Numerical simulation of the plasma parameters of a low-pressure arc discharge in helium

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Abstract. A global integral model of an arc discharge in a helium medium with a sputtered graphite anode is presented. The main feature of the model is the simultaneous consideration of the plasma of the discharge gap, the cathode and anode layers, the current transfer, the thermal regime of the electrodes, and the evaporation of the anode. Both the calculated and experimental results show that the discharge voltage linearly increases with the inter-electrode distance, as in a classical positive column where the voltage drop is proportional to its length with a constant electric field. The calculated data also show a good agreement with the experimental and calculated data of other authors, in particular, a good correspondence between the absolute values of electron density and temperature, the discharge voltage and the anode ablation rate as a function of the discharge current.

1. Introduction
Arc discharge is one of the most simple and inexpensive facilities to produce nano-materials [1,2]. In particular, fullerenes were discovered in an arc discharge by Kretchmer in 1990 [3], and carbon nanotubes (CNT) were first discovered with the deposition of carbon in an arc discharge by Iijima in 1991 [4]. There are a number of papers devoted to modeling of the arc discharges for the synthesis of carbon nano-materials (see review [5]). Models with cathode sputtering [6] or models for discharge gap plasma [7] were developed. The one-dimensional model (in the axial direction) of synthesis the CNT in the arc discharge with sputtered anode was presented in [8]. The experimental efforts were directed toward studying mechanisms associated with the anode sputtering [9], current-voltage characteristics of the anodic arc discharges [10], and deposition of material at the cathode [11]. In particular, it was shown that anode erosion increases with decreasing anode radius, current-voltage characteristics are V-shaped, and the radius of the deposited material at the cathode increases with increasing discharge current.

There are only a few theoretical models devoted to the synthesis of nano-particles in anodic arc discharges. A one-dimensional model of an arc discharge with evaporating graphite anode was presented in [12]. The thermodynamics of the formation of droplets of a carbon melt in a metallic catalyst was considered in [13]. Recently, a self-consistent integral model of an arc discharge with a sputtered anode [14] was presented. The model takes into account the relationship between discharge plasma parameters and electrodes, the continuity of current at the electrodes, the thermal regime of the electrodes, and the erosion rate of the anode. Two-dimensional model of an anodic arc discharge with the ablation of composite anode materials consisting of carbon particles and metallic particles was presented in [15]. Based on the above-mentioned arc discharge models, the following numerical model
of the arc discharge was developed to describe the main processes occurring in the synthesis of carbon nano-materials.

2. Model

In order to take into account the basic physical processes occurring in the synthesis of nano-particles in an arc discharge, a global integral model is constructed. The main features of the model are the simultaneous consideration and interconnection between the discharge gap plasma, the cathode and the anode layers, the current transfer, the thermal regime of the electrodes, and the evaporation of the anode. The stationary arc burning mode with the graphite cathode of radius \( R_c = 10 \) mm and the graphite anode of radius \( R_A = 3.5 \) mm is considered. In the burning mode, the anode material is constantly evaporated due to excessive heating, which is determined by the surface temperature of the anode \( T_A \). In turn, the surface temperature of the anode is determined by the energy flux from the discharge gap, which is related to the saturated pressure of the ablated particles from the anode.

During the arcing the erosion of the anode material occurs, and the length of the anode decreases from \( L_A = 65 \) to 30 mm. The anode is mechanically moved towards the cathode, so that the distance between the electrodes \( L_{gap} \) remains constant. Experimental observations show that erosion of the cathode does not occur in the process of discharge burning, and a slight redeposition of the anode material onto the cathode may take place. Surface flux of carbon particles, evaporated from the anode, propagates into the discharge gap and interacts with the buffer gas (helium), whose pressure is kept constant \( p_{He} = 24 \) Torr. During the discharge arcing, the discharge voltage and the discharge current are specially maintained to be constants, \( U_d = 22 \) V, \( I_d = 100 \) A. The radius of the discharge plasma \( R_{arc} \) is considered here to be equal to the radius of the anode \( R_{arc} = R_A \).

For the inter-electrode gap, the following energy balance equation was considered [14-16]:

\[
J_{gap} = Q_e + Q_n + Q_{gap,i} + Q_{gap,n} + Q_{gap,rad}, \tag{1}
\]

\[
J_{gap} = I_d U_d, \tag{1a}
\]

\[
Q_{gap,i} = I_{gap,i} (\xi_i U_{i,C} + \xi_{Me} U_{i,Me}), \tag{1b}
\]

\[
Q_{gap,n} = \frac{m_i}{m_e} (T_e - T) n_e \sigma \varepsilon T^{3/2} I_{gap}, \tag{1c}
\]

\[
Q_{gap,rad} = \varepsilon \omega T^{3} I_{arc}. \tag{1d}
\]

It was assumed that the Joule heat \( J_{gap} \) released in the discharge gap goes to heating the cathode (the heat flux to the cathode) \( Q_e \), to heating the anode (heat flux to the anode) \( Q_n \), to the ionization in the discharge gap \( Q_{gap,i} \), to heating by the electrons of the neutral particles \( Q_{gap,n} \), and to the radiation \( Q_{gap,rad} \). Here, the total discharge current (arc current) \( I_a \) is equal to the sum of the electron current \( I_e \) and the ion current \( I_i \) (at the cathode), \( I_d = I_e + I_i \). \( U_{i,C} \) is the ionization potential of carbon, \( T_e \) is the electron temperature in the discharge gap, \( n_e \) is the electron density, and \( \nu_i \) is the collision frequency of electrons with neutral atoms. It is assumed that heavy particles (ions and neutrals) are in equilibrium and have the same temperature equal to the anode temperature, \( T_i = T_A \).

The energy balance at the cathode surface is determined by the heat flux from the discharge gap \( Q_e \), brought by the ion flux to the cathode, and the loss of heat by the radiation \( Q_{c,rad} \) and the thermal conductivity \( Q_{c,cond} \) and the thermal emission of electrons from the cathode \( Q_{c,emission} \) [17]:

\[
\Delta Q_c = Q_c - Q_{c,rad} + Q_{c,cond} + Q_{c,emission}, \tag{2}
\]

\[
Q_c = \pi R^2_c j_{i,j} (U_{i,C} - \theta_{in,j}), \tag{2a}
\]

\[
Q_{c,rad} = \pi R^2_c \varepsilon \sigma T_e^3 \tag{2b}
\]

\[
Q_{c,cond} = (T_e - T) \lambda_c \pi^{3/2} R_c \tag{2c}
\]
$Q_{\text{emission}} = \pi R_c^2 \cdot j_{e,c} \cdot \varphi_{W,C}$, \hspace{1cm} (2d)

where $U_{l,c}$ is the voltage drop in the cathode layer, $\varphi_{W,C}$ is the work function for carbon, $T_e$ is the temperature of the cathode surface, $T_0$ is the chamber wall temperature ($\approx 300$ K), $j_{e,i}$ and $j_{e,c}$ are the ion and the electron flux densities at the cathode. The electron flux density from the heated cathode is determined by thermal emission:

$j_{e,e} = AT_e^2 \exp \left( -\frac{e\varphi_{W,C}}{kT_e} \right)$, \hspace{1cm} (3)

where $A$ is a constant determined by the cathode material. The motion of ions in the cathode layer can be considered to be collisionless, and, consequently, the Bohm criterion for the ion flux density at the cathode can be used [18]:

$j_{i,i} = 0.6en_i \sqrt{\frac{eT_e}{m_i}}$. \hspace{1cm} (4)

The total discharge current at the cathode consists of the ion and electron components:

$I_a = I_{e,e} + I_{e,i} = \pi R_c^2 (j_{e,e} + j_{e,i}) = \pi R_c^2 \left[ AT_e^2 \exp \left( -\frac{e\varphi_{W,C}}{kT_e} \right) + 0.6en_i \sqrt{\frac{eT_e}{m_i}} \right]$. \hspace{1cm} (5)

It should be noted that the parameter $n_i$ (electron density) connects two regions of the solution to the problem (two submodels), i.e. the cathode layer model and the entire discharge gap model. The discharge current is determined by the voltage drop over the length of the discharge gap and its electric conductivity $\sigma_{gap}$. The discharge voltage can be defined as:

$U_d = \frac{I_{top}}{\sigma_{gap} \pi R_c}$. \hspace{1cm} (6)

It is assumed that the discharge plasma reaches a local thermodynamic equilibrium, and the plasma composition and the ionization components can be calculated using the Saha equation:

\begin{equation}
\frac{n_{i,l}}{n_{0,l}} = \left( \frac{2m_i kT_e}{\hbar^2} \right)^{3/2} \exp \left( -\frac{U_{i,l}}{kT_e} \right)
\end{equation}

where $n_{i,l}$ and $n_{0,l}$ are the densities of ions and neutrals of particles of type $l$, $U_{i,l}$ is the ionization potential of particles of type $l$, and $\hbar$ is the Planck constant. The paper considers the Saha equation for carbon particles, $l = C$. In the discharge gap, it is assumed that the condition of electrical neutrality is fulfilled:

$n_e + n_{e,C} \approx 0$. \hspace{1cm} (8)

The anode layer of the discharge is produced in such a way as to ensure continuity of the current at the anode. As will be shown later, the voltage at the anode is negative, so that the electron flux to the anode decreases. The voltage drop at the anode $U_a$ is determined as follows [14]:

$U_a = -T_e \ln(I_{th} / I_a)$, \hspace{1cm} (9)

where $I_{th} = \pi R_c^2 n_e v_{Te}$ ($v_{Te}$ is the thermal velocity of electrons at the anode). The heating of the anode by the electrons leads to a significant heating of the anode surface. In the stationary state, the balance of the heat flux on the surface of the anode is fulfilled:

$\Delta Q_e = Q_e - Q_{e,inl} - Q_{e,rad}$, \hspace{1cm} (10)

where the heat flux $q_e$ coming from the plasma per unit surface of the anode per unit time:

$q_e = Q_e / (\pi R_c^2) = I_e (2T_e + U_a + \varphi_d) / (\pi R_c^2)$ \hspace{1cm} (10a)

is consumed to heat the sublimated material from the unit surface of the anode per unit time from the initial temperature $T_0$ to the melting point equal to the surface temperature of the anode $T_a$:

$q_{e,m} = \Gamma c_p (T_a - T_0)$ \hspace{1cm} (10b)

and subsequent vaporization of the molten material:
\[ q_{\text{evap}} = \Gamma \Delta H_{\text{evap}}. \]  

(10c)

Here \( \Gamma \) is the erosion rate of the anode surface (ablation flux) expressed in units [kg/(m\(^2\)s)]. \( c_p \) is the specific heat of the heated material of the anode. It should be noted that in the stationary process of evaporation of the anode material, both fluxes (10b) and (10c) are proportional to the rate of erosion of the anode material \( \Gamma \). \( \Delta H_{\text{evap}} \) is the latent heat of vaporization. Sublimation of the anode was calculated in the framework of the Langmuir evaporation model [15]:

\[ \Gamma = p_{\text{sat}}(2\pi R T_a^2 / M)^{1/2}, \]  

(11)

where \( p_{\text{sat}} \) is the saturated vapor pressure of the ablated anode material, \( R \) is the gas constant, \( M \) is the molecular weight.

3. Results

The parameters of the arc discharge in a helium medium with a sputtered graphite anode were calculated to verify the constructed global integrated model. For this purpose, experimental measurements of the dependence of the inter-electrode gap \( L_g \) on the discharge voltage \( U_d \) were made at different helium pressures \( p_{\text{He}} = 25, 50, 100 \) Torr, at a given discharge current \( I_d = 100 \) A. The measurements were carried out in a standard experimental setup described in [19,20] with usual discharge geometry (cathode and anode radius) and discharge parameters. At this stage, a pure graphite anode was used without metal additions to its core. After the steady-state burning mode (constant current \( I_d \) and discharge voltage \( U_{d,i} \), buffer gas pressure \( p_{\text{He}} \)) was achieved, the discharge was turned off and the inter-electrode distance \( L_g \) was measured. The results of the experimental measurements are represented by the symbols in figure 1. It is seen that the minimum discharge voltage under these conditions is approximately equal to \( U_{d,\text{min}} \approx 15 \) V at a minimum inter-electrode distance \( L_{g,\text{min}} \approx 1 \) mm. The maximum received voltage \( U_{d,\text{max}} \approx 35 \) V was at inter-electrode distance \( L_{g,\text{max}} \approx 17 \) mm. The voltage limit is explained by the physical limitation of the inter-electrode distance in the installation, i.e. the maximum shift of the anode rod is 17 mm. The practically linear increase in the discharge voltage with increasing inter-electrode distance indicates the presence of a classical positive column (PC) of the discharge, when the voltage drop in the PC is proportional to the length of the PC with a constant electric field. The numerical calculations of the \( L_g \) versus \( U_d \) for a given helium pressure \( p_{\text{He}} = 25 \) Torr and a discharge current of \( I_d = 100 \) A have been performed. It can be seen that the calculated and experimental data agree qualitatively within a wide range of the discharge voltage \( U_d \) from 15 to 35 V and the inter-electrode distance \( L_g \) from 1 to 17 mm.

**Figure 1.** Experimental data on the mutual dependence of the inter-electrode gap \( L_g \) and the discharge voltage \( U_d \) (squares for \( p_{\text{He}} = 25 \) Torr, circles for \( p_{\text{He}} = 50 \) Torr, triangles for \( p_{\text{He}} = 100 \) Torr). The solid line corresponds to the calculated results for \( p_{\text{He}} = 25 \) Torr. The discharge current is 100 A.
The parameters of the arc discharge in a helium medium with a sputtered graphite anode were calculated for the discharge conditions and parameters realized experimentally in [21] and simulated in [12]. The parameters of the arc discharge were as follows: both graphite electrodes 6 mm in diameter, inter-electrode gap 6 mm, helium pressure 100 Torr, arc current from 50 to 150 A, discharge voltage 20 V. The most important and determining characteristic of the arc discharge is the rate of erosion of the anode \( \alpha = \Delta m/\tau \), usually measured in units of mg/s. Figure 2a shows the experimental data of [21] and the calculated data of [12], and figure 2b shows simulation results using the presented model. It can be seen that all the data on the anode evaporation rate \( \alpha \) coincide very well within the whole range of discharge currents. With the help of the developed model, the integral characteristics of the plasma parameters (temperature \( T_e \) and density \( n_e \) of electrons, voltage on the discharge gap \( U_d \)) were calculated as a function of the discharge current \( I_d \) (from 50 to 100 A). The calculated results are in good agreement with their absolute values obtained in [12,21], i.e. the electron temperature \( T_e \) within the range from 0.6 to 0.8 eV, and electron density \( n_e \) from \( 10^{15} \) to \( 10^{16} \) cm\(^{-3}\), \( U_d \sim 15 \) V.

![Figure 2](image)

**Figure 2.** Dependencies of the anode erosion rate \( \alpha \) on the discharge current \( I_d \). a) Solid line corresponds to numerical calculations [12]. Dashed line shows experimental data [21]. b) Solid line with circles corresponds to the data calculated by the present model. \( P_{He} = 100 \) Torr, \( L_g = 6 \) mm.

4. Conclusions
A global integral model of an arc discharge in a helium medium with a sputtered graphite anode was constructed. The main features of the theoretical model are the simultaneous consideration and interconnection between the plasma of the discharge gap, the cathode and anode layers, the current continuity, the thermal regime of the electrodes, and the evaporation of the anode. The model permits to calculate the heat balance in the entire discharge gap (radiation losses, heating of materials), the temperature of the electrodes surfaces, the electrons and carbon ions concentrations in the discharge gap, the electron temperature, the voltage drops in the anode and cathode layers, the ratio of ion and electron currents at the cathode, and the ablation rate of the evaporated anode material.

The dependence of the inter-electrode gap on the discharge voltage was calculated for helium pressure of 25 Torr and discharge current of 100 A. The calculated data were compared with the experimental measurements. It was experimentally and theoretically shown that the discharge voltage linearly increases with the inter-electrode distance, which indicates the presence of a classical positive column when the voltage drop is proportional to the length of the positive column with a constant electric field.

The parameters of the arc discharge were also calculated for the discharge conditions and parameters realized experimentally in [21] and simulated in work [12]. The results of the simulation with the help of the presented model showed good agreement with the experimental and calculated data of these studies, in particular, a good correspondence between the absolute values of electron
density and temperature, the discharge voltage and the ablation rate of the anode as a function of the discharge current.

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