Monte Carlo Model for the Argon Ions and Fast Argon Atoms in a Radio-Frequency Discharge

Annemiek Bogaerts and Renaest Gijbels

Abstract—A three-dimensional Monte Carlo model has been developed for the argon ions and fast argon atoms in the RF sheath of a capacitively coupled RF glow discharge in argon. Our interest in the argon ions and fast argon atoms in the RF sheath arises from the fact that the glow discharge under study is used as a sputtering source for analytical chemistry. This source operates at typical working conditions of a few Torr pressure and about 10 W incoming power. The argon ion and atom Monte Carlo model has been coupled to a hybrid Monte Carlo-fluid model for electrons and argon ions developed before to obtain fully self-consistent results. Typical results of this model include, among others, the density and fluxes, collision rates, mean energies, and energy distributions of the argon ions and atoms. Moreover, we have investigated how many RF cycles have to be followed before a periodic steady state is reached, and the effects of all previous RF cycles are correctly accounted for. This is found to be the case for about 20-25 RF cycles.

Index Terms—Argon ions, fast argon atoms, glow discharge, hybrid model, modeling, Monte Carlo, plasma, radio frequency.

I. INTRODUCTION

Glow discharges are finding increased interest in a number of applications, like the microelectronics industry, materials technology, lasers, light sources, and plasma display panels. Moreover, an additional application field is analytical chemistry [1], [2]: the material to be analyzed is used as the cathode of the glow discharge, which is being sputtered by energetic plasma particles. The sputtered (analytically important) atoms arrive in the plasma where they are subject to collisions. The ionization collisions create ions of the material to be analyzed, which can be measured with a mass spectrometer [glow discharge mass spectrometry (GDMS)]. The excitation collisions and the subsequent decays to lower levels give rise to characteristic photons of the material, which can be detected with an optical emission spectrometer [glow discharge optical emission spectrometry (GD-OES)]. The simplest operation mode of an analytical glow discharge is the direct current mode. However, because the material to be analyzed is used as the cathode of the glow discharge, this seems to restrict the applications to the analysis of nonconducting materials. To extend the application range to the analysis of nonconductors as well, a radio-frequency voltage is applied to prevent charging up of the nonconducting materials. Because of its much larger applicability, capacitively coupled radio-frequency discharges are becoming more and more popular in analytical chemistry.

To improve the results in the various application fields of glow discharges, a good insight in the glow discharge processes is desirable. We try to obtain this by mathematical modeling. During the last few years, we have developed a comprehensive modeling network for a dc glow discharge in argon, consisting of a number of sub models for the various plasma species, i.e., electrons, argon ions, fast argon atoms, argon atoms in various excited levels, sputtered argon, and argon ions in the ground state and in various excited levels. More information can be found in [3]–[9] and references therein. Recently, we started with modeling a capacitively coupled RF discharge. In a previous paper [10], a hybrid Monte Carlo fluid model was presented for the electrons and argon ions. However, from our dc models, it became obvious that the fast argon ions and atoms play a nonnegligible role in the ionization processes of argon at the high dc voltages under consideration (typically 600–1000 V). We found that these processes had to be included in the dc model, in order to be able to predict the correct current–voltage characteristics [6]. Moreover, these species result in sputtering at the cathode (or RF powered electrode), which makes them of great importance from an analytical point of view.

A number of models have been presented in the literature describing the behavior of ions explicitly and kinetically in an RF discharge, either by using a Monte Carlo method [11]–[16] or by applying a self-consistent particle-in-cell model [17]–[21]. However, these models apply to glow discharges operating at much lower pressures (mtr regime) than our analytical glow discharges (typically a few Torr). Moreover, except for [14], they describe only the behavior of the ions and do not take into account the fast atoms. Nevertheless, it has been demonstrated that the fast atoms often play a more important role than the ions for ionization and sputtering, not only in analytical glow discharges [3], [6], but also for etching and materials processing applications [14]. Therefore, in the present paper, a Monte Carlo model is described for both the argon ions and fast argon atoms in the sheath in front of the RF powered electrode. The model is made self-consistent by coupling it to our hybrid Monte Carlo fluid model for electrons and argon ions [10]. In the following section, the model will be described. First, a brief explanation of the previously developed hybrid model will
be given, because the present model is linked to it. Next, the present ion/atom Monte Carlo model will be explained in detail. Finally, the coupling between the ion/atom Monte Carlo model and the hybrid model will be outlined.

II. DESCRIPTION OF THE MODEL

A. Brief Explanation of the Previous Hybrid Model

The previous hybrid model consists of a Monte Carlo model for the electrons, and a fluid model for the electrons and argon ions. In the electron Monte Carlo model, a large number of electrons are followed during successive time steps. Their trajectory is calculated by Newton’s laws, whereas their collisions (i.e., occurrence of a collision, kind of the collision, and new energy and direction after collision) are determined by random numbers (similar to the ion/atom Monte Carlo model; see below). Besides electrons starting at the RF electrode as a function of time, electrons created in the plasma by ionization collisions are also followed. Collision processes incorporated in this model are total electron impact excitation and ionization from the argon atom ground state, elastic collisions with argon atoms, and electron-electron Coulomb scattering. The electrons are followed during a number of RF cycles until periodic steady state is reached, i.e., when the results do not change anymore from one RF cycle to the next (analog to the ion/atom Monte Carlo model; see below).

Beside this Monte Carlo model, the electrons are also treated in a fluid model, together with the argon ions. The equations are the first three velocity moments of the Boltzmann equation, i.e., the balance equations for particle density, for momentum density, and for energy density. Furthermore, these equations are coupled to Poisson’s equation to obtain a self-consistent potential and electric field distribution. The method we used for solving these coupled equations is based on the Scharfetter–Gunn–Munzel exponential scheme (see [10]). The input parameters in this fluid model are the geometry, the boundary conditions, the pressure, and the discharge power, as well as the creation rates for ions and electrons, and the energy loss rate for the electrons, both obtained from the electron Monte Carlo model. The RF voltage and the dc bias voltage are calculated self-consistently from this model. This is based on the fact that the power dissipated in the discharge should be equal to the given input power (for the RF voltage) and that the ion and electron fluxes to the RF electrode should be equal to each other when integrated over the entire RF cycle, which is imposed by the capacitive coupling and the difference in size of RF powered and grounded electrodes (for the dc bias voltage). More detailed information about this hybrid model can be found in [10].

B. Detailed Explanation of the Ion/Atom Monte Carlo Model

The argon ions and atoms are followed as a function of time in the Monte Carlo model, in the sheath adjacent to the RF powered electrode. The region where the ions and atoms are followed is assumed constant and is taken a little further than the farthest point of the sheath. In practice, a region of about 1.5 mm was considered. The time-varying electric field is adopted from the fluid model [10]. Since the ions cannot follow the rapidly fluctuating electric field, they feel only an effective electric field, which approaches the time-averaged electric field (see [10]). It is calculated from the real electric field as follows [22]:

$$\frac{\partial E^{\text{eff}}}{\partial t} = v_m(E - E^{\text{eff}})$$

where $E$ and $E^{\text{eff}}$ are the real and effective electric field, respectively, and $v_m$ is the momentum transfer frequency

$$v_m = \frac{e}{\mu m_A + m^{\ast} A^+}$$

with $\mu$ and $m^{\ast}$ being the mobility and mass of the argon ions, and $e$ being the electron charge.

Fig. 1 shows the one-dimensional axial electric field distributions in the first mm in front of the RF electrode at four different times in the RF cycle. The solid lines symbolize the real electric field, whereas the effective electric field as felt by the ions is represented by the dashed lines. The real electric field varies strongly as a function of time: it is almost zero at $\omega t = \pi/2$, but it is extremely negative at the RF electrode at the other times (i.e., in the order of $-15$ kV/cm at $\omega t = \pi$ and $2\pi$ and even $-25$ kV/cm at $\omega t = 3\pi/2$). The effective electric field, however, does not vary so much in time (i.e., only a factor of two: between $-10$ and $-20$ kV/cm at the RF electrode). Moreover, since the ions cannot follow the real electric field due to their high mass, they seem to feel the electric field with a slight delay. This explains why the effective electric field is lowest at $\omega t = \pi$ instead of at $\omega t = \pi/2$. Similarly, the effective electric field felt by the ions is highest at $\omega t = 2\pi$, whereas the real electric field was highest at $\omega t = 3\pi/2$.

All the argon ions and fast argon atoms are followed during successive time steps. Besides continuing with the ions from
the previous time step, it is also checked whether ions have to be followed, which: 1) are entering the sheath from the bulk plasma at that time (the ion flux as a function of time is obtained from the fluid model), 2) are formed by electron impact ionization at that time (taken from the electron Monte Carlo model), 3) are created by fast ion impact ionization, and 4) arise from fast atom impact ionization (both adopted from this model). It is found that the majority of ions are formed by electron impact ionization in the RF sheath (nearly 97.5%); about 2.5% originate from the bulk plasma, and only a small fraction is formed in the RF sheath by fast argon ion and atom impact ionization.

Similarly, beside the atoms from the previous time step, it is checked whether additional atoms have to be followed, which are created at that time-step by: 1) charge transfer of argon ions, 2) momentum transfer of the argon ions, and 3) momentum transfer of the argon atoms. However, in order to reduce the calculation time, the argon atom is removed from the Monte Carlo calculations when its energy becomes lower than 1 eV, because it is not really "fast" anymore and cannot give rise to atom impact ionization, excitation, and sputtering anymore.

A typical time step in the order of 1.15 \times 10^{-9} s was used in the model, which corresponded to 64 time-steps within one RF cycle. This time step was found to be small enough. Indeed, we have checked that exactly the same calculation results were obtained when using a time-step of $10^{-10}$ and $10^{-11}$ s.

During this time-step, the trajectories of all the ions and atoms are computed with Newton's laws:

$$
x = x_0 + v_{x0} \Delta t + \frac{qE_{ax}(z, r, t)}{2m}(\Delta t)^2
$$

$$
y = y_0 + v_{y0} \Delta t + \frac{qE_{ay}(z, r, t)}{2m}(\Delta t)^2
$$

$$
z = z_0 + v_{z0} \Delta t + \frac{qE_{az}(z, r, t)}{m}(\Delta t)^2
$$

where $x_0$, $y_0$, $z_0$, $v_{x0}$, $v_{y0}$, $v_{z0}$, and $x$, $y$, $z$ are the position coordinates before and after $\Delta t$, $v_{x0}$, $v_{y0}$, $v_{z0}$, and $x$, $y$, $z$ are the velocities before and after $\Delta t$, $E_{ax}$ and $E_{ay}$ are the axial and radial electric field, as a function of axial and radial position and time in the RF cycle (obtained from the fluid model; see above), $\alpha$ is the azimuthal angle of the radial position (i.e., the angle of the radial position coordinates with respect to the x-axis), and $q$ and $m$ are the charge and mass, respectively. For the atoms, $v_{z0}$ is of course zero, and the last term in the equations disappears.

Then, the collision probability is calculated and compared to a random number between zero and one. If the probability is lower than the random number, no collision takes place. However, if the probability is higher, a collision takes place, and the kind of collision is determined, based on the partial collision probabilities and a second random number. The collisions taken into account for the argon ions include charge transfer and momentum transfer collisions and fast argon ion impact ionization and excitation. For the fast argon atoms, momentum transfer collisions, fast argon atom impact ionization, and excitation were included. The cross sections for all these processes were obtained from Phelps [23], [24]. Finally, the new energy and direction after collision are determined based on formulas for scattering angles and energy conservation. More details about the Monte Carlo procedure and about the scattering theories used can be found in [3], [10], and [25], respectively.

This procedure is repeated during a large number of time steps (and RF cycles) until periodic steady state is reached, i.e., when the effect of all previous RF cycles is taken into account. In practice, it was found that periodic steady state was reached after 20–25 RF cycles (see below). Indeed, the maximum distance for the ions and atoms to travel is equal to the maximum sheath length and is typically 1 mm [10]. The velocity of the ions and atoms is in the order of $10^{-6}$–$10^{-7}$ cm/s. Hence, to cross the entire sheath, they need $10^{-6}$–$10^{-7}$ s. Since one RF period lasts $7.4 \times 10^{-8}$ s, at least 14 RF cycles have to be followed. Hence, 20–25 RF cycles are safe numbers, and they correspond well with the number of 25, predicted in [17]. In practice, we followed the ions during 20 RF cycles to be sure that periodic steady state will also be reached at other discharge conditions (i.e., other RF sheath thicknesses) under investigation. Later in this paper, the effect of the number of RF cycles to be followed will be discussed in more detail.

C. Coupling Between the Two Models

As mentioned before, this argon ion and fast argon atom Monte Carlo model is coupled to our hybrid model [10] to reach self-consistent results. The procedure is as follows. First, the fluid equations are solved, assuming arbitrary values for the ion and electron production rates. The results of this fluid model are the electric field as a function of position and time in the RF cycle, as well as the ion flux bombarding the RF electrode and entering the RF sheath from the bulk plasma. The electric field and the ion flux at the RF electrode are used as inputs in the electron Monte Carlo model to determine the electron trajectory in the electric field and the number of electrons starting at the RF electrode as a function of time, respectively. This Monte Carlo model yields, among others, the electron impact ionization rate. The latter is used as input in the argon ion/fast argon atom Monte Carlo model to determine the number of ions created by this process as a function of time. Other input values for this model include the electric field as a function of position and time, as well as the ion flux entering the RF sheath from the bulk plasma, both derived from the fluid model. Outputs of the latter model are, among others, the fast argon ion and atom impact ionization rates. Hence, the electron Monte Carlo model is then again calculated, including the electrons formed by the latter two processes. Next, the argon ion/fast argon atom Monte Carlo model is again run, etc. This procedure of successive electron and argon ion/atom Monte Carlo models is repeated until the
total ionization (due to electron, fast argon ion, and atom impact) remains constant. After that, the fluid calculations are again solved using the sum of the electron, fast argon ion, and fast argon atom impact ionization rates as input (i.e., for the creation rates of argon ions and electrons as a function of position and time). This fluid model yields new values for the electric field distribution and for the ion fluxes at the RF electrode and at the RF sheath, which are again used in the electron, and argon ion/fast argon atom Monte Carlo models. This whole procedure is repeated until convergence is reached, which typically occurs after five iterations.

III. RESULTS AND DISCUSSION

A. Argon Ion and Fast Argon Atom Densities

Fig. 2 shows the argon ion density at $\omega t = \pi$. The results at the other time steps in the RF cycle are practically the same. The argon ion density is in the order of $10^{12}$ to $10^{18}$ cm$^{-3}$. It is rather low at the RF electrode and reaches its maximum of $10^{13}$ cm$^{-3}$ at about 0.12 cm from the RF electrode. This result is in fair agreement with the outcome of the fluid model, as is illustrated in Fig. 3 (both the results in the entire discharge region and the detail of the RF sheath are presented). The absolute values are nearly identical, but the fluid model yields a maximum slightly further away from the RF electrode (i.e., at $0.15-0.2$ cm).

The fast argon atom density distribution at $\omega t = \pi$ is presented in Fig. 4. It is in the order of $10^{12}$ to $10^{18}$ cm$^{-3}$, with its lowest value at the RF sheath-bulk plasma interface and increasing toward the RF electrode. It is also more or less constant during the entire RF cycle. By comparing Figs. 2 and 4, it becomes clear that the fast argon atom density is higher than the argon ion density by about one order of magnitude. However, compared to the overall argon gas atom density of nearly $10^{17}$ cm$^{-3}$ (at a pressure of 6 torr and a gas temperature of 600K), the contribution of the fast argon atom density is negligible, i.e., most argon atoms are still thermalized. Nevertheless, it is expected that these fast argon atoms play a significant role in the sputtering process, as was also the case in the dc glow discharge [3].

B. Argon Ion and Fast Argon Atom Mean Energies and Energy Distributions

The mean argon ion and fast argon atom energies, as a function of axial position in the RF sheath, are depicted in Fig. 5, at four different times in the RF cycle. The mean argon ion energy (solid lines) increases toward the RF electrode, since the ions gain energy from the electric field. The energy is not constant during the entire RF cycle; it is highest at $\omega t = 2\pi$ (about 33 eV at the RF electrode) and lowest at $\omega t = \pi$ (around 13.5 eV at the RF electrode), which corresponds exactly to the variations in the effective electric field distributions, illustrated in Fig. 1. At first sight, this result is surprising, because the ions need many RF cycles to traverse the RF sheath, and one would expect that the effect of the varying electric field and hence the ion energy would be averaged out. However, the argon ions undergo many charge transfer collisions, after which they start again from thermal energy. Next, they again receive energy from the instantaneous electric field (which can be considerably close to the RF electrode, see Fig. 1). When
The ion data are sampled at this moment, the variation of the effective electric field is clearly felt.

The mean fast argon atom energies at the four different times, represented by the dashed lines in Fig. 5, do not vary so much during the RF cycle (although they are a little bit higher at $\omega t = \pi/2$ and $2\pi$), and they stay also more or less constant in the RF sheath, only slightly increasing in the last 0.5 mm in front of the RF electrode. The mean fast argon atom energy is somewhat lower than the argon ion energy because the fast argon atoms cannot gain energy from the electric field, they can only lose energy by collisions. It should be mentioned, however, that only atoms with energy higher than 1 eV are called "fast," and that the mean fast argon atom energy looks artificially high, especially between 0.5 and 1.5 mm from the RF electrode, because it includes only fast argon atoms. Of course, the overall mean argon atom energy will be very close to thermal, as discussed before.

The flux energy distributions of argon ions bombarding the RF electrode, at four different times in the RF cycle, are shown in Fig. 6. It should be noticed that the energy axis is truncated at 60 eV, i.e., lower than the total dc or RF voltage, because the energy axis beyond 60 eV gives no additional information. The energy distributions exhibit some pronounced features, characterized by a series of peaks, superimposed on a rapidly decreasing curve. This characteristic structure has also been observed in other calculation results (e.g., [11], [15], [18], [19], [26]), as well as in experimental measurements (e.g., [25]-[29]). At very low pressure (a few mTorr), when the ions do not undergo collisions in the sheath, the ion energy distribution is characterized by a saddle-shaped structure, due to RF modulation. However, as the pressure increases, the saddle shape disappears gradually, and secondary peaks appear on top of the energy distribution, as a result of thermal ions formed in the sheath due to charge transfer collisions, and which gain only a fraction of the RF electric field. The relative heights and positions of these secondary peaks depend on the discharge conditions (dc bias voltage, pressure) and are directly related to the number of ions which have undergone charge transfer collisions at different distances from the RF electrode. Moreover, the energy distribution is shifted toward lower energies due to charge transfer and momentum transfer collisions. Although the models and measurements described in the literature apply to glow discharges operating at lower pressures (e.g., 1–100 mtorr), the same feature is also observed in our modeling results for 6 torr.

Fig. 7 presents the flux energy distribution of the fast argon atoms bombarding the RF electrode at $\omega t = \pi$. The results at the other time steps were nearly identical. Again, the energy axis is truncated at 100 eV, because the number of atoms with higher energy is negligible. In contrast to the argon ion flux energy distribution, the fast argon atom energy distribution does not exhibit a peaked structure, but decreases monotonically toward higher energies, because the atoms, when they have lost energy due to collisions, cannot gain energy anymore from the electric field. The same decreasing curve was also obtained in the modeling results presented in [14]. Finally, it is also worth mentioning that the energy distribution peaks at 1 eV and is zero at still lower energies. The reason, of course, is the fact that atoms with energies below 1 eV are removed from the fast argon atom Monte Carlo model (see above).

C. Argon Ion and Fast Argon Atom Fluxes

Another feature which becomes apparent from Figs. 6 and 7 is that the fast argon atom flux bombarding the RF electrode is
much higher than the argon ion flux. Indeed, a large number of these fast argon atoms are created due to charge transfer and momentum transfer collisions. Hence, although the mean fast argon atom energy is lower than the mean ion energy, it is expected that the fast argon atoms still play a dominant role in sputtering, due to their much higher flux, as is also the case in the dc glow discharge [3].

The total argon ion and fast argon atom fluxes bombarding the RF electrode, as a function of time in the RF cycle, are depicted in Fig. 8. It is indeed clear that the fast argon atom flux is much higher than the argon ion flux (i.e., a factor of 20, see left and right axes). Moreover, the fast argon atom flux is more or less constant in time, whereas the argon ion flux is at its maximum around \( \omega t = 0 = 2\pi \) and has a minimum around \( \omega t = \pi \) (see negative scale). Hence, there is a phase shift with regard to the RF voltage applied to the RF electrode (which is at its maximum around \( \omega t = 3\pi/2 \) and at its minimum around \( \omega t = \pi/2 \), in agreement with results in the literature (e.g., [18]-[20]). The argon ion flux at the RF electrode, calculated with our fluid model, is also illustrated in Fig. 8 (short dashed line). It reaches also a minimum at \( \omega t = \pi \) and a maximum at \( \omega t = 0 = 2\pi \), in good correspondence with our Monte Carlo results. However, the variation between the minimum and the maximum value seems to be somewhat less pronounced in the Monte Carlo model: it varies between \(-8.5 \times 10^{16} \) s\(^{-1}\) and \(-1.5 \times 10^{17} \) s\(^{-1}\), whereas the result of

![Fig. 6](image_url)  
Fig. 6. Flux energy distributions of the argon ions bombarding the RF electrode, at four different times in the RF cycle. Note that the x-axis is truncated at 60 eV.

![Fig. 7](image_url)  
Fig. 7. Flux energy distribution of the fast argon atoms bombarding the RF electrode at \( \omega t = \pi \). The results at the other times in the RF cycle are practically identical. Note that the x-axis is truncated at 100 eV.
D. Collisions of the Argon Ions and Fast Argon Atoms

Fig. 8 illustrates the ionization rates, due to fast argon ion and fast argon atom impact, integrated over the entire RF sheath region, as a function of time in the RF cycle. The ionization is at its minimum around \( \omega t = \pi \), and reaches a maximum at \( \omega t = 0 = 2\pi \), which follows the trend of the effective electric field (Fig. 1). This is expected because the above mentioned processes become important only at high ion and atom energies [23]. The fast argon atoms seem to produce more ionization than the argon ions (by a factor of 3-4), as is explained by the much higher argon atom flux (see Fig. 8). This outcome is in correspondence with our previous dc results [6].

For comparison, the electron impact ionization rate, integrated over the entire discharge, is also illustrated (dashed line, right axis). It reaches a minimum around \( \omega t = \pi/2 \) and a maximum at \( \omega t = 0 = 2\pi \), but it is much higher than the argon atom impact ionization rate (by a factor of 50) and than the argon ion impact ionization rate (by a factor of 150). Integrated over the entire discharge region and RF
cycle, the electrons contribute for about 97.7% to the total ionization of argon (of which 60% occurs in the RF sheath and 40% in the plasma bulk). The relative contributions of the fast argon atoms and ions to the total ionization were calculated to be about 1.8% and 0.5%, respectively. Hence, the fast argon atoms and ions seem to play only a minor role for ionization at the present discharge conditions. This is not surprising, because the calculated RF voltage and dc bias voltage are in the order of 600 V and −345 V, respectively (see below). We have found that in the dc case, the effect of fast argon ions and atoms on the total ionization (and on the calculated current–voltage characteristics) started to become important for voltages higher than 600 V [5]. Hence, it is expected that ion and atom impact ionization become more important at higher RF and dc bias voltages, which give rise to higher ion and fast atom energies, and hence, more efficient fast argon ion and atom impact ionization.

E. Effect of the Argon Ions and Atoms on the Electrical Characteristics

In our previous hybrid Monte Carlo-fluid model [10], the RF and dc bias voltages were calculated self-consistently, when the input power, the gas pressure, and gas temperature were given. For the present conditions of 10 W incoming power, 6 torr argon pressure and 600 K gas temperature \( V_{RF} \) was calculated to be 900 V and \( V_{dc} \) was −610 V [10]. Now, when the ion/atom Monte Carlo model is added to this hybrid model, \( V_{RF} \) and \( V_{dc} \) become clearly lower (i.e., in the order of 600 V and −345 V, respectively). The reason for this is that fast argon ion and atom impact ionization make the glow discharge self-sustained at lower voltages. The explanation that ion and atom impact ionization are really responsible for the lower voltages, and that this was not due to another phenomenon, was checked by comparing the results of the hybrid ion/atom Monte Carlo and electron Monte Carlo-fluid model, with and without taking into account these processes.

Hence, although the above processes have only a minor contribution to the total amount of ionization, they do have a significant effect on the electrical characteristics due to the so-called “snowball-effect” (i.e., the ions and atoms cause ionization, thereby creating more electrons; these electrons can also give ionization, creating more ions; the ions, and the atoms formed by collisions of the ions, yield again ionization, giving rise to more electrons, etc.) and it is expected that this effect will become even more important at higher dc bias voltages.

F. Effect of the Number of RF Cycles to Be Followed on the Calculated Results

As mentioned before, it was investigated how many RF cycles had to be followed before periodic steady state was
reached, and it was found that this was the case for 20-25 RF cycles. To demonstrate this, Figs. 10(a) and (b) shows the argon ion and fast argon atom fluxes at the RF electrode, for a range of different RF cycles followed. It is indeed apparent that below 20 or 25 RF cycles (for the ions and atoms, respectively), the results increase with the number of RF cycles, because the previous RF cycles still have noticeable effect on the last cycle (i.e., the one from which the output data are sampled). However, after 20 or 25 RF cycles, the results do not change anymore, which shows that periodic steady state is indeed reached after 20-25 RF cycles. However, to be sure that this steady state would be reached for all discharge conditions to be investigated, we ran the calculations for 30 RF cycles. Fig. 11 illustrates the number of ions (solid line, left axis) and atoms (dashed line, right axis), followed as a function of time, during the successive RF cycles. It appears indeed that the number of ions and atoms remains periodically constant after about 20 and 23 RF cycles, respectively.

**IV. CONCLUSION**

A Monte Carlo model has been developed for the argon ions and fast argon atoms in the sheath adjacent to the RF electrode, and this model has been coupled to a hybrid Monte Carlo fluid model for the electrons and argon ions, which was developed before [10]. Typical results of the model presented here comprise the argon ion and fast atom densities, fluxes, collision rates, mean energies, and energy distributions. The latter are very important for our application (spattering glow discharge for analytical chemistry), because it determines the amount of sputtering. Moreover, this quantity is also of great significance from the point of view of materials processing, as appears from the large number of publications in the literature (e.g., [11], [14], [18], [19], [26]-[29]). Since the fast argon atom flux bombarding the RF electrode is considerably higher than the argon ion flux, it is expected that these fast argon atoms give the main contribution to sputtering, in spite of their slightly lower energy.

Further, it was demonstrated that the argon ions and fast argon atoms play a role in the total degree of ionization. Although their relative contribution, compared to electron impact ionization, was rather low, these species do have a significant effect in lowering the RF and dc bias voltages. Moreover, it is anticipated that both the contribution of the fast argon ions and atoms to the overall ionization, and the effect of these species on the RF and dc bias voltages, will still increase with rising $V_{dc}$.

Finally, it was investigated how many RF cycles have to be followed before periodic steady state is reached: and this was found to be the case for about 20-25 RF cycles.

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Annie Bogaerts was born in Belgium on October 25, 1971. She received the M.S. degree in chemistry and the Ph.D. degree in science from the University of Aalst, Belgium, in 1993 and 1995, respectively. She is now working as a postdoctoral researcher at the University of Antwerp. Her current research interest is in the modeling of glow discharge plasmas, as well as in experimental plasma diagnostics to validate the modeling results.

Renat Gijbels was born in Belgium on January 1, 1959. He received the Ph.D. degree in science from the University of Ghent, Belgium, in 1985. He became a Professor of Physical Chemistry at the University of Aalst, Belgium, in 1975. His research interests are in micro surface and trace analysis by electron, ion, and laser beam techniques: fundamentals, methodology, and applications.