PRODUCTION OF COATINGS FROM POWDER MATERIALS WITH THE USE OF A SHORT ARGON ARC. 1. EXPERIMENTAL STUDY OF PLASMA HEATING OF MACROPARTICLES

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Consideration is given to the processes of acceleration and heating of metallic particles in the device proposed by the authors for deposition of metallurgical coatings with the use of a short argon arc. The coatings produced are compared to those deposited by the traditional method of plasma spraying. Results of spectroscopic determinations of the temperature fields of the arc are given. The heat-flux density and the temperature of macroparticles are evaluated by the parameters of their destruction in the plasma found experimentally. The high efficiency of the technology proposed for heating of metallic powders, including high-melting ones, is shown.

Introduction. Deposition of protective coatings is widely used in engineering to decrease the influence of mechanical and chemical actions on the surfaces of elements of machines and mechanisms. One of the most widespread methods of producing them is gas-thermal (gas-plasma, plasma, detonation) spraying of powder materials. The maximum temperature of heating of particles is attained in plasma spraying. The high adaptability and universality of this method make it possible to produce elements and deposit coatings of various functional purposes in a wide range of their thicknesses. However the traditional methods of spraying have substantial drawbacks. As a consequence of the rapid hardening of the particles sprayed and the formation of oxide films on their surface and of the presence of high tensile stresses between the particles in their shrinkage during crystallization, the most important properties (density, homogeneity, strength) of the elements produced are much poorer than those of forged or molded pieces. The subsequent heat treatment of products with plasma-deposited coatings makes it possible to improve adhesion and cohesion; however, this process can lead to inadmissible changes in the geometric shape of a product.

The results of investigations [1, 2] show that the parameters of cohesion and adhesion of a coating are improved with increase in the temperature in the zone of contact of a sprayed particle with the surface of a product. This is attained either at high velocities of the particles (detonation spraying, $V_p > 1000$ m/sec) or in their heating much above the melting temperature. The degree of heating of the particles mainly depends on the plasma parameters in the zone of a plasma device into which the sprayed powder is introduced.

In plasma sprayers, one usually employs a d.c. electric arc to heat the working medium. The particles are introduced into the peripheral turbulent zone of a plasma jet with a temperature of 5000 to 7000 K. In this case they can be heated to temperatures only insignificantly higher than the melting temperature [2–4]. The temperature of the particles can substantially be increased as compared to the temperature realized in traditional technologies of plasma spraying in their heating in the cathode region of the arc, where, according to [5–8], the plasma temperature can be higher than 20,000 K.

The possibility in principle of forming dense homogeneous coatings with the use of a short argon arc has been shown in [9]. The high cohesion and adhesion parameters are attributed to the presence of a metallurgical bond between the particles and between the particles and the base (when the transition layer between them represents a metal alloy). The plasma of the considered arc has been diagnosed in [10, 11]. To elucidate the physical processes occurring when the proposed technology is employed and to establish its prospects for solution of practical problems the

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Fig. 1. Diagram of the experimental stand (a) and photographs of the metallographic sections of coatings deposited by the traditional method of plasma spraying (b) and produced on the stand (c). P, plasmatron; L, laser; O_1 , O_2 , and O_3 , objectives; Sh₁ and Sh₂, optical shutters; F, optical filter; S, slit; PF, photographic film, SG, spectrograph.

authors carried out a complex of experimental and theoretical investigations. In the present work, representing the first part of the investigations, we describe the experimental setup, compare the properties of the coatings produced to those attained in traditional plasma spraying, give results of spectroscopic determinations of the temperature fields in the plasma formation employed, and evaluate the heat-flux density and the temperature of particles by the parameters of their destruction in the plasma found experimentally.

Experimental Stand and Structure of the Coatings Produced. A diagram of the experimental stand for production of coatings by spraying of powder materials using a short argon arc is given in Fig. 1a. The water-cooled cathode unit contains a nozzle for supplying a plasma-forming gas and a powder and a cathode in the form of a tungsten rod 6 mm in diameter sharpened as a cone with an angle of 60° C and having a flat site 1 mm in diameter. The structure of the nozzle ensures local supply of a powder to the zone near the cathode ($z \sim 2$ mm) without the particle sticking to the cathode surface and the nozzle walls. The anode is a cylinder 95 mm in diameter whose rotation and reciprocating movement ensures a linear velocity of motion of its surface of 0.2 m/sec relative to the discharge axis.

As a consequence of the high temperature of the plasma the particles of powder materials are heated to the formation of liquid droplets. Because of evaporation, much of their mass changes to a vapor phase, which contributes to the formation of dense homogeneous coatings with high cohesion and adhesion parameters. The laminarity of the conveying flow ensures protection of a heated particle against contact with the ambient air atmosphere, which prevents the surface oxidation of the particle and of the layer formed on the anode-product. Overheating of the product is eliminated by rapid scanning of the attachment of the arc on its surface and by forced cooling. The basic characteristics of the technology proposed are as follows:

strength of cohesion of the coating with the base >150 MPa; porosity of the coating <0.5%; powder-utilization factor 0.95; productivity of the process 2–5 kg/h; arc-current strength 100–200 A; power of the plasmatron 2–6 kW; consumption of the gas 5–20 liters/min; specific expenditure of energy <4kJ/g.



Fig. 2. Isotherms of the argon arc for P = 1 atm, I = 200 A, and $R_{Ar} = 5$ liters/min. The temperature contours are in units of 1000 K. *z*, *r*, mm.

The experiments have been conducted with a bronze powder (90% Cu + 10% Sn); the average particle size was $\sim 90 \,\mu\text{m}$. The most optimum regime for forming homogeneous coatings with a high strength of cohesion with the product surface is as follows: current strength 200 A, consumption of the argon 5 liters/min, consumption of the powder 2 kg/h, and distance between the cathode and the anode 10 mm.

Figure 1 shows the structure of a bronze coating on steel deposited by the traditional method of plasma spraying (b) in the case of introduction of a powder onto the cut of an arc-plasmatron nozzle [1] and produced on the stand described (c).

The porosity evaluated by the ratio of the pore area to the coating area under study amounts to $\sim 15\%$ in traditional spraying. Upon heat treatment, the pores acquire the shape of globules 10 to 50 µm in size. The strength of cohesion of such coatings with the base of 45 steel without a sublayer is about 3 MPa.

In deposition by a short argon arc, the porosity of the coatings decreases by more than an order of magnitude as compared to the traditional plasma spraying and it amounts to ~0.5%. The cohesive strength is higher than 150 MPa, and the powder-utilization factor is 0.95. The analysis of the structure of the coatings produced demonstrates that the thickness of the intermediate layer between the coating and the base is no higher than 10^{-5} m.

Parameters of the Arc Plasma. As has been indicated above, the parameters of the proposed technology are prescribed primarily by the high temperature of the plasma formation employed. The temperature profiles of the arc plasma were determined by the Lorentz–Fowler–Milne method from the relative intensities of the 695.5-nm ArI spectral line [10, 11]. We recorded intensities integral over the observation beam for the arc cross sections at different distances from the cathode (z = 1, 2, 3, 4, 6, and 8 mm); these intensities were transformed to the radial values of the emission coefficient with the use of the Abel inversion. The data obtained enabled us to construct the isotherms of the arc (Fig. 2). The region of the cathode jet is pronounced in its structure, and a certain contraction of the arc channel toward the anode is observed. The plasma temperature is maximum on the arc axis near the cathode and it is ~22,000 K.

The analysis of the results obtained in the work demonstrates that the parameters of the plasma formation in question are much higher than the parameters of the working region that determines heating of the particles in traditional plasma sprayers. If a comparison is made on the basis of the average plasma temperature, it is about 15,000 K, i.e., almost twice as high, in the present work. As a consequence, high homogeneity and adhesion of coatings are attained in short-arc spraying. At the same time, the data on plasma parameters obtained can be used as the basis for calculating heat fluxes to metallic particles with the aim of optimizing the processes of deposition of coatings.



Fig. 3. Photographs of the particles in a plasma flow (a) (frame spacing $5 \cdot 10^{-5}$ sec, A and C, positions of the anode surface and the nozzle cut), diffraction tracks of the particles (b), and scheme of formation of the track on a moving film (c).

Structure of a Heterogeneous Plasma Flow and Parameters of Particles. The properties of the coatings produced are determined by the parameters of the metallic particles in a plasma flow. The structure of a heterogeneous plasma flow has been studied using high-speed chambers operating in the regimes of continuous photorecording (Fig. 1a, I) and frame-by-frame filming (Fig. 1a, II) with a time resolution of $5 \cdot 10^{-5}$ and 10^{-6} sec respectively.

Figure 3a shows the individual frames of filming of the arc. It is seen that the particles sprayed are surrounded by radiating shells of a shape extended to 3 mm in the direction of motion of the plasma flow (toward the anode). The average transverse dimension of the shell is 0.5 to 1 mm, which is an order of magnitude higher than the initial size of the particles supplied to the plasma.

The size of particles in the plasma flow was determined by the method of observation of the continuous scans of their diffraction images resulting from the shadowing of the laser beam. The recorded diffraction track of a particle (Fig. 3b) has the form of alternating light and dark bands located symmetrically about the central light band with a strong maximum beyond its geometrical shadow. The narrowing of the tracks at the ends is related to the presence of a certain angle between the slit and the direction of motion of a particle, and it is governed by the input and output of its image in the slit space. The size sought was restored from the distance d_m between narrow diffraction minima after the first maximum beyond the geometrical shadow of the particle.

The scheme of formation of a diffraction track on the photographic film is shown in Fig. 3c. It follows that

$$d_{\rm m} = \frac{h_{\rm t}}{\cos\alpha} - \Delta d_{\rm s} \,, \tag{1}$$

where $\Delta d_{\rm s} = h_{\rm s} \tan \alpha$. It is clear that $\tan \alpha = V_{\rm p}/V_{\rm f}$.

The measuring system was calibrated against the diffraction images of objects of known dimensions in the range 50–200 μ m. It should be noted that the diffraction pattern of a cylindrical thread and the diffraction image of the track of a macroparticle in the plasma are the same in a qualitative sense, which demonstrates the absence of the refraction of the laser beam on the plasma shell around the particle. It was established that its actual size and the distance $d_{\rm m}$ are related by the relation

$$R_{\rm p} = \frac{d_{\rm m} - \Delta d_{\rm dif}}{2\beta}.$$
 (2)

Combining (1) and (2), we obtain the expression to determine the particle size from the width of the track of the particle's diffraction image:



Fig. 4. Size (a) and velocity (b) distribution of particles: 1) initial size of the particles; 2) and 3) distribution functions referring to the arc cross sections at a distance of 5 and 9 mm from the cathode respectively. $R_{\rm p}$, μ m. $V_{\rm p}$, m/sec; D, rel. units.

$$R_{\rm p} = \frac{1}{2\beta} \left(\frac{h_{\rm t}}{\cos \alpha} - h_{\rm s} \tan \alpha - \Delta d_{\rm dif} \right). \tag{3}$$

From the slope of the tracks on the high-speed scans we determined the velocity of macroparticles in the plasma flow $V_p = V_f \tan \alpha$, which is 4 to 10 m/sec. On the continuous scans of the axial cross section of the arc, in addition to the tracks of the macroparticles, we observe the tracks of plasma bunches of higher-than-average brightness. The velocity of the plasma flow evaluated from the slopes of these tracks is $V_g \sim 200$ m/sec. This is in agreement with the general ideas of the value of the plasma velocity in the cathode zone of arc discharges [12].

As a result of the statistical processing of several hundred tracks we found the size- and velocity-distribution functions of macroparticles in different cross sections of the arc channel (Fig. 4). The relative error of determination of the velocity and size of an individual particle was no higher than 3 and 15%, respectively. The analysis of the distributions obtained has shown that a particle in the plasma flow moves with an acceleration of $2.8 \cdot 10^3$ m/sec² during the time $t_{\rm h} = 2 \cdot 10^{-3}$ sec, losing up to 50% of its mass. At the moment of collision with the anode surface, the average velocity of particles is $V_{\rm p} = 6.7$ m/sec.

Consideration of the balance of the ion and electron flows from the plasma (and of the thermoelectron flows) yields that a bronze particle with a radius of $4 \cdot 10^{-5}$ m in the plasma acquires a negative charge of $q = -Ze = -2 \cdot 10^5 e$, where $e = 1.6 \cdot 10^{-19}$ C. Negatively charged particles in the electric field of the arc are accelerated in the direction to the anode. The velocity required by them can be evaluated by the formula

$$V_{\rm p}^{\rm e} = (2eZ\Delta U/m_{\rm p})^{1/2} = (3Ze\Delta U/2\pi\rho_{\rm p}R_{\rm p}^3)^{1/2}.$$
(4)

On the basis of the density of bronze $\rho_p = 8.8 \cdot 10^3 \text{ kg/m}^3$ and taking $\Delta U = 20 \text{ V}$ as the accelerating potential difference, we obtain the value $V_p^e = 2.3 \cdot 10^{-2} \text{ m/sec}$ for the particle velocity as a consequence of electrostatic acceleration.

Under the assumption $V_p \ll V_g$, to calculate the velocity of spherical particles accelerated as a result of the dynamic action of a plasma flow we can employ the following expression:

$$V_{\rm p}^{\rm g.d} = V_{\rm g} \left(\Delta z \, \frac{3\rho_{\rm g} A}{4\rho_{\rm p} R_{\rm p}} \right)^{1/2},\tag{5}$$

where Δz is the distance on which a particle is accelerated, and the coefficient of resistance to particle motion for the Reynolds numbers $2 \le \text{Re} < 21$ can be represented as [13]

$$A = (24/\text{Re}) (1 + 0.11 \cdot \text{Re}^{0.81}) .$$
(6)

TABLE 1. Densities of the Heat Flux to a Particle and the Corresponding Evaporation Temperature, Time of Heating to T_v , Evaporation Rate, and Change in the Particle Radius

$Q \cdot 10^{-8}, \text{ W/m}^2$	<i>T</i> _v , K	$t_{\rm v} \cdot 10^3$, sec	μ , kg/(sec·m ²)	$\Delta R \cdot 10^6$, m
0.44	2400	3.5	7	0
1.5	2600	1.1	25	2.5
4.3	2800	0.4	70	12.5
11	3000	0.2	170	35.0

TABLE 2. Parameters of Heating and Destruction of Particles of Different Materials

Material	<i>T</i> _v , K	$t_{\rm v} \cdot 10^4$, sec	$\mu(T_v)$, kg/(sec·m ²)	$\Delta R \cdot 10^6$, m
Bronze	2800	3.4	70	13
Titanium	3475	3.9	45	16
Iron	3005	7.5	50	8
Tungsten	6280	7.4	84	6

The corresponding calculations show that $\text{Re} \sim 10$ and $V_p \sim 7$ m/sec for the argon plasma at T = 15,000 K, which is no different in essence from the value measured experimentally. Thus, the main contribution to the acceleration of the particles is made by the gasdynamic action of the plasma flow.

Discussion of the Results. Let us consider a decrease (as a consequence of evaporation) in the size of a particle moving in the plasma flow. For a spherical particle at T_p the change in the radius over the period Δt is

$$\Delta R = \frac{\Delta t \mu \left(T_{\rm p}\right)}{\rho_{\rm p}} \,. \tag{7}$$

In accordance with [14], the dependence of the evaporation rate on the temperature of the particle has the form

$$\log \mu (T_{\rm p}) = C - 0.5 \log T_{\rm p} - B/T_{\rm p}, \qquad (8)$$

where C and B are constants that are equal to, respectively, 9.63 and 16,980 for copper. The same values of the constants were employed for bronze, since the parameters of evaporation of copper and tin are approximately equal.

A particle in the plasma can be heated to a certain limiting temperature of evaporation $T_p = T_v$ at which the total heat flux on its surface vanishes. Without allowance for the heat loss by melting and radiation that, according to the evaluations, amounts to less than 10% of the heat flux Q from the plasma to the particle, we can write the equation of balance of the heat fluxes as

$$Q = \mu \left(T_{\rm v} \right) \left(L_{\rm v} + cT_{\rm v} \right) \,. \tag{9}$$

The removal of mass in evaporation of the particle during the time t_v of heating to the temperature T_v is insignificant. Consequently, the time during which it is evaporated is

$$\Delta t = t_{\rm h} - t_{\rm v} \,. \tag{10}$$

The value of t_v can be found by solution of the heat-conduction equation for a bronze plate of thickness R heated on both sides by the heat flux Q when T_v and Q are related by relation (9).

The results obtained are presented in Table 1.

If the value $\Delta R \sim 10^{-5}$ m recorded in the experiment is determined by the evaporation of a metallic particle, the heat flux from the plasma to its surface is $4.3 \cdot 10^8$ W/m². The temperature at the center of the particle and on its surface differs by no more than 10%.

On the basis of the heat-flux density determined from experimental data on measuring the size of a bronze particle we can consider heating of powders of different materials in a plasma flow.

Based on the data on evaporation [14] we find the limiting temperature T_v from formulas (8) and (9). Next, considering the thermal problem of heating of a plate of thickness *R* by the heat flux *Q*, we determine the time of heating t_v to the temperature T_v . The calculation results for particles of different materials of radius $4 \cdot 10^{-5}$ m are presented in Table 2 under the assumption of equal time of heating in the plasma, $t_h = 2$ msec.

Thus, the degree of heating of powder materials in a short argon arc is much higher than that attained in jet plasma spraying.

The proposed method of determination of the heat flux and the surface temperature of a particle to be found in the short-arc plasma takes no account of the expenditure of energy on melting it. The evaluations show that the contribution of this mechanism under conditions of an argon-plasma flow with a temperature of $\sim 15,000$ K is much smaller than the error of experimental measurements of the removal of mass. The assumption on the destruction of a particle just by evaporation is basic in this method. This issue necessitates further detailed consideration.

Conclusions. In the work, we describe a plasma unit designed to deposit coatings with the use of a short argon arc. It has been shown that the coatings produced by the method proposed have higher technical indices, including the values of adhesion and cohesion, than those obtained in traditional jet plasma spraying. We have determined, by spectroscopic methods, the temperature fields in the plasma formation employed. The procedure for evaluating the density of the heat flux to the surface of a macroparticle has been proposed and implemented based on the study of the kinetics of its destruction in a plasma flow. It has been established that the value of the heat flux in the device proposed is much higher than the value attained in jet plasma spraying. The temperature of the particles is much higher than the melting temperature in the parameters of their destruction in the plasma found experimentally. We have evaluated the degree of heating of different powder materials in the argon plasma. The results obtained demonstrate the prospects of the equipment and technology proposed for production of coatings for use in practice.

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NOTATION

V, velocity; h_t , track width; Δd_s , correction for the finiteness of the slit width; h_s , slit width; d_m , distance between the minima of the diffraction image; Δd_{dif} , diffraction correction; R, particle radius; D, probability density; T, temperature; m, mass; q, particle charge; Z, particle charge in the units of the electron charge; e, electron charge; ΔU , potential difference; P, pressure; I, current strength; A, coefficient of resistance to particle motion; Re, Reynolds number; Δt , time of evaporation of a particle; Q, density of the heat flux from the plasma to a particle; T_v , limiting evaporation temperature; L_v and c, latent heat of evaporation and heat capacity of the particle material; ΔR , change in the particle radius; z, coordinate reckoned from the cathode surface; r, coordinate reckoned from the arc axis; t, time; α , slope of the track; β , magnification of the optical system; μ , evaporation rate; ρ , density. Subscripts: p, particle; g, plasma; h, heating; v, evaporation; f, film; t, track; m, minimum; dif, diffraction; s, slit; e, electrostatic; d, dynamic.

REFERENCES

- 1. V. V. Kudinov and G. V. Bobrov, *Deposition of Coatings by Spraying. Theory, Technology, and Equipment* [in Russian], Moscow (1992).
- M. F. Zhukov and O. P. Solonenko, in: High-Temperature Dust-Laden Jets in the Processes of Treatment of Powder Materials [in Russian], Novosibirsk (1990), pp. 363–371.
- 3. G. S. Antonov, V. S. Loskutov, B. M. Solov'ev, V. D. Shimanovich, and A. K. Shipai, in: *Generators of a Low-Temperature Plasma* [in Russian], Moscow (1969), pp. 490–500.
- 4. A. I. Zolotovskii, A. F. Il'yushchenko, L. E. Krat'ko, N. N. Naumenko, S. B. Sobalevskii, and V. D. Shimanovich, in: *Ext. Abstr. of Papers presented at Conf. "Theory and Practice of Coating Deposition"* [in Russian], Dmitrov (1983), p. 17.
- 5. K. S. Hsu, K. Etemadi, and E. Pfender, J. Appl. Phys., 54, 1293–1301 (1983).

- 6. G. N. Haddad and A. G. D. Farmer, J. Phys. D: Appl. Phys., 17, 1189–1196 (1984).
- 7. P. Kovitya and L. E. Cram, Welding J., 65, 34-39 (1986).
- 8. V. V. Azharonok, A. I. Zolotovsky, G. P. Lizunkov, and V. D. Shimanovich, in: *Proc. 8th Int. Symp. Plasma Chemistry*, Vol. 3, Tokyo (1987), pp. 1906–1911.
- 9. D. V. Bagino, A. F. Il'yushchenko, V. D. Shimanovich, I. P. Smyaglikov, and A. I. Zolotovsky, in: *Progress in Plasma Processing of Materials*, Wallingford, New York (1997), pp. 771–778.
- 10. I. P. Smyaglikov, A. I. Zolotovsky, and V. D. Shimanovich, in: Proc. 3rd Int. Conf. Plasma Physics and Plasma Technology, Vol. 1, Minsk (2000), pp. 309-312.
- 11. A. I. Zolotovsky, I. P. Smyaglikov, and V. D. Shimanovich, Zh. Prikl. Spektrosk., 69, No. 3, 402-405 (2002).
- 12. W. Finkelnburg and H. Maecker, in: *Elektrische Bogen und Thermisches Plasma. Handbuch der Physik* [Russian translation], XXII, Moscow (1961), pp. 254–444.
- 13. I. S. Burov, Fiz. Khim. Obrab. Mater., No. 2, 42-49 (1979).
- 14. S. Dushman, Scientific Foundations of Vacuum Technique [Russian translation], Moscow (1964).