Simulation of cathode surface sputtering by ions and fast atoms in Townsend discharge in argon-mercury mixture with temperature-dependent composition

Bondarenko G.G.[1], Kristya V.I.[2], Savichkin D.O.[2], Żukowski P.[3]

[1]*National Research University Higher School of Economics,*
Myasnitskaya str., 20, Moscow 101000, Russia

[2]*Bauman Moscow State Technical University, Kaluga Branch,*
Bazhenov str., 2, Kaluga 248000, Russia

[3]*Lublin University of Technology,*
Nadbystrzycka str., 38D, Lublin 20-001, Poland

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Abstract

The mixture of argon and mercury vapor is used as the background gas in different types of gas discharge illuminating lamps. The aim of this work was development of a model, describing transport of electrons, ions and fast atoms in the one-dimensional low-current gas discharge in argon-mercury mixture, and determination of the dependence of their contributions to the cathode sputtering, limiting the device service time, on the temperature.

For simulation of motion of electrons we used the Monte Carlo method of statistical modeling, whereas the ion and metastable excited atom motion, in order to reduce the calculation time, we described on the basis of their macroscopic transport equations, which allowed to obtain their flow densities at the cathode surface. Then, using the Monte Carlo method, we found the energy spectra of ions and fast atoms, generated in collisions of ions with mixture atoms, at the cathode surface and also the effective coefficients of the cathode sputtering by each type of particles.

Calculations showed that the flow densities of argon ions and fast argon atoms, produced in collisions of argon ions with slow argon atoms, do not depend on the temperature, while the flow densities of mercury ions and fast argon atoms generated by them grow rapidly with the temperature due to an increase of mercury content in the mixture.

There are represented results of modeling of the energy spectra of ions and fast atoms at the cathode surface. They demonstrate that at low mercury content in the mixture of the order of 10^{-3} the energies of mercury ions exceed that of the other types of particles, so that the cathode is sputtered mainly by mercury ions, and their contribution to sputtering is reduced at a mixture temperature decrease.

Keywords: gas discharge lamp, low-current discharge, argon-mercury mixture, ion and atom energy spectra, cathode sputtering.

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Адрес для переписки:
Кристя В.И.
Московский государственный технический университет имени Н.Э. Баумана, Калужский филиал,
ул. Баженова, 2, г. Калуга 248000, Россия
e-mail: kristya@bmstu-kaluga.ru

Address for correspondence:
Kristya V.I.
Bauman Moscow State Technical University, Kaluga Branch,
Bazhenov str., 2, Kaluga 248000, Russia
e-mail: kristya@bmstu-kaluga.ru

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Моделирование распыления поверхности катода ионами и быстрыми атомами в таунсендовском разряде в смеси аргон-ртуть с зависящим от температуры составом

Бондаренко Г.Г.[1], Кристя В.И.[2], Савичкин Д.О.[2], Жуковский П.[3]

[1]Национальный исследовательский университет «Высшая школа экономики», ул. Мясницкая, 20, г. Москва 101000, Россия

[2]Московский государственный технический университет им. Н.Э. Баумана, Калужский филиал, ул. Баженова, 2, г. Калуга 248000, Россия

[3]Люблинский технологический университет, ул. Надбыстрицкая, 38Д, г. Люблин 20-001, Польша

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Смесь аргона и паров ртути используется в качестве рабочего газа в различных типах газоразрядных осветительных ламп. Целью данной работы являлось построение модели, описывающей перенос электронов, ионов и быстрых атомов в слаботочном разряде в смеси аргон-ртуть, а также определение зависимости их вкладов в распыление катода, ограничивающее срок службы прибора, от температуры.

Для моделирования движения электронов мы применяли метод статистического моделирования Монте-Карло. Перенос ионов и возбужденных атомов с целью сокращения затрат расчетного времени описывали на основе макроскопических уравнений, что позволило найти плотности их потоков у поверхности катода. Затем с использованием метода Монте-Карло находили энергетические спектры ионов и быстрых атомов, образующихся при столкновениях ионов с атомами смеси, у поверхности катода, а также эффективные коэффициенты распыления катода каждым типом частиц.

Расчеты показали, что плотности потоков ионов аргона и быстрых атомов аргона, возникающих при столкновениях ионов аргона с медленными атомами аргона, не зависят от температуры, в то время как плотности потоков ионов ртути и быстрых атомов аргона, образуемых ими, быстро возрастают при увеличении температуры вследствие увеличения содержания ртути в смеси.

Представлены результаты моделирования энергетических спектров ионов и быстрых атомов у поверхности катода. Они демонстрируют, что при малом содержании атомов ртути в смеси порядка 10^{-3} распыление катода происходит, главным образом, ионами ртути, так как их энергии существенно превосходят энергии других типов частиц, причем их вклад в распыление уменьшается со снижением температуры смеси.

Ключевые слова: газоразрядная осветительная лампа, слаботочный разряд, смесь аргон–ртуть, энергетические спектры ионов и атомов, распыление катода.

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Адрес для переписки:
Кристя В.И.
Московский государственный технический университет имени Н.Э. Баумана, Калужский филиал, ул. Баженова, 2, г. Калуга 248000, Россия
e-mail: kristya@bmstu-kaluga.ru

Address for correspondence:
Kristya V.I.
Bauman Moscow State Technical University, Kaluga Branch, Bazhenov str., 2, Kaluga 248000, Russia
e-mail: kristya@bmstu-kaluga.ru

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Introduction

In different types of gas discharge illuminating lamps, the mixture of argon and mercury vapor is used as the background gas [1, 2]. The argon atom number density in it does not depend on the temperature, whereas the mercury atom number density is decreased under its reduction. The most intensive sputtering of the cathode surface in the discharge proceeds directly after the lamp ignition, because its lifetime in the continuous operation mode exceeds considerably that in the periodic turning on and off mode [3]. The mercury ion flow density near the cathode surface at the stage of lamp turning on should increase with the ambient temperature due to rising of the mercury atom number density in the discharge volume. Moreover, in the argon-mercury mixture, besides of the direct ionization of gas atoms by electrons, ionization of mercury atoms by metastable exited argon atoms takes place (the Penning reaction) [4–6], which also increases the mercury ion number density. Therefore, at quite small mercury content in the mixture, its ions can make a significant contribution to the lamp electrode sputtering.

The distributions of ions and fast atoms by energy in gas discharges, as well as the cathode sputtering by them, were studied in a number of works both experimentally and theoretically. However, only discharges in pure noble gases or their mixtures with temperature-independent composition were usually investigated in them [7–15]. For the discharge in the argon-mercury mixture with the temperature-dependent composition and under the existence of the Penning ionization of mercury atoms, though, this question was studied insufficiently. The distributions of ions of the mixture components by energy in the low-current discharge was calculated only in [16] on the basis of the approximation of continuous slowing down of mercury ions in argon without taking into account the stochastic nature of ion-atomic collisions. Besides, the energy spectrum of fast atoms, generated under the charge exchange and elastic scattering of ions on slow argon atoms, which can contribute substantially to the cathode sputtering, was not found in [16].

In this work, a model describing the ion and fast atom motion in the low-current (Townsend) discharge in an argon-mercury mixture, based on the Monte Carlo method, is used. The energy distributions of ions and fast atoms at the cathode surface are calculated and their contributions to its sputtering are found as functions of the mixture temperature.

Mathematical model

Let the gap of length d between the parallel planar electrodes with the large transverse dimensions be filled with a mixture of argon with density n_{Ar} and saturated mercury vapor with density n_{Hg}, and the voltage U sufficient for ignition of the Town-send discharge is applied to it. If the z-axis is directed along the normal to the electrode surfaces, then, since the space charge is rather small in such discharge [16], the electric field in all points is parallel to axis z and its strength is equal to $E = U/d$.

Electrons generated in the discharge gap under the ionization of atoms of the mixture components are accelerated by the field in the direction of the anode, and ions are accelerated to the cathode, colliding with neutral atoms. Simulation of their transport is fulfilled in this work on the basis of the hybrid discharge model [17, 18]. At the first stage, motion of primary electrons (emitted from the cathode and produced in electron-atomic collisions) is calculated using the Monte Carlo method, whereas the ion and metastable excited atom motion, in order to reduce the calculation time, is described on the basis of their macroscopic transport equations. As a result of their solution by the finite-difference method, numbers Δn_e of electrons, appearing in a unit time in a unit volume in collisions of heavy particles, are found in each of s intervals of length $\Delta z = d/s$, into which the interelectrode gap is divided. Then the corresponding numbers of secondary electrons, which should be added in the cells of the length Δz under modeling of the electron kinetics by the Monte Carlo method, are found. After that, simulation of electron motion in the discharge gap is performed again taking into account the additional electrons, as well as calculation of the ion and metastable transport. Such cycle is repeated until the relative difference between values of the quantities in successive iterations becomes sufficiently small. As a result, numbers $\Delta n_{i\mathrm{Ar}}$ and $\Delta n_{i\mathrm{Hg}}$ of argon and mercury ions, appearing in a unit time in a unit discharge volume in each of s intervals of length Δz, are obtained, as well as the argon and mercury ion flow densities J_{Ar^+} and J_{Hg^+} at the cathode surface.

At the second stage, the energy spectra of ions and fast atoms, produced in collisions of ions with argon atoms, at the cathode surface are found using the Monte Carlo method and the obtained earlier values of $\Delta n_{i\mathrm{Ar}}$ and $\Delta n_{i\mathrm{Hg}}$. In the process of calculation, it is being taken into account that when an ion collides with an atom of its parent gas, the resonant charge exchange can occur, resulting in generation of a slow ion with zero energy and a fast atom with the energy equal to the ion energy before the charge exchange. Besides of the charge exchange, the elastic scattering of ions and fast atoms on slow atoms can take place, in which they lose a fraction of their energy and slow atoms become fast ones, i. e. as a result of each elastic collision a new fast atom is produced. Between collisions with slow atoms the fast atoms move rectilinearly and uniformly.

Since the relative content of mercury in the discharge at the stage of lamp ignition is usually small ($n_{Hg}/n_{Ar} = 10^{-5} - 10^{-2}$) [6, 18], only collisions of ions and fast atoms with argon atoms can be taken into account. The trajectory of each fast heavy particle is calculated starting from the point of its formation by solving the equation of its motion sequentially at each time step Δt. The step value is chosen small enough so that the length of the trajectory section passed by the particle during time Δt is much less than its mean path length between collisions with atoms. Whether the particle collides with an atom in such section, its type, as well as the direction of motion and energy ε of the particle after the collision, are determined from the corresponding formulas of the collision theory with using of random numbers [12]. The energy dependencies of the cross sections of argon ion charge exchange on argon atom and isotropic elastic scattering of argon ion and atom on argon atom, taken from [19], are used, as well as the cross section of isotropic elastic scattering of mercury ion on argon atom, found using of the argon and mercury atom gas-kinetic radii. The trajectory of each ion is calculated until it reaches the cathode, and the trajectory of each fast atom is calculated until it reaches the cathode or its energy becomes less than 10 eV, since such atoms do not contribute to the cathode sputtering.

As a result of calculations, the energy distribution functions of ions and fast atoms $f_{Ar^+}(d,\varepsilon)$, $f_{Hg^+}(d,\varepsilon)$ and $f_{Ar}(d,\varepsilon)$ at the cathode surface are formed. Using them, the effective (averaged over particle energies) coefficients of the cathode surface sputtering by each type of particles are found, defined by expressions [16]:

$$R_{Ar^+} = \int_{\varepsilon_{tAr}}^{eU} Y_{Ar^+}(\varepsilon) f_{Ar^+}(d,\varepsilon)\,d\varepsilon;$$
$$R_{Hg^+} = \int_{\varepsilon_{tHg}}^{eU} Y_{Hg^+}(\varepsilon) f_{Hg^+}(d,\varepsilon)\,d\varepsilon; \qquad (1)$$
$$R_{Ar} = \int_{\varepsilon_{tAr}}^{eU} Y_{Ar}(\varepsilon) f_{Ar}(d,\varepsilon)\,d\varepsilon;$$

where $Y_{Ar^+}(\varepsilon)$, $Y_{Hg^+}(\varepsilon)$ and $Y_{Ar}(\varepsilon) = Y_{Ar^+}(\varepsilon)$ are the yields of cathode material sputtering by argon and mercury ions and fast argon atoms with energy ε, ε_{tAr} and ε_{tHg} are the corresponding threshold sputtering energies, e is the elementary charge. Then the flow densities of cathode material atoms, sputtered by different types of particles, are obtained as follows:

$$J_{sAr^+} = R_{Ar^+} J_{Ar^+}\ ; \quad J_{sHg^+} = R_{Hg^+} J_{Hg^+}\ ; \quad J_{sAr} = R_{Ar} J_{Ar}, \qquad (2)$$

where J_{Ar} is the fast argon atom flow density at the cathode, which is expressed via J_{Ar^+} and J_{Hg^+} with using of the results of simulation of ion and fast atom motion in the discharge.

Results and discussion

Calculations were performed for the low-current discharge in the interelectrode gap of the length $d = 10^{-3}$ m. It was considered to be filled with a mixture of argon and mercury, in which the number density of argon atoms was assumed to be constant and equal to $n_{Ar} = 6.6\cdot10^{22}$ m^{-3}, which corresponds to its pressure $p = 266$ Pa at the room temperature, whereas the mercury atom number density was dependent on the temperature T [18]. The discharge voltage was equal to 200 V, so that value of the reduced electric field strength E/n in the discharge was $3\cdot10^{-18}$ Vm^2 (where $n = n_{Hg} + n_{Ar}$). In the process of simulation, trajectories of 10^6 argon and mercury ions were calculated with using of the coordinates of their generation, found taking into account the calculated distributions of quantities Δn_{iAr} and Δn_{iHg} along the discharge gap at $s = 100$. After that, the trajectories of fast atoms, produced in collisions of ions and generated earlier fast atoms with slow argon atoms, were calculated.

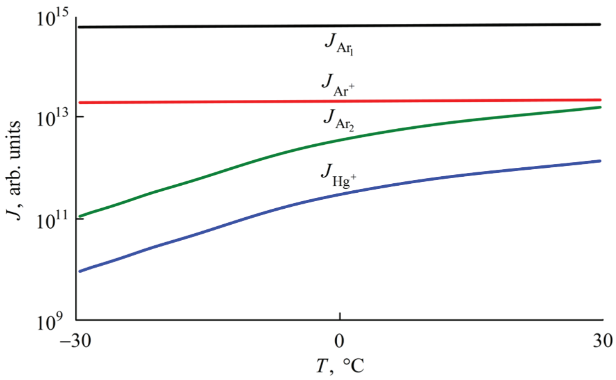

Figure 1 – The flow densities of argon ions (Ar^+), mercury ions (Hg^+), fast argon atoms, produced by argon ions (Ar_1), and fast argon atoms, produced by mercury ions (Ar_2), at the cathode surface

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The obtained flow densities of ions and fast atoms, bombarding the cathode surface, as function of the mixture temperature are presented in Figure 1. It is seen that the flow densities of argon ions and fast argon atoms, arising in collisions of argon ions with slow argon atoms, do not depend on the temperature, whereas the flow densities of mercury ions and fast argon atoms, produced by them, grow rapidly with temperature due to rising of the mercury content in the mixture by three orders of value at a temperature increase from –30 °C to +30 °C [18].

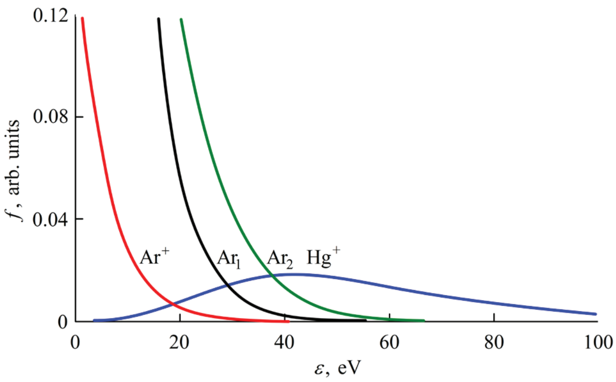

Figure 2 – The energy spectra of ions and fast atoms at the cathode surface. Designations are the same as in Figure 1

The calculated energy distributions of argon ions, mercury ions and fast argon atoms at the cathode surface are presented in Figure 2. It can be seen in it that most of mercury ions have energies exceeding considerably that of argon ions, because they lose only a fraction of their energy in elastic collisions with argon atoms, whereas argon ions lose all their energy under the charge exchanges on slow argon atoms, and the elastic scattering cross section is less than the charge exchange cross section.

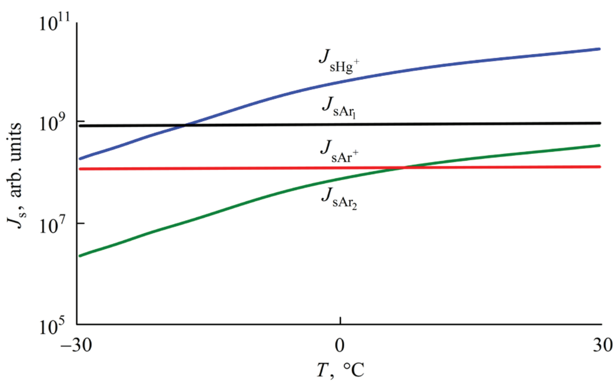

Figure 3 – The flow densities of tungsten atoms, sputtered from the cathode surface by ions and fast atoms. Designations are the same as in Figure 1

It follows from results of calculations that at a temperature increase the energies of mercury ions, as well as the energies of argon ions, are changed insignificantly, because their energy losses occur in collisions of ions with argon atoms, which number density does not depend on the temperature. It is seen also in Figure 2 that under the charge exchange and elastic scattering a large number of fast argon atoms are generated, and mercury ions make a main contribution to this process as their substantial fraction has energies exceeding that of argon ions. In Figure 3, the temperature variation of the flow densities of atoms, sputtered from the tungsten cathode surface by different types of particles, obtained with using of expressions (1), (2) and the experimental dependencies $Y_{Ar^+}(\varepsilon)$ and $Y_{Hg^+}(\varepsilon)$ [20], are shown. It can be seen that at low temperatures near –30 °C the main contribution to the cathode sputtering make fast argon atoms generated by argon ions. At temperatures exceeding 0 °C, though, the cathode is sputtered predominantly by mercury ions, because, as it follows from figures 1, 2, their flow density approaching that of argon atoms and their energies are considerably higher than the argon atom energies. Therefore, this factor must be taken into account under investigation of the cathode sputtering in gas discharge lamps.

Conclusion

In this work, simulation of transport of electrons, ions and fast atoms in the one-dimensional low-current gas discharge in argon-mercury mixture, used in gas discharge illuminating lamps, is fulfilled. It is taken into account that at the stage of lamp ignition mercury content in the mixture is small and collisions of fast heavy particles with argon atoms only can be considered. The main types of such collisions are the resonant charge exchange of argon ions on argon atoms and the elastic scattering of argon ions, mercury ions and fast argon atoms on slow argon atoms.

The flow densities of ions and fast atoms bombarding the cathode surface, their energy spectra, the effective rates of tungsten cathode sputtering by ions of both types and fast atoms, and also the flow densities of atoms, sputtered from the cathode by them, are found as functions of the temperature. It is shown that at low mercury content in the mixture of the order of 10^{-3} energies of mercury ions exceed that of the other types of particles, so that the cathode is sputtered mainly by mercury ions, and their contribution to sputtering is decreased at a mixture temperature reduction.

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