Simulation of charged and excited particle transport in the low-current discharge in argon-mercury mixture

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Abstract. Simulation of the electron, ion and metastable excited atom transport in the argon-mercury mixture low-current discharge is fulfilled. Distributions of the particle densities along the discharge gap under different mixture temperatures are obtained and it is demonstrated that the principal mechanism of mercury ion generation is the Penning ionization of mercury atoms by argon metastables, which contribution grows sharply with the mixture temperature due to mercury density increase. Calculations show that the mercury and argon ion flow densities near the cathode are of the same order already under the relative mercury content of about $10^{-4}$ corresponding at the argon pressure $10^3$ Pa to the mixture temperature 30°C. Therefore, at the room temperature the electrodes of mercury illuminating lamps at the stage of their ignition are sputtered predominantly by mercury ions.

1. Introduction
Mixtures of argon with mercury vapor are widely used in gas-discharge illuminating lamps [1, 2]. After lamp turning on, the gas breakdown occurs under the applied voltage and the low-current (Townsend) discharge is initiated between the electrodes, which then transits to the glow and arc modes [3, 4]. Electrons are accelerated by the electric field from the cathode to the anode, ions are accelerated in the opposite direction and excited atoms diffuse in the gas in the direction of their density reduction. As a result of the cathode bombardment by ions, electrons necessary to maintain the discharge are emitted from its surface and the cathode material sputtering takes place, which terminates the electrode lifetime [1, 5, 6]. In the argon-mercury mixture, besides of direct ionization of gas atoms by electrons, ionization of mercury atoms in collisions with metastable excited argon atoms (the Penning reaction) proceeds as well [1, 2]. Therefore, the charged and excited particle densities and flows in the interelectrode gap depend substantially on the mixture temperature, as the density of saturated mercury vapor grows with its increase. But the influence of the temperature on the transport of charged and excited particles in the argon-mercury mixture low-current discharge plasma under conditions typical for high pressure illuminating lamps at the stage of ignition is studied insufficiently. In particular, the gas ionization coefficient and contributions of the main types of particle interactions

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in it under different relative mercury contents in the mixture and electric field strength values were found in [7], where the electron density was obtained by fitting the results of calculations to experimental data of Ref. [8] and the densities of ions and excited atoms were estimated from their balance equations averaged over the discharge volume. The charged particle distributions in the lamp tube were computed in [9] under specific discharge conditions with using of the macroscopic description of their motion.

In this work, simulation of electron, ion and metastable excited atom motion and interactions in a low-current discharge between the flat electrodes in argon-mercury mixture is fulfilled. Computation of electron trajectories is carried out by the Monte Carlo method [3-5], whereas ion and metastable excited atom motion is described on the basis of their transport equations. An influence of the mixture temperature on the particle densities and flows in the interelectrode gap is studied.

2. Mathematical model
Let the low-current discharge be burning in the argon-mercury mixture between the flat parallel electrodes operating as the cathode and the anode, which dimensions substantially exceed the distance \( d \) between them. Suppose that axis \( z \) of the coordinate system is directed perpendicularly to the electrodes, and the cathode and the anode coincide with planes \( z = 0 \) and \( z = d \), respectively. In the low-current mode, which can exist at quite low discharge current density \( j \) values, the space charge in the interelectrode gap is negligible and does not distort the external electric field in it, which is directed along \( z \)-axis and equals to \( E = U / d \) in each point, where \( U \) is the discharge voltage [5].

Simulation of electron motion in the gas is carried out with the Monte Carlo method. Calculation of the trajectories of electrons emitted from the cathode (the primary electrons) is executed at each time step, which value is taken small enough to ensure the length of the trajectory section passing by an electron during this time being much less than its mean path length between collisions with gas atoms. The probability of an electron-atom collision at the trajectory section, its type, as well as the electron direction of motion and energy after the collision, are obtained from the corresponding expressions of the collision theory with using of random numbers [10-14]. The trajectory of an electron emitted from the cathode is calculated until it reaches the anode or returns to the cathode. Motion of the secondary electrons generated in the interelectrode gap due to ionization of atoms by electrons and metastables is simulated in a similar way. As a result, the electron velocity distribution function \( f_z(z, v, v_z) \) is found in each of \( s \) subintervals of the length \( \Delta z = d / s \), into which the interelectrode distance is divided, where \( z_i \) is the coordinate of the central point of the \( i \)-subinterval, \( v \) and \( v_z \) are the electron velocity and its \( z \)-component.

Main types of heavy charged particles in the low-current discharge in argon-mercury mixture under the pressure \( 10^3 - 10^4 \) Pa at the room temperature, typical for high-pressure mercury lamps, are atomic ions of the both gases \( \text{Ar}^+ \) and \( \text{Hg}^+ \), whereas the density of metastable argon atoms \( \text{Ar}^* \) and \( \text{Hg}^* \) exceeds considerably the density of atoms excited at the resonant levels (\( \text{Ar}^* \) и \( \text{Hg}^* \)) [7, 9].

The following particle interactions are taken into account:
- electron elastic scattering on an atom;
- electron ionization of a ground-state atom
  \[ \text{Ar} + e \rightarrow \text{Ar}^+ + 2e, \]  
  \[ \text{Hg} + e \rightarrow \text{Hg}^+ + 2e; \]  
- electron excitation of a ground-state atom at the metastable and resonant levels
  \[ \text{Ar} + e \rightarrow \text{Ar}^* + e, \]  
  \[ \text{Ar} + e \rightarrow \text{Ar}'' + e, \]  
  \[ \text{Hg} + e \rightarrow \text{Hg}^* + e, \]  
  \[ \text{Hg} + e \rightarrow \text{Hg}'' + e; \]
- ionization of a metastable atom

\[
\text{Ar}^* + \text{Ar}^* \rightarrow \text{Ar}^+ + \text{Ar} + e, \quad (7)
\]

\[
\text{Hg}^* + \text{Hg}^* \rightarrow \text{Hg}^+ + \text{Hg} + e; \quad (8)
\]

- Penning ionization

\[
\text{Ar}^* + \text{Hg} \rightarrow \text{Ar} + \text{Hg}^+ + e; \quad (9)
\]

- metastable quenching

\[
\text{Ar}^* + \text{Ar} \rightarrow 2\text{Ar}, \quad (10)
\]

\[
\text{Hg}^* + \text{Ar} \rightarrow \text{Hg} + \text{Ar} \quad (11)
\]

\[
\text{Hg}^* + \text{Hg} \rightarrow 2\text{Hg}. \quad (12)
\]

At high electric field strength values typical for the low-current discharge, the charged particles move in the gas in the drift mode [5], and the main mechanism of excited atom motion is diffusion in the direction of their density reduction. Therefore, their transport equations can be presented as follows:

\[
\frac{d}{dz}\left(n_{\text{Ar}^*} - \mu_{\text{Ar}^*} E + \Delta n_{\text{Ar}^*} + k_1 n_{\text{Ar}^*}^2 = 0, \right. \quad (11)
\]

\[
\frac{d}{dz}\left(n_{\text{Hg}^*} - \mu_{\text{Hg}^*} E + \Delta n_{\text{Hg}^*} + k_2 n_{\text{Hg}^*}^2 + k_3 n_{\text{Ar}^*} n_{\text{Hg}} = 0, \quad (12)
\]

\[
D_{\text{Ar}^*} \frac{d^2 n_{\text{Ar}^*}}{dz^2} + \Delta n_{\text{Ar}^*} - 2k_1 n_{\text{Ar}^*}^2 - k_2 n_{\text{Ar}^*} n_{\text{Hg}} - k_4 n_{\text{Ar}^*} n_{\text{Ar}^*} - k_6 n_{\text{Hg}} n_{\text{Hg}} = 0, \quad (13)
\]

\[
D_{\text{Hg}^*} \frac{d^2 n_{\text{Hg}^*}}{dz^2} + \Delta n_{\text{Hg}^*} - 2k_2 n_{\text{Hg}^*}^2 - k_5 n_{\text{Ar}^*} n_{\text{Ar}^*} - k_6 n_{\text{Hg}} n_{\text{Hg}} = 0, \quad (14)
\]

where \( n_{\text{Ar}^*}, n_{\text{Hg}^*}, n_{\text{Ar}^*} \) and \( n_{\text{Hg}} \) are the densities of argon ions, mercury ions, argon metastables and mercury metastables, \( \mu_{\text{Ar}^*} \) and \( \mu_{\text{Hg}^*} \) are the argon and mercury ion mobilities, \( D_{\text{Ar}^*} \) and \( D_{\text{Hg}^*} \) are the argon and mercury metastable diffusion coefficients, \( k_1 - k_6 \) are the constants of interactions (7) – (12). The number of \( p \)-type particles (\( \text{Ar}^+, \text{Hg}^+, \text{Ar}^* \text{Hg}^* \)) generated in a unit volume in a unit time in collisions of electrons with atoms of the mixture \( l \)-component (\( \text{Ar}, \text{Hg} \)) equals to

\[
\Delta n_p = n_l \int \sigma_l^p (v) f_e(z,v,v_z) v dv d v_z, \quad \text{where} \quad \sigma_l^p (v) \text{ is the cross-section of generation of a } p \text{-type particle in collision of an electron having energy } v \text{ with an atom of the mixture } l \text{-component having density } n_l \text{ (reactions (1) – (6))}. \]

Boundary conditions for the equations are zero values of the ion densities at the anode and the metastable atom densities at the both electrodes:

\[
n_{\text{Ar}^*} (d) = n_{\text{Hg}^*} (d) = 0, \quad n_{\text{Ar}^*} (0) = n_{\text{Ar}^*} (d) = n_{\text{Hg}^*} (0) = n_{\text{Hg}^*} (d) = 0. \quad (15)
\]

Solution of the system (11) - (14) with the conditions (15) is found numerically by the finite difference method. Then the number of electrons generated in a unit volume in a unit time by argon metastables in reactions (7) - (9) is calculated

\[
\Delta n_e = k_4 n_{\text{Ar}^*}^2 + k_5 n_{\text{Hg}^*}^2 + k_6 n_{\text{Ar}^*} n_{\text{Hg}}; \quad (16)
\]

and the corresponding number of the secondary electrons, which must be added to the primary electrons in each subinterval of the length \( \Delta z \), is found.

After that, simulation of the electron velocity distribution function is carried out again taking into account the secondary electrons and the ion and metastable transport is computed. This cycle is repeated until the relative differences of values obtained in two successive iterations become sufficiently small.
3. Results and Discussion
Calculations have been performed for the discharge in the interelectrode gap of the length \( d = 1.5 \times 10^{-3} \) m under the sustaining voltage \( U = 255 \) V in the mixture of argon with the constant density \( n_{\text{Ar}} = 6.58 \times 10^{23} \text{ m}^{-3} \) corresponding to its pressure 2660 Pa at the temperature 293 K and saturated mercury vapor, which relative density \( n_{\text{Hg}}/n_{\text{Ar}} \) takes values in the interval \( 10^{-7} - 10^{-3} \) under temperature variation from -30 to 60 C [15] (this corresponds to the typical situation when the lamp bulb is filled with argon with an addition of liquid mercury and is sealed off indoor at the room temperature, and then the lamp operates outdoor at different temperatures). The current density value \( j \approx 10^{-5} \) A m\(^{-2}\) is supposed, ensuring the low-current discharge mode under the indicated conditions [5].

In the process of electron motion simulation with the Monte Carlo method, the interelectrode distance and the intervals of \( v \) and \( v_z \) variation are divided into 100 subintervals, the number of primary electrons is \( 10^4 \), their initial energies are considered as uniformly distributed in the range from 0 to 4 eV and the initial direction of motion is assumed to be isotropic. The cross-sections of the main types of electron collisions with argon and mercury atoms presented in [16-18] are used, as well as the constants \( k_1 - k_6 \) of the heavy particle interactions (7) – (12), the ion mobilities and the metastable atom diffusion coefficients taken from [9, 13, 19-24]. The calculated dependences of the gas ionization coefficient and the electron drift velocity on the electric field strength in pure argon and mercury, as well as the dependence of the ionization coefficient on the relative content of mercury in the mixture are in agreement with experimental data [14].

![Figure 1](image-url)  
**Figure 1.** Distributions of the electron, ion and metastable densities in the discharge gap at different mixture temperature values: -30 C (a), 0 C (b), 30 C (c) and 60 C (d).
In figure 1 the obtained distributions of the charged and excited particle densities between the electrodes at different temperature values are presented. It follows from it that the density of electrons is exponentially increased from the cathode to the anode as a result of generation of secondary electrons in ionizing collisions (1) – (2) and (7) – (9), and its value grows substantially with the temperature because of more intensive Penning ionization at higher mercury vapor densities (this fact can be ascertained by comparison of figures 1c and 2, in which distributions of the particle densities at the temperature 30°C, calculated with and without taking into account the Penning ionization (9), are shown). Argon metastables arise under excitation of argon atoms by electrons (in reaction (3)) and are destroyed mainly in the Penning and quenching reactions, whereas diffusion is of no importance for them. Therefore, distribution of their density is similar to that of electrons and its value is varied insignificantly with temperature increase. Mercury metastables appear under electron excitation of mercury atoms and are lost due to diffusion to the electrodes, therefore, their density distribution in the central area of the discharge gap is quite uniform. Argon ions are produced under ionization of argon atoms by electrons, whereas the dominant mechanism of mercury ion generation is the Penning ionization of mercury atoms by argon metastables, and ions of the both types are lost mainly due to their drift to the cathode under the influence of the electric field. Therefore, the mercury ion density, which at the temperature -30°C is by a factor of a hundred lower than the argon ion density, grows considerably with the temperature and at 60°C becomes comparable with the argon ion density.

As a result, the mercury and argon ion flow densities at the cathode are of the same order already under the relative mercury content in the mixture of about $10^{-4}$, corresponding to the mixture temperature 30°C (see figure 3). Because the mean path length of a mercury ion in the mixture between the resonant charge exchanges on parent gas atoms is much more than that of an argon ion, the energies of mercury ions exceed considerably the energies of argon ions, and they make the main contribution to the cathode sputtering, which reduces the lamp service time [1].

**4. Conclusions**

In this work, simulation of the electron, ion and metastable excited atom transfer in a low-current discharge in the argon-mercury mixture between the flat electrodes is carried out. Calculation of electron trajectories is performed with the Monte Carlo method, and motion of ions and metastable excited atoms is described on the basis of their transport equations, which are solved by the finite difference method. Distributions of the particle densities along the discharge gap are calculated under...
different mixture temperature values, and the main mechanisms of their generation and loss are determined. It is shown that the principal mechanism of mercury ion production is the Penning ionization of mercury atoms by argon metastable excited atoms, and its contribution grows sharply with the mixture temperature. It follows from the obtained results that the mercury and argon ion flow densities near the cathode are of the same order already under the relative mercury content of about $10^{-4}$, corresponding at the argon pressure $10^3$ Pa to the mixture temperature 30 C. Therefore, under the room temperature the electrodes of mercury illuminating lamps at the stage of their ignition are sputtered predominantly by mercury ions.

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