PLASMOCHEMICAL EFFECTS AND SOME FUNDAMENTAL PROBLEMS OF THE PHYSICS OF GAS DISCHARGES

V.A. ZHOVTYANSKY

The Gas Institute, Nat. Acad. Sci. of Ukraine
(39, Degtyarivs'ka Str., Kyiv 03113; e-mail: zhovt@ukr.net)

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Some fundamental problems of investigations of gas discharges are analyzed from the viewpoint of their energotechnological applications. As examples of the specific real or potential application, this work considers the technologies of surface modification of constructional materials and those of the plasma conversion of fuel. For the realization of the former, one widely uses low-pressure glow or arc discharges, while the latter require atmospheric-pressure arc discharges. A general comparison of the properties of glow and arc discharges from the viewpoint of their energy efficiency is performed. It is shown that the key problem for both understanding the physical processes running in these objects and the target searches for the most efficient technological solutions lies in the study of ion-molecular reactions in the region of the interaction of plasma with a solid-body surface or the environment, where the electric arc is maintained.

1. Introduction

The logic of the development of up-to-date plasma technologies puts the necessity of the fundamental investigations in the field of the physics of gas-discharge and low-temperature plasma on the agenda such that go beyond the scope of the traditional approaches in plasma physics. It's indicative that, for various kinds of the gas discharge discussed in the given paper, – low-pressure glow and arc discharges as well as atmospheric-pressure arcs – they are reduced to the study of ion-molecular reactions running in the region of the interaction of plasma with a solid-body surface or the environment, where the electric arc is maintained. This fact objectively testifies to the importance of plasmochemical investigations that were widely performed during the second half of the last century in the former Soviet Union and in Ukraine, in particular. Nowadays, a deficiency of such investigations in Ukraine is obvious.

By the way, this fact is reflected on the formal level by the substitution of the former name of speciality 01.04.08 "plasma physics and chemistry", in which theses for scientific degrees are defended, by "plasma physics".

2. Peculiarities and Problems of Technological Applications of the Plasma of Low-Pressure Glow and Arc Discharges

An important place in the contemporary arsenal of high-technology plasma applications is held by etching technologies in the production of microelectronic devices and the surface modification of constructional materials in mechanical engineering. They are characterized by low pressures ($p \approx 10^{-1} \div 10^2$ Pa) and relatively low degrees of ionization of plasma-forming gases, i.e. the conditions, at which one can use all the advantages of the non-equilibrium state of plasma. As for the surface modification of constructional materials, the most wide-spread operation here is nitriding that provides the improvement of their most important properties [1]. Low pressures condition a high ecological purity of the process, while the technology itself is remarkable for the possibility to control its parameters in such a way as to form nitrided layers with optimal structure.

The extensive material of publications in this field, which are available for today, is mainly restricted to the empirically obtained dependences characterizing plasma as a technological atmosphere (see references in [1]). This fact doesn't allow one to perform a strict analysis of the efficiency factors. In this connection, an urgent problem lies in the study of physical processes in gas-discharge plasma applied to the creation of such technological atmospheres.

Generally speaking, the nitriding methods are realized on the basis of two basic kinds of electric discharges in low-pressure gases: volume arc and glow ones. As is known, the arc discharges include those where the cathode potential drop has a relatively small magnitude of the order of atomic ionization potentials, i.e. ~ 10 V [2]. They are characterized by significant currents ($I \sim 1 \div 10^5$ A) that essentially exceed the typical currents in other kinds of discharges. In contrast, the currents in glow discharges are usually much lower

($I \sim 10^{-4} \div 1$ A); they are also characterized by a high cathode potential drop ($\sim$ 100 V and higher).

Thus, one of the doubtless advantages of the electric arc in the technological applications discussed here, as well as numerous other ones, is its high energy efficiency. Indeed, the arc allows one to transmit considerable currents at relatively small voltages or, correspondingly, power losses. These advantages are conditioned by the optimal combination of the near-electrode and volume properties of the plasma forming the current-carrying arc channel [3].

Though yielding to arc discharges in this respect and also allowing one to obtain much less concentrations of charged particles N_e in plasma (due to insignificant currents), glow discharges still have one of the preferences lying in the possibility of the maximal localization of the technological influence of the discharge on the treated product. It is achieved by means of using the latter as the cathode of a glow anomalous discharge. In this case, the efficiency of the technological influence is reached locally at the expense of the formation of the region of near-cathode potential drop between the plasma and the treated product. As for the arc discharge, it's practically impossible to uniformly close the discharge current on the treated material surface due to the basically different mechanism of running of near-cathode processes. Here, the technological influence can be realized only by the volume discharge of an electric arc. However, a peculiar compromise is often realized: plasma is rather effectively created in the arc discharge, the treated product is placed in the region of the positive glow, while the additional intensification of the process of material treatment is performed at the expense of applying a significant negative potential with respect to plasma to the product. At the Gas Institute of the National Academy of Sciences of Ukraine, nitriding processes in the both kinds of discharges are investigated [1].

We think that, from the viewpoint of the purposeful optimization of the modification processes as a whole (and nitriding, in particular) in these discharges, it's very important to study the peculiarities of ion-molecular reactions accompanying such processes. Such problems involve the generalization of the classic theory of glow discharge developed for the cylindrical geometry to the more general case that combines the consideration of both the diffusion and drift motions of charged particles under the conditions of (for example) the spherical geometry [4].

The theory of glow discharge was developed by Engel and Steenbeck in the first half of the 20th century [5] and is of great importance for low-temperature plasma and gas-discharge physics. The further refinement and the deeper penetration into the course of the process don't conflict with the basic essence of the points considered in [5]. The authors essentially simplified the solution of the problem, which is, generally speaking, described by a system of nonlinear differential equations, and practically divided it into two separate problems concerning the cathode potential drop and the positive glow. The physical ground for such a separation is presented by the experimental facts testifying to the relative independence of these two regions of the discharge.

As for the cathode region, the authors [5] used the theory of gas breakdown in the electric field, according to which electrons ionize neutral particles with the ionization coefficient α (the first Townsend coefficient) depending on the local value of the electric field intensity E:

$$\alpha = Ap \exp[-B/(E/p)],$$

where p denotes the gas pressure, and A and B are constants, whose values for various gases are experimentally determined for certain ranges of E/p. Based on the experiment, it is also assumed that the electric field distribution in the near-cathode region is determined exclusively by the ion space charge and is linear. This fact allows one to theoretically determine the thickness of this region and the characteristic values of the field.

It's considered that positive ions don't ionize the gas in bulk and release electrons only during collisions with the cathode. In this case, the probability of the knocking-out of electrons by ions, $\gamma \sim 10^{-1} \div 10^{-3}$, depends on a material of the cathode, the state of its surface, and the nature of a gas but doesn't depend on the ion velocity. It's also assumed that the recombination of charged particles takes place at the wall, whereas the volume recombination is of secondary importance due to the large field intensity and high velocities of ions and electrons. Finally, this fact allows one to determine the current-voltage characteristic of the region of cathode potential drop [2,5,6].

To a certain extent, the positive column is considered as a secondary part of a glow discharge, whose role consists in the closure of the current from the cathode region to the anode. The fact that the glow discharge is usually investigated in long tubes essentially simplifies the problem, because it is reduced, in the case of the positive column, to the cylindrical geometry. Obtaining the energy from the longitudinal electric field, the electrons are heated up to a high temperature T_e, especially since the energy exchange between these two components is inessential due to low pressures and the difference of masses of electrons and the atomic component. Under the conditions of high T_e, the electron component has a large heat conductivity and, in addition, is sufficiently heat-isolated from the walls of a discharge chamber at the expense of the inevitably arising electric field retarding the electrons. All these points allow one to assume $T_e(r)$ = const, which considerably simplifies the solution of the problem. Based on the balance of the rate of ionization processes from the ground atomic state that exponentially depend on T_e and the losses of charged particles at the walls of the tube under ambipolar diffusion conditions (they depend on the tube radius R and the pressure), one derives the value of $T_e = f(p, R)$ and the relative distribution described by the Bessel function: $N_e(r) = N_{e0}J_0/(2.405r/R)$. The disadvantage of such a solution is the impossibility to determine both the potential drop across the discharge gap and the absolute values of N_{e0} at the cylinder axis. The latter is conditioned by the fact that the parameter N_e is cancelled generally in the course of consideration of the particle balance.

The further development of the glow-discharge theory allowed one to take into account a number of nuances related, for example, to the generation of charged particles only near the cylinder axis, the influence of volume recombination, the formation of the distribution $N_e(r,t)$ in the decaying plasma, the flat geometry of the problem, the determination of the radial distribution of the concentration of metastable atoms, and the account of the step-like ionization of atoms [2, 6, 7]. By the way, in the latter case, the equation for the electric field intensity in a discharge can be included into the consideration. However, all these solutions in their basic features agree with assumptions made in [5].

In the theory of cathode potential drop, one doesn't take into account that the unique correspondence between α and E is possible only in the case where the value of E remains constant at a distance much larger than the electron free path. If the electron moves under the condition of a decrease of the field intensity, as it occurs in the cathode-drop region, then the actual value of α will be larger than that corresponding to the field intensity at the given point. Thus, the actual space charge and the current density must exceed the calculated ones. Correspondingly, the cathode potential drop itself necessary for the generation of a sufficient number of ions must be somewhat lower [2, 8].

The practice of investigations of the processes of surface modification [1] poses more complicated problems concerning the regularities of the distribution of plasma parameters in glow discharges, where the validity of assumptions [5] is violated or unobvious. The solution of these problems is necessary for the purposeful search for the most efficient technological solutions. In particular, such a situation arises in the study of the processes of surface modification in the glow-discharge geometry that approaches the spherical one. Here, it's already impossible to separate the problems of the drift motion of charged particles in the electric field of the discharge and their diffusion to the wall. Moreover, due to a decrease of the discharge current density along the radius of such a spherical diode, there are the doubts about the validity of the assumption $T_e(r)$ = const. The problem is also complicated by the fact that the cathode is heated up to a temperature of ~500 °C, that's why the concentration of gas particles in such a system changes along the radius.

Such a complicated problem can be considered by means of the successive development of the models describing it. At the first stage, we considered it under the simplest assumptions typical of [5], namely $T_e(r)$ = const [9]. It was discovered that, for the cathode and anode radii of 2 and 33 cm, T_e changes within the limits $(2.14\text{–}1.06)\times10^4$ K for the pressure range 1–150 Pa, correspondingly; the electron concentration in the case of $p = 150$ Pa, $I = 4$ A amounts to $N_e(r) \leq 3.3\times10^{11}$ cm^{-3}. Here, the heat-conduction problem is also considered to determine the radial distribution of the gas temperature; the basic temperature drop occurs in the region $r \leq 4$ cm.

The following stage of investigations is presented by work [4], where the problem of the determination of the parameters of a stationary glow discharge in the same spherical diode is considered in the assumption $T_e(r)$= const = 1.16×10^4 K. Its disadvantage lies in the absence of the account of bopth the ionization processes in the "positive glow" region and the self-consistency with respect to the parameter T_e.

It's expedient to determine the peculiarities of the influence of significant temperature changes within the limits of the cathode region taking into account that their characteristic dimensions are comparable; therefore, it's difficult to use approximate estimates in this case. It's also worth studying the possibility of the separate analysis of the ionization processes in the cathode-drop region, where the drift of charged particles prevails, and in the external region, where the processes of bulk ionization can be dominant; this will also require the self-consistent determination of $T_e(r)$.

The following stage of such investigations can be presented by a rather complicated problem of studying the ion-molecular reactions in the region of plasma treatment of a solid-body surface in the positive glow of a low-pressure arc discharge or in the cathode-drop region of a glow discharge with regard for the advantages of using plasma-forming mixtures on the basis of Ar– N_2 empirically proved for today (see, e.g., [1]).

The present publication is based of the theses (see [10]) of my report presented at the XXVIII International Conference on Phenomena in Ionized Gases (Prague, July 15–20, 2007). It turned out that tens of the reports in this representative scientific forum were consonant with this part of the publication and devoted just to both the investigation of the dissociation of molecular gases and the study of the atomic component in the plasmas of nitrogen, oxygen, their mixture with argon, etc. As a rule, they were performed in the low-pressure low-temperature plasma mainly under the conditions of high-frequency discharges characteristic of industrial plasmochemical technologies. In particular, the report of Japanese authors T. Nakano et al. [11] emphasizes the advantages of using N_2 and O_2 diluted with inert gases in microelectronics. The processes of their dissociation are especially important. It was stressed that the „key player" in these technologies is the metastable atom of nitrogen or oxygen. In order to determine the degree of dissociation of N_2 in the discharge with a frequency of 13.56 MHz and a power of 0.3– 1 kW, the emission spectroscopy in the vacuum ultraviolet region was used. The degree of dissociation was changed in the range of 0.006–0.044 (a pressure of 0.13–1.3 Pa). The error of these measurements amounted to 27%. More detailed investigations were performed in the oxygen plasma that also could be diluted by argon. They were carried out with the help of the absorption spectroscopy in the visible and ultraviolet spectral regions. One observed a double increase of the concentration of metastable oxygen atoms up to 7.7×10^{-3} cm^{-3} in the case of the one-percent oxygen admixture as compared to 3.2×10^{-3} cm^{-3} in pure oxygen. It is explained by the increase of the electron concentration, the degree of dissociation of oxygen molecules, and the concentration of metastable argon at a low content of oxygen in the mixture. In this case, the ratio of the concentration of metastable oxygen to that in the ground state increased by a factor 7 – up to 7.7×10^{-2}.

Investigations of such a kind require the use of the methods of actinometry, absorption spectroscopy, or laser-induced fluorescence. In the technical respect, it's most expedient to use frequency-tuned lasers in such experiments. Unfortunately, after the appearance of monograph [12], relatively few works oriented to the investigation of plasma by laser methods were published in Ukraine. However, even in spite of a considerable cost of the laser equipment, this way is rather promising.

3. Perspectives of the Expansion of Technological Applications of the Plasma of Atmospheric-Pressure Arc Discharges and Further Increase of Their Efficiency

The appearance of separate questions concerning the properties of atmospheric-pressure arc discharges and, correspondingly, dense low-temperature plasma is rather unexpected taking into account the more than two-century history of investigations of arcs and their wide application [2, 13]. However, this object appeared to be so "friendly" to experimenters at the initial stages of the study of its potential applications, that it was possible in many cases to restrict oneself to empirically determined properties of arcs. In addition, a part of the questions related, for example, to the instability of the atmospheric-pressure arc conditioned by the problem of removal of heat released in its channel was taken away for a certain period by the plasmatron technique, where the arc was stabilized, by definition, by the plasma-forming gas stream. Thus, figuratively speaking, properties of the arc for a long time remained a some kind of the Brest Fortress (which, as is known, for a long time was unoccupied at the enemy's rear) in the plasma physics. Indeed:

1) Starting from the commonly used explanation of the term "arc" (a form of the discharge conditioned by its emersion under the action of the Archimedean force in the case of the horizontal position of electrodes [2]), it's worth indicating its incorrectness. Indeed, the arc doesn't form an envelope where it could emerge. It only acquires the form promoting to the optimal heat removal from the channel: since the upper part of the arc in the convective stream borders upon the heated air that passed through its channel, one can improve the heat transfer by increasing both the surface area of the arc from this side and its curvature; on the contrary, the lower part of the arc obtains a stream of cold air, that's why, with heat transfer conditions being equal, one can decrease the surface area and its opening for heat radiation.

2) The arc that is freely maintained between electrodes in air represents an extremely inconvenient object for the simulation. For example, by modeling it with the help of an infinite cylinder, it's impossible to provide the

removal of thermal or diffusion flows from it to "infinity" in the framework of the mathematical heat conduction or diffusion problems (in more details, see, e.g., [14]). That's why one should perform modeling with regard for the effects of convection, special cooling, or the so-called stabilizing wall that are not characteristic of an arc proper.

3) In the framework of the equilibrium-based model, even a short arc with the elliptic geometry that describes it well can't remove the heat released in the channel at high discharge currents [14].

4) After accurate measurements of plasma parameters in the atmospheric-pressure arc channel, it turns out that the pressure calculated in the assumption of equilibrium states differs significantly from the atmospheric one (however, the assumption about equilibrium properties of the electric arc plasma is absolutely dominant in the world's scientific literature) [13].

5) The commonly used estimates of the role of diffusion processes $\tau = (R/2.4)^2/D_a$, where τ denotes the characteristic diffusion time, R is the radius of a tube confining the discharge, and D_a is the ambipolar diffusion coefficient, are inadmissible in the arc plasma theory, as a rule, because diffusion is "gripped" by near-wall layers of plasma [3,15].

The further analysis of the problems concerning investigations of the physical properties of arcs is given in the recent publication [13]. An essential part of them can be solved with the help of the model of nonequilibrium properties of the electric arc plasma. They are conditioned by the radiation transfer from the axis (the hottest) region of the arc to the periphery. This results in the overpopulation of the resonance level of plasma-forming atoms [3]. Due to such nonequilibrium state, the arc channel seems to "clear" from the viewpoint of the possibility to transmit the electric current. Indeed, the overpopulation of the resonance level can be considered as a decrease of the effective ionization potential of the plasma-forming atoms, which finally corresponds to a decrease of the resistance of the arc channel and the reduction of the electric current power released in the arc discharge gap almost by 25% [16]. Taking into account that the individual powers of the plasma equipment used in the processes of fuel conversion reach hundreds of kilowatts or even unities of megawatts, it is a significant reserve to increase its effectiveness [17].

Omitting a comprehensive consideration of the experimental and theoretical aspects of similar investigations of an electric arc freely maintained in the atmosphere between melting copper electrodes (they were discussed in the publications cited above), let's indicate typical difficulties that accompany the further analysis of the processes characteristic of such arcs. One of the most complicated problems is the adequate consideration of atom-molecular processes at the arc periphery. Indeed, in the case where plasma is formed in the mixture of copper vapors and atmospheric air, the dominant plasma-forming component is copper atoms that have the lowest ionization potential in the mixture. In the process of diffusion expansion from the arc channel, copper atoms, being in the plasma state, have a possibility to populate a metastable level; this process is also promoted by the flux of the resonance radiation of plasma that permanently runs through the indicated diffusion flow. In fact, the absorption of this radiation represents the prime cause of the overpopulation of the resonance state of copper atoms within the arc channel at its periphery. At the macrolevel, an analog of the latter effect is the fireplace one – the heating of objects under conditions of a generally cold room due to the absorption of heat radiation. This fact was convincingly proved by the direct experiment, where the radiation produced by a copper vapor laser with a wavelength of 510.5 nm (which corresponds to the spectral transition between the resonance and metastable levels) was absorbed by the arc [18]. This experiment allowed one to determine the radial distribution of metastable copper atoms. It turned out that the region, where this population was considerable, exceeded the region of radiation of the arc itself in the radial coordinate by a factor of 3–5. Thus, in this rather extended region, there must take place the processes of excitation by the resonance radiation and deactivation of the copper metastable state, in which an essential role can be played by molecular nitrogen (as a certain analog, one can consider work [19]). However, as far as the author knows, parameters of such an interaction weren't investigated.

It's interesting that the discovery of this effect in the copper-air plasma of an electric arc occurred, so to speak, in the second turn of the investigations of important plasma properties related to the resonance radiation transfer, at the same laboratory. The first discovery was made as early as in the 1960s [20]; it consisted in the formation (without participation of electric or magnetic fields) of plasma with $N_e \sim 5{\times}10^{11}$ cm^{-3} in the confined volume filled with cesium at an optimal pressure of 13 Pa due to its external irradiation by means of the electric discharge also burning in cesium. The latter discharge was maintained in the pressure range of 5–25 Pa ($N_e \sim 10^{14}$ cm^{-3}) in the space between two concentric tubes enclosing the mentioned volume. The physical reason of the origination of this plasma lies in the absorption of the resonance radiation emitted by the external discharge. The existence of such plasma is a proved fact. (Later it was registered as the discovery “Phenomenon of formation of photoresonance plasma”, authors: A.M. Klyucharev, Yu.P. Korchevoy, N.D. Morgulis, A.M. Przhonskii, M.S. Ryazanov; Russian Academy of Natural Sciences, diploma N 106, 1999). The mentioned publication [20] gave an incentive to the further detailed investigation of atom-molecular processes in cesium plasma and its mixtures (see [19, 21–23]).

Returning to the “clearing” of plasma with respect to the resistance to the electric current in the arc channel, it’s worth noting that, in [16], a model – in fact, limiting – equation of state of nonequilibrium plasma was used. A strict analysis of the character of this nonequilibrium requires again (similarly to the above-mentioned case of low-temperature low-pressure plasma) to consider the role of the processes of atom-molecular reactions between Cu and N_2 in the arc channel in the deactivation of excited copper atoms.

Still more complicated (but important and large-scale) problems for energotechnological applications of dense plasma are those concerning the kinetics of atom-molecular processes in technologies of the plasma conversion of carbon-containing raw materials [17]. The dynamics of the world prices of primary energy resources of the basic kinds observed in the recent years creates the necessary prerequisites for their development. In turn, it only reflects the actual situation concerning the ratio of the world’s reserves and the availability of the basic kinds of primary fuel-energy resources on the economical level. Indeed, fossil fuel will remain the basis of mankind’s economics during the next decades. However, with the consumption rates remaining the same, the oil reserves will last for 40, natural gas – for 65, and coal – for 250 years [24]. As compared to the data as of the 1970s (see, for example, [25]), the basic relations of the long-term analytical predictions didn’t essentially change; only the time moment of the depletion of the reserves somewhat shifted to the future by absolute estimates. In addition, coal reserves are distributed rather uniformly among the basic regions and countries of the world making no stress between states concerning the availability of this kind of minerals. This fact provides a relative stability of its price indices. In contrast, the world prices of oil dramatically changed during the last year; the same concerns prices of natural gas supplied to Ukraine.

The technologies of the plasma conversion of carbon-containing raw materials practically allow one to reanimate, on the modern level, the production of synthetic gas-like and liquid fuels that rapidly developed in the world since the 19th century (starting from the illumination of cities at that time) but were replaced in the second half of the 20th century by oil and natural gas that were cheap and available in that period [26].

The plasma gasification is characterized by the fact that the heat necessary for the reaction of formation of the synthesis-gas $H_2 + CO$ from a carbon-containing raw material (endothermic by its character)

$$C + H_2O = H_2 + CO - 131.5 \text{ kJ/mole}, \qquad (1)$$

is introduced into the system, for example, with water plasma rather than at the expense of coal combustion [27]. It allows one to essentially intensify the process by decreasing its dependence on the raw material quality and by increasing the quality of the obtained synthesis-gas – first of all, by reducing the influence of detrimental impurities on next technological processes of its use or processing – at the expense of the realization of the conversion process in the high-temperature region.

Today, E.O. Paton Electric Welding Institute of the National Academy of Sciences of Ukraine and the Gas Institute of the National Academy of Sciences of Ukraine perform the construction of an experimental setup for the realization of such technologies. However, the designing stage has already testified to the fact that the processes in such setups can now be analyzed only on the basis of the so-called weighted-mean indices, when the plasma processes are calculated by using their integral energy characteristics without regard for the peculiarities of plasmochemical reactions. This fact again doesn’t allow one to purposefully optimize the operation of such setups.

4. Conclusions

Real needs for the development of high-tech productions based on the use of plasma technologies in industry and power engineering accentuate the necessity of the activation of investigations in the field of plasmochemistry.

The execution of such investigations requires the continuation of the activity traditions of the scientific schools led by N.D. Morgulis (Taras Shevchenko Kyiv National Unversity) and Yu.P. Korchevoy (Institute of Coal Energy Technologies, Nat. Acad. Sci. of Ukraine).

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ПЛАЗМОХІМІЧНІ ЕФЕКТИ І ДЕЯКІ ФУНДАМЕНТАЛЬНІ ПРОБЛЕМИ ФІЗИКИ ГАЗОВОГО РОЗРЯДУ

В.А. Жовтянський

Р е з ю м е

Аналізуються деякі фундаментальні проблеми досліджень газових розрядів з точки зору їх енерготехнологічних застосувань. Як конкретні реальні або потенційні застосування у цій роботі розглядаються технології модифікації поверхні конструкційних матеріалів та плазмової конверсії палива. Для реалізації перших широко застосовуються жевріючі або дугові розряди низького тиску, а для других – дугові розряди атмосферного тиску. Проведено загальне порівняння властивостей жевріючих та дугових розрядів з точки зору їх енергетичної ефективності. Показано, що ключовою задачею для розуміння фізичних процесів у цих об'єктах та цілеспрямованих пошуків найефективніших технологічних рішень є вивчення іонно-молекулярних реакцій в області взаємодії плазми з поверхнею твердого тіла або ж в навколишній атмосфері, в якій підтримується електрична дуга.