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Study of plasma-solid interaction in electronegative gas mixtures at low and medium pressures

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1. Introduction

Low-temperature reactive plasmas employing electronegative gases are often used for material processing. Negative ions in such plasmas affect the transport of charged species from the plasma to immersed substrates and in this way influence the corresponding plasma chemical technologies. The same situation holds for probe diagnostics in plasmas containing negative ions, therefore the understanding of processes in the boundary layer between plasma and immersed substrates is very important.

In low-temperature plasma physics, the new challenging problem started to be the surface treatment at medium and high pressures, including the atmospheric pressure plasma. However, the detailed description of processes taking place during plasma-solid interaction at higher pressures is rather difficult and theories derived for collisionless or slightly collisional plasmas lose their validity, so the computer experiments are being widely used now, based either on the technique of fluid modelling [1], [2] or on various types of particle simulation codes.

As the particle modelling gives deeper insight into physical processes taking part in plasma-solid interaction, in the present contribution the combination of molecular dynamics method and Monte Carlo approach was used for the study of plasma containing negative ions at low and medium pressures. The following questions were studied by the computer modelling: the influence of plasma composition on the distribution of electric potential near the metal substrate, the influence of pressure on the energy and angular distributions of charged particles in the vicinity of plasma-solid boundary and a special attention was given to the technique of modelling and performance of computer codes at higher pressures.

2. Experimental technique

The properties of discharges in inert gases are intensively studied for a very long period. In our laboratory the measurements were performed in a positive column of dc glow discharge in rare gases – argon, neon and helium – both pure and in their mixtures with oxygen. For the study of plasma properties optical, microwave and Langmuir probe diagnostics were used. Some of our experimental results can be found e.g. in [3], [4].

The input data for our computer experiment were obtained from argon and oxygen plasma and from their mixtures. The main experimental parameters were the gas pressure, which varied between 67 and 800 Pa, the discharge current in limits 5 to 30 mA and the concentration of oxygen in the mixture in limits 0 to 100 %. During the measurement the following plasma characteristics were obtained: concentrations of electrons and their temperatures, intensity of axial electric field and collisional frequency. An example of experimental results can be seen in Fig. 1. Other data important for modelling, especially about scattering of charged particles by neutrals, were obtained from the literature [5].

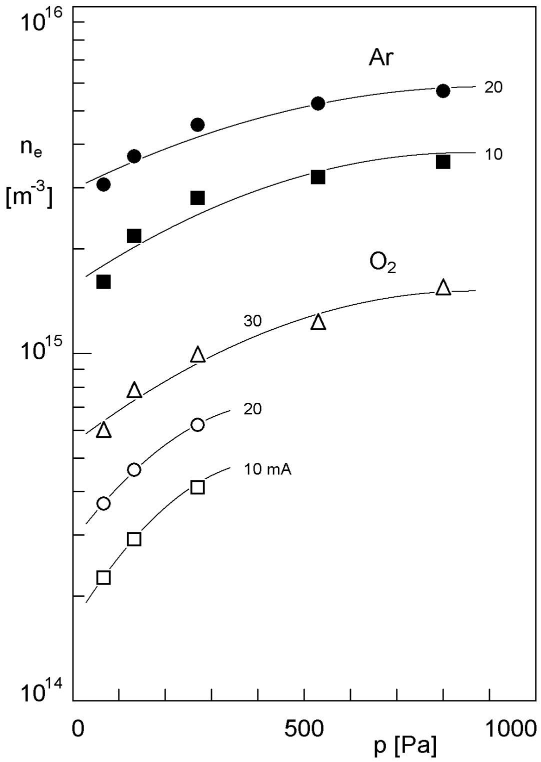

Figure 1: Electron densities in positive column of dc glow discharge in argon and oxygen. Discharge current 10 to 30 mA.

3. Model

In our computer experiment the main attention was devoted to two basic questions – to the role of negative ions in the formation of the sheath region and to the influence of plasma parameters, especially pressure, on this process. The particle simulation in self-consistent modification, i.e. the standard PIC-MC approach, was used, when the movement of charged particles in both external and local electric fields is studied by the deterministic molecular dynamics technique and the interactions between charged and neutral particles are treated stochastically by the Monte Carlo method. The details can be found e.g. in [6].

The main ideas of our model were:

- The geometry of the model was simplified in accordance with problems studied – planar substrates in plasma technologies and planar or cylindrical probes in plasma diagnostics. The first modification was one-dimensional with one spatial coordinate (x-axis) and two velocity coordinates (v_x, v_{yz}). The second one, used for cylindrical geometries, was two-dimensional with two spatial (x and y axes) and three velocity coordinates (v_x, v_y, v_z).
- Source of particles was the undisturbed plasma consisting of basic charged species in oxygen-argon discharge, i.e. electrons, positive argon ions and negative oxygen ions. The Maxwell distribution of velocities was supposed with different temperatures of electrons and ions.

- Interactions of charged particles with neutrals were simulated stochastically including the null-collision technique in order to compensate the energy dependencies of individual reactions. For electrons elastic collisions, excitation and ionization, for argon ions elastic collisions and charge transfer interactions and for oxygen ions elastic collisions were supposed.
- For the solving of Newton's equations of motion the second-order Verlet algorithm was used with discrete time steps Δt. In order to speed-up calculations, the steady-state simulation was performed with different time steps for electrons (10^{-11} to 10^{-13} s) and ions (10^{-8} to 10^{-10} s). In the dynamic regime the electron time steps had to be used for all particles.

The model was written in Fortran 90 programming language and processed by the PC microcomputer. The number of particles treated simultaneously was 2×10^5 to 2×10^6 and the total number of time steps was about 1×10^5. For the determination of energy and angular distributions of electrons the effective number of particles in model reached the number 1×10^9, however this increase was achieved by some sophisticated programming techniques.

4. Results and discussion

In order to analyse the pressure dependence of plasma-solid interaction, the experimental data for the electron and ion densities were applied (see Fig. 1) and it was supposed that the cross-sections of interactions of charged particles with neutrals depend on the pressure linearly. In Fig. 2 the results for the sheath region for various concentrations of negative ions and for various pressures are summed up. It can be seen that the sheath thickness decreases with increasing pressure and increases with the relative number of negative ions.

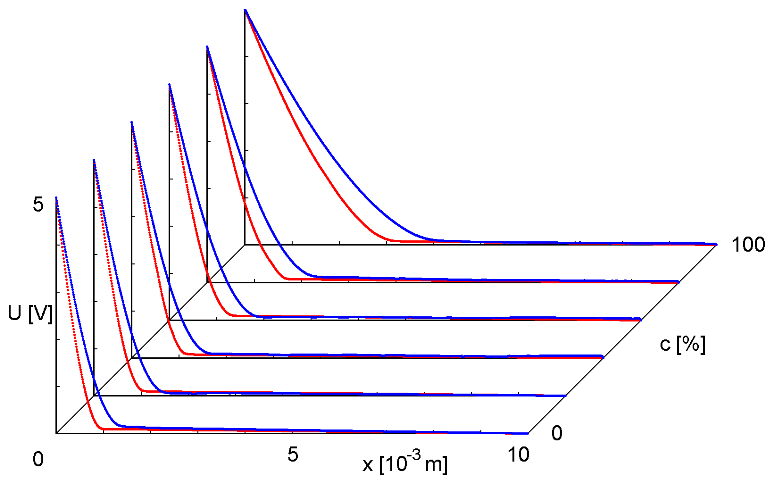

Figure 2: Potential distributions *U(x)* in the sheath for various relative concentrations of negative ions *c*. Pressure of plasma: blue – 133 Pa, red – 1330 Pa. Planar geometry.

Further, the energy and angular distributions of electrons in the vicinity of the probe immersed into plasma were calculated. In Fig. 3 the energy distributions of electrons are demonstrated for two pressures (133 Pa and 1330 Pa) and two relative concentrations of negative ions (blue, red: $c = 0\,\%$, black: $c = 50\,\%$). From this figure it can be seen that while the pressure changes the velocities of electrons in the sheath region, the influence of plasma electronegativity is negligible. The same situation was observed in angular distributions, too.

From the point of view of computational physics, the efficiency of particle modelling decreases rapidly with increasing pressure and new approaches of simulation must be introduced.

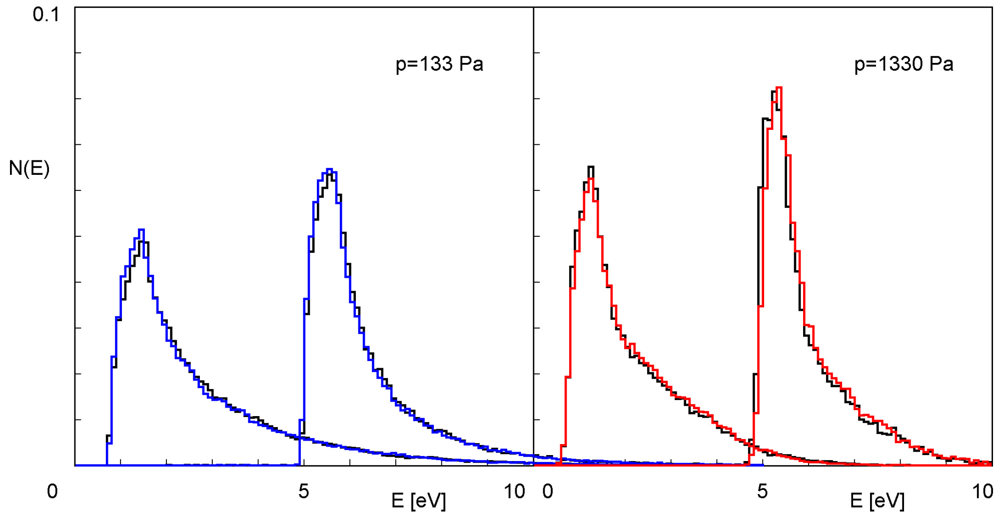

Figure 3: Energy distributions of electrons at two plasma pressures, both in two positions in the sheath – on the boundary sheath-presheath (left characteristics) and on the surface of immersed solid (right). Planar geometry, applied voltage bias +5 volts. Original electron temperature 2 eV.

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