GAS-DYNAMIC SPRAYING. AN EXPERIMENTAL STUDY OF THE SPRAYING PROCESS

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We present results of an experimental study of the specific features of the gas-dynamic formation of coatings from metallic powders ($d_p < 50 \ \mu m$) on substrates of various materials depending on the particle velocity (200-1200 m/sec), the jet temperature (300-700 K), and other parameters. Results of a prospecting study of the implementation of the methods of particle acceleration in supersonic (M = 2.0-3.0) rectangular nozzles are described. The rate of bond formation in a cold particle-cold substrate contact occurring in gas-dynamic spraying is estimated within the framework of the concepts applied in analysis of gas-dynamic spraying.

The essence of the traditional gas-thermal methods for application of powder coatings implies the necessity of heating a dispersed material up to the melting point or higher and to accelerate and transport this material to the surface by high-temperature jets. This causes complex physicochemical processes, including the reactions of oxidation, the burn-out of fine powders, the decomposition of some materials, etc. [1, 2]. Thus, along with positive processes, some negative processes, which are associated with the use of high-temperature gas or plasma flows, occur. Some approaches that allow one to reduce the influence of adverse factors on the physicotechnical characteristics of coatings were proposed and implemented. These approaches are mainly associated with the optimization of gas-dynamic spraying processes.

The method of "cold" gas-dynamic spraying (CGDS) [3] offers a different solution of this problem. It employs the kinetic rather than the thermal energy of the deposited particles, which are accelerated by a cold gas, thus eliminating adverse high-temperature effects.

Description of a Setup. To carry out experiments for a study of the fundamental laws of coating formation by the CGDS, the need arose to develop experimental setups that ensure a controlled variation of the main parameters of a gas-powder flow. The methodical studies performed in [4] underlay the design of the experimental setup shown schematically in Fig. 1. The main elements of the setup are a spraying unit 1, which consists of a plenum chamber and a plane supersonic nozzle, a gas heater 2, a particle feeder 3, a compressed air source 4, a helium source 5, a spraying chamber 6, a traverse gear 7 to displace the substrate to be sprayed, a control panel 8 for flow parameters (the gas pressure in the plenum chamber and the feeder and the gas temperature in the plenum chamber), and a particle separator 9. The setup ensured the acceleration of particles of diameter $d_p = 1-50 \ \mu m$ and different concentrations up to a velocity $v_p = 200-1200 \ m/sec$ in supersonic nozzles.

The majority of the powders used for cold gas-dynamic spraying had a particle size of 1-50 μ m. These particles have poor looseness, which hinders the use of many traditional methods of dispensing the powder by means of valves, gates, screw conveyers, and even vibrators. The main problems in developing this setup were, therefore, related to the formation of jets with a prescribed controlled concentration of particles. Based on the conducted studies, mixer-feeders of barrel type were developed. During the tests, the mixer-feeders demonstrated high reliability in the absence of noticeable fluctuations of the particle concentration in the jet. A proportioning feeder was also used. It allowed us to introduce a prescribed portion of powder M_0 into the

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gas flow for the time $\Delta t < 1$ sec, which is important if expensive helium is used as the accelerating gas. In addition, this design of the feeder allowed us to change the powder rapidly if only the possibility of deposition of various powders on this or that substrate was checked.

Based on the experiments in [4], we chose plane nozzles to use the CGDS method. These nozzles ensured the minimum thickness of the compressed gas layer ahead of the target. For the supersonic section of the nozzle of length L = 80-120 mm and Mach number M = 2.0-3.0, the smallest size of the nozzle exit section h = 3 mm allowed one to obtain a two-phase supersonic jet with the necessary particle velocity.

Two methods based on the variation of the speed of sound of the working gas were used for particle acceleration. In the first method, a mixture of gases with different molecular weights, namely, air and helium, was used at room temperature as the accelerating gas. The outflow velocity of the working gas was controlled by changing the concentration of the components in the mixture.

Figure 2 shows the calculated velocities of the aluminum particles near the substrate surface as a function of the helium concentration k_{He} for L = 0.1 m, M = 2.35, and $p_0 = 1.5$ MPa. It is seen that changing the mixture composition from pure air to pure helium, we can change the velocity of particle-substrate collisions from 200 to 1200 m/sec. This allowed us to study experimentally the specific features of coating application in a two-phase jet-moving target regime at room temperature and, which is particularly important, to examine the influence of the particle velocity on this process in an explicit form.

It should be noted, however, that the use of gases (helium and hydrogen) with a speed of sound higher than that of air restricts the possibilities of using the CGDS technique in practice because of their shortage, higher cost, and, for hydrogen, higher hazard. In this connection, another method of particle acceleration by an air jet with moderate heating was considered [5]. The gas was heated by an electric heater, which made it possible to vary the gas-jet temperature from 300 to 700 K. The main portion of the working gas was heated up to a given temperature and was mixed in the plenum chamber with a small amount of cold gas (~1%) carrying the particles from the feeder. As the air temperature in the plenum chamber was increased, the speed of sound in the jet, the velocity of its effusion, and the particle velocity also increased. This is clearly seen in Fig. 3, which shows the calculated (for L = 0.1 m, M = 2.35, and $p_0 = 1.5 \text{ MPa}$) velocities of aluminum and copper particles near the substrate surface versus the air temperature in the plenum chamber.

The designed setup allowed us to perform a cycle of experimental studies on coating application by the CGDS method in a two-phase jet-moving target regime.

Experimental Results. Experimental results on determination of (depending on the particle velocity) the coefficient of particle deposition $\Delta m/M_0$, which characterizes the ratio of the increment of substrate amount Δm to the total amount of the consumed powder M_0 , are presented. The coatings were applied to motionless substrates under conditions of strictly dosed portions of various metallic powders with a particle size of 1-50 μ m. The amount of deposited material Δm was measured in each experiment by an analytical balance as the difference of the substrate amount prior to and after deposition. The experimental values of Δm and M_0 being known, the dependence $\Delta m/M_0 = f(v_p)$ was constructed. The experimental values of the







deposition coefficient for the aluminum, copper, and nickel powders versus the particle velocity, which were obtained by accelerating the particles by a mixture of air and helium, are shown in Fig. 4 (curves 1-3). It is seen that, for the examined metallic particles ($d_p < 50 \mu m$), there exists the critical velocity $v_{cr} \approx 500-600 m/sec$ of their interaction with the substrate. The classical process of erosion is observed for $v_p < v_{cr}$ [6]; for $v_p > v_{cr}$, it Lecomes the process of deposition, i.e., a dense metallic layer on the substrate surface is formed, and the character of coating formation changes abruptly as the velocity is further increased. In particular, the value of the deposition coefficient for the examined powders increases from zero to 0.4-0.8 for $v_p \approx 1000 m/sec$.

The effect of the erosion-to-deposition transition is clearly illustrated by Fig. 5, which shows photographs of the trajectories of the incoming aluminum particles and those reflected from the substrate. The processing of these photographs and the examination of the substrate surface showed that, for $v_p \leq v_{cr}$ ($v_p \approx 250 \text{ m/sec}$, Fig. 5a), all the single particles were reflected. As the velocity increased within the range $v_p > v_{cr}$, the character of the particle-substrate interaction changed abruptly: a rapidly growing layer of metallic coating was formed on the substrate surface ($v_p \approx 900 \text{ m/sec}$, Fig. 5b).

Thus, the above results show that, using a supersonic gas jet with a stagnation temperature of ~ 300 K, it is possible to obtain coatings from most of the metals with particle size $d_p < 50 \ \mu$ m.

Figure 6 shows experimental values of the coefficient of deposition on the copper substrates for the aluminum, copper, and nickel powders, depending on the air temperature in the plenum chamber. For these materials, curves 4-6 in Fig. 4 show, respectively, the same data, but depending on the particle velocity calculated for the corresponding temperatures of air heating. Having compared these plots with the curves obtained when a mixture of air and helium at $T_0 = 300$ K was used as the accelerating gas, we concluded that the particle and substrate temperatures significantly affect the spraying process. Otherwise these two

families of curves would coincide. An increase in the air temperature in the plenum chamber increases not only the velocity, but also the temperature of the particles and the substrate. Therefore, the abrupt increase in the deposition coefficient is, apparently, explained by the increase of both the velocity of the deposited particles (which increases the pressure and the temperature in the contact site at the moment of impact) and the temperature of the deposited particles and the substrate (which leads to a change in their plasticity, a temperature rise in the particle-substrate contact, and, hence, a shift of the critical velocity v_{cr} to lower values).

Thus, the studies showed that using a supersonic (M = 2.0-3.0) air jet with moderate heating ($\Delta T \leq 400$ K), it is possible to obtain coatings from most of the metals and many alloys (Al, Cu, Ni, Zn, Pb, Sn, V, Co, Fe, Ti, bronze, brass, etc.) on various metal and dielectric substrates (in particular, glass, ceramics, etc.). Heating a helium jet and thus providing $v_p \ge 1200$ m/sec, we managed to obtain coatings of refractory metals (Nb, Mo, and W). The deposition coefficient of powders can reach 0.5-0.8, which is important in designing particular technological processes.

Discussion of the Results. We consider some specific features of the formation of gas-dynamic coatings within the framework of the approach widely used for analysis of gas-thermal coatings [1, 2, 7]. The rate of formation of chemical bonds in the particle-substrate contact is written as

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$$\frac{dN}{dt} = (N_0 - N)\nu \exp\left(-E_{\rm a}/kT_{\rm c}\right),\tag{1}$$

where T_c is the temperature in the contact zone, N_0 is the total number of atoms per unit surface of the substrate or particle surface in contact, ν is the frequency of eigenoscillations of atoms in the lattice (it is usually assumed that $\nu \approx 10^{13} \text{ sec}^{-1}$), and E_a is the activation energy of the interaction process.

Using Eq. (1), we can estimate the relative number of bonds formed by the contact reaction (assuming the mean values of E_a and T_c during the contact time t_c):

$$N/N_0 = 1 - \exp\left[-\nu t_c \exp\left(-E_a/kT_c\right)\right].$$
 (2)

It follows from (2) that, for $T_c \ge E_a/k \ln(\nu t_c)$, the relative number of bonds is $N/N_0 \ge 1 - 1/e \approx 0.63$, and it increases rapidly with increasing T_c . This number of bonds is usually considered sufficient for deposition.

The activation energy of the process E_a depends on the pressure in the contact zone: as the pressure increases, the activation energy decreases and approaches the theoretically calculated value as a limit. For example, the limiting value of the activation energy for aluminum is $0.5 \cdot 10^{-19}$ J, and it is reached when $p \ge 100$ MPa [8].

We estimate the mean dynamic pressure p_{mean} over the contact area using the momentum equation $t_c p_{\text{mean}} S_{\text{dyn}} = \rho_p(\pi d_p^3/6) v_p$, where S_{dyn} is the mean value of the area on which the dynamic pressure acts, ρ_p is the density of the particle material, and v_p is the velocity of the particle-substrate collision. We estimate the contact time as the time of equally decelerated motion of the back point of the particle from $v = v_p$ to v = 0, i.e., $t_c = 2\varepsilon_p d_p/v_p$. When an aluminum particle with characteristic velocity $v_p \approx 500$ m/sec hits the substrate, its deformation determined from microscopic observations is $\varepsilon_p \approx 0.5$. The particle mid-section area can be taken for these deformations as the area of action of the dynamic pressure: $S_{\text{dyn}} = \pi d_p^2/4$. Substituting all the data into the momentum equation, we obtain $p_{\text{mean}} \approx \rho_p v_p^2/3\varepsilon_p$. The substitution of $\rho_p = 2700 \text{ kg/m}^3$ and $v_p \approx 500 \text{ m/sec}$ yields $p_{\text{mean}} = 450 \text{ MPa}$ and, therefore, we can use the limiting value of the activation energy $E_a = 0.5 \cdot 10^{-19} \text{ J}$ in the estimates. Substituting $E_a = 0.5 \cdot 10^{-19} \text{ J}$, $k = 1.38 \cdot 10^{-23} \text{ J/deg}$, $\nu \approx 10^{13} \text{ sec}^{-1}$, and the above value of t_c into the relation $T_c \ge E_a/k \ln(\nu t_c)$, we obtain $T_c \ge 260 \text{ K}$ for $d_p = 50 \ \mu\text{m}$, $T_c \ge 300 \text{ K}$ for $d_p = 10 \ \mu\text{m}$, and $T_c \ge 365 \text{ K}$ for $d_p = 1 \ \mu\text{m}$.

Thus, it follows from these estimates that the temperature in the contact zone $T_c \ge 365$ K is sufficient for the formation of the necessary number of chemical bonds during the contact time for all particle sizes and for a pressure in the zone of contact between a single particle and the substrate which is typical of the gas-dynamic spraying. This temperature is provided by local heating with plastic deformation in the particlesubstrate contact zone. Assuming that all the kinetic energy of the particle transforms upon deformation



into the thermal energy equally distributed over the particle volume, one can determine the temperature rise owing to deformation as $\Delta T_c \approx v_p^2/2c_p$ (c_p is the heat capacity of the particle material). For example, for an aluminum particle, we have $\Delta T_c \approx 140$ K for $v_p \approx 500$ m/sec and $\Delta T_c \approx 560$ K for $v_p \approx 1000$ m/sec. Taking into account that the heat release depends on the degree of deformation, which is most intense in the contact zone, we can assume that $\Delta T_c \geq v_p^2/2c_p$.

The estimates show that the formation of a junction in the cold particle-cold substrate contact realized in gas-dynamic spraying can be quite well described within the framework of the concepts applied in gasthermal deposition analysis [1, 2, 7].

Finally, we determine the status of the CGDS method among the other methods of gas-thermal spraying. Figure 7 shows a chart of the characteristic ranges of particle temperatures and velocities for the method of cold gas-dynamic spraying in comparison with the known gas-thermal methods: 1) powder gas-flame technique; 2) wire gas-flame technique; 3) electric arc method; 4) plasma method; 5) detonation method; 6) high-speed gas-flame method; 7) gas-dynamic spraying. Clearly, the distinctive feature of the CGDS method is the possibility of coating formation at a jet temperature of 0-500°C, which is lower than the melting point of the particle material. Thus, there are no effects of high-temperature oxidation, evaporation, melting, crystallization, gas production, etc., which are typical of the gas-thermal methods. The advantages of the CGDS method are as follows:

• the possibility of formations of coatings with properties similar to the properties of the material of the initial particles;

• the possibility of obtaining composite coatings from mechanical mixtures of powders having different physicochemical properties;

• formation of coatings from powders with a particle size smaller than 1 μ m;

• no significant thermal action on the workpiece;

• higher safety of works owing to the absence of high-temperature jets and radiation, simplicity of technical realization;

• the absence of energy-stressed units, which ensures prolonged serviceability;

• the possibility of particle collection and the reusability of them in deposition.

Thus, the studies have shown that for coating formation the particles should have a temperature close to the melting point of the particle material, which is typical of the traditional gas-thermal methods [1, 2, 7]. Sometimes, it is reasonable to reduce the particle temperature and to increase their velocity simultaneously, which allows one to employ devices for deposition that are simpler from the engineering viewpoint, thus providing the implementation of the technological processes of material compacting and coating formation from substances (alloys) easily decomposed under the conditions of gas-thermal spraying.

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