The role of metastables in the formation of an argon discharge in a two-pin geometry

Citation for published version (APA):

DOI:
10.1109/TPS.2010.2056934

Document status and date:
Published: 01/01/2010

Document Version:
Publisher’s PDF, also known as Version of Record (includes final page, issue and volume numbers)

Please check the document version of this publication:
• A submitted manuscript is the version of the article upon submission and before peer-review. There can be important differences between the submitted version and the official published version of record. People interested in the research are advised to contact the author for the final version of the publication, or visit the DOI to the publisher’s website.
• The final author version and the galley proof are versions of the publication after peer review.
• The final published version features the final layout of the paper including the volume, issue and page numbers.

Link to publication

General rights
Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

• Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
• You may not further distribute the material or use it for any profit-making activity or commercial gain
• You may freely distribute the URL identifying the publication in the public portal.

If the publication is distributed under the terms of Article 25fa of the Dutch Copyright Act, indicated by the “Taverne” license above, please follow below link for the End User Agreement:
www.tue.nl/taverne

Take down policy
If you believe that this document breaches copyright please contact us at:
openaccess@tue.nl
providing details and we will investigate your claim.

Download date: 08. Jun. 2020
The Role of Metastables in the Formation of an Argon Discharge in a Two-Pin Geometry

Ana Sobota, Freddy Manders, E. M. van Veldhuizen, Jan van Dijk, and Marco Haverlag

Abstract—The breakdown process in gases is a versatile research topic. Numerous processes play more or less important roles in discharge formation, strongly depending on the gas mixture, the electrode configuration, the applied electric field, the size of the geometry, and even on the structures surrounding the active volume where the breakdown takes place. We focus our research on the breakdown process in argon at 700 mbar, in a pin–pin (point-to-point) electrode geometry, with increasing positive voltage at the active electrode. The voltage rises by 100 V/ns. We use a 2-D fluid model to examine the formation of a charged channel between the electrodes under given conditions. We find that the results describe previous experiments reasonably well. We also explore the role of excited argon atoms at (4s) metastable levels in the breakdown process, and we conclude that the ionization path with an intermediate step containing the metastables does indeed make a notable difference in the breakdown process.

Index Terms—Argon, atmospheric pressure, breakdown, fluid model, gas discharges, modeling, plasmas.

I. INTRODUCTION

ELECTRICAL discharges in gas (gas discharges) are widely used in the modern society. They have been studied in different ways—both fundamental and practical knowledge has been obtained to satisfy both the industrial needs [1]–[5] and the needs of the scientific community [6]–[8]. The lighting industry drives extensive research efforts in this field [9], both application driven and the research into fundamental processes. A lot has been understood thus far, but even more new questions have been raised. In particular, the breakdown process in lamps is still not completely understood [10]. It has been researched and modeled for low-pressure lamps [11], [12], experimental studies have been carried out for high-pressure lamps [3], [4], and the modeling was done for the breakdown process in metal halide lamps with low noble gas pressure [13]–[16].

Modern high-intensity discharge (HID) lamps are used in many ways; all of the applications require high efficacy, good color rendering, and high-energy output from a pea-sized volume. For that reason, the lamps are filled with noble gas at high pressure (0.1–12 bar) and salts. The noble gas provides a lot of light during the warm-up phase of operation, and the salts take over the light emission after they have evaporated. Due to the high pressure of the noble gas, the lamps are difficult to ignite. It typically takes a pulse between 5 and 20 kV to build a conductive channel in the gas volume between the electrodes, while the latter stage of lamp operation requires only a fraction of that value.

Attempts have been made to lower the breakdown voltage in HID lamps. This can be accomplished in several ways. Lay et al. [13], [14] showed that a small amount of Hg can improve the HID breakdown characteristics, i.e., lower the breakdown voltage, thanks to the Penning process. Bhoj et al. [15], [16] showed that small admixtures of Xe in an Ar atmosphere also help lower the breakdown voltage due to the lower ionization potential of Xe, while the electron energy distribution function is not significantly altered with respect to the pure-Ar atmosphere. These modeling efforts have described the ignition process in noble gas pressures ranging from 10 to 90 torr (13–120 mbar), where the discharge is glowlike and not constricted. Research in the field of high pressures of noble gases has been mainly experimental [4], [17]–[19], and it has been shown that, due to high voltages, the criterion for glow-to-streamer transition is fulfilled and that the discharges forming under these conditions are filamentary streamerlike ionizing channels. The experiments have also shown that the breakdown voltage rises with pressure and that the speed of propagation of the discharge rises with applied voltage and decreases with the increase in pressure.

Recently Wendt et al. [19] qualitatively described the discharge in Xe at 0.1–5 bar at an electrode distance of 5 mm. The pulse rise rate was varied between 10 and 1000 V/ns. In their research, they used a 1-D fluid model with a standard part where they solved Poisson’s equation, the species continuity equation, and the energy balance equation. The electron transport and reaction rate coefficients were functions of local electric field. The starting particle densities were quite low ($10^8$ m$^{-3}$ for electrons and $10^{12}$ m$^{-3}$ for atomic ions), but they corresponded to the particle densities in reality. The model showed an ionization front propagating toward the cathode, with its speed and breakdown voltage being highly dependent on the pulse rise rate. The electric field at the discharge tip was on the order of magnitude of $10^6$ V/m.

The aim of this paper is to show the importance of metastables in near-atmospheric argon discharge, because the
ionization paths containing metastable states are sometimes neglected in the models, irrespective of the noble gas or the complexity of the chemistry used. To this end, we characterized the breakdown process between two pin electrodes in 700-mbar argon atmosphere. We focused on the breakdown process during a rising voltage profile (the ramp in the rest of this paper), like the one used for the experiments described in [18]. The reference measurements for such a system gave a breakdown voltage of around 14 kV [20] and a discharge speed between $2.5 \times 10^5$ and $3 \times 10^5$ m/s [18]. We found that our results are in reasonable agreement with the experiments. Next, we examined the influence of atomic metastables on discharge formation. We concluded that, even though they did not play a decisive role, the inclusion of processes of atomic metastable ionization did make a notable difference in the timing of the discharge evolution.

II. DESCRIPTION OF THE MODEL

The plasma that is characterized by this model is not in thermal equilibrium; during breakdown, the mean electron energy can reach several electron volts, while the temperatures of ions and neutrals stay close to room temperature. In fact, given the high gas pressure, the temperature, and the low ionization degree of the gas, its density and temperature are assumed constant throughout the discharge evolution in this model. The species described by the equations in the following section are electrons, atomic and molecular ions ($\text{Ar}^+$ and $\text{Ar}_2^+$), and atomic metastables ($\text{Ar}^*$). The plasma is described in a 2-D cylindrically symmetric fluid model. We used a limited set of reactions to characterize the discharge formation, including direct ionization of argon atoms and ionization via the $(4s)$ metastable levels, ionization and production of molecular ions through heavy-particle collisions, and two- and three-particle recombination.

The model used in this paper is a part of the Plasimo modeling toolkit [21] and was originally developed by Hagelaar and Kroesen and documented in [22]. It was used to describe plasmas in display devices. The model was further developed by Brok et al. for the description of breakdown in fluorescent tubes and described in [11] and [12]. We adapted parts of the model to characterize a breakdown event in a high-pressure system; the details of the model are discussed in the following.

A. Fluid Model

A fluid model describes a physical system by using balance equations for particles and energy. The balance equation for particle species $p$ in terms of species density $n_p$, flux $\Gamma_p$, and source term $S_p$ as a function of time and space

$$\frac{\partial n_p}{\partial t} + \nabla \cdot \Gamma_p = S_p.$$ 

(1)

Source term $S_p$ depends on the volume reactions included in the model. It consists of positive contributions from reactions that create new particles of type $p$, and of negative contributions from reactions in which the corresponding particles are destroyed

$$S_p = \sum_r c_{r,p} R_{r,p} = \sum_r \left[ c_{r,p} k_r \prod_i n_i \right].$$

(2)

The total number of particles $p$ created in a reaction of type $r$ is marked by $c_{r,p}$, and $R_{r,p}$ is the associated reaction rate. The reaction rate is a product of reaction coefficient $k_r$ and densities of the reacting species $n_i$.

The flux in (1) is given by the momentum transport equation, approximated by the drift–diffusion equation

$$\Gamma_p = \pm \mu_p E n_p - D_p \nabla n_p.$$ 

(3)

The first term is the drift flux, with particle mobility $\mu_p = e/(m_p \nu_m)$, and $\nu_m$ is the frequency of momentum transfer collisions in interactions with the background gas. The $\pm$ sign in the drift term is positive for the positively charged particles and negative for the negatively charged ones in the model. The second term is the diffusive flux, with $D_p = u_p^2/(3\nu_m)$ as the diffusion coefficient of species $p$.

The temperature of the neutral species in the model (namely, argon metastables) is assumed to be the same as that of the surrounding gas. The ion temperature $T_i$ is related to the background gas temperature $T_g$ and to the electric field $E$ as follows [23]:

$$k_B T_i = k_B T_g + \frac{m_i + m_g}{5m_i + 3m_g} m_g (\mu_i E)^2.$$ 

(4)

The ion mobilities $\mu_{i,\text{ion}}(E/p)$ are given in a table, and the diffusion coefficients are calculated according to the Einstein relation (Table I)

$$D_p = \frac{k_B T_i \mu_p}{e}.$$ 

(5)

The same assumptions for the temperature do not typically hold for electrons, because of their small mass, which causes poor energy transfer in electron–neutral collisions. The parameters concerning electrons are instead given as a function of local mean electron energy; the same holds for the reaction coefficients, because electrons are considered the most energetic particles in reactions in which they take part. The local mean electron energy is given by the energy balance equation

$$\frac{\partial (n_e \epsilon)}{\partial t} + \nabla \cdot \Gamma_\epsilon = -e \Gamma_e \cdot E + S_\epsilon.$$ 

(6)

The electron energy density is related to the electron density by $n_\epsilon = \pi n_e$. The first term on the right-hand side represents...
the heating by the electric field. The second term is the energy gained or lost in reactions and is given by

$$S_r = - \sum_r c_r \varepsilon_r R_r.$$  \hfill (7)

The aforementioned summation is performed only over reactions with electrons as among the reacting species. The value $\varepsilon_r$ is the energy gained or lost in reaction $r$.

The electron temperature can be related to its energy by $T_e = (3/5) \varepsilon_e$. We use the Einstein relation to calculate the diffusion coefficient for electrons as well.

The mean energy flux in (6) is given by

$$\Gamma_r = -\frac{5}{3} \mu_e E n_e \varepsilon_e - \frac{5}{3} n_e D_e \nabla \varepsilon_e.$$  \hfill (8)

The second term in the aforementioned equation is the heat conduction flux, which is proportional to the mean electron energy gradient.

The electric field in the system is calculated by solving Poisson’s equation

$$\nabla \cdot \epsilon \nabla \phi = -\nabla \cdot \epsilon E = - \sum_p q_p n_p.$$  \hfill (9)

The boundary conditions used in the model have been described in detail by Hagelaar [24]. Here, we will present them in short. For the boundary conditions for Poisson’s equation, we use the electrode potentials. For particle densities $n_p$’s and electron energy density $n_e$, we employ homogenous Neumann boundary conditions at open boundaries and at the symmetry axis

$$\nabla n_p \cdot e_n = 0 \quad \nabla n_e \cdot e_n = 0$$  \hfill (10)

where $e_n$ is the unit vector normal to the boundary. At the electrode surface, we do not define reflection or secondary electron emission coefficients, and the boundary conditions are the same for all species

$$\Gamma_p \cdot e_n = (2a - 1) \text{sgn}(q) \mu_p E \cdot e_n + \frac{1}{2} v_{th} n_p$$  \hfill (11)

$$a = \begin{cases} 1, & \text{sgn}(q) \mu E \cdot e_n > 0 \\ 0, & \text{sgn}(q) \mu E \cdot e_n \leq 0. \end{cases}$$  \hfill (12)

The coefficient $a$ is set to one if the drift velocity of the given species has a component directed toward the boundary; otherwise, it is set to zero.

The model uses the control volume method for solving equations for densities of the species, electron energy density, and electrostatic potential. We use a uniform 2-D grid with rectangular elements $\Delta x$ and $\Delta y$ in size. Each cell is defined as either a part of the plasma region or an electrode element, in which case we assign a voltage to the cell. Poisson’s and transport equations are solved in the plasma region of the defined grid.

<table>
<thead>
<tr>
<th>No.</th>
<th>Reaction</th>
<th>Reaction coefficients $k_r$</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>$e + Ar \rightarrow e + Ar^*$</td>
<td>BOLSIG+</td>
<td>[25]</td>
</tr>
<tr>
<td>2</td>
<td>$e + Ar^* \rightarrow 2 e + Ar^+$</td>
<td>BOLSIG+</td>
<td>[25]</td>
</tr>
<tr>
<td>3</td>
<td>$e + Ar \rightarrow 2 e + Ar^+$</td>
<td>BOLSIG+</td>
<td>[25]</td>
</tr>
<tr>
<td>4</td>
<td>$Ar^+ + 2 Ar \rightarrow Ar^+_2 + \gamma$</td>
<td>$2.5 \times 10^{-13} m^3 s^{-1}$</td>
<td>[27]</td>
</tr>
<tr>
<td>5</td>
<td>$2 Ar^+ \rightarrow Ar^+_2 + Ar^+$</td>
<td>$1.2 \times 10^{-13} m^3 s^{-1}$</td>
<td>[27]</td>
</tr>
<tr>
<td>6</td>
<td>$e + Ar^* \rightarrow e + Ar$</td>
<td>BOLSIG+</td>
<td>[25]</td>
</tr>
<tr>
<td>7</td>
<td>$2 e + Ar^+ \rightarrow e + Ar$</td>
<td>$5.4 \times 10^{-59} m^3 s^{-1}$</td>
<td>[28]</td>
</tr>
<tr>
<td>8</td>
<td>$e + Ar^+ \rightarrow e + Ar + h\nu$</td>
<td>$1 \times 10^{-17} m^3 s^{-1}$</td>
<td>[28]</td>
</tr>
<tr>
<td>9</td>
<td>$Ar^+_2 + e \rightarrow Ar^+ + 2 e$</td>
<td>$2.496 \times 10^{-58} m^3 s^{-1}$</td>
<td>[29]</td>
</tr>
<tr>
<td>10</td>
<td>$e + Ar^+_2 \rightarrow Ar^* + Ar$</td>
<td>$7 \times 10^{-13} (300K/T_e) m^3 s^{-1}$</td>
<td>[27]</td>
</tr>
</tbody>
</table>

**B. Input Data**

The model described in this paper deals with a pure-argon discharge. Given high pressure, low ionization degree, and very quick timing (our process develops over about 150 ns), the density and temperature of the ground-state argon atoms are considered constant. We describe the time evolution of electrons, argon atomic and molecular ions ($Ar^+$ and $Ar_2^+$), and argon metastables ($Ar^*$) representing the $4s$ atomic metastable states.

The reactions initially included in the model are given in Table II. We included the reactions that are commonly considered in simpler argon models. After careful consideration of results and testing, we concluded that, due to the very short development time of the discharge (around 150 ns) and low particle densities, the recombination reactions made no difference in the model. As a consequence, we decided to the present results using a reduced model, which considers only reactions 1–6 in Table II. The rates for excitation and ionization of argon atoms, as well as for collisional deexcitation of metastable levels, were obtained from Bolsig+ [25] as a function of mean electron energy. Bolsig+ was designed for use in fluid modeling of discharges in a large range of conditions, making the data that we needed for our model available. The amount of metastables in the mixture for Bolsig+ calculations was assumed to be zero.

We have tested the model with tables generated for a metastable amount equal up to $10^{-4}$ with respect to the amount of ground-state atoms in the gas and obtained exactly the same results. In reality, as will be shown in the next section, the average metastable density in the gas mixture is at least five orders of magnitude lower than the neutral gas density. Reaction rates calculated by Bolsig+ and the reaction rate for molecular ion dissociative recombination are given as a function of local mean electron energy. They are shown in Fig. 1. The other reaction coefficients are constant and were taken from the literature.

**C. Description of the System Under Consideration**

The system under consideration is a model representation of the setup used in the experiments where we measured the speed of discharge in argon [18]. We use a cylindrically symmetric geometry to describe a system of two electrodes (0.06 mm in diameter and work function of 4.5 eV) in a 700-mbar argon atmosphere at 300 K. The distance between the electrode tips is 30 mm. An extra space of 10 mm is provided at the sides of
The electrodes in order to diminish the effects of computational boundary.

The computational domain consists of 1200 × 322 square grid cells, which are (0.03 × 0.03) mm² in size. We found that the optimal ratio between the cell sizes in the radial and axial directions is around one, even though the discharge propagates in the axial direction. This is due to the fact that the streamer that propagates through the gas volume has a certain width, which should be bigger than one cell size in the radial direction. The smallest radii measured in the experiments [30] were 0.1–0.2 mm wide, so a cell size of 0.03 mm in the radial direction should be sufficient. The cell size (therefore, the total number of cells) is chosen for optimal output and calculation time, considering the lack of dynamical grid refinement in the model.

In the experiment, we used tungsten-rod-shaped electrodes, which were 0.6 mm in diameter. However, in the model, the sharp edges of the electrodes cause effects that do not appear in the experiments. In the experiments, the discharge in argon in near-atmospheric pressure initiates at the tip of the electrode. Even if the electrode does not have a tip, there is always some asymmetry in the system, like slight misalignment of the electrodes, or, more commonly, microfractures that make the discharge favor one region or one spot on the electrode. The geometry in our model is 2-D cylindrically symmetric, which is why we cannot introduce any asymmetry. The modeled discharge starts at the region with highest electric field, which is at the electrode edge. However, being 2-D cylindrically symmetric, the point at the electrode edge in the model represents a ring at the electrode edge in reality. After such discharge initiation, we get propagation of the discharge in the direction of the externally applied electric field, where the particle densities describe a cylinder around the symmetry axis that diverges as it approaches the cathode, instead of a channel with highest density in its center, whose profile does not diverge. This is indeed an artifact that is not observed in the experiments. When obtaining the results that we will show in the next section, we used electrodes whose dimensions are 3 mm in length and just one cell (0.03 mm) in radius.

### Table III

<table>
<thead>
<tr>
<th>Species</th>
<th>Initial density $n_p$ [m⁻³]</th>
</tr>
</thead>
<tbody>
<tr>
<td>e</td>
<td>$10^9$</td>
</tr>
<tr>
<td>Ar⁺</td>
<td>0</td>
</tr>
<tr>
<td>Ar⁺⁺</td>
<td>$10^9$</td>
</tr>
<tr>
<td>Ar⁻</td>
<td>0</td>
</tr>
</tbody>
</table>

The voltage of the following form is applied at the electrodes [24]:

$$V_{\text{electrode}} = \frac{\partial V}{\partial t} t - I_{\text{electrode}} R_{\text{electrode}}$$  \hspace{1cm} (13)

$$I_{\text{electrode}} = \oint_{\text{electrode surface}} (\mathbf{j} \cdot E_{\text{surf}} - \epsilon \frac{\partial E}{\partial t} n) d^2 S$$  \hspace{1cm} (14)

where $\partial V/\partial t$ is the rise rate of the ramp; $t$ is the time; $I$ is the current through the electrode; $R$ is the resistance in series with the electrode, which is always present in the experiments; and $\mathbf{j}$ is the plasma current density (sum of all the fluxes of the charged species multiplied by the appropriate unit of charge). The second term in (14) is the displacement current. The cathode is connected to ground ($\partial V/\partial t = 0$ V/ns) with a resistance of $R = 10 \, \Omega$, and there is a ramp applied to the anode ($V_R = 100$ V/ns). The serial resistance at the anode is set to $R = 1000 \, \Omega$. These values were chosen to accurately describe the experimental conditions [18].

The starting conditions for the model are given in Table III. The starting particle densities are sufficiently low to represent the starting particle densities in a gas. We have tested the influence of starting particle densities up to $10^{12}$ m⁻³ for electrons and atomic argon ions. We found that the initial electron density governs the speed of the discharge growth (propagation): the higher the initial density, the lower the speed of propagation. The difference in maximum speed of the discharge between models with initial electron densities of $10^9$ and $10^{12}$ m⁻³ is 9%.

## III. Results

In this section, we present the results of simulations. First, we discuss the simulation of a 700-mbar discharge in argon between two pin electrodes at 3-cm distance. Afterward, we analyze the influence of reactions involving argon metastables on the overall process.

### A. Breakdown Process

The experimentally observed breakdown process in near-atmospheric pressure argon has been documented and described before [3], [17], [18], [30]. It features bright constricted channels propagating through the gas volume at high velocities. Branching occurs, and the path that the discharge takes is straight only in segments between the branching points. However, the general direction of the discharge is always in the direction of the externally applied electric field. The described appearance differs greatly from the glowlike discharges at lower pressures. The explanation for the narrow channel profile is...
that, at high pressures, the density of the background gas is quite high (1.7 $\times$ 10$^{25}$ m$^{-3}$ in our case of 700 mbar); the creation and destruction processes in plasma are thereby localized to a small space at the maximum of the reduced electric field. At the beginning of the discharge formation, the electric field maximum is at the electrode tip, and during the propagation, the maximum is at the discharge tip due to the space charge in the head of the discharge.

Our model, being 2-D cylindrically symmetric, cannot simulate the branching of the discharge, or its erratic path, as observed in the experiments [17], [30]. We do, however, show the propagation of a thin channel along the direction of the electric field. Fig. 2 shows a 2-D distribution of argon metastable density during the propagation of the discharge. The metastable density profile cannot be directly compared to the experimental observations because, in the experiments, one observes a light emitted from the gas volume, not particle density. However, the light is emitted in radiative recombination or relaxation of excited atoms in plasma, and the light intensity is proportional to the density of the species involved. Visible and near-visible lights that come from relaxation of neutral atoms are most frequently observed in the experiments, which is why we choose to show the density of metastable atoms.

In the figure, there are two discharge formations growing from the electrode tips. At the cathode, there is a glowlike structure of lower particle density. At the anode side, first, there is charge accumulation at the electrode tip, followed by fast growth of a thin channel of high particle density, directed toward the cathode. Please note that the metastable density in the figure is presented in logarithmic scale in order to better show the details of the discharge formations forming between the electrodes. For the same reason, the ratio of the units of distance on the $x$- and $y$-axes in the figure is not unity; each part of the figure shows a (30 mm) $\times$ (0.96 mm) part of the modeled discharge between the electrode tips and starting from the symmetry axis. In the figure, the discharge speeds up on its way through the gas—an effect previously observed in the experiment [18].

The figure shows that the channel propagating from the anode reaches the glowlike cloud around the cathode very shortly after 13 kV, i.e., 130 ns after the beginning of the simulation. The actual voltage breakdown takes place 134 ns after the beginning of the simulation, as shown in Fig. 3. During the few remaining nanoseconds, a high-particle-density channel is formed in the remaining space between the electrodes close to the cathode tip, and fast charge multiplication takes place until the moment when the current through the anode is high enough to cause a drop in the anode voltage, as predicted by (13) and shown in Fig. 3.

Fig. 3 shows the average density of particles in the whole computational volume for all particle types and as a function of time. The voltage at the anode is also shown as a function of time. The average densities in the model depend on the size of the computational volume; even so, the main contributor to the values described earlier is the ionized channel that propagates through the gas, because densities in the channel are orders of magnitude larger than that in the other regions of the computational volume. In other words, we can track the evolution of the species in the ionized channel by observing the average particle densities in the whole volume as a function of time.

During the first 7 ns of the simulation, the electron density stays at the initial value, while the density of atomic ions slightly drops, mainly due to inelastic collisions with the background gas, in which molecular ions are produced (reaction 4 in Table II). As a result, the molecular ion density sharply increases. Argon metastables exhibit a somewhat slower increase, but their density quickly reaches the initial electron density. When the densities of all particle types reach approximately 10$^9$ m$^{-3}$, a steep increase in all densities occurs. The same
behavior and timing are observed for every combination of initial particle densities below $10^{12}$ particles/m$^3$, which is the highest initial density that we have tested. During this steep increase, particle buildup takes place at the electrode tips. During this time, a streamer has not yet begun to form.

At the end of the steep increase, at roughly 15 ns after the beginning of the simulation, the electron density reaches a value that is slightly above $10^{13}$ m$^{-3}$, the ion density is at $4.7 \times 10^{13}$ m$^{-3}$, the molecular ion is the lowest of the four at $5 \times 10^{12}$ m$^{-3}$ and the metastables are the most dense at $10^{14}$ m$^{-3}$. Around this time, a fast narrow channel starts to form. The increase in particle densities continues to be exponential but much slower.

All particle densities show further increase with time. The metastables remain the most dense species in the computational volume throughout the simulation. The densities of both ion species are at roughly the same level for most of the discharge propagation. Shortly before the breakdown moment, electron and atomic ion densities show a slightly faster increase. We define breakdown moment as the instance when the anode voltage starts to drop because of the increase in current, according to (13). With the given resistance parameters in the model (see Section III for details), the anode voltage starts to drop when the anode current reaches around 0.5 A. Breakdown happens around 134 ns after the beginning of the simulation at 13.4 kV.

Fig. 2 shows that the metastable density is highest at the symmetry axis, along which the discharge propagates. This is true for all particle species in the model. The densities of all particle types along the symmetry axis are shown in Figs. 4–7 for various points in time. Even though the metastables are the most dense species in the computational volume as a whole, electrons and atomic ions have the highest density at the symmetry axis, along the axis of the discharge. This means that the metastables are more diffuse around the charged channel than the electrons and atomic ions. The same is valid for molecular ions.
Fig. 8. Axial component of the electric field, at the symmetry axis, for different points in time during the discharge evolution. The position at the $x$-axis indicates the gap between the electrodes: The anode is at position zero, while the cathode is at 3 cm. Each line represents electron density at a given time in the discharge formation; lines are drawn 10 ns apart. The peaks are caused by the net charge at the streamer tip. The axial electric field strength at the peak increases with time, and the position of the peak moves toward the cathode at increasing speed.

There is a sharp increase in particle density at the discharge tip in all four graphs, as expected, and the density remains roughly constant throughout the body of the channel. The electron density shows a sharp fall near the cathode at the beginning of the discharge formation. This is due to the initial drift of electrons away from the cathode tip. A closer look at the densities at the discharge tip reveals additional differences in density distribution for the four species: While electron and atomic ion densities exhibit a sharp “corner” at the tip of the discharge, the densities of the other two species are more rounded, which is particularly true for the molecular ions. From this, we can conclude that the positive net charge at the tip of the streamer is largely contributed to by the atomic ions, even though the molecular ions do play a significant role in the chemistry of the whole discharge formation.

The positive net charge at the discharge tip causes a high peak in the axial electric field in front of the streamer tip. The axial component of the electric field along the symmetry axis is shown in Fig. 8.

B. Role of Metastables

We saw already in Fig. 3 that the average density of argon metastables in the model rises above the level of any other species. Figs. 4–7 show that the density of the metastable argon atoms in the charged channel is also not negligible. However, the role of metastables in discharge formation is sometimes downplayed or even neglected. Here, we would like to prove their importance in the breakdown process.

To that end, we made a simple adaptation of our standard model described in the previous section. We removed the two ionization reactions that convert argon metastables to atomic ions (reactions 2 and 5 from the tables of reactions in II) while leaving everything else the same. Argon metastables are still created in the model by excitation of the ground-state atoms and recombination of the molecular ions, and they still relax back to the ground state by inelastic collisions with electrons.

Figs. 9–12 show the average particle densities as a function of time for all four species and compare them to the densities in the standard model. The figures show that the breakdown moment in the adapted model comes approximately 41 ns after the breakdown in the standard model; this also means that the breakdown voltage is roughly 4.1 kV higher in the model without $\text{Ar}^* \rightarrow \text{Ar}^+$ reactions (breakdown occurs at 17.5 kV). A duration of 41 ns is quite a large difference within the timescales that we are considering: The discharge in the adapted model takes about 31% more time to develop than the discharge in the standard model.

Surprisingly, the evolution of the density of argon metastables is almost the same for the two models. However, electron and atomic ion densities are quite different in the two
Fig. 11. Average molecular ion density as a function of time for two different simulations. The solid curve — denotes the molecular ion density in the standard model, and the dashed curve — shows the average density in the simulation without ionization of argon metastables. The curves stop at respective breakdown moments.

Fig. 12. Average metastable density as a function of time for two different simulations. The solid curve — denotes the metastable density in the standard model, and the dashed curve — shows the average density in the simulation without ionization of argon metastables. The curves stop at respective breakdown moments.

Simulations. A steeper increase with time is exhibited in average electron and Ar\(^{+}\) densities in the standard model. The density profile of Ar\(^{+}\) ions is slightly lower in the adapted model; the main production channel for this particle type is in collisions of atomic ions and background gas (see Table II), so the decrease in the adapted model is mainly the result of atomic ion density decrease.

Given the effect that the removal of Ar\(^{*}\) → Ar\(^{+}\) reactions has on average electron and Ar\(^{+}\) densities, we expect an impact on density profiles along the symmetry axis for the same species. Figs. 13 and 14 show the behavior of electron and atomic ion densities in the charged channel as a function of time. In quite the same manner as that in the standard model, steep rises in densities are present at the tip of the charged channel propagating between the electrodes. The electron density stays within the same order of magnitude throughout the channel, like that in the standard model. The main effect of the adapted model is that the peak value of electron density is an order of magnitude lower than that in the standard model. The Ar\(^{+}\) density profile shows a significant decrease in density throughout the charged channel, which is roughly two orders of magnitude for a channel at 145 ns after the beginning of the simulation.

Fig. 13 shows the velocities of the discharges in the two simulations as a function of position in the discharge gap. The velocities were calculated from the positions of the discharge tip as a function of time. For the purposes of this calculation, we define discharge tip as the position of the peak of the axial component of the electric field, resulting from the net charge at the head of the charged channel. Velocity is calculated as the first time derivative of the peak position. Both velocities rise with time, but the maximum velocity of the discharge in the standard model is 27% higher than that of the discharge in the model without ionization of argon metastables.
The experiments done under similar conditions [18] have shown that discharge velocities increase on their way through the gas, but the increase observed in the experiments was much slower than the one obtained by both simulations. The velocity in the experiment increases from $2.3 \times 10^5$ m/s at 0.4 cm from the anode to $3 \times 10^5$ m/s at 2.4 cm from the anode. One must remember that the experimentally determined velocities were obtained from fast photography measurements of the discharge, in visible and near-visible regions. As stated before in this section, what we observe in the experiments is a light emitted mainly from the excited argon atoms that relax to lower the atomic states. In the experiments, we cannot determine the position of the peak net-charge density. On the other hand, in our model, we cannot calculate the position in the channel where there is enough visible light emitted for an iCCD camera to register. This is one of the reasons why the velocity profile along the discharge path is not the same in the experiment and in the model. Another probable reason for the discrepancy between the modeled and the measured velocity is that, in the experiments, it is nearly impossible to do measurements in pure gas. Experiments in pure gases typically contain traces of nitrogen and oxygen, and it has already been established by Aleksandrov et al. [31] that even a very small amount of oxygen in the discharge changes the dynamics of discharge development, drastically reduces the conductivity of the streamer channel, and increases the average electric field required to bridge the discharge gap by a streamer. The proposed mechanism for this effect was quenching of excited argon atoms by oxygen molecules, which leads to the deceleration of two-step ionization in the streamer channel in argon and to the slowing down of discharge growth.

IV. DISCUSSION AND CONCLUSION

In this paper, we have modeled the prebreakdown events in 700-mbar argon, using a 2-D fluid model. Our aims were to make an overview of the processes that take place during the initiation and propagation of a high-pressure argon discharge and to determine the role of atomic metastables in the breakdown process.

The experiments have shown that the discharge in 700-mbar argon forms thin channels that propagate from the anode to the cathode at high velocities [17], [18], [30]. The breakdown voltage for the conditions that we simulated was measured at 14 kV [20]. We show the propagation of the discharge as calculated by our model. The discharge propagates from the anode toward the cathode in a narrow charged channel with a positive net charge at its tip. The net charge is a result of charge separation in the head of the channel, and it causes a high peak in the electric field. There is a glowlike formation growing around the cathode tip, as previously observed by Sobota et al. [17] and Briels [30], with particle densities that are orders of magnitude lower than that in the propagating channel.

We have not implemented photoionization in our model; the discharge forms and grows solely due to background ionization. We start with a uniform electron density of $10^9$ m$^{-3}$. At the cathode side, during the initiation stage of discharge development, the electrons drift away from the electrode tip, causing a sharp drop of electron density, as shown in Figs. 4 and 13. At the anode side, charge multiplication and accumulation take place, and the accumulated charge modifies the electric field at the tip, causing the electrons around the electric field peak to gain energy and engage in ionization processes. As a result, the discharge starts growing in the direction of the cathode, driven by the high electric field peak at its tip.

The growth velocity of the discharge increases with time, like that observed in the experiments [18]. The increase in discharge velocity is a direct consequence of the increase of electric field strength at the discharge tip, and the two mechanisms that are responsible for the increase of electric field are as follows.

1) A high electric field causes a sharp increase in local ionization rates by electron impact at the discharge tip, which follows from the table of reactions II and reaction coefficients in Fig. 1. High ionization rates cause further increase in particle densities, higher net charge at the discharge tip, and, thus, further increase of the electric field. This, in turn, facilitates further increase in ionization, higher electric field, and so on.

2) Also, the simulation involves the potential at the anode, which is rising at 100 V/ns. This causes extra increase of the electric field imposed from the electrical part of the setup.

The two mechanisms work toward higher ionization rates at the discharge tip and an increase in discharge speed. One could say that, after inception, the discharge pushes itself forward by causing favorable conditions for ionization processes at its tip.

Figures featuring particle densities in the standard model show that the density of argon metastables is quite high in the charged channel, and it is highest when it is averaged over the whole computational volume. Given this high density, we wanted to test the importance of argon metastables in discharge formation. The processes that ionize argon metastables were taken out of the model, and the results of the adapted model were compared to that of the standard simulation. Averaged over the whole volume, the metastable and molecular ion densities are almost the same in the two simulations, even though we expected the metastable density to be visibly affected by the
adaptation. Careful consideration reveals that the process that collisionally deexcites the metastable atoms back to the ground state is the most efficient process at low electron energies in the model (see Table II and Fig. 1), which could explain no extra metastables in the adapted model, where Ar* is not used to produce atomic ions. The effect is, however, visible in electron and atomic ion densities, which are lower in the adapted model. This is due to the fact that, in the adapted model, atomic ions are no longer produced from the metastable atoms.

In addition, the discharge in the adapted model took 31% more time to reach breakdown; this also shows in its velocity. The speed of the propagating discharge in the standard model is roughly twice as high as the speed of the discharge in the adapted model. The mean value of velocity obtained in the standard model with Ar* → Ar+ reactions is comparable to the velocity obtained in the experiments.

The significant difference in particle densities and the discharge speed in the two simulations leads to the conclusion that the metastable levels are indeed an important step in the ionization process in near-atmospheric-pressure argon. This ionization path is responsible for an order-of-magnitude increase in electron and atomic ion densities at the discharge axis and for a 27% increase of the maximum propagation speed.

The model predicts the discharge behavior quite accurately, but there is more room for improvement. First, there is a problem with the size of computational cells in the model. If the cells are too big in the direction of the propagation of the discharge, the streamer head is not properly described. For example, errors in electric field can easily occur. On the other hand, if the computational cells are too big in the direction perpendicular to the propagation of the discharge, the charged channel will be too wide, or the radial profile of the channel will be poorly described, and the discharge will again not be properly characterized. An implementation of local dynamical grid refinement, which is a technique already implemented in many simulations, would solve our problem of cell size while keeping the requirements on computer memory and computational time at a reasonable level.

The second improvement that we would suggest is to describe the electrodes in an analytical way instead of assigning them grid cells. This is a technique already used by Celestin et al. [32]. In this way, we would avoid the artificial enhancement of electric field at the rough corners, which are not present in reality. These artificial electric field peaks can cause side discharges to appear at the electrode tips, which would not have been there if the electrode surface was smooth. One can rightfully argue that side discharges appear in reality as well, but in a cylindrically symmetric geometry of our model, side discharges are really “umbrellalike” structures rather than thin ionized channels. This technique of implementing the physical boundaries in an analytical way could also be considered for the implementation of dielectric boundaries in the model.

One could argue that the chemistry used in the model is too simple to conclusively show the importance of (4s) metastable states in the development of a near-atmospheric-pressure argon discharge. We did consider possible additions to the chemistry. As already stated in Section II, the consideration of various recombination processes leads us to the conclusion that, in a short time frame and low densities in which our discharge develops, the recombination processes play no role in discharge formation. Additional processes of metastable ionization, such as metastable–metastable associative collisional ionization, could make a slight difference in metastable and ion densities; however, this would not change the overall conclusion that the ionization path of argon atoms using metastables as an intermediate step does make a notable difference in the modeled breakdown process. If anything, the extra ions produced via this additional process of metastable ionization would make our point even clearer. A possible improvement to the chemistry in our model would be the addition of another excited species, which would represent the (4p) and higher excited states in argon. In this way, we could couple the higher excited argon atoms in the model with the other species and more correctly determine the influence of excited species on discharge formation.

In conclusion, this paper has summarized the results of a 2-D fluid model of a discharge in 700-nbar argon. We have used a two-pin geometry and a ramp at the anode to describe the initiation and growth of the discharge. The results of the model are in good agreement with that of the experiments previously performed. We have also demonstrated the importance of the role of argon atoms in metastable (4s) states during the breakdown process.

REFERENCES


Ana Sobota, photograph and biography not available at the time of publication.

Freddy Manders, photograph and biography not available at the time of publication.

E. M. van Veldhuizen, photograph and biography not available at the time of publication.

Jan van Dijk, photograph and biography not available at the time of publication.

Marco Haverlag, photograph and biography not available at the time of publication.

This article has been accepted for inclusion in a future issue of this journal. Content is final as presented, with the exception of pagination.