NUMERICAL MODELLING OF DC ARGON GLOW DISCHARGE AT LOW PRESSURE WITHOUT AND WITH Ar \((^{3}\text{P}_2)\) METASTABLE STATE

M. N. STANKOV\textsuperscript{1}, M. D. PETKOVIĆ\textsuperscript{2}, V. LJ. MARKOVIĆ\textsuperscript{1}, S. N. STAMENKOVIĆ\textsuperscript{1}, A. P. JOVANOVIĆ\textsuperscript{1}

\textsuperscript{1}Department of Physics, Faculty of Sciences and Mathematics, University of Niš, PO Box 224, 18001 Niš, Serbia, E-mail: marjansstankov@gmail.com
\textsuperscript{2}Department of Computer Science, Faculty of Sciences and Mathematics, University of Niš, PO Box 224, 18001 Niš, Serbia

Received September 16, 2013

One-dimensional extended fluid model of DC glow discharge in argon at low pressure is applied without and with Ar \((^{3}\text{P}_2)\) metastable state. The profiles of particle number densities, mean electron energy, potential and ionization source in the stationary state of glow discharge for 300V and 500V are presented.

Key words: glow discharge, elementary processes in plasmas, computer modelling and simulation.

1. INTRODUCTION

The gas discharges are widely used in different areas of science and technology, \textit{i.e.} for surfaces etching, sputtering deposition, in light sources, in medicine, etc. [1–5]. For better understanding of physical processes in various types of gas discharges numerical models are often applied. Among different models, the most common are fluid and particle models or their combination. The fluid model consists of the system of partial differential equations that could be solved by using the finite difference, finite element or finite volume method [6-8]. Even in 1971 computer modelling was applied for the simulation of rapidly developing gaseous discharge [9]. Later, Winkler and Wilhelm [10], Lymberopoulos and Economou [11] and Passchier and Goedheer [12] used the fluid model for the analysis of RF gas discharges in their papers. In the past ten years the fluid model has been encountered in a large number of papers treating DC or RF discharges and their applications [13–25].

In this paper one-dimensional extended fluid model is applied for the modelling of DC glow discharge in argon in the voltage range of \(300\text{V}–700\text{V}\), the inter-electrode distance of \(d = 1\text{ cm}\) and the pressure of \(p = 1.33\text{ mbar}\). From the emission spectrum of glow discharge in argon [26], the existence of metastable
state Ar ($^3P_2$) is confirmed, so the modelling is performed for the case without and with metastable state Ar ($^3P_2$) and the results are compared. The applied model is solved by the finite difference method. The system of equations obtained by the discretization of fluid equations is solved by applying the Thomas’s algorithm. The theoretical description of extended fluid model is given in section 2. In section 3 the transport and rate coefficients used in argon glow discharge modelling are reviewed. Section 4 presents detailed numerical analysis for solving the fluid equations, while section 5 describes applied boundary conditions. In section 6, the results of modelling are presented and discussed and finally the short conclusion is given in section 7.

2. EXTENDED FLUID MODEL

The fluid model is based on equations which are derived from the moments of the Boltzmann equation. The continuity equation for charged and excited particles (1) is obtained from the zero momentum of the Boltzmann equation. From the continuity equation the number density of particles can be calculated. Beside the continuity equation for particles, the extended fluid model contains the equation of energy balance (2) obtained from the second momentum of Boltzmann equation. The Poisson’s equation (3) is also a part of the fluid model from which the electrical field and potential are calculated, and the whole system reads:

\[
\frac{\partial n_{\text{ele,ion}}}{\partial t} + \nabla \cdot \Gamma_{\text{ele,ion}}^{\text{ele}} = k_{1\text{io}} n_{\text{ele}} N,
\]

\[
\frac{\partial (u_{\text{ele}} n_{\text{ele}})}{\partial t} + \frac{5}{3} \nabla \Gamma_{E}^{\text{ele}} = -e \Gamma_{E}^{\text{ele}} E - u_{\text{ion}} k_{1\text{io}} n_{\text{ele}} N,
\]

\[
\nabla^2 \varphi = -\frac{e}{\varepsilon_0} \left(n_{\text{ion}} - n_{\text{ele}}\right),
\]

where \( n \) is the number density of particles, \( \Gamma \) and \( \Gamma_E \) are fluxes of particles and electron energy, respectively, \( u_{\text{ele}} \) is mean electron energy, \( \varphi \) is potential, \( N \) is number density of gas, \( k_{1\text{io}} \) is the rate coefficient for electron impact ionization, \( u_{\text{ion}} = 15.8 \text{ eV} \) is the ionization energy of argon atoms, \( e \) is the electron charge and \( \varepsilon_0 \) is the dielectric constant. Superscripts \( \text{ele, ion} \) are used for the electrons and ions, respectively.

Fluxes for electrons and ions have the form:

\[
\Gamma_{\text{ele,ion}} = \pm \mu \Gamma_{\text{ele,ion}} n_{\text{ele,ion}} E - D_{\text{ele,ion}} \nabla n_{\text{ele,ion}},
\]

while the flux of electron energy is given by:
\( \Gamma_E = \pm \mu \nabla \times n_{ele} E - D n_{ele} \nabla \left( \mu n_{ele} E \right), \)  
\hspace{1cm} (5) 

where \( \mu \) is the mobility of charged particles, \( D \) is the diffusion coefficient, and \( E \) is the vector of electric field.

**Table 1**

<table>
<thead>
<tr>
<th>Reactions</th>
<th>Name of reactions</th>
</tr>
</thead>
<tbody>
<tr>
<td>( e + Ar \rightarrow e + Ar^+ )</td>
<td>Excitation to metastable state ( ^3P_2 )</td>
</tr>
<tr>
<td>( e + Ar^+ \rightarrow e+e+Ar^+ )</td>
<td>Stepwise ionization</td>
</tr>
<tr>
<td>( e+Ar^+ \rightarrow e+Ar )</td>
<td>Deexcitation of metastable state ( ^3P_2 )</td>
</tr>
<tr>
<td>( Ar^+ + Ar^+ \rightarrow e+Ar^+ + Ar )</td>
<td>Penning ionization</td>
</tr>
<tr>
<td>( Ar^+ + Ar \rightarrow 2Ar )</td>
<td>Two-body quenching collisions with ground-state atoms</td>
</tr>
<tr>
<td>( Ar^+ + Ar + Ar \rightarrow Ar_3^+ + Ar )</td>
<td>Three-body quenching collisions with two ground state atoms</td>
</tr>
</tbody>
</table>

By including the Ar \((^3P_2)\) metastable state into the extended fluid model the processes from Table 1 must be taken into account, thus the equations of extended fluid model take the form:

\[
\frac{\partial n_{ele,ion}}{\partial t} + \nabla \nabla n_{ele,ion} = k_e n_{ele} N + k_w n_{ele} n_{met} + k_{Pen} n_{ele} n_{met},
\]
\hspace{1cm} (6)

\[
\frac{\partial n_{met}}{\partial t} + \nabla \nabla n_{met} = k_{ex} n_{ele} N - k_{de} n_{ele} n_{met} - k_{io} n_{ele} n_{met} - 2 k_{Pen} n_{ele} n_{met} - k_{qu_2} n_{ele} n_{met} n_{met} N^2,
\]
\hspace{1cm} (7)

\[
\frac{\partial (u_{ele} n_{ele})}{\partial t} + \frac{5}{3} \nabla \nabla n_{ele} = -e \nabla \nabla E - \nabla \nabla n_{ele} N - k_{Pen} n_{ele} n_{met} + k_{Pen} n_{ele} n_{ele} n_{met},
\]
\hspace{1cm} (8)

\[
\nabla^2 \phi = \frac{e}{\epsilon_0} \left( n_{ion} - n_{ele} \right),
\]
\hspace{1cm} (9)

where superscripts \( met \) are used for metastable state of argon \((^3P_2)\). The notations of rate coefficients from the relations (6-9) and the Table 1 are following: \( k_{ex} \) - excitation to metastable state \( ^3P_2 \), \( k_{io} \) - stepwise ionization, \( k_{de} \) - deexcitation of metastable state \( ^3P_2 \), \( k_{Pen} \) - Penning ionization, \( k_{qu_2} \) - two-body quenching in collisions with ground-state atoms and \( k_{qu_3} \) - three-body quenching in collisions with two ground state atoms. The excitation energy of Ar atom to metastable state
3P₂ is \( u^{\text{net}} = 11.5 \text{ eV} \), while \( u^{\text{ion}} - u^{\text{net}} \) is the energy loss due to stepwise ionization. The energy which electron gains in Penning ionization process is \( u^{\text{Pe}} = 2u^{\text{net}} - u^{\text{ion}} \). The equation (7) is the continuity equation for metastable state which flux has the following form:

\[
\Gamma^{\text{met}} = -D^{\text{met}} \nabla N^{\text{met}}.
\]  

3. THE TRANSPORT AND RATE COEFFICIENTS

Since the mean electron energy is not constant, the mobility and diffusion coefficients for electrons are taken as a function of the mean electron energy [17], obtained by fitting the data from paper [23]. The ion mobility is taken as a function of the reduced electric field [24]:

\[
\mu^{\text{ion}} = \frac{4.411 \times 10^{19}}{(1 + (7.721 \times 10^{-3} (E/n))^{1.5})^{0.33}} \frac{\text{cm}^2 \text{V}^{-1} \text{s}^{-1}}{n}.
\]

The ion diffusion coefficient is expressed by the Einstein’s relation

\[
D^{\text{ion}}/\mu^{\text{ion}} = kT^{\text{ion}}/e \quad \text{(assuming ion temperature } T^{\text{ion}} = 0.025 \text{ eV)} \quad [17, 24].
\]

It can be noticed that the diffusion coefficients and mobility for electrons could be taken as constant values

\[
D^{\text{ele}} = 3 \times 10^{5} / p \quad \text{[cm}^2 \text{s}^{-1}], \quad \mu^{\text{ele}} = 3 \times 10^{5} / p \quad \text{[cm}^2 \text{V}^{-1} \text{s}^{-1}],
\]

respectively, as reasonable approximation [24, 25]. The constant value for the diffusion coefficient of metastable atoms in their parent gas is taken from [27]

\[
D^{\text{met}} = 54.9 \quad \text{[cm}^2 \text{s}^{-1}].
\]

The rate coefficients \( k_{1}^{\text{io}}, k_{2}^{\text{i0}}, k^{\text{ex}}, k^{\text{de}} \) are taken as functions of mean electron energy and obtained by fitting the data from [23]. These dependencies are similar to data from Bolsig+ program [28, 29]. The rate coefficients for the Penning ionization, two-body quenching in collisions with ground-state atoms and three-body quenching in collisions with two ground state atoms are constant [17, 27] and take the following values

\[
k^{\text{Pe}} = 6.2 \times 10^{-10} \quad \text{cm}^3 \text{s}^{-1}, \quad k^{\text{i0}} = 2.1 \times 10^{-15} \quad \text{cm}^3 \text{s}^{-1}
\]

and \( k^{\text{de}} = 1.1 \times 10^{-32} \quad \text{cm}^6 \text{s}^{-1} \), respectively. For the secondary electron yield the value of \( \gamma = 0.06 \) was used as in the papers [17, 23, 30], where the glow discharge in argon was modelled under the similar conditions.

4. NUMERICAL PROCEDURE

The equations (1-3, 6-9) are partial differential equations and the numerical procedure based on finite difference method is used for their solving. The distance between the electrodes is divided into the equal spatial intervals \( \Delta x \), thus
generating one dimensional spatial mesh. The values of variables are defined in \(i\) and \(i+1/2\) point of the mesh. The number of interior points of mesh is \(N\), while \(i = 0\) and \(i = N + 1\) are points at the boundaries. After discretization, the equations (6-9) have the form:

\[
\frac{n_{ele}^i - n_{ele}^j}{\Delta t} + \frac{\Gamma_{ele}^{j} - \Gamma_{ele}^{i+1/2}}{\Delta x} = k_{io}^{i} n_{ele}^{i} N + k_{io}^{j} n_{ele}^{j} n_{ele}^{i} + k_{pe} n_{ele}^{i} n_{ele}^{j}, \tag{12}
\]

\[
\frac{n_{ion}^{i} - n_{ion}^{j}}{\Delta t} + \frac{\Gamma_{ion}^{j} - \Gamma_{ion}^{i+1/2}}{\Delta x} = k_{io}^{i} n_{ion}^{i} N + k_{io}^{j} n_{ion}^{j} n_{ion}^{i} + k_{pe} n_{ion}^{i} n_{ion}^{j}, \tag{13}
\]

\[
\frac{n_{ele}^{i+1} - n_{ele}^{i}}{\Delta t} - D \frac{n_{ele}^{i+1} - n_{ele}^{i}}{\Delta x^2} = k_{en}^{i} n_{ele}^{i} n_{ele}^{i} - k_{de} n_{ele}^{i} n_{ele}^{j} + k_{eq}^{i} n_{ele}^{i} N - k_{eq}^{j} n_{ele}^{j} N^2, \tag{14}
\]

\[
\frac{(u_{ele} n_{ele}^{i})_{j+1} - (u_{ele} n_{ele}^{i})_{j}}{\Delta t} + \frac{5 \Delta x}{3} \left[ \Gamma_{ele}^{j+1/2} - \Gamma_{ele}^{j-1/2} \right] = -e \Gamma_{ele}^{i} E_{i} - u_{ion} n_{ele}^{i} + u_{eq} k_{de} n_{ele}^{i} - \frac{\varphi_{i+1/2}}{\Delta x^2}, \tag{15}
\]

In the equations (12-16) \(j\) is a time index while \(i\) is a spatial index, where \(j = 0, 1, 2, \ldots\) and \(i = 0, 1, \ldots, N + 1\). The time step \(\Delta t\) is the time between two time moments \(j\) and \(j+1\), while the spatial step \(\Delta x\) is the distance between \(i\) and \(i+1\) points of the mesh. Fluxes for electron and ion densities and electron energy flux are discretized by the exponential scheme of Scharfetter-Gummel [31, 32]:

\[
\Gamma_{ele, ion}^{i+1/2} = \frac{1}{\Delta x} \left[ n_{ele, ion}^{i+1/2} D_{ele, ion}^{i+1/2} \exp(M) - n_{ele, ion}^{i} D_{ele, ion}^{i} \exp(M) \right], \tag{17}
\]

\[
\Gamma_{ele, ion}^{i-1/2} = \frac{1}{\Delta x} \left[ n_{ele, ion}^{i-1/2} D_{ele, ion}^{i-1/2} \exp(M) - n_{ele, ion}^{i} D_{ele, ion}^{i} \exp(M) \right], \tag{18}
\]

where

\[
M_1 = \frac{-\mu_{ele, ion}^{i} + \mu_{ele, ion}^{j}}{D_{ele, ion}^{i} + D_{ele, ion}^{j}} (\varphi_{i+1/2} - \varphi_{i-1/2}),
\]

\[
M_2 = \frac{-\mu_{ele, ion}^{i} + \mu_{ele, ion}^{j}}{D_{ele, ion}^{i} + D_{ele, ion}^{j}} (\varphi_{i+1/2} - \varphi_{i-1/2}).
\]
Flux for electron energy has the same form like fluxes for electron and ion. By inserting the relations (17, 18) into the equations (12, 13, 15) we get:

\[
\begin{align*}
\Delta t A_1 n^{ele}_{i,j+1} &+ (\Delta x + \Delta t A_2) n^{ele}_{i,j+1} + \Delta t A_3 n^{ele}_{i-1,j+1} = \Delta x n^{ele}_{i,j} + \Delta t \Delta k n^{ele}_{i,j} N + \\
&+ \Delta x \Delta k n^{ele}_{i,j} n^{met}_{i,j} + \Delta x \Delta t \Delta k \phi n^{met}_{i,j} n^{met}_{i,j}, \\
\Delta t B_1 n^{ion}_{i,j+1} &+ (\Delta x + \Delta t B_2) n^{ion}_{i,j+1} + \Delta t B_3 n^{ion}_{i-1,j+1} = \Delta x n^{ion}_{i,j} + \Delta t \Delta k n^{ion}_{i,j} n^{ele}_{i,j} N + \\
&+ \Delta x \Delta k n^{ion}_{i,j} n^{met}_{i,j} + \Delta x \Delta t \Delta k \phi n^{met}_{i,j} n^{met}_{i,j},
\end{align*}
\]

where

\[
A_1 = \frac{-M_1 \left( \frac{1}{\Delta x} \right) D^{ele}_{i,j+1}}{\exp(M_1) - 1},
\]

\[
A_2 = \frac{M_1 \left( \frac{1}{\Delta x} \right) D^{ele}_{i,j+1}}{\exp(M_1) - 1} \cdot \frac{M_2 \left( \frac{1}{\Delta x} \right) D^{ele}_{i,j+1}}{\exp(M_2) - 1},
\]

\[
A_3 = \frac{-M_2 \left( \frac{1}{\Delta x} \right) D^{ele}_{i,j+1}}{\exp(M_2) - 1} \cdot \exp(M_2).
\]

The coefficients \( B_1, B_2, B_3 \) for ions have the same form as \( A_1, A_2, A_3 \) for electrons. After arranging, the equation for metastable state (14) and Poisson’s equation (16) take the form:

\[
\begin{align*}
-\Delta t D^{met} n^{met}_{i,j+1} &+ (\Delta x^2 + 2 \Delta t D^{met}) n^{met}_{i,j} - \Delta t D^{met} n^{met}_{i-1,j+1} = \Delta x^2 n^{met}_{i,j} + \\
&+ \Delta t \Delta x^2 k^{met}_{i,j} n^{ele}_{i,j} N - \Delta t \Delta x^2 k^{met}_{i,j} n^{ele}_{i,j} n^{met}_{i,j} - \Delta t \Delta x^2 k^{met}_{i,j} n^{ele}_{i,j} n^{met}_{i,j} - \\
&- 2 \Delta t \Delta x^2 k^{met}_{i,j} n^{met}_{i,j} - \Delta t \Delta x^2 k^{met}_{i,j} n^{met}_{i,j} N - \Delta t \Delta x^2 k^{met}_{i,j} n^{met}_{i,j} N^2, \\
\phi_{j+1,j+1} - 2 \phi_{j+1,j} + \phi_{j-1,j+1} = - \left( \Delta x \right)^2 \frac{e}{\varepsilon_0} \left( n^{ion}_{i,j} - n^{ele}_{i,j} \right).
\end{align*}
\]
The procedure for solving equations (12-16) consists of solving the Poisson’s equation, the continuity equations for electrons, ions and metastable atoms and at the end the energy balance equation for electrons. The solving procedure for equations (1-3) is identical as in the case of equations (6-9). The equations (19, 20, 21, 25, 26) are the system of linear equations which are solved by Thomas’s algorithm [33].

5. BOUNDARY AND INITIAL CONDITIONS

In our extended fluid model it is assumed that ion number density on the electrodes satisfies Neumann boundary conditions \( \frac{dn^{ion}}{dt} = 0 \). In order to determine the electron number densities at the cathode, the secondary electron yield \( \gamma \) is included into the boundary condition \( \Gamma^{ele} = -\gamma \Gamma^{ion} \), which is further substituted into the continuity equations for electrons (1, 6). The value of electron number densities at the anode is \( n^{ele} = 0 \). The number density of metastable state on both electrodes is assumed to be equal to zero. Boundary condition for electric potential at the cathode is \( V_c = 0 \) and at the anode \( V_a \) is equal to the applied voltage. The value for the mean electron energy at cathode is taken to be 5 eV as in the paper [23]. Since the emission of electrons occurs in the process of Auger neutralization, this energy can be determined from the difference \( u^{ion} - 2w_i \), where \( u^{ion} \) is the ionization energy of argon atom and \( w_i \) is the work function of the cathode material. As the number density of electrons at the anode is equal to zero, then the mean electron energy at the anode is taken to be 0 eV [13, 23]. The initial value of mean electron energy in the model is 1 eV [13, 14, 23], while the values of \( 10^3 \text{ cm}^{-3} \) are used for the initial number densities of particles.

6. RESULTS AND DISCUSSION

Numerical modelling based on one dimensional extended fluid model was carried out for the voltage range 300 V – 700 V, the pressure \( p = 1.33 \text{ mbar} \) and the inter-electrode distance \( d = 1 \text{ cm} \). This analysis is valid for the parallel plate electrodes with diameters much larger than the inter-electrode space. The detailed analysis for voltages of 300 V and 500 V, without and with metastable state will be presented. The mobility for ions are expressed by the relations (11) and diffusion coefficients by the Einstein’s relation, while these coefficients for electrons are taken as the function of mean electron energy [23].
The number density profiles of electrons, ions and metastable state, mean electron energy and ionization source in stationary state of glow discharge at voltage of 300 V are shown in the figure 1. The cathode fall region is shown in the figure 1c. The electron energy has the maximum value in this area (figure 1c), leading to the highest production of electrons and ions in the inter-electrode space (figure 1b). In the case when metastable state is included in the model, the production of electrons and ions is enhanced due to stepwise and Penning ionization. However, these processes have a lower contribution to the electron production than the electron impact ionization in the cathode fall area (figure 1b). The electron and ion number density profiles without and with metastable state are of similar shape and their maximum values are approximately equal (figure 1a).

When the voltage is 500 V (figure 2), the cathode fall and the mean electron energy are higher than in previous case. The maximum values of number densities of electrons and ions are higher than in the case when applied voltage was 300 V due to increased production of electrons and ions. From the figure 2b it can be noticed that in cathode fall region the influence of stepwise and Penning ionization is significant and production of electrons and ions from these process is comparable with production from direct electron impact ionization. Because of the
increased importance of these two processes the maximum number density of the electrons and ions obtained from the model with metastable state is higher than without them (figure 2a), which was not the case when applied voltage was 300 V.

Fig. 2 – Calculated profiles for a) electron and ion number densities, b) ionization source, c) mean electron energy and potential and d) number density of metastable state, at the voltage of 500 V.

Fig. 3 – Ratio of maximum values of number density with and without metastable state as a function of voltage for the voltage range 300–700 V.
The ratio of the maximum values of electron and ion number densities without and with metastable states as a function of the applied voltage is shown in figure 3. It can be noticed that this ratio is increasing as voltage increases, which is the consequence of enhanced Penning and stepwise ionization. It should be noted that the production of metastable state of argon Ar \((^3P_2)\) can be controlled by changing the working conditions (e.g. voltage, pressure, etc.), as well as by adding the quenching gases (e.g. N\(_2\), Cl\(_2\), etc.) [34, 35, 36].

7. CONCLUSION

Fluid models are often used for modelling the processes in various types of gas discharges. In this paper the extended fluid model is applied which is solved by the finite difference method. The system of equations obtained by the discretization of fluid equations is solved by the Thomas’s algorithm. The detailed numerical scheme describing numerical procedure used in solving governing equations is presented. The results of one dimensional extended fluid model for argon DC glow discharge without and with Ar \((^3P_2)\) metastable state accompanied by detailed analysis for 300 V and 500 V voltages are presented. The fluid model without metastable state includes only the process of electron impact ionization, thus neglecting the Penning and stepwise ionization. For the lower voltages the difference between two cases is neglectable and maximum number densities of electrons and ions are approximately the same. With the increase of the voltage, the number densities obtained in the case when metastable state is included are slightly higher compared to the ones without metastable state (due to Penning and stepwise ionization), but they remain of the same order of magnitude. Therefore, the model without metastables can give estimation of number densities, but the one with included metastable state can separate the contributions of different physical processes and mechanisms and more properly describes the characteristics of gas discharges.

Acknowledgments. The authors are grateful to the Ministry of Education, Science and Technological development of the Republic of Serbia for the financial support (projects 171025 and 174015).

REFERENCES