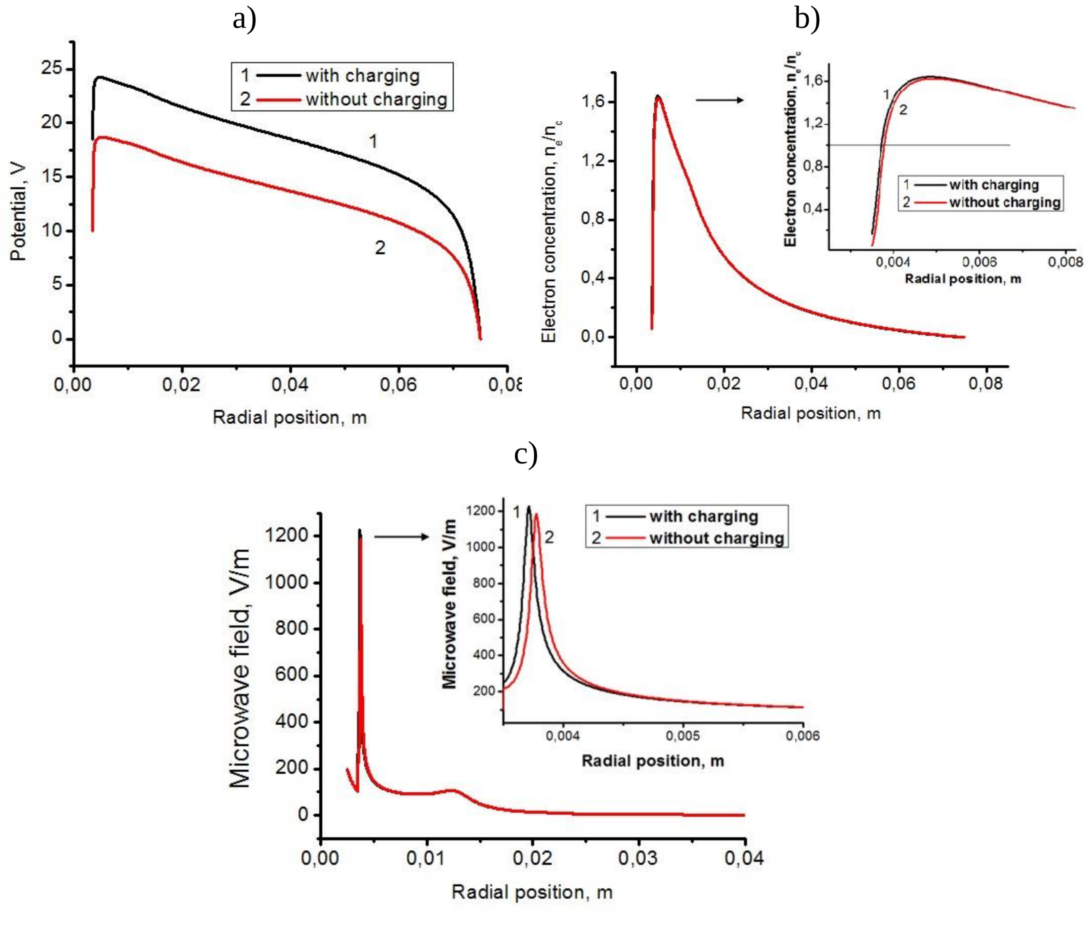

Figure 2. Radial distributions of DC potential (a), electron density (b) and microwave field (c) with 1 (black curves) and without 2 (red curves) charge deposition on quartz cover of thickness of 1 mm (results of 1-D modeling).

As it should be, on the boundary of the dielectric and gas there is a field discontinuity, the microwave field experiences a jump in accordance with the difference of dielectric permeability. It is seen also that the presence of an insulator with appropriate ε and thickness allows to obtain more uniform distribution of the field in the region under the dielectric, than without it. There is also a certain increase of the field at the edge on the end face of the dielectric.

3.2. *The deposition of charges on the dielectric surface in the plasma*

Let us consider in the framework of the one-dimensional model, the impact of deposition of charges on the dielectric-plasma interface on the plasma parameters. Since the dielectric is ideal (lossless), the influence of charging of its surface is considered only for one thickness of the dielectric (1 mm). The results of the calculations are shown in Fig.2.

Charging leads to an increase in surface potential and the potential difference between the electrode and plasma, though the profile of spatial potential distribution changes slightly (see Fig.2a).

The maximum value of the electron density is modified slightly, but the position of its maximum slightly shifts to the dielectric surface (Fig.2b). The changes are noticeable only near the surface, which occurs due to changes in the potential of the surface when it is charging. The change in the distribution of electron density near the surface leads to the fact that the region of plasma resonance approaches the surface and the magnitude of the resonance field slightly increases (Fig.2c). The position of the plasma resonance, $n_e / n_c = 1$ ($n_c \sim 7 \times 10^{11}$ cm^{-3}) lie on a straight line, marked in Fig.2b parallel to the x-axis.

Thus the accumulation of the charge on the surface of the electrode covered with dielectric has little effect on the plasma parameters in the volume of discharge.

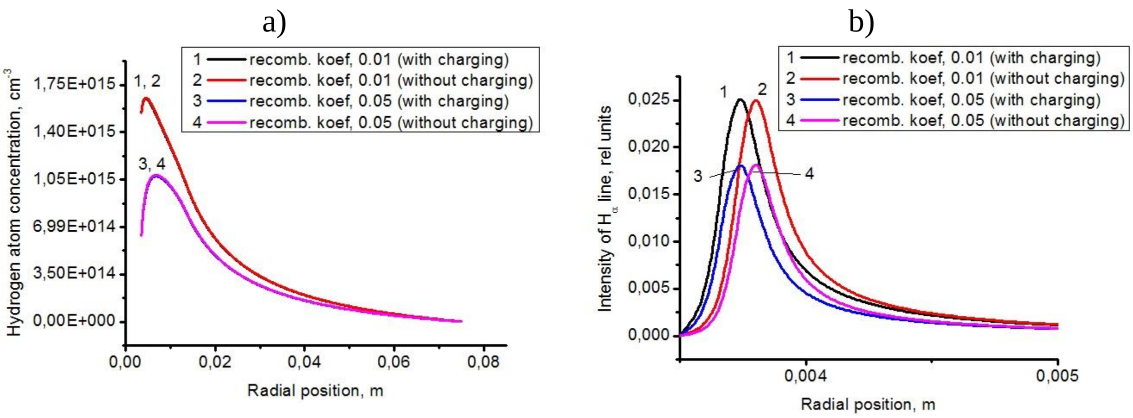

Figure 3. Radial distributions of hydrogen atom concentration (a) and H_α-line intensity (b) with (1, 3 curves) and without (2, 4 curves) charge on quartz cover of thickness 1 mm for different values of H-atom recombination coefficient (results of 1-D modeling).

3.3. *The change of catalytic properties of the surface of the antenna covered with dielectric.*

The calculations within the one-dimensional model showed that the catalytic activity affects neither the magnitude nor the distribution of the electron density and microwave field, and they coincide with those given in Fig.3b,c. Two values for coefficients of surface recombination of atoms have been used in calculations: 0.05 for copper and 0.01 for quartz. At the same time, the concentration of hydrogen atoms increases significantly at the transition from the copper to quartz. This is due to the fact that in considered conditions the main channel of loss of atoms is diffusion towards the surface with subsequent recombination. The deposition of charges on the surface of the electrode does not practically affect the concentration of the atoms (curves 1, 2 and also 3, 4). The difference is small and has a spatial scale, determined from the inset in Fig.2b. The absolute values of the emission intensity of line H_α also increase with changing recombination coefficient of hydrogen atoms from 0.05 to 0.01. Fig.3b shows the radial distribution of the emission intensity of the line H_α in the vicinity of the electrode, where the microwave field is high (see Fig.2c). Unlike for atoms the emission curves reveal the influence of the charges deposition on the electrode surface: as in the case of the microwave fields, the deposition of charges leads to a shift of the emission maximum to the surface of the electrode

It can be expected that at high pressures and high concentrations of the atoms the channel of volume losses of atoms begins to dominate and the role of the catalytic activity of the wall will decrease.

4. Conclusion

Modeling of the influence of the dielectric cover (the change in electrodynamics of the discharge, deposition of charges on the dielectric surface, changing the catalytic properties of the surface of the antenna) on plasma parameters of the microwave discharge in hydrogen at reduced pressure have shown that: (a) the use of dielectric covers of different thickness and different dielectric permeability makes it possible to control the spatial distribution of microwave field intensity near the antenna; (b) modification of electrodynamics and the deposition of charges on the dielectric surface only affect the surface layer of plasma and practically does not affect the characteristics of plasma in the plasma volume (e.g., the maximum value of the electron density); (c) catalytic properties of the dielectric surface strongly affect the maximum value of the concentration of hydrogen atoms in the case if their loss occurs due to the processes of diffusion and recombination on the surface; (d) deposition of charges on the dielectric surface leads to an increase in surface potential relative to the plasma, but the form of the spatial profile of the potential remains the same, and also leads to the shifting of the region of plasma resonance towards the surface of the electrode. It can be expected that when increasing the role of volume recombination of hydrogen atoms, the role of the recombination coefficient on the surface of the dielectric on the plasma parameters would be small.

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