

Research on modes of plasma generation in low-pressure discharge for thermal radiation processes

T V Koval¹, I V Lopatin², Nguyen Bao Hung³

¹ Professor, National Research Tomsk Polytechnic University, 30, Lenin Ave., Tomsk, 634050, Russia.

² Junior researcher, Institute of High Current Electronics of Siberian Branch of Russian Academy of Sciences, 2/3, Akademicheskoy Ave., Tomsk, 634055, Russia.

³ Post-graduate student, National Research Tomsk Polytechnic University, 30, Lenin Ave., Tomsk, 634050, Russia.

E-mail: lopatin@opee.hcei.tsc.ru

Abstract In this work a theoretical research on modes of low-pressure glow discharge generation and its parameters in a large area hollow cathode are carried out. The relations describing the dependence of the burning voltage on the gas pressure and the geometry of the system were obtained. It is shown that it is possible to adjust the ion current density and burning voltage by external current injection independently of the surface area and material of the processed parts. This work also carries out a numerical study of the influence of plasma parameters on the heating of processed parts. Theoretical results are compared with experimental data.

1. Introduction

Thermal radiation processing enhances the surface properties and lifetime of machine parts, while decreasing their cost. A target for plasma treatment is immersed in gas plasma in which ions are accelerated toward the target in the electric double layer formed between the target and discharge column. The saturating element (gas atoms) in the treated target volume propagates due to heat-activated diffusion whose rate depends mostly on the treatment temperature and saturating element gradient. The main parameters of plasma treatment are the gas composition or gas mixture ratio, temperature and time of the process, operating pressure, discharge parameters, dissociation and ionization degrees of the working gas, ion energy, and ion current density at the surface of a treated target [1]. Most of these parameters are interrelated. So, the gas composition or gas mixture ratio affects the discharge initiating voltage and hence the ion energy, and the voltage and current density are limited by the temperature allowable for treatment and by the gas pressure. Nitriding at 260–510 °C provides the formation of hard nitride layers of thickness ranging up to 200–300 µm [2, 3].

The vacuum plasma technology used for surface modification of materials and large-size objects is based on the generation of low-temperature glow discharge plasmas [4–9] for which it is required to provide an ion current density of ~1 mA/cm² to a treated surface and a discharge operating voltage of hundred volts [6–9]. One of the important problems is to control the discharge current and discharge operating voltage irrespective of the gas kind, gas pressure, and treated surface area.

The paper presents numerical simulation results on plasma generation modes (self-sustained and non-self-sustained) in a low-pressure discharge with a large-area hollow cathode. The effect of the plasma



parameters, material and geometry of treated parts on their heating rate and temperature is investigated. The theoretical research results are compared with experimental data.

2. Hollow-cathode gas discharge

The cylindrical hollow cathode used in experiments on plasma generation is shown in Figure 1 [6, 7]. Anode 1 in the form of two tubes of total area S_a is located at the side wall of the cathode. The anode area S_a is varied by displacing the anode deep into the cathode cavity the volume of which is $V_c = 2 \times 10^5 \text{ cm}^3$, $S_a = 200 \dots 500 \text{ cm}^2$. The plasma is generated both in the main (self-sustained) mode and in the mode in which the discharge is sustained by an electron beam extracted from the plasma of auxiliary arc discharge 3. The plasma is formed inside the volume of the hollow cathode. The cathode potential fall U_c is almost equal to the discharge voltage U : $U_c = U - U_a \approx U$, where U_a is the negative anode potential fall. The plasma ions accelerated in the cathode layer provides ion-electron emission from the cathode surface. The electrons accelerated and set in oscillatory motion in the hollow cathode (with about the same probability of ionization at any point of the cathode cavity) lose their energy in collisions with neutrals and ensure a self-sustained discharge. This provides high homogeneity of the ion current density, which is defined by the properties of the discharge plasma.

The hollow-cathode discharge is described by equations of energy balance of charged particles and current continuity which allow us to relate the internal parameters (temperature, plasma potential) to the external parameters (working gas pressure, discharge current and voltage, system geometry). From the balance equation for fast electrons we can obtain the discharge operating condition. In the general case of auxiliary discharge current injection I_{ext} , the balance equation for fast electrons is written in the form

$$\frac{\gamma I_i}{e} + \frac{\delta I_i}{e} = \frac{n_f V v_i}{u} + \frac{S_a}{4} n_f \langle v \rangle \quad (1)$$

Here γ is the ion-electron emission coefficient which depends on the discharge voltage and gas kind; $I_i = e v_i n_f V$ is the ion current which is defined in terms of energy loss by fast electrons in gas ionization in the cathode cavity $V = (V_c - V_a)(1 - h/2D)$, h is the anode height, D is the length and diameter of the cathode cavity; $\delta = (1 - \alpha)(I_{ext}/I_i)$, α is the fraction of fast electrons not involved in ionization; n_f and $\langle v \rangle$ are the density and average velocity of fast electrons; $v_i = n_g \sigma_i v$ is the ionization rate; and e is the electron charge. From equation (1) we can obtain the condition of the main discharge in the non-self-sustained mode:

$$u = \frac{P}{P(\gamma + \delta) - 1}, \quad (2)$$

which gives a relation between the dimensionless discharge operating voltage $u = e U_c / W$, gas pressure $P = (p/kT_e)\sigma_i L$, and effective length of the cathode cavity $L = 4V/S_a$; W is the total electron energy expended in gas ionization. At high pressures $p \gg (kT_e/\sigma_i L)(\gamma + \delta)^{-1}$, the discharge operating voltage does not depend on the effective length of the cathode cavity; the glow discharge condition is defined by the relation $(\gamma + \delta)u = 1$, and at $S_a \ll S_c$ it coincides with the relation derived from energy balance [8] and at $\delta = 0$ with the relation obtained elsewhere [10].

To determine the spatial distribution of the electron temperature and plasma density and to study the effect of the geometry, size, and material of targets placed in the cathode cavity, we used a hydrodynamic model which describes the density of charged particles and their average energy as a function of time and space [7]. The transport coefficients (discharge coefficients) dependent on the electron energy distribution function are output parameters for the hydrodynamic model and are calculated using the BOLSIG+ program [11]. The plasma generation in the hollow cathode is simulated without considering the cathode region in which emission electrons are accelerated. When the current continuity condition

is valid at the computational domain boundaries, the output parameters for the hydrodynamic model are the discharge characteristics obtained from (1)–(2).

Self-sustained mode. In the self-sustained mode, the high voltage of the main discharge is a shortcoming for the hollow cathode, because it leads to intense ion etching of a treated surface [3]. Figure 2 shows calculated (formula (2)) and experimental dependences [6, 7] of the discharge operating voltage on the gas pressure (argon, nitrogen) in the self-sustained mode at different effective lengths of the hollow cathode $L = 4V/S_a$; the discharge current is 30 A; the experimental values correspond to $S_a = 500 \text{ cm}^2$. Decreasing the operating pressure of the main discharge causes a substantial increase in discharge operating voltage at a pressure below 0.65 Pa. At these pressures, a large fraction of ionizing electrons whose energy is only partly expended in ionization reaches the anode. The decrease in ionization efficiency is compensated by a corresponding increase in the coefficient of secondary electron emission $\gamma = \gamma(u)$ due to the increase in discharge voltage; in the range of voltages (200...1000) V, $\gamma_{Ar} = 0.03...0.11$ and $\gamma_{N_2} = 0.04...0.2$, ($\sigma_{iAr}/\sigma_{iN_2} \approx 1.5$).

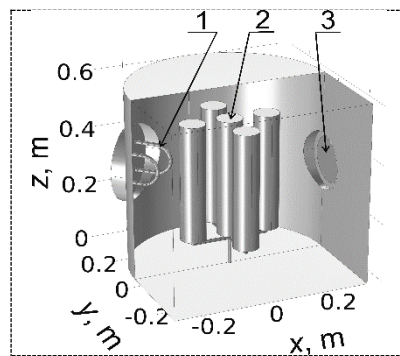


Figure 1. Hollow cathode model: 1 – anode; 2 – targets; 3 – auxiliary discharge window.

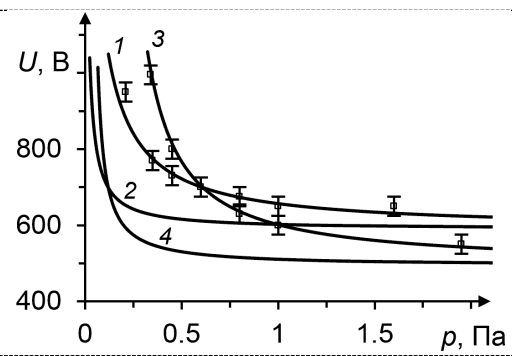


Figure 2. Discharge operating voltage vs the gas pressure in the self-sustained mode for nitrogen (1, 2) and argon (3, 4) at $L=1.6 \times 10^3 \text{ cm}$ (1, 3) and $L=6.4 \times 10^3 \text{ cm}$ (2, 4); $\bar{}$ – experiment, $I = 30 \text{ A}$.

From relation (2) and Figure 2 it is seen that decreasing the effective length of the hollow cathode $L = 4V/S_a$ (increasing the anode area S_a or decreasing the cathode volume due to targets, $V_c - V_d$) increases the discharge voltage. This owes to capture of fast electrons by the anode, and the self-sustained discharge current is kept at the specified level due to the emission current (the coefficient γ), i.e., due to the voltage in the cathode layer.

The current to the anode is defined by plasma electrons and anode fall. Figure 3 shows calculated gas pressure dependences of the argon plasma density in the center of the cathode cavity, plasma temperature, and plasma potential in the self-sustained mode at a constant discharge current of 30 A. The discharge current and current density at the cathode are kept constant due to the increase in anode fall with decreasing plasma density ($S_a = 220 \text{ cm}^2$).

Non-self-sustained mode. Additional electron injection into the cathode cavity is through the window of the auxiliary discharge (Figure 1). As can be seen from relation (2), the auxiliary discharge shifts the characteristics of the main discharge toward lower voltages and gas pressures. This decreases the energy gained by an electron in the cathode layer of the discharge and the number of its ionization events.

Studies of the main discharge operating voltage U depending on the relative volume of targets (Figure 4) show that in both modes, the voltage U varies slightly at a nitrogen pressure of $p_{N_2} > 0.65 \text{ Pa}$. In the non-self-sustained mode at an operating pressure of $p_{N_2} < 0.65 \text{ Pa}$ and discharge voltage of $U > 350 \text{ V}$, the main discharge operating voltage can be stabilized by varying the auxiliary discharge current.

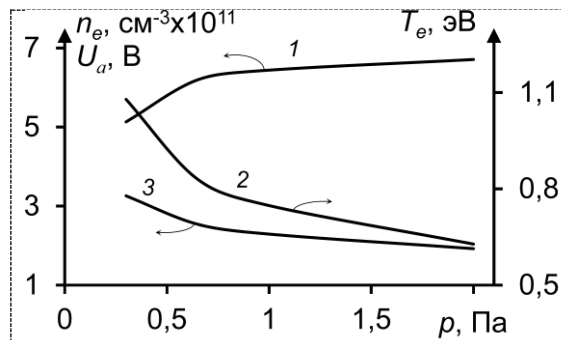


Figure 3. Calculated pressure dependences of the argon plasma in the self-sustained mode: 1 – plasma density; 2 – plasma temperature; 3 – plasma potential; $S_a = 220 \text{ cm}^2$.

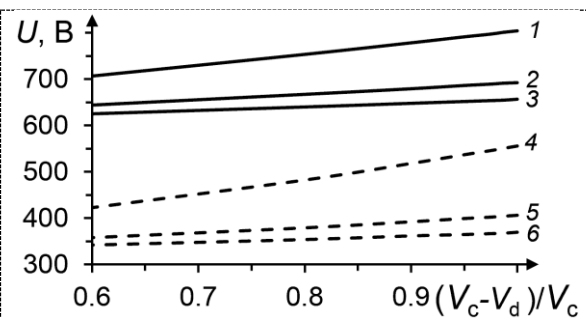


Figure 4. Discharge voltage in argon vs the relative volume in the self-sustained (solid lines) and non-self-sustained (dashed lines) modes at different pressures p : 0.35 (1, 4), 0.65 (2, 5), and 1 Pa (3, 6); $\delta = 0.11$.

The external injection current and plasma potential vs the number of targets N (shaped as cylinders of dimensions $\varnothing 10 \text{ cm} \times 40 \text{ cm}$) for $p_{N_2} = 0.65$ are shown in Figure 5. Stabilization of the discharge voltage at 370 V and ion current density with increasing the number of targets ($N=1 \div 8$) takes place as the current of additional emission is increased from 9 to 14 A. The plasma potential increases slightly, because the relative volume $(V_c - V_d)/V_c$ varies in the range of unity up to 0.9 (Figure 4), the total volume of targets is $(0.25 \div 2) \times 10^4 \text{ cm}^3$, and their relative surface area is $S_d/S_c = 0.071 \div 0.57$.

Using the hydrodynamic model, we obtained plasma density distributions in the hollow cathode for nitrogen. Figure 6 shows calculated lines of the plasma density in the non-self-sustained mode at $I_{ext} = 14 \text{ A}$ and $U = 370 \text{ V}$ in two sections of the hollow cathode with targets. The pressure gradient due to gas supply through the electron source increases the plasma density gradient in the region of the emission window.

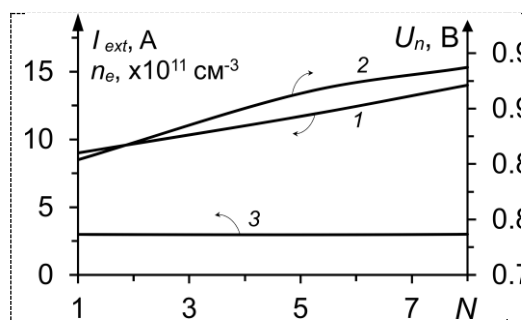


Figure 5. External injection current (I), plasma density (3), and plasma potential (2) vs the number of targets; nitrogen.

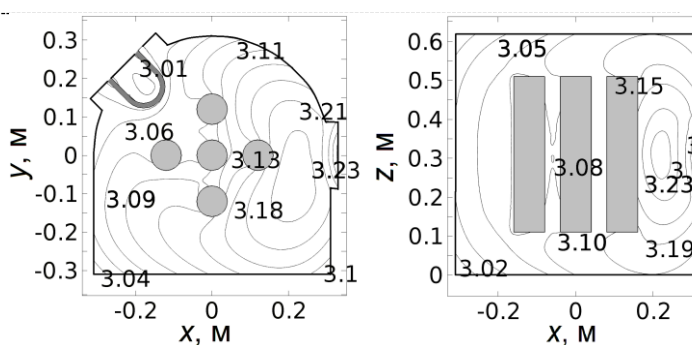


Figure 6. Plasma density distribution for nitrogen ($\text{cm}^{-3} \times 10^{11}$) in the lateral and longitudinal sections of the hollow cathode with targets; non-self-sustained mode.

The properties of the cathode material, configuration of a treated target, and its size can affect the discharge characteristics and the degree of treatment [12]. When treated, a target serves as a cathode; between the plasma and treated target, a space charge layer arises and its electric field provides acceleration of plasma ions. Due to ion bombardment, the target is heated. The heating time depends on the material and dimensions of a treated target. The heat conduction problem with regard to water cooling of the hollow cathode walls (Figure 1) was numerically solved for titanium and copper parts of

dimensions $\varnothing 5 \div 13$ cm and $20 \div 40$ cm. The calculations show that with no shield, the specimen temperature is no greater than 330°C , which is lower than 530°C optimal for nitriding [3].

With an active shield surrounding the treated targets, the shield creates active particles and ensures additional heating of the targets. The times of heating to 400°C for targets of different geometries with an active shield placed at a distance of 2 cm from the cathode are presented in Table 1. The heating time of targets depends on their material and dimensions.

Table 4. The times of heating to 400°C for targets of different geometries with an active shield placed at a distance of 2 cm from the cathode.

Dimension of a part	Heating time, min	
	Fe	Ti
$\varnothing 10\text{cm} \times 40$ cm	123	86
$\varnothing 10\text{cm} \times 20$ mm	113	75
$\varnothing 5\text{cm} \times 40$ mm	66	43
$\varnothing 13\text{cm} \times 40$ mm	155	108
$\varnothing 13\text{cm} \times 20$ mm	140	93

It is seen from the table that the difference in heating time for the parts is more than one hour. This defines the difference in nitriding time and hence in nitrogen penetration depth. The energy expended in heating several targets occupying the entire cathode volume can be the same and even lower than the energy expended in heating a single target. This owes to a decrease in discharge voltage and mutual thermal radiation of targets, which impairs heat removal from them.

The ion current density and ion energy are not decisive factors for nitriding, and the process efficiency is defined to a greater extent by the dose of implanted nitrogen ions at an ion current density to a treated target higher than 1 mA/cm^2 [3]. In this case, thermal diffusion dominates and ion bombardment is secondary. Experiments show that there are two competitive processes: penetration of nitrogen through diffusion and reduction of its depth due to sputtering [14]. An increase in nitrogen concentration correlates well with an increase in surface microhardness, and an increase in nitrided layer thickness provides an increase in wear resistance [3].

The Arrhenius curve characterizes the rate of increase in nitride layer thickness $\ln(d^2/t)$ depending on the temperature T [3]. The linear form of this dependence evidences that the growth of nitride layers is controlled by diffusion of nitrogen atoms. The diffusion is described by the formula

$$\frac{d^2}{t} = \exp\left(-\frac{E_a}{kT}\right), \quad (3)$$

where d is the nitride layer thickness, t is the treatment time, E_a is the diffusion activation energy determined from experimental data, T is the treatment temperature.

3. Conclusion

Thus, we studied the plasma generation in a low-pressure gas discharge with a large-area hollow cathode in the self-sustained mode and in the mode with additional electron injection.

Using the theoretical model in which fast electrons are responsible for the birth of charged particles in the hollow cathode plasma, dependences of the discharge operating voltage on the gas pressure and on the geometry of the hollow cathode and treated targets were obtained. It is shown that the discharge operating voltage decreases more than two times due to the current of additional electron injection. It is demonstrated that whatever the surface area and material of treated targets, it is possible to control the ion current density and discharge operating voltage by varying the current of additional electron injection.

Using the hydrodynamic model, it is shown that at a discharge operating voltage of 300–400 V, gas pressure of 0.65 Pa, and plasma temperature of about 1 eV, the plasma density for nitrogen and argon reaches 3×10^{11} and $6 \times 10^{11} \text{ cm}^{-3}$, respectively.

The active shield provides uniform heating of targets of different shapes and dimensions to a temperature required for nitriding. The heating time of targets depends on their material and dimensions and decreases with increasing the number of targets due to their mutual thermal radiation. The temperature conditions of treated targets are controlled by varying the auxiliary discharge current and gas pressure.

The theoretical and numerical models agree with experimental data [6, 7].

This work was carried out at the expense of Russia research foundation (Grant no. 14-29-00091)

References

- [1] Hosseini, Ashrafizadeh F 2009 Accurate measurement and evaluation of the nitrogen depth profile in plasma nitrided iron *Vacuum* **83** 1174–1178
- [2] Zagonel L F, Figueroa C A, Droppa Jr. R, Alvarez F 2006 Influence of the process temperature on the steel microstructure and hardening in pulsed plasma nitriding *Surf. Coat. Tech.* **201** 452–457
- [3] Berlin E V, Koval N N, Seidman L A 2012 *Plasma thermochemical surface treatment of steel parts* (Moscow: Tekhnosfera) p 462 (in Russian)
- [4] Lopatin I V, Akhmadeev Yu Kh, Koval N N, Schanin P M, Yakovlev V V 2010 Generation of plasma in non-self-sustained glow discharge with hollow cathode *Modification of Materials with Particle Beams and Plasma Flows: Proc. of 10th Int. Conf. (Tomsk: Russia, 19-24 september 2010) (Publishing house of the IOA SB RAS)* pp 35–38
- [5] Lopatin I V, Akhmadeev Yu Kh, Koval N N, Schanin P M, Shnaider A V 2008 Plasma Production in a Low-pressure Hollow-cathode Non-selfsustained Discharge *Proc. of 9 Int. Conf. on Modification of Materials with Particle Beams and Plasma flows (Tomsk: Russia 21-26 september 2008) (Publishing house of the IOA SB RAS)* pp 312–315
- [6] Lopatin I V, Schanin P M, Akhmadeev Y H, Kovalsky S S, Koval N N 2012 Self-sustained low pressure glow discharge with a hollow cathode at currents of tens of amperes *Plasma Phys. Rep.* **38** 7 583–589
- [7] Koval T V, Lopatin I V, Ogorodnikov A A, Nguyen Bao Hung 2014 Research on low-temperature plasma generation with a large area hollow cathode glow discharge *Izv. Vussh. Uchebn. Zaved. Fiz.* **57** 3/2 122–125
- [8] Korolev Y D, Frants O B, Landl N V, Shemyakin I A, Geyman V G 2013 High-Current Stages in a Low-Pressure Glow Discharge With Hollow Cathode *IEEE Trans. Plasma Sci.* **41** 8 2087–2096
- [9] Korolev Yu D, Frank K Discharge formation processes and glow-to-arc transition in pseudospark switch 1999 *IEEE Trans. Plasma Sci.* **27** 5 1525–1537
- [10] Metel A S, Grigoriev S N 2005 *Glow discharge with electrostatic confinement of electrons: physics, engineering, applications.* (Moscow: MSU IC STANKIN, Janus-K)
- [11] Hagelaar G J M, Pitchford L C 2005 Solving the Boltzmann equation to obtain electron transport coefficients and rate coefficient for fluid models *Plasma Sources Sci. Technol.* **14** 722
- [12] BOLSIG+ Electron Boltzmann equation solver: <http://www.bolsig.laplace.univ-tlse.fr>.
- [13] de Ataíde A R P, Alves Jr. C, Hajek V, Leite J P 2003 . Effects during plasma nitriding of shaped materials of different sizes. - Surface and Coating Technology *Surf. Coat. Tech.* **167** 52–58
- [14] Galdikas A, Moskaliuviene T 2011 Modeling of stress induced nitrogen diffusion in nitrided stainless steel *Surf. Coat. Tech.* **205** 3742–3746