Fluid simulations of radio frequency glow discharges: Two-dimensional argon discharge including metastables

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Two-dimensional self-consistent fluid simulations of a 13.56 MHz argon glow discharge including metastable species were performed as examples of a coupled glow-discharge/neutral-transport-and-reaction system. The electron density was found to peak in the radial direction. The metastable density profiles showed "hot spots" in both axial and radial directions.

Etching and deposition of thin films using low-pressure nonequilibrium gas discharges are crucial for the fabrication of advanced microelectronic devices.[1] During the past decade there has been growing interest in modeling and simulation of reactive plasma processes.[2] The goal is to develop a robust plasma reactor simulator [at least two dimensional (2D)] which can execute in a short time and can be used with confidence to predict reaction rate and uniformity as well as anisotropy and selectivity. Development of such a simulator has been hampered by (a) the nonequilibrium nature of the discharge which necessitates calculation of the electron energy distribution function, (b) the complexity of the plasma chemistry, especially the surface reactions, and (c) the numerical complexity of the problem due to stiffness in both space and time.

Numerical complexity results from the fact that electrons respond on a nanosecond time scale while neutral reactions can happen on time scales of hundreds of milliseconds. On the other hand, the applied plasma excitation frequency has a period of 74 ns (13.56 MHz). Therefore one may need to integrate for millions of rf cycles (with at least a few hundred time steps per cycle) to reach the periodic steady state. Another complexity is introduced because of the presence of the plasma sheath near the reactor walls (a "boundary layer") which also happens to be highly convective and introduces instabilities in the numerical schemes. Neglecting neutral species chemistry reduces the stiffness in the time domain as well as the number of dependent variables. Thus, it comes as no surprise that most glow discharge models reported to date are one dimensional and do not include neutral transport and reaction.[3-5] Recently, glow discharge models including plasma chemistry[6] as well as 2D geometries[7-9] have started to appear in the literature.

In a previous report,[10] we presented a one-dimensional fluid simulation of an argon discharge including metastables as a case study of a coupled glow-discharge/neutral-transport-and-reaction problem. Due to the long time scale of metastable species reactions, integration must be carried out for some 10^5 rf cycles to achieve convergence. In order to avoid the tedious direct-time integration, an algorithm based on the Newton–Raphson method was employed to speed up convergence by orders of magnitude. It was shown that metastables play an important role in the discharge despite the fact that their mole fraction was only $\sim 10^{-5}$. Specifically, metastable (two-step) ionization was found to be the main source of electrons sustaining the discharge. In addition, the (axial) metastable density profile showed distinct peaks near the electrodes and a valley in the midplane. It was concluded that neutral species transport and reaction must be accounted for in a self-consistent manner in order to obtain a correct picture of the discharge. The simulation was extended to a 2D reactor consisting of two parallel disk electrodes surrounded by a radial confining wall.[11] Hot spots in the metastable density were observed near the corners of the electrodes with the radial wall. In this letter, a 2D simulation is reported in a more practical reactor geometry, that of the Gaseous Electronics Conference (GEC) reference cell. Riley *et al.*[12] presented a one-dimensional glow discharge simulation in He including metastable species self-consistently. Preliminary results on a 2D helium discharge simulation in the GEC reference cell including metastables were presented at the 45th GEC in 1992 by Pak and Riley.[12] We are not aware of any published 2D glow discharge simulations of the GEC reference cell with or without neutral transport and chemistry.

A schematic of the reactor is shown in Fig. 1. Dimen-

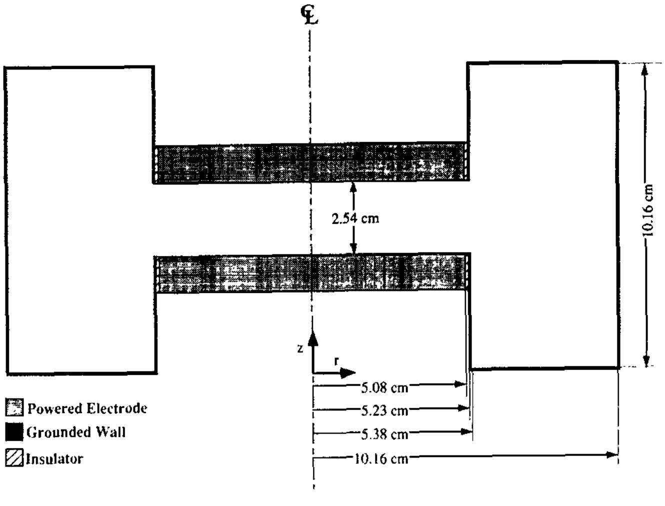

FIG. 1. Schematic of the GEC reference cell studied. The origin of the axisymmetric system is shown.

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sions of the axisymmetric system are typical of the GEC reference cell. Power at 13.56 MHz was applied in a push–pull fashion to preserve plasma symmetry. A two-moment fluid approach was followed as before.[10] The assumptions of the model are discussed in our previous report.[10] The same set of equations was used except that (a) a term to account for elastic collision losses was added to the electron energy equation, and (b) an equation was added for the effective electric field to which ions respond

$$\frac{\partial \mathbf{E}^{\text{eff}}}{\partial t} = \nu_{\text{ion}}(\mathbf{E} - \mathbf{E}^{\text{eff}}).$$

In this equation, $\mathbf{E}$ is the electric field, $\mathbf{E}^{\text{eff}}$ is the effective electric field, and ν_{ion} is the ion-neutral collision frequency. The electron–particle reaction rate coefficients were found as before.[10] Model equations and boundary conditions will be discussed in detail in a forthcoming paper.[13]

The problem consists of determining the electron, positive ion, and metastable atom density, electron energy, potential, and effective electric field as a function of space and time for a given set of system parameters. The gradient and divergence operators in the conservation equations[10] were written in cylindrical coordinates (r,z). The equations were discretized in space by using the Galerkin finite element method. The physical domain was divided into 2604 bilinear elements having a total of 2709 nodes. Spatial discretization resulted in a system of 18 673 coupled ordinary differential equations which must be integrated in time. This is a very demanding computational problem. In order to reduce the computational load, a modular approach was followed. Different modules were used for the charged species density and potential, electron temperature, effective electric field, and neutral (metastable) density. Necessary information was cycled back-and-forth among the modules until convergence. Time integration was performed using the stiff integrator LSODI.[14] In order to avoid the tedious direct-time integration, an "acceleration" scheme was employed based on the Newton–Raphson method. Details of the method are provided elsewhere.[10] Since the neutral density module did not present much stiffness in space, a staggered mesh was employed with a coarser grid for the neutrals. The simulation was run for 500 rf cycles.

The operating conditions used for calculation were: argon gas number density $N=3.22\times10^{16}$ cm^{-3} (1 Torr, 300 K), excitation frequency $f=13.56$ MHz, applied rf voltage 60 V peak to peak. Dimensions are shown in Fig. 1 which also shows the origin of the coordinate system (r, z) used. Results of the 2D simulation at the reactor axis ($r=0$) differed by less than 4% from the one-dimensional results under otherwise identical operating conditions.

Figure 2 shows a contour plot of the time-average electron density distribution. Note that only a portion of the reactor domain is shown. Axial location $z=3.81$ cm corresponds to the surface of the bottom electrode. The upper electrode is 2.54 cm apart from the bottom electrode (at $z=6.35$ cm, see Fig. 1). The axial distribution of electron density is paraboliclike. Strong electron gradients are evident in the sheath regions. A peak in electron density develops in the radial direction. This peak can be enhanced by applying a higher rf voltage. Similar peaks in plasma density have been observed in other 2D discharge models with a confining radial wall.[7,8,11] Sharp electron density gradients develop as one moves radially beyond the plasma zone into the surrounding chamber. The electrons are trying to diffuse out of the plasma zone, but are impended by a radial space-charge field. The presence of this field is evident from the potential distribution in the reactor (Fig. 3). The field is higher in regions of rapid potential change. The radial electric field is very weak near the reactor axis ($r=0$). The radial field opposes the outward motion of negatively charged particles (the field is pointing away from the plasma). High (axial) fields develop near the electrode surfaces (in the sheath) and also in the corner regions near the electrode edges. As the rf voltage increases or the pressure decreases, the electrons are expected to "leak out" further away from the interelectrode space.

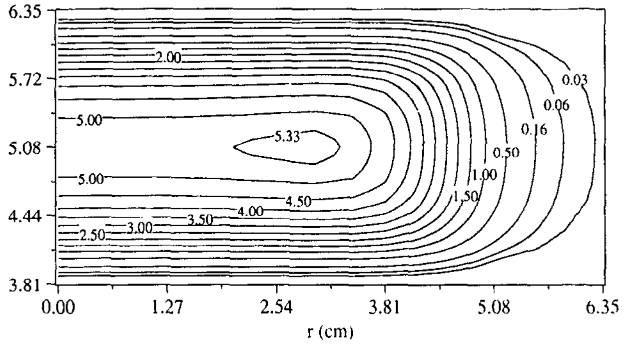

FIG. 2. Time-average electron density profile. Only part of the reactor domain is shown for clarity. To obtain the actual electron density multiply the numbers shown on the contours by 10^9 cm^{-3}.

The electron mean energy (not shown) near the reactor axis was close to that predicted by a one-dimensional model. Away from the plasma zone the electrons cool down as they diffuse against the radial space-charge field. The minimum electron energy was found to be near the

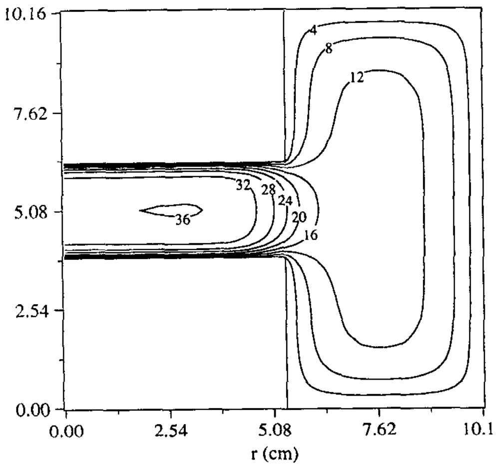

FIG. 3. Time-average electric potential profile. Values of potential in volts are shown on the contours.

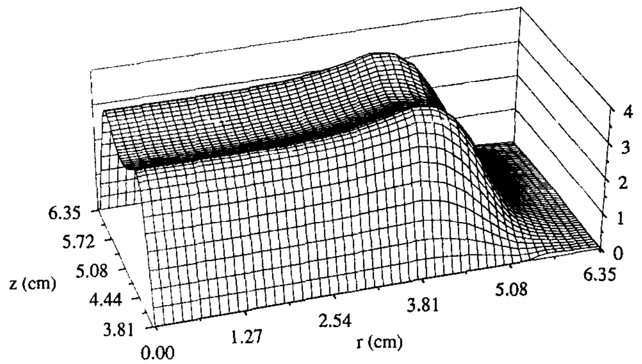

FIG. 4. Metastable density profile. Only part of the reactor domain is shown. The metastables are not modulated in time. To obtain the actual metastable density multiply the numbers shown on the vertical axis by 10^{11} cm^{-3}.

corners of the surrounding grounded chamber at $(r,z) = (10.16,0)$ and (10.16,10.16) cm.

The metastable density profiles are shown in Fig. 4. The axial peaks in metastable density in the interelectrode space are due to (a) enhanced production of metastables near the glow/sheath interface due to higher electron energy in that region, and (b) enhanced losses of metastables by electron quenching in the central plane of the discharge due to higher electron density there (Fig. 2). The metastables are quenched effectively on solid surfaces and their density is zero all around the wall. Metastables are not affected by the electric field and can diffuse outside the plasma zone unimpeded. For this reason the metastable density is substantial beyond the plasma edge (beyond $r = 5.08$ cm). Peaks in the radial profile of metastable density are observed near the plasma edge. These hot spots were also seen in a 2D simulation of parallel disk electrodes with a radial confining wall.[11] Metastable density profiles similar to those shown in Fig. 4 were obtained experimentally by Greenberg and Hebner[15] for a He metastable state in the GEC reference cell.

In summary, a 2D fluid simulation of a coupled glow-discharge/neutral-transport-reaction system was carried out in the GEC reference cell geometry with 13.56 MHz excitation. In particular, an argon discharge including metastable chemistry was studied. The radial profile of the plasma density and potential was nonuniform. This has practical implications regarding the uniformity of ion flux bombarding the electrode and device damage. The metastable density profiles showed hot spots in both axial and radial directions. In a chemically reactive discharge, such hot spots may contribute to nonuniformity of the radical flux bombarding the wafer. Using the model, one can modify the reactor or optimize the operating conditions to achieve uniform ion and radical fluxes.

Multidimensional simulations of plasma reactors can serve as powerful tools for improving our understanding of reactive plasmas and for helping in the design of new and improved plasma processes. Such simulations have only recently started to appear. Further understanding of plasma physics and chemistry as well as developments in numerical methods and in parallel computing will have a profound impact on multidimensional simulations of glow discharges coupled with neutral transport and reaction.

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