

ENERGY CHARACTERISTICS OF SPHERICAL GLOW DISCHARGE

*V.A. Zhovtyansky, O.V. Anisimova, V.O. Khomych, Yu.I. Lelyukh,
V.G. Nazarenko, Ya.V. Tkachenko*

*Institute of Gas, National Academy of Sciences of Ukraine, Kiev, Ukraine
E-mail: zhovt@ukr.net*

The ultimate goal of this study is optimization of the modification processes, namely the constructional details' surface nitriding, based on the using of glow discharge (GD). These processes are studied both experimentally and theoretically in spherical abnormal GD plasma in N₂-Ar mixture. The balance equations for the density of charged particles and Poisson equation added with equation for heat conductivity are taken into account in the theoretical part of investigation. The last equation describes the influence of the hot cathode on the processes in discharge volume. As is shown, the correct account of the anode fall of potential plays a key role to represent adequately the volt-ampere characteristic of the spherical GD.

PACS: 51.50.+v, 52.25.Dg, 52.80.Hc

1. ENERGY EFFICIENCY CRITERION FOR THE NITRIDING PROCESS

The abnormal glow discharges (GD) are widely used in the processes of metal surface nitriding as they provide maximal localization of the technological action on a treated surface. Unfortunately, a lot of publications in this area are limited, mainly, to empirically obtain general rule regarding application of plasma as technological atmospheres. It does not allow making the strict analysis of energy efficiency parameters. A basis of analytical methods for optimization mentioned should be the careful analysis of physical processes in the plasmas as technological atmospheres.

There is no common viewpoint concerning the mechanism of diffusion saturation of metal surface with nitrogen. Now, the substantial attention is attracted to studying the ion-molecular reactions which run in the processes of surface modification. It was pointed out that the efficiency of such processes in N₂ plasma is mostly governed by metastable atomic nitrogen N* [1] (see also papers [2, 3]), having two characteristic radiation lifetimes of 1.4×10^5 and 6.1×10^4 s in their two metastable states. Therefore, N* is the best agent to transfer the excitation energy obtained in plasma to the surface of a solid. Really, a typical free path of neutral particles in N₂ atmosphere at the pressures 0.1...150 Pa changes from about 35 to 2.3×10^{-2} cm, respectively. Hence, N* has plenty of chances to get from the plasma region to the surface of a solid without collisions. Just collisions are able to transit an atom from a metastable into the ground state.

Thus, as the energy efficiency criterion of nitriding process the ratio between atomic nitrogen stream diffusion on a surface of material to be processed and electric discharge power may be considered. The higher the ratio, the higher is the energy efficiency of nitriding.

Plasma forming mixes of N₂ with Ar impurity have considerable advantages in nitriding processes as atomic nitrogen can effectively be generated in this mix [4]. GD plasma is sharply nonequilibrium and the strict analysis of atomic nitrogen producing is a challenge. Moreover, essential meaning have nonlocal effect in near cathode GD area which are a subject of active research during last years (see, for example, [5]). In turn, methods of direct

experimental definition of nitrogen atoms density N_a in plasma are difficult enough, as application of vacuum spectroscopy methods [1] demands. Besides, they do not allow predicting character of relationship between N_a and GD parameters.

2. EXPERIMENT

The discharge plasma is generated in nitrogen or N₂-Ar mixture at the pressure 50...250 Pa and discharge current $I \leq 120$ mA. The constructional details to be modified (samples) were placed on the metal plate (cathode) 5 cm in diameter at the central part of the vacuum camera (anode) with a volume of 0.1 m³ [3]. The temperature of cathode was controlled by a thermocouple. The density of charged particles N_e and the electric field E were measured by double probes that could be moved along the radius of the chamber. GD was powered by rectified voltage U up to 1500 V and the volt-ampere characteristics (VAC) were measured too.

The process of nitriding was performed after evacuation of the chamber and preliminary treatment (cleaning) of the samples at the satellite discharge in pure argon. Actually nitriding was effected by GD in a N₂-Ar mixture. The temperature of the plate (and, hence, of the samples) during the process was maintained within 810...820° K due to the energy supply from GD powered at $UI \sim 60$ Wt.

This system was approximated as spherical diode in numerical simulation [6, 7]. The following values of parameters were considered in calculations: $r_K = 1.5$ cm and $R = 33$ cm are the radii of the internal and the external spheres of diode, respectively. The calculations were performed for values of the discharge current density 2...20 mA/cm² that is corresponding with the interval of calculations carried out in [8]. In turn, in some cases for experimental studies spherical cathode of molybdenum was also used.

3. VAC OF THE SPHERICAL GD

As it is well known, despite the influence of nonlocal effects, the estimated VAC of GD in fluid model is well correspond to the real. This is due to the fact that only a small part of the fast electrons from their total number is responsible for these effects [5].

Previously we simulated spherical GD, paying special attention to the problem of adequate description of the diffusion processes [7]. The role of the latter can be very significant at low pressures. The system of fluid equations was solved

$$\frac{1}{r^2} \frac{d}{dr} (r^2 J_e) - \alpha(E) J_e = 0, \quad \frac{1}{r^2} \frac{d}{dr} (r^2 J_i) + \alpha(E) J_e = 0, \quad (1)$$

$$-D_e \frac{dN_e}{dr} + \mu_e N_e E = J_e, \quad -D_i \frac{dN_i}{dr} + \mu_i N_i E = J_i, \quad (2)$$

$$\frac{1}{r^2} \frac{d}{dr} (r^2 E) = \frac{e}{\varepsilon_0} (N_e - N_i), \quad (3)$$

where J_e and J_i are the densities of the electron and ion flows, respectively ($J = e(J_i + J_e)$); $\alpha(E)$ is the first Townsend coefficient; D_e , μ_e , D_i , μ_i are the diffusion and mobility coefficients of electrons and ions, respectively; e is the electron charge, and ε_0 is the dielectric constant. The boundary conditions for problem (1) – (3) were initially formulated in a manner like [8] as follows:

$$J_e = \gamma J_i, \quad eJ_e = \gamma J_K / (1 + \gamma), \quad \varphi = 0, \quad (4)$$

$$N_i = 0, \quad eJ_e = J_A, \quad dN_e/dr = 0, \quad (5)$$

at the cathode and the anode, respectively; here J_K and J_A are current densities at the cathode and the anode, $\gamma = 0,02$ denotes the coefficient of electron secondary emission from the cathode. The problem (1) – (5) was solved by modified method of continuation of the solution with respect to a parameter [6, 7].

The set of solutions of upper equations allows to determine theoretically obtained VAC of DC and to compare it with experimental one. As may be seen in Fig.1 this comparison demonstrates only slight quantitative consistency between two groups of VAC.

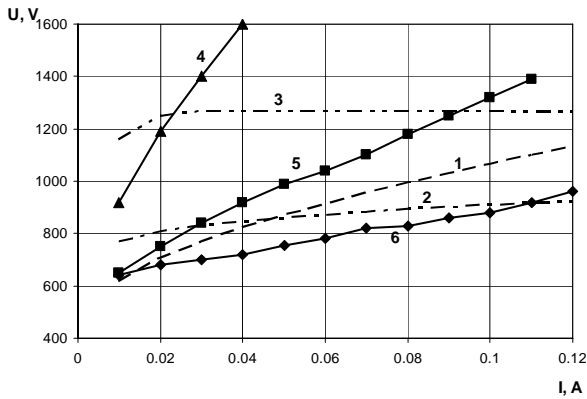


Fig. 1. Volt-ampere characteristics of spherical glow discharge ($r_c = 1,5$ cm): experimentally obtained (solid lines) and numerically calculated (dotted lines): $p = 50$ (1, 4), 120 (2, 5) and 250 Pa (3, 6)

In particular, numerical simulation (1) – (5) demonstrates a significant level of potential fall on the positive column GD that is about half of the total voltage drop on the discharge gap. This contradicts to the results of experimental determination of potential distribution along the diode radius by floating probe method. It does not fix appreciable electric field in the positive column. In this connection the role of the processes near the anode was specified by method of numerical experiment. In this case the principle of minimum power supply in GD was used in fact [9]. The setting of numerical experiment become possible due to high efficiency of computational procedure for GD mathematical model, proposed in [6, 7].

96

During the numerical experiment was changed last of the boundary conditions (5) on the anode. Namely, instead of condition $dN_e/dr = 0$ was introduced value of electron density on the anode N_{ea} . It was varied during the process of numerical experiment. The radial distribution of potential in positive column was radically changed under threshold value of this condition N_{ea} in simulation ($N_{ea} \sim 3.8 \times 10^9 \text{ cm}^{-3}$ at a pressure 133 Pa , $j_k = 20 \text{ mA/cm}^2$). In this case the electric field was decreased on the order of value in the positive column and the positive fall of potential value of several volts took place near the anode. The nature of this phenomenon is generation of charge particles in anode fall of potential (AF) [10]. Due to admitting of ions from AF region into positive column is providing ionic component of discharge current that is resulted in the lowering of potential on the GD. The set of GD VAC is shown in Fig. 2 as illustration of the role of adequate account of the processes on the anode.

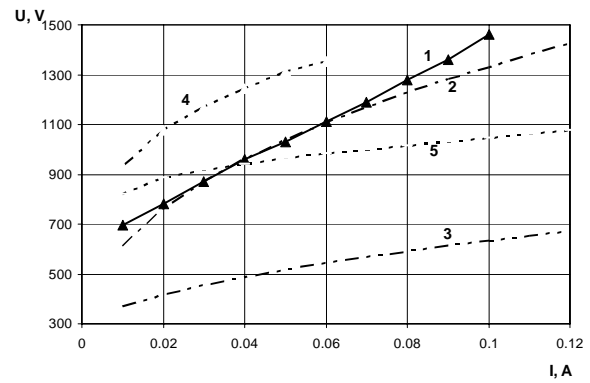


Fig. 2. Volt-ampere characteristics of spherical glow discharge at a pressure 100 Pa ($r_K = 1,5$ cm): experimentally obtained (solid line 1) and numerically calculated (dotted lines): with account AF ($T_K = 800 \text{ K}$ (2) and $T_K = 300 \text{ K}$ (3)) and without AF ($T_K = 800 \text{ K}$ (4) and $T_K = 300 \text{ K}$ (5))

The reason of the numerical experiment was the fact that the processes on the anode were presented as the anode spots located on the sharp edges of vacuum chamber. This system is quite complex for its formal description. However, the results obtained during the numerical experiment reflect adequately the major role that performs these spots – filling a gap discharge with positive ions that compensate space charge. All the basic laws of so introduced AF in our case correspond well to their experimental study [10].

The influence of the cathode temperature on the processes in discharge volume might be taken into account to further improvement in agreement experimentally obtained and numerically calculated VAC. For this purpose the set of equation (1) – (5) was added with equation for heat conductivity:

$$\frac{1}{r^2} \frac{d}{dr} \left(r^2 \kappa \frac{dT}{dr} \right) = 0. \quad (6)$$

Here κ is heat conductivity coefficient [8]:

$$\kappa = \frac{8,334 \cdot 10^{-4}}{\sigma^2 \Omega^{(2,2)}} \sqrt{\frac{T}{M}} \left(0,115 + 0,354 \frac{c_p M}{R} \right), \quad (7)$$

where M – molar mass, $\Omega^{(2,2)} = 1,157(71.4/t)^{0.1472}$, $\sigma = 3.68 \text{ \AA}$, $R = 8,314 \text{ J/(mol K)}$.

With account of this equation the temperature of gas in discharge volume is changed from 810...820 K at the cathode to 400 K at the distance 5 cm from it.

As may be seen from Fig. 2 by comparing the curves 1 and 2, the system of equation (1) – (2) allows describing adequately enough real VAC of GD in the process of nitriding. The further improving may be consisted with the account of current change of the temperature of cathode (plate) due to variation of energy supply with grow of voltage U on the discharge gap.

4. CONCLUSIONS

The complex study of glow discharge presented in this paper as a whole may be used for optimization of the modification processes from the view point of energy efficiency. Strictly speaking the electric discharge power is not parameter of the technology presented in this paper. Really, in the process of nitriding energy supply of GD in maintained on the level of $UI \sim 60$ Wt to stabilize the temperature of samples to be processed within 810...820 K. Nevertheless these results allow determining quantitatively the basic parameters of GD depending of pressure. The way how to use these parameters as well as to determine the percentage composition of N_2 -Ar mixture as technological atmosphere to optimize the process of nitriding is presented in paper [11].

REFERENCES

1. T. Nakano, T. Kitajima, S. Samukawa, T. Makabe. Diagnostics of N_2 and O_2 dissociation in RF plasmas by vacuum ultraviolet emission and absorption // *Abstracts of the XXVIII Int. Conf. on Phenomena in Ionized Gases*, Prague 15-20 July, 2007 / Institute of Plasma Physics AS CR, Prague, 2007, p. 42.
2. V.A. Zhovtyansky. Plasma-chemistry effects and some fundamental problems of the gas discharge physics // *Ukr. J. Phys.* 2008, v. 53, N 5, p. 490 – 496.
3. O.G. Didyk, V.A. Zhovtyansky, V.G. Nazarenko, and V.A. Khomich. Plasma modification of the surface of constructional materials // *Ukr. J. Phys.* 2008, v. 53, N 5, p. 482-489.
4. *Principles of Laser Plasmas* / Ed. by G. Bekefi. New York: “Wiley Interscience”, 1976.
5. A.A. Kudryavtsev, A.V. Morin, L.D. Tsendin. The role of nonlocal ionization in short glow discharges // *Tech. Phys.* 2008, v. 53, p. 1029-1040.
6. V.A. Zhovtyansky, Yu.I. Lelyukh. Mathematical modelling of plasma in a glow discharge of spherical geometry // *Ukr. J. Phys.* 2008, v. 53, N5, p. 497-503.
7. V.A. Zhovtyansky, Yu.I. Lelyukh. Numerical simulation of stationary processes in glow discharge plasma // *Tech. Phys. Let.* 2009, v. 35, p. 725-729.
8. A.S. Petrushev, S.T. Surzhikov, J.S. Shang. Two dimensional model of glow discharge with account of the vibration excitation of molecular nitrogen // *Teplotfiz. Vys. Temp.* 2006, v. 44, p. 814-822 (in Russian).
9. Yu.P. Raizer. *Physics of gas discharge*. M.: “Nauka”, 1987 (in Russian).
10. Yu.S. Akishev, A.P. Napartovich, P.I. Peretyat'ko, N.I. Trushkin. Near electrodes regions of glow discharge and normal current density on the anode // *Tech. Phys.* 1980, v. 18, p. 873-876 (in Russian).
11. V.A. Zhovtyansky, V.G. Nazarenko, V.O. Khomych, et al. Efficiency of the nitriding process in glow discharge plasma // *This volume*, p. 92-94.

Article received 27.10.10

ЭНЕРГЕТИЧЕСКИЕ ХАРАКТЕРИСТИКИ СФЕРИЧЕСКОГО ТЛЕЮЩЕГО РАЗРЯДА

В.А. Жовтянский, О.В. Анисимова, В.А. Хомыч, Ю.И. Лелюх, В.Г. Назаренко, Я.В. Ткаченко

Конечной целью данной работы является оптимизация процессов модификации, основанных на использовании тлеющего разряда (ТР), а именно азотирования поверхности конструкционных деталей. Эти процессы изучены экспериментально и теоретически в плазме аномального сферического ТР в смеси N_2 -Ar. В теоретической части работы рассмотрены уравнения баланса для плотности заряженных частиц и уравнение Пуассона, дополненные уравнением теплопроводности. Последнее из них описывает влияние горячего катода на процессы в разрядном объеме. Показано также, что корректный учет прианодного падения потенциала играет ключевую роль для адекватного моделирования вольт-амперной характеристики сферического ТР.

ЕНЕРГЕТИЧНІ ХАРАКТЕРИСТИКИ СФЕРИЧНОГО ЖЕВРІЮЧОГО РОЗРЯДУ

В.А. Жовтянский, О.В. Анисимова, В.О. Хомич, Ю.И. Лелюх, В.Г. Назаренко, Я.В. Ткаченко

Кінцевою метою цієї роботи є оптимізація процесів модифікації на основі використання жевріючого розряду (ЖР), а саме азотування поверхні конструкційних деталей. Ці процеси вивчені експериментально і теоретично в плазмі аномального сферичного ЖР у суміші N_2 -Ar. У теоретичній частині роботи розглянуто рівняння балансу концентрації заряджених частинок і рівняння Пуассона, доповнені рівнянням теплопровідності. Останнє з них описує вплив гарячого катода на процеси в розрядному об'ємі. Показано також, що коректне урахування прианодного падіння потенціалу відіграє ключову роль для адекватного моделювання вольт-амперної характеристики сферичного ЖР.