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MACROPARTICLE ACCELERATION IN A CARBON ARC

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Consideration is given to the mechanisms of acceleration of carbon macroparticles forming a cathode coating in a carbon arc under the conditions of fullerene formation. Based on experimental data and theoretical estimates, the macroparticle velocity is determined. It is shown that on collision of particles with the cathode surface the conditions are realized in the zone of contact where graphite fusion can occur.

Recently the carbon arc has aroused substantial interest owing to its application for obtaining fullerenes and nanotubes. For this, an a.c. arc discharge at a reduced pressure of the inert atmosphere is usually employed with use of graphite rods as electrodes. In the course of a discharge, an anode undergoes intense destruction. The destruction products condense on the walls of a discharge chamber in the form of fullerene-containing carbon black and form a coating on the cathode. The fullerene-containing carbon black consists of amorphous carbon particles sized from 0.02 to 0.05 μm, graphite particles of micron size, and crystallites of fullerite C_{60} comparable in size to the amorphous particles [1].

An investigation of the processes of fullerene synthesis [2] has shown that an arc discharge at a reduced pressure of the buffer gas has two modes, the transition between which proceeds spontaneously and does not depend on the discharge parameters. The first mode is a steady arc with uniform destruction of the anode, while the second one represents an unsteady plasma formation with quasiperiodic pulsations of brightness and voltage. Based on the results obtained, a hypothesis has been proposed about the role of the superheat-induced instability in formation of the unsteady mode of the arc.

For the working regimes considered, almost half the mass of the anode sputtered in the arc deposits at the cathode in the form of a coating that possesses unique physical and chemical properties. The fact that the material of anode erosion deposits at the cathode in a great amount is indicative of the existence of directivity of the flow of carbon macroparticles toward the cathode. The main reasons for the existence of such a directivity can include the influence of an electric field between the anode and the cathode that accelerates the positively charged carbon particles toward the cathode and the gasdynamic force induced by rotation of a spiral arc [3].

On the basis of the results of an all-round study of the plasma parameters by the optical and spectroscopic methods and the properties of carbon structures obtained by the microscopic and x-ray structural methods, in [4] a model of formation of a cathode coating is suggested. It is based on the physical and chemical changes that occur on collision with the cathode surface of the carbon macroparticles charged as a consequence of electronic thermal emission and accelerated by an electric field of the arc.

The present work is aimed at elaboration of the suggested model. It involves an experimental determination of the velocity of carbon macroparticles, an investigation of the mechanisms of macroparticle acceleration in an arc plasma and evaluation of the parameters of a boundary layer on collision of particles with the cathode.

In the considered model of formation of a cathode coating, the velocity of motion of macroparticles to the cathode is the governing parameter. To determine its value, scanning of the arc emission in time was carried out by a high-speed photographic camera operating in the regime of photorecording. An image of the axial section of the carbon arc was formed with the aid of a narrow slot parallel to the discharge axis and located either in the plane of an intermediate image or immediately in front of the moving film. The experiments were carried out at the following parameters of a discharge: current I = 80–100 A, initial pressure of helium in the discharge chamber P_0 = 10–500 torr, length of the discharge gap L = 1–5 mm, arc voltage U = 20–40 V, and rate of destruction of the graphite anode of diameter 6 mm R_a = 4–24 mm/min (3–18 mg/sec).

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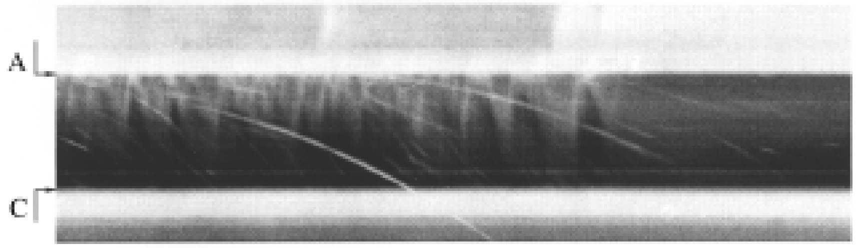

Fig. 1. Continuous scanning of the axial section of the carbon arc (arrows A and C indicate the positions of the anode and cathode surfaces, respectively).

It has been found that on continuous scannings of the axial section of the carbon arc at the end of the unsteady mode and at the beginning of the steady one the tracks of particles are always observed (Fig. 1). They pertain to sufficiently large particles whose size exceeds 100 μm. The velocity of these particles determined by the slope of the tracks exceeds 20 m/sec near the cathode surface.

In experiments, we have not managed to record reliably tracks of the particles with a size less than 100 μm. At the same time, as is known from [4, 5], on the coating surface of the cathode there are quasispherical 20–40-μm formations consisting of the particles sized to several micrometers that, in turn, are formed by smaller particles. We can assume that coalescence of the small particles proceeds in a plasma flow, while the coating is formed by the macroparticles with a diameter of ≈20 μm colliding with the cathode surface. Since it is impossible to determine experimentally the velocities of such macroparticles, we have evaluated the roles of the mechanisms of their acceleration in a plasma flow.

In a plasma of the considered arc, the heated carbon macroparticles emit electrons and, becoming charged positively, can increase substantially the electron concentration in the plasma. On the contrary, if the particles absorb electrons from the plasma, they are charged negatively and decrease the number of free electrons.

We will consider the charge acquired by a spherical particle of radius R that is in the plasma, whose electron and ion concentrations in the distance from a particle are equal to each other, $n_{e\infty} = n_{i\infty}$. In [6], it has been shown that in the case where a particle leads to strong inhomogeneity of the plasma in its neighborhood, it turns out to be a strongly screened plasma. The thickness of the screening layer is small as compared to the particle radius. Moreover, at large distances ($r >> R$) the particle is perceived as the point one with an effective charge (in units of the electron charge)

$$Z^* = \frac{4kT_pR}{e^2} \tanh \frac{\Phi_s}{4} = \frac{4kT_pR}{e^2} \frac{\sqrt{n_{e.s}} - \sqrt{n_{e\infty}}}{\sqrt{n_{e.s}} + \sqrt{n_{e\infty}}}, \tag{1}$$

where $\Phi_s = \Phi(R\kappa_\infty) = \ln (n_{e.s}/n_{e\infty})$ is the value of the potential on the particle surface, $\kappa_\infty^{-1} = (kT/8\pi e^2 n_{e\infty})^{1/2}$.

The electron concentration near the surface of a sufficiently large particle ($R\kappa_\infty >> 1$) is determined by the Richardson–Dushman formula

$$n_{e.s} = 2 \left(\frac{mkT_p}{2\pi \hbar^2} \right)^{3/2} \exp (W/kT_p), \tag{2}$$

where for graphite W = 4.7 eV.

The true charge of the particle is

$$Z = \frac{2kT_pR^2}{e^2} \kappa_\infty \sinh \frac{\Phi_s}{2}. \tag{3}$$

If the potential of the particle is great, the effective charge Z^* can be substantially smaller than Z.

TABLE 1. Charge of a Solitary Particle as a Function of T_{pl} at $T_p = 4000$ K

T_{pl}, K		4000	6000	8000	10 000
Z^*	$2R = 20$ μm	$9.3 \cdot 10^3$	$4.8 \cdot 10^3$	$-1.4 \cdot 10^3$	$-4.5 \cdot 10^3$
	$2R = 4$ μm	–	$9.6 \cdot 10^2$	$-2.7 \cdot 10^2$	$-8.9 \cdot 10^2$
Z	$2R = 20$ μm	$3.0 \cdot 10^5$	$2.2 \cdot 10^5$	$-1.6 \cdot 10^5$	$-1.2 \cdot 10^6$
	$2R = 4$ μm	–	$8.6 \cdot 10^3$	$-6.6 \cdot 10^3$	$-4.9 \cdot 10^4$

The plasma temperature T_{pl} at different instants of time in different spatial zones of the arc changes within the temperature range of 4000–10,000 K. Results of the calculation of a charge in the plasma of a solitary particle of size 20 and 4 μm with a surface temperature of 4000 K by formulas (1) and (3) are given in Table 1.

Thus, in the carbon arc plasma of reduced pressure such conditions are realized that the carbon crystallites formed on erosion of the graphite anode and the particles formed as a result of their agglomeration can acquire both positive and negative charges and a charge can have a very large value. In this case, the plasma temperature $T_{pl} \sim 7000$ K corresponds to that zone of the arc where a carbon particle changes the sign of its charge.

In an electric field of the arc, the positively charged particles are accelerated in the direction of the cathode. The velocity acquired by particles can be evaluated by the formula

$$V_p = (2ZeU/m_p)^{1/2} = [3ZeU/(2\pi\rho R^3)]^{1/2} . \tag{4}$$

Proceeding from a density of the initial graphite anode of $\rho = 1.6$ g/cm^3 and assuming a voltage drop across the discharge gap of $U \sim 30$ V to be the accelerating difference of potentials, we obtain $V_p \sim 0.7$ m/sec for the particle with a diameter of 20 μm and a charge of $Z = 3.0 \cdot 10^5$.

In accordance with (3), for $T_p = T_{pl}$ with the surface temperature of the 20-μm-diam. particle increasing from 4000 to 5000 K, which is quite realistic for such a material as graphite, its true charge increases fivefold. One more reason for the higher value of the electric charge of the macroparticle can be its extended surface. If the particle represents a dense packing of smaller spherical particles, the actual area of its surface will be at least twofold larger. This must lead to a twofold increase in the concentration of thermal emission electrons near the particle surface. As a result, the acquired charge will be approximately one and a half times higher. The higher value of the macroparticle charge will be attained due to photoemission of electrons under the action of the high-power ultraviolet radiation of the arc plasma. However, accounting for this phenomenon is a complicated problem and requires its own consideration. Thus, adopting a value of $2.2 \cdot 10^6$ for Z, we obtain from (4) still a very low value of $V_p \sim 1.8$ m/sec for the particle velocity. This is apparently indicative of the prevailing role of the gasdynamic mechanism of particle acceleration.

In [3], it has been established that in the unsteady mode the arc represents two spiral channels rotating around the lateral surfaces of cylindrical electrodes with a frequency of about 10^4 Hz. To increase the stability of the spiral channel rotating with a large speed, the latter undergoes splitting into two symmetrical channels stemming from an anode spot. Figure 2 schematically shows a model of such an arc and continuous scannings obtained by computer simulation (Fig. 2a and b) that correspond to those observed in the experiment (Fig. 2c and d). Fundamental agreement of the experimental and calculated data is indicative of the possible use of the suggested model for describing the behavior of the rotating arc in the considered range of discharge regimes.

It is known that as a result of eddy motion of a gas, rarefaction develops on the vortex axis. An investigation the vortex-type apparatuses [7] has shown that the degree of rarefaction in this case can be substantial. Assume that under the conditions of our experiments the pressure drop in the discharge gap is equal to the pressure in the discharge chamber P_0. In this case, the upper case of the velocity acquired by particles is

$$V_p = \left(2L\,\frac{\pi R^2 P_0}{m_p}\right)^{1/2} = \left(\frac{3LP_0}{2\rho R}\right)^{1/2} . \tag{5}$$

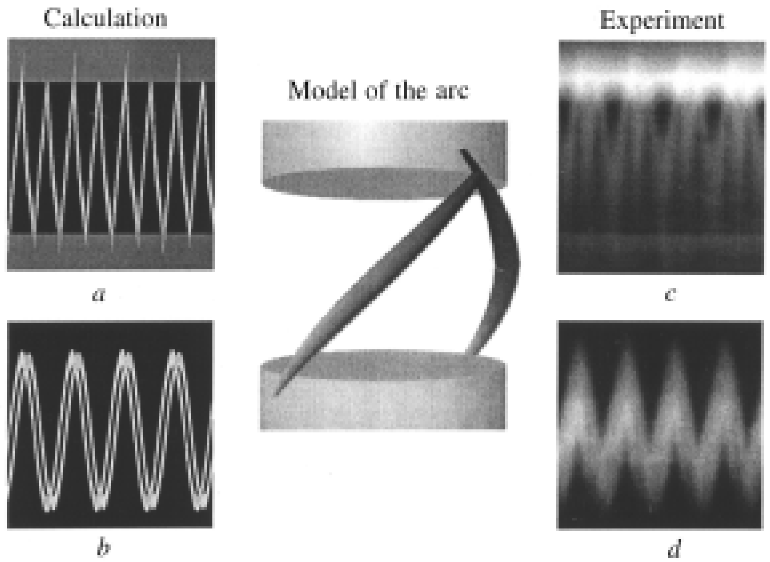

Fig. 2. Model of the arc between the graphite electrodes at a reduced pressure and continuous scannings of radiation of the longitudinal (a, c) and transverse (b, d) sections of the arc.

At a length of the discharge gap of L = 5 mm and a pressure of P_0 = 100 torr we obtain $V_p \sim 80$ m/sec for the 20-μm-diam. particle.

In our experiments, even for coarse carbon particles (~100 μm) velocities of $V_p > 20$ m/sec are recorded. It is obvious that for both the electric and gasdynamic mechanisms of acceleration the dependence of the particle velocity on its radius is of the form $V_p = f(R^{-1/2})$. Thus, based on the experiment and theoretical estimates for particles sized to 20 μm, the expected velocity must exceed 50 m/sec.

On collision of a particle with the cathode surface its kinetic energy converts into heat. The time of energy conversion is equal to the time of particle deceleration that can be evaluated as $\tau = 2R/V_{sound}$. For V_{sound} = 200 m/sec we obtain $\tau = 2 \cdot 10^{-8}$ sec. For this time the thermal front propagates to a depth of $\Delta x = (a\tau)^{1/2} \sim 1.4 \cdot 10^{-7}$, where $a = \lambda/(c\rho)$ ($\lambda = 6$ J/(sec·m·K) and $c = 3000$ J/(kg·K) are taken for polycrystalline graphite at a temperature of 3800 K [8]).

Let us assume that on collision only one-fourth of the particle surface makes contact with the cathode. Consequently, the share of the particle volume in which heat releases is $\delta = 2(\pi R^2 \Delta x)/(4\pi R^3/3) = 3\Delta x/2R \sim 10^{-2}$. The increase in the temperature in the layer of thickness δ is

$$\Delta T = V_p^2/2c\delta \, . \tag{6}$$

On collision of the particle with the cathode in the zone of contact the pressure also increases; the value of this increase can be evaluated as

$$\Delta P = \frac{m_p V_p}{\pi R^2 \tau} = \frac{2}{3} \rho V_p V_{sound} \, . \tag{7}$$

For a particle of size 20 μm, on its collision with the cathode at a speed of 50 m/sec the increment of the temperature and pressure in the boundary layer is $\Delta T \sim 40$ K and $P \sim 10^8$ Pa, respectively. Consequently, in the zone of contact conditions are realized at which graphite can fuse in accordance with the phase diagram of the state of carbon [9].

The analysis of the conditions in the heterogeneous plasma of the carbon arc of reduced pressure shows that the largest contribution to the acceleration of carbon macroparticles is apparently made by the dynamic action of the plasma flow as a consequence of rotation of the spiral arc and intense erosion of the anode. The velocity of the macroparticles forming the cathode coating can exceed 50 m/sec. On collision of particles with the cathode surface in the zone of contact, temperatures and pressures sufficient for passing solid graphite into the fused state are realized.

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NOTATION

I, current strength; U, voltage; P_0, initial pressure of helium; L, length of the discharge gap; R_a, rate of anode destruction; $n_{e\infty}$ and $n_{i\infty}$, electron and ion concentrations in the plasma; $n_{e.s}$, electron concentration near the particle surface; R, particle radius; r, distance from the center of the particle; Z and Z^*, true and effective particle charges in units of the electron charge; T, temperature; e, electron charge; k, Boltzmann constant; Φ, value of the potential; κ_∞^{-1}, Debye length of screening; W, electronic work function; V, velocity; ρ, density; m, mass; τ, time of particle deceleration; V_{sound}, sound velocity in the particle material; Δx, depth of propagation of the thermal front; a , λ, and c, thermal diffusivity, thermal conductivity, and heat capacity; δ, share of the particle volume in which heat release occurs; ΔT and ΔP, increments of the temperature and pressure in the zone of contact of the particle with the surface. Subscripts: p, particle; pl, plasma; s, surface; a, anode; e, electron; i, ion; sound, sound.

REFERENCES

1. V. D. Shimanovich, V. B. Shipilo, G. K. Zubovich, I. P. Smyaglikov, and I. V. Gasenkova, *Vestsi Nats. Akad. Navuk Belarusi, Ser. Fiz.-Mat. Navuk*, No. 1, 109–113 (2000).
2. V. D. Shimanovich, I. P. Smyaglikov, and A. I. Zolotovskii, *Inzh.-Fiz. Zh.*, **71**, No. 4, 669–674 (1998).
3. V. D. Shimanovich, I. P. Smyaglikov, A. I. Zolotovskii, S. M. Pankovets, N. I. Chubrik, and S. V. Goncharik, *Pis'ma Zh. Tekh. Fiz.*, **63**, Issue 11, 80–83 (2000).
4. V. M. Anishchik, I. A. Pyzhov, I. P. Smyaglikov, and V. D. Shimanovich, *Inzh.-Fiz. Zh.*, **74**, 115–119 (2001).
5. V. M. Anishchik, I. A. Pyzhov, I. P. Smyaglikov, and V. D. Shimanovich, in: *Proc. III Int. Conf. on Plasma Physics and Plasma Technology*, Vol. 1, Minsk, Belarus (2000), pp. 381–384.
6. D. I. Zhukhovitskii, A. G. Khrapak, and I. T. Yakubov, in: B. M. Smirnov (ed.), *Chemistry of Plasma* [in Russian], Collection of Papers, Issue 11, Moscow (1984).
7. M. G. Dubinskii, *Izv. Akad. Nauk SSSR, Otd. Tekh. Nauk*, No. 8, 3–10 (1955).
8. J. Abrahamson, *Carbon*, **12**, 111–114 (1974).
9. *Encyclopedia of Chemistry* [in Russian], Vol. 1 (1988), pp. 1189–1192.