# Modeling of magnetron and glow discharges

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*Pour améliorer les résultats des décharges magnétron (et des décharges luminescentes, en général), il est important d'avoir une bonne compréhension des mécanismes intervenant dans la décharge. Ceci peut être obtenu par la modélisation numérique. On a entrepris le développement d'un modèle hybride Monte Carlo-fluide pour une décharge magnétron en Argon et pour la description du comportement des électrons rapides et lents et des ions $Ar^+$. Nous présentons, pour une décharge luminescente en argon avec un cathode de cuivre, un "réseau des modèles" comprenant des modèles Monte Carlo, fluide, collisionnel-radiatif et un modèle de transfert de chaleur pour les différentes espèces de plasma : atomes d'argon, électrons, ions d'argon, atomes d'argon rapides, atomes excités d'argon, atomes et ions de cuivre. Ce réseau des modèles est intéressant pour les décharges magnétron auxquelles nous avons l'intention de l'appliquer.*

***Mots-clé : Modélisation - Décharge luminescente - Décharge magnétron - Modèle hybride - Pulvérisation de cuivre - Modèle Monte Carlo - Modèle fluide.***

## Introduction

To improve the results of magnetron discharges used for the sputter-deposition of thin films, a good insight in the discharge mechanisms is desirable. This can be obtained by numerical modeling. Different kinds of models have been applied already in the literature to magnetron discharges, i.e., analytical approaches [1], fluid modeling [2], collisional-radiative models [3], Boltzmann equations [4], Monte Carlo methods [5], particle-in-cell simulations [6] and hybrid Monte Carlo – fluid models [7]. The advantage of the latter is the accuracy at low pressure (in comparison to a pure fluid model) in combination with a reduced calculation time (compared to particle-in-cell simulations). Therefore, we try to develop ***a hybrid Monte Carlo – fluid model for a planar magnetron discharge in argon***. In Section 2, this hybrid model will be briefly outlined.

Whereas our modeling activities of magnetron discharges started only recently, we have much more experience with the modeling of glow discharges used for the sputtering of solid materials in analytical spectrometry, either in direct current (dc) [8], radio-frequency (rf) [9] or pulsed mode [10]. Typical operating conditions are a pressure of 0.5-5 Torr, a discharge voltage of 500-1500 V, and an electrical current ranging from 1 mA to 100 mA. The discharge gas is typically argon, and the cathode material is mostly assumed to be copper. For this type of glow discharges, we have ***developed a hybrid modeling network, consisting of Monte Carlo, fluid and collisional-radiative models as well as a heat-transfer model, for the different species assumed to be present in the plasma***. Section 3 of this paper will deal with this hybrid modeling network. The different submodels will be briefly described, and some typical results, which are relevant for magnetron discharges as well, will be presented.

## Hybrid Monte Carlo – fluid model for a planar magnetron discharge in argon

The plasma species considered in this hybrid Monte Carlo – fluid model, include argon gas atoms (assumed to be thermal and uniformly distributed in the plasma), argon ions and slow electrons. The electrons are subdivided in two groups, based on their energy, i.e., the fast electrons with total (i.e., sum of potential + kinetic) energy above 11.55 eV (which are able to cause excitation and ionization of the argon atoms) and the slow electrons with lower energy.

The ***fast electrons are simulated with a three-dimensional (3D) Monte Carlo algorithm***. They are treated as individual particles, and their trajectories are traced, starting from when they are created (either at the cathode by secondary electron emission, or in the plasma by electron impact ionization) until they are absorbed at the electrodes or transferred to the slow electron group (see above). For each electron, the Newton equation with a Lorentz force term is solved at each time-step :

$$m\frac{\vec{V}_{new}-\vec{V}_{old}}{\Delta t}=\vec{F}_{old} \qquad \frac{\vec{X}_{new}-\vec{X}_{old}}{\Delta t}=\vec{V}_{new}$$

$$\vec{F}=\vec{F}_{elec}+\vec{F}_{magn}=qE+q(\vec{V}\times\vec{B})$$

An axisymmetric magnetic field, formed by two concentric cylindrical magnets located behind the cathode (see below), is used as input in the model. The magnetic field values as a function of position are adopted from the literature [11] and are depicted in ***figure 1***.

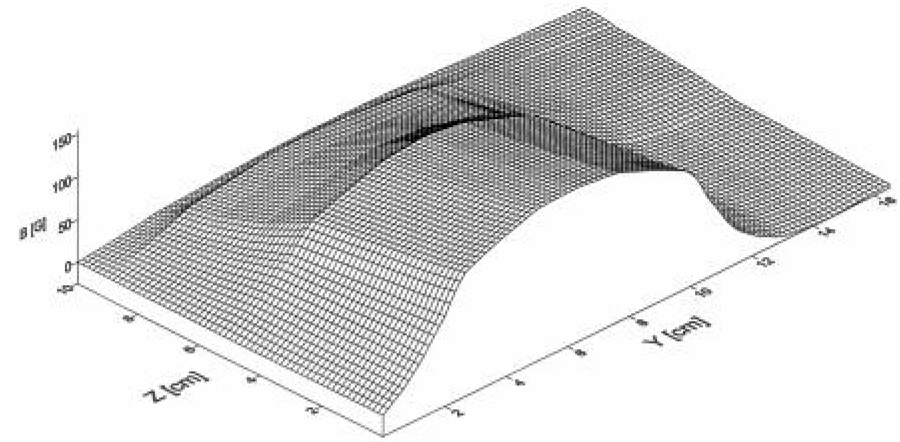


*Fig. 1*
*Magnetic field values used as input in the Monte Carlo model [11].*

The electric field as a function of position is normally taken from the fluid model (see below), but in the first iteration, an initial guess is used. The numerical method used is "leap frog", which is a time-centered method with second order of accuracy. The collisions taken into account are elastic scattering with argon gas atoms, ionization and excitation of the argon ground state. The output of the Monte Carlo code includes the fast electron density, the ionization rate and the slow electron creation rate, as a function of space, which are used as input in the fluid model.

The ***fluid model treats the slow electrons and argon ions*** in the plasma. They are considered as non-interacting particles, which allows us to treat them as a medium rather than as "individuals". The correct way to treat them is to apply equations of magneto hydrodynamics. Doing this in 3D, unfortunately, is not a straightforward problem, due to the fact that the magnetic field is not curl-free, which leads to coupling between the components of mass flux. At this stage, we are developing a 2D model, assuming a 2D electric field (in the z and y direction) and a 1D constant magnetic field (in the x-direction, i.e., perpendicular to the electric field). These assumptions make it possible, after some mathematical manipulations, to uncouple the components of the mass flux, and to solve the transport equation of the electrons self-consistently with Poisson's equation for the electric field, using the exponential scheme [12]. Hence, the equations are :

• the mass balance equations for electrons and ions :

$$\frac{\partial n^{e^-,Ar^+}}{\partial t} + \mathrm{div}\vec{\Gamma}^{e^-,Ar^+} = R^{e^-,Ar^+}$$

• the transport equation for electrons and ions :

$$\Gamma_y^{e^-} = \frac{1}{1+\frac{\omega_x^2}{\nu^2}}\left(-\frac{\omega_x}{\nu}\mu^{e^-} n^{e^-} E_z - \mu^{e^-} n^{e^-} E_y - \frac{\omega_x}{\nu} D^{e^-} \frac{\partial n^{e^-}}{\partial z} - D^{e^-} \frac{\partial n^{e^-}}{\partial y}\right)$$

$$\Gamma_z^{e^-} = \frac{1}{1+\frac{\omega_x^2}{\nu^2}}\left(-\mu^{e^-} n^{e^-} E_z + \frac{\omega_x}{\nu}\mu^{e^-} n^{e^-} E_y - D^{e^-} \frac{\partial n^{e^-}}{\partial z} + \frac{\omega_x}{\nu} D^{e^-} \frac{\partial n^{e^-}}{\partial y}\right)$$

$$\Gamma_y^{Ar+} = \left(-\mu^{Ar^+} n^{Ar^+} E_y - D^{Ar^+} \frac{\partial n^{Ar^+}}{\partial y}\right)$$

$$\Gamma_z^{Ar+} = \left(-\mu^{Ar^+} n^{Ar^+} E_z - D^{Ar^+} \frac{\partial n^{Ar^+}}{\partial z}\right)$$

• Poisson equation for the electric field :

$$\Delta V = -\frac{q}{\varepsilon_0}\left(n^{Ar^+} - n_{fast}^{e^-} - n_{slow}^{e^-}\right)$$

The electric field resulting from the fluid model, is used again as input in the Monte Carlo code. The two models are solved iteratively until final convergence is reached.

The model is applied to a dc planar magnetron, as is illustrated in ***figure 2***, which operates at the following conditions : an argon gas pressure of 5 mTorr, a dc voltage of 500 V and a magnetic field in the order of 50-150 G.

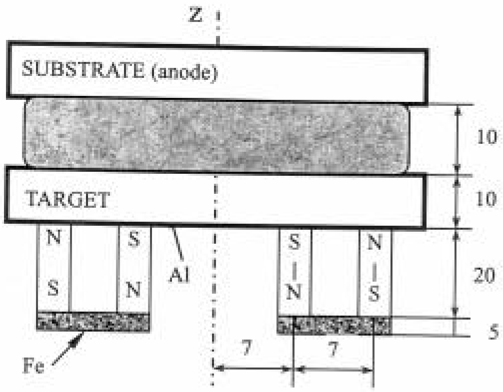


*Fig. 2*
*Schematic diagram of the magnetron setup.*

Typical results of the model include the electron and argon ion density profiles, the electric field distribution, the ionization and excitation rates, the electron and argon ion mass fluxes, and the energy distribution of electrons. Due to the complexity of the fluid model for a magnetron discharge, we are not yet able to present results, but they are expected for the near future.

# Hybrid modeling network for a glow discharge in argon with copper cathode

The hybrid modeling network that we have developed for a glow discharge in argon with copper cathode (in dc, rf and pulsed mode) consists of a number of Monte Carlo, fluid and collisional-radiative models as well as a heat-transfer model, for the different species assumed to be present in the plasma.

The ***argon gas atoms*** are either assumed to be with thermal energy, uniformly distributed throughout the plasma, or alternatively, the heating of the argon gas (due to collisions of energetic plasma species with the argon atoms) is calculated with a ***heat transfer equation*** [13] :

$$\frac{\partial^2 T_g}{\partial z^2} + \frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial T_g}{\partial r}\right) = -\frac{P}{\kappa}$$

Here, $T_g$ is the gas temperature, $\kappa$ is the thermal conductivity, and P is the power input into the argon gas, due to collisions of the energetic plasma species (electrons, $Ar^+$ ions, fast Ar atoms and sputtered Cu atoms). This power input is calculated in Monte Carlo models (see below) [13].

With this heat transfer model, gas temperatures were calculated to range from room temperature to 1000 K, depending on the discharge conditions ***(fig. 3)***, and with a maximum value near the cathode ***(fig. 4)***. Hence, this implies that the argon gas can be "depleted" (e.g., by a factor of 2-3) near the cathode.

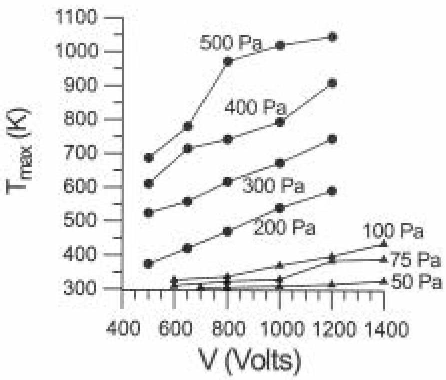


***Fig. 3***
*Calculated gas temperature at the maximum of its profile, as a function of voltage and pressure.*

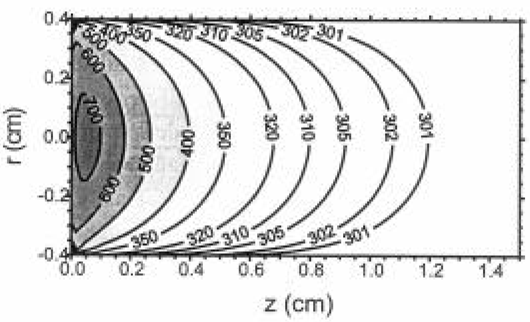


***Fig. 4***
*Calculated gas temperature distribution at 800 V, 50 mA and 400 Pa. The cathode is found at z = 0 cm, whereas the other borders of the figure represent the cell walls at anode potential.*

The other species present in the plasma, i.e., electrons, $Ar^+$ ions, fast Ar atoms, Ar atoms in various excited levels, sputtered Cu atoms and the corresponding $Cu^+$ ions, both in the ground state and in various excited levels, are described with Monte Carlo, fluid and collisional-radiative models.

The electrons are split up in two groups, depending on their energy. The ***fast electrons***, which are not in equilibrium with the electric field, are simulated with a ***Monte Carlo method***, whereas the slow electrons are treated, together with the ***argon ions***, in a ***fluid approach***. This model consists of the continuity equations and transport equations (based on diffusion and on drift in the electric field) as well as Poisson's equation for a self-consistent electric field. The latter means that the electric field used to calculate the transport of the charged particles (based on migration) is calculated itself using the charged particle densities.

Because the argon ions are not really in equilibrium with the strong electric field in the cathode dark space (CDS), the fluid approach is only an approximation here. Therefore, we simulate the behavior of the ***argon ions in the CDS*** also with a ***Monte Carlo model***, in addition to the fluid model. This ***Monte Carlo model*** is also used to treat the ***fast argon atoms in the CDS***, which are created from elastic collisions of the argon ions with the argon gas atoms. The most important output of these two Monte Carlo models is the flux energy distribution of the argon ions and fast argon atoms bombarding the cathode, which is needed to calculate the sputtering (see below).

The ***argon atoms in various excited levels*** are described with a ***collisional-radiative model***, consisting of a set of balance equations (one for each excited level) with different production and loss terms. 64 excited argon atom levels were taken into account in this model, and the production and loss processes include, among others, excitation, de-excitation, ionization, recombination, radiative decay, Hornbeck-Molnar associative ionization, Penning ionization of sputtered atoms by argon metastable atoms, and trapping of radiation to the ground state.

As mentioned above, the ***flux of sputtered atoms*** is calculated from the flux energy distributions of the argon ions, fast argon atoms as well as copper ions bombarding the cathode, which are all calculated with Monte Carlo models, multiplied with an ***empirical formula*** for the sputter yield as a function of energy of the bombarding species.

When the ***copper atoms*** are sputtered, they have typical energies in the order of a few eV. They lose this energy, however, quite rapidly at the pressure of about 1 Torr by collisions with the argon gas atoms, until they are thermalized. This ***thermalization process*** is described with a ***Monte Carlo algorithm***.

Once the ***copper atoms*** are thermalized, their further transport is diffusion-dominated. The copper atoms are also subject to collisions in the plasma, of which excitation and ionization are the most important. The behavior of the ***copper atoms and corresponding copper ions, both in the ground state and in various excited levels***, is described with a

***collisional-radiative model***. Eight copper atomic levels, seven $Cu^{+}$ ion levels as well as the $Cu^{2+}$ ions are considered in this model. Their level populations are again calculated with a set of balance equations with different production and loss processes, including excitation, de-excitation, ionization, recombination, Penning ionization by argon metastable atoms and asymmetric charge transfer with argon ions.

Finally, the ***$Cu^{+}$ ions*** are also simulated with a ***Monte Carlo model in the CDS***, which calculates, among others, the flux energy distribution of copper ions bombarding the cathode, needed to calculate the sputtering (see above).

All these models are coupled to each other due to the interaction processes between the various species, and they are solved iteratively until final convergence is reached. This takes typically several days on today's fast computers. A flowchart of the coupling of the models is presented in ***figure 5***.

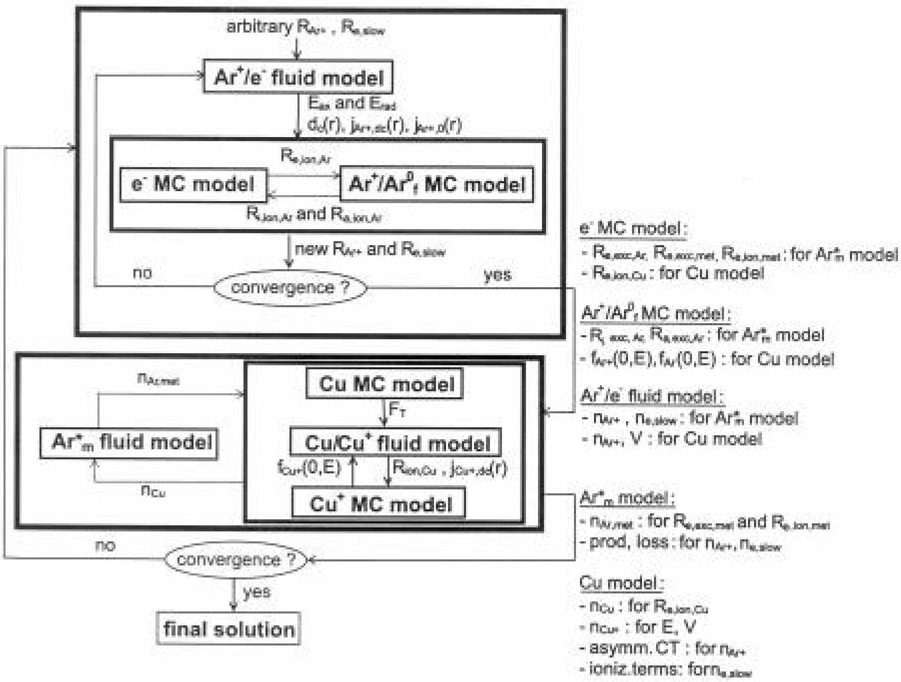


*Fig. 5*

*Flowchart of the modeling network.*

Typical output of this model includes, among others, the electrical characteristics (i.e., electrical current as a function of voltage and pressure, and in the case of an rf or pulsed discharge also as a function of time), the electric field and potential distribution, the densities, fluxes and energies of the various plasma species, information about the various collision processes in the plasma, and about sputtering at the cathode (i.e., erosion rates and crater profiles due to cathode sputtering), optical emission intensities, etc. Some of these results, which are also relevant for magnetron discharges, will be presented here, to show the possibilities of this modeling network.

***Figure 6a*** shows a calculated crater profile as a result of sputtering. It appears that the crater is much deeper at the sides than in the center, which is attributed to focussing of the ions by the electric field to the sides of the crater. Moreover, the crater walls are not very steep, and there is a small rim observed outside the crater. The latter is due to redeposition of sputtered atoms. These features of the calculated crater profile are also found back in the typical measured crater profiles, for the same conditions, as is clear from ***figure 6b***. This example shows that our model is able to reproduce the experimentally observed crater profiles, and to predict how the crater profiles can be optimized by changing cell geometry or discharge conditions [14].

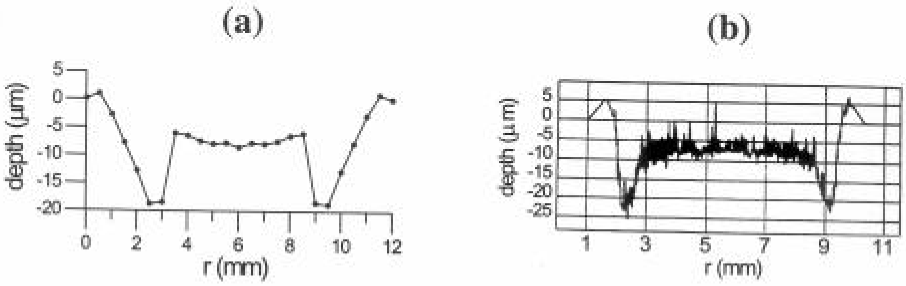


***Fig. 6***
*(a) Calculated crater profile as a result of cathode sputtering, in comparison with*
*(b) the measured crater profile at exactly the same conditions (1000 V, 0.5 Torr, 3 mA).*

***Figure 7*** shows the calculated thermalization profiles of sputtered atoms (i.e., the fraction of sputtered atoms thermalized as a function of distance from the cathode), at different pressures. It is clear that the thermalization profile becomes much more narrow at increasing pressure, i.e., the sputtered atoms are thermalized much closer to the cathode at increasing pressure, due to the occurrence of more collisions. At typical glow discharge conditions used for analytical spectrometry (pressure in the order of 1 Torr), most of the sputtered atoms are thermalized already at 2 mm from the cathode. For typical magnetron operating conditions, on the other hand, (i.e., pressure of a few mTorr), most of the sputtered atoms can travel a distance of 20-50 cm before being thermalized, as appears from ***figure 7.***

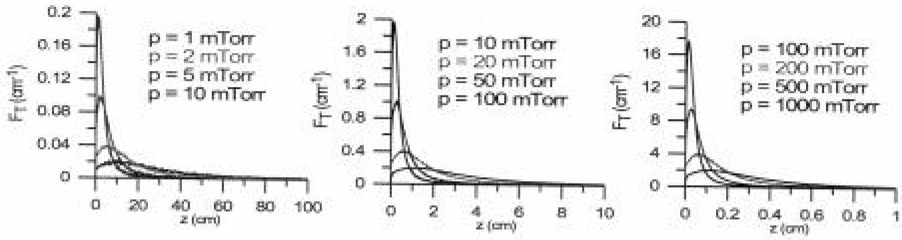


***Fig. 7***
*Calculated thermalization profiles of sputtered atoms, at different pressures.*

A comparison of the calculated and measured density profiles of sputtered Ta atoms, for typical glow discharge spectrometry conditions of 1000 V discharge voltage, 1 Torr argon pressure and 2 mA electrical current, is given in ***figure 8*** [15]. The cathode is represented by the thick black line at z = 0 cm. The calculation result ***(fig. 8a)*** shows a maximum density of about 3 x $10^{12}$ $cm^{-3}$ at 1 mm from the cathode, and the density drops toward the cell walls. The sputtered Ta atom density profile was also measured by a combination of laser-induced fluorescence spectrometry (to obtain the relative density profile) and atomic absorption spectrometry (to put an absolute number on the density values), and the result is shown in ***figure 8b***. The measured density shows a maximum of 1.6 x $10^{12}$ $cm^{-3}$, and drops also toward the cell walls. The observed difference in contour lines at z = 0 cm is due to some simplifications in the model, i.e., the cell was assumed to be closed at z = 0 cm in the model, whereas the real cell extended behind the position of the cathode (at z = 0 cm). Beside this difference, the agreement between calculated and measured profiles was found to be quite satisfactory, which demonstrates that our model gives a realistic picture of cathode sputtering.

Finally, based on the computed level populations of the excited levels, the model is also able to calculate optical emission intensities. ***Figure 9*** shows the calculated (left side) and measured (right side) optical emission intensities of some Ar(I), Ar(II) and Cu(I) lines, as a function of distance from the cathode, at a pressure of 0.6 Torr and at different conditions of voltage and current [16]. The good agreement between calculated and measured results shows that the correct processes are taken into account in the collisional-radiative models.

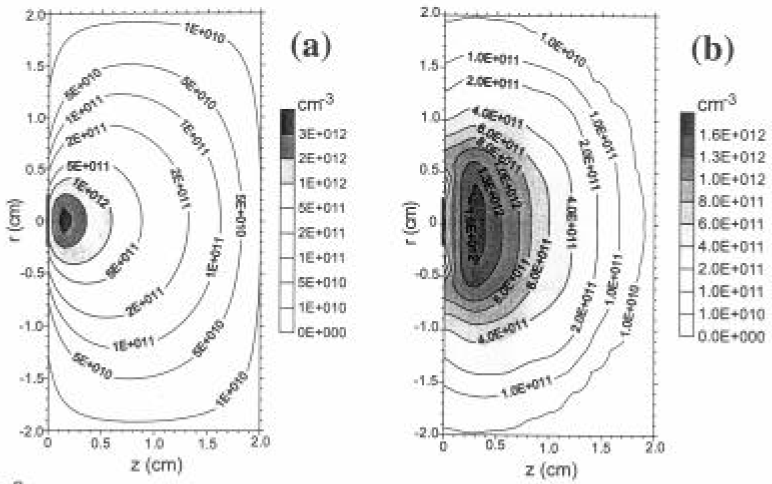


*Fig. 8*

*Calculated **(a)** and measured **(b)** density profiles of the sputtered Ta atoms, at 1000 V, 1 Torr and 2 mA. The cathode is represented by the thick black line at z = 0 cm, whereas the other borders of the figure symbolize the cell walls.*

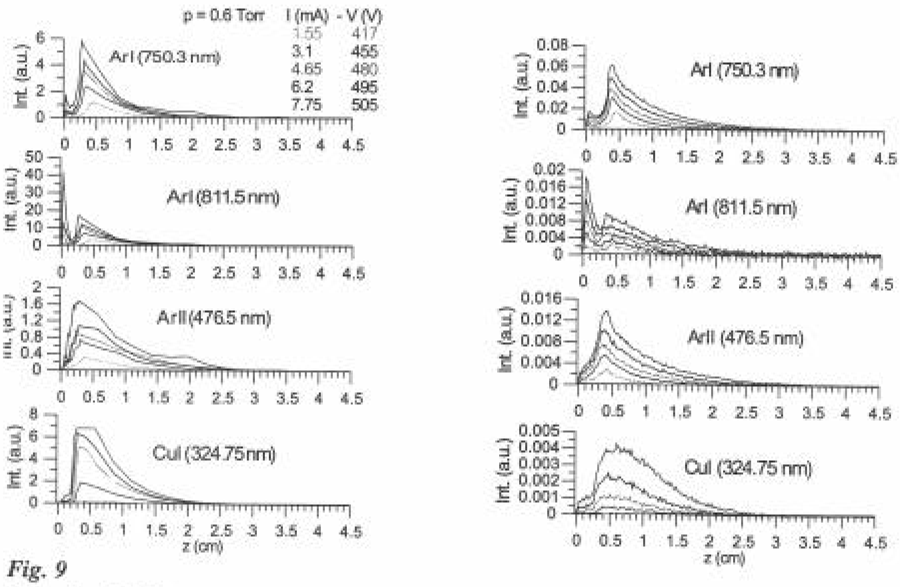


*Fig. 9*

*Calculated **(left)** and measured **(right)** optical emission intensities of some Ar(I), Ar(II) and Cu(I) lines, as a function of distance from the cathode.*

## Conclusion

The hybrid modeling network developed for the argon glow discharge with copper cathode at typical conditions for analytical spectrometry, which is presented in Section 3, describes the behavior of the sputtered species, as well as the heating of the background argon gas (which can also be important for magnetron discharges). Hence, it is of interest to magnetron sputtering discharges as well. ***Therefore, we plan to apply this hybrid modeling network to the magnetron discharge in the near future***. The first, and most difficult step is the development of the hybrid Monte Carlo – fluid model for electrons and $Ar^+$ ions, as is described in Section 2. Once that model works properly, which will be achieved in the near future, the other models will be applied to the magnetron discharge as well.

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