JTTEE5 2:159-164

Stochastic Approach to the Modeling and Optimization of Thermal Spray Coating Formation

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Application of the stochastic process theory is proposed for the modeling and optimization of the thermal spray process (TSP). The advantages of the approach suggested are illustrated by estimation of the "integral" thermal conditions of TSP coating formation, as well as the formation characteristics of the first monolayer coating. Modeling results are in reasonable agreement with bond strength test data for the plasma-, arc-, and flame-spray processes as well as wear resistance data for arc-sprayed steel coatings.

1. Introduction

IMPROVEMENT of coating properties and increasing their realm of application are inseparably linked with the problem of spray optimization. For the thermal spray process (TSP), as for any complex multiparameter process, the standard optimization problem can be subdivided as follows: (1) to determine the primary governing parameters to be optimized, i.e., those parameter values that have the most impact on determining coating properties, and to determine a preliminary range of parameter variation; (2) to determine the optimal values and tolerances of these parameters, which can provide the optimum quality of a coating for a given application and material; and (3) to determine ways in which the process can be changed to provide further improvement in coating properties.

As a rule, it is relatively easy to obtain information on item 1 above in papers, manuals, etc. However, due to a great deal of specific data on thermal spray coatings, together with the numerous different coating applications, reviewing the literature for information described in item 2 raises major problems. The third step of the optimization process generally can be performed after analysis, or the results of step 2.

To solve the second part of the optimization process, an extensive experimental study of TSP would be prohibitively expensive due to the complexity and number of the process parameters. In addition, the primary physical characteristics of spray particles and their interaction with the substrate are described by various fields that are stochastic in nature. This fact creates additional problems for the experimental optimization of TSP. Therefore, constructing a model of the physical conditions should be a necessary step in a detailed study of the spraying process.

Traditional approaches to the modeling of TSP are based on the use of various types of single-particle models (SPM), i.e., on the analysis of interactions of one single spray particle with the projecting medium jet or with the substrate. However, the SPM, as Smith and Novak[1] state, "while leading to improved process knowledge and reinforcing intuitive results, never developed predictive or process control." The SPM are incomplete because they do not take into account the following important features of TSP.

First, the coating is formed by a vast number of particles. The concentration of the particles (Fig. 1), as well as the spatial distribution of their temperature and velocity, is nonuniform within any cross section of the carrier jet. Second, the substrate surface temperature in the region where the coating is being formed is also nonuniform.

Keywords: bond strength, modeling, optimization, stochastic approach, temperature conditions, wear resistance

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Nomenclature

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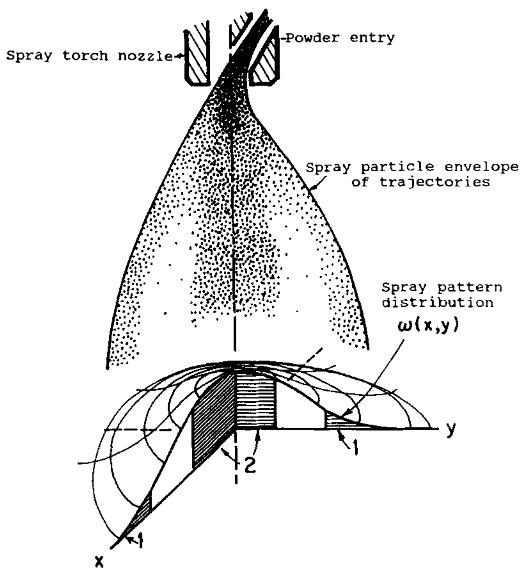

Fig. 1 Schematic of the thermal spray process and spray pattern distribution $\omega(x,y)$. 1, peripheral zone; 2, central zone.

Thus, particle properties, as well as conditions of their interaction with the substrate, differ considerably. Therefore, the local properties of the coating differ substantially from point to point on the sprayed surface. For example, a difference of 10 to 15% in the coating density from the central to the peripheral zones of a spray pattern (Fig. 1) can exist in the case of alumina spraying.[2]

One of the main requirements of TSP coatings is to ensure that the integral coating properties remain constant over the entire sprayed surface. To satisfy this requirement, spraying can be carried out so that the coating formation on any part of the surface takes place by deposition of many layers whose axes are displaced relative to each other. On a macroscopic scale, this procedure will homogenize the properties of the coating. However, on a microscopic scale, the coating consists of a complex mixture of particles that experience vastly different thermal histories and interactions with the substrate.

The conditions of formation and the properties of such a complex system can be described by a statistically based model.[3] At the same time, the fact that any microscopic part of the coating comprises a "particle mixture" indicates that mean effective values of the various parameters can be used to determine the dependence of integral properties of the coating on the parameters of the substrate and sprayed particles.

This article discusses the advantages of using the stochastic theory for TSP modeling and optimization. This approach allows one to calculate the pertinent mean values of the major stochastic fields that determine the physical parameters of the spray particles and their interaction with the substrate.

2. Statistical Description

Generally, to describe a system consisting of N-particles, it is necessary to have an M-dimensional distribution function ($M = Nm$, where m is the number of parameters that determines the state of one particle). It should be evident that it is almost impossible to obtain such a complex distribution function. At the same time, the probabilities of spray particle interaction and collisions in a plasma/gas jet are very small.[4] This fact allows one to apply an m-dimensional distribution function for the statistical description of the spray particle system.

The main parameters that determine the state of a particle at the instant that it collides with the substrate are velocity, temperature distribution, and mass of the particle. For TSP, especially in automatic spraying systems, primarily powders with narrow size distributions are used. In this case, the velocity and temperature of the particles depend chiefly on their position in the jet or in a spray pattern (at the instant of collision). Therefore, it is sufficient to use the spray pattern distribution, ω (Fig. 1), as an approximate distribution function of the particle system.

If the normalized distribution function is known, then it is possible to calculate the mean value $\langle\varphi\rangle$ of any parameter φ that describes the condition of the interaction between particles and substrate. For a Cartesian coordinate system, which is considered below, it is possible to write:

$$\langle\varphi\rangle = \iint_{\text{Surface}} \varphi(x,y)\omega(x,y)dxdy \tag{1}$$

In addition to this "integral" mean value $\langle\varphi\rangle$, it is sometimes important to know the conditions of formation for different monolayers in the coating, i.e., to take into account the stratified structure of the coating. In this case, one can regard subsequent "stacking" of the particles forming the coating during a single passage of a spray torch as the transition of various points of the spray surface from one state to another. For instance, if a particle arrives at the point with the Cartesian coordinates (x,y) at which k particles have already arrived and bonded, the point (x,y) is said to pass from the state k to the $(k+1)^{\text{th}}$ state.

The event consisting of the arrival of a particle at the point that has already stacked $k-1$ particles is a set of two events. The first event, with the probability density $p_{k-1}(x,y,t)$, is that $k-1$ particles have arrived at point (x,y). The second event, with the probability density $\omega(x,y,t)$, is the arrival of the k^{th} particle in the vicinity of point (x,y) at time t. Because these two events are statistically independent, the mean value $\langle\varphi^{(k)}\rangle$ of the formation of the k^{th} monolayer of the coating has the form:

$$\langle\varphi^{(k)}\rangle = \frac{1}{\zeta_N}\iint_{\text{Surface}} \varphi(x,y)\omega(x,y)p_{k-1}(x,y)dxdy \tag{2}$$

where ζ_N is the normalization factor.

In this article, the advantages of the approach are illustrated by estimation of the "integral" thermal conditions of TSP coating formation, as well as the formation characteristics of the first monolayer. The reasons for this choice are as follows. According to current views, the thermal conditions of TSP coating formation have a significant effect on residual stresses, hardness of the coating, etc. Also, failure of the TSP coatings takes place mainly at the boundary between the coating and the substrate surface or at the boundary between the layers formed during different passes of the spray torch. Therefore, the conditions of the first monolayer formation are of particular interest.

3. Integral Thermal Conditions

For the estimation of the integral mean substrate surface temperature $\langle T \rangle$, the surface temperature field description described previously has been used.[5-7] These surface temperature field calculations were based on the following approach. Primarily, the spraying surface is heated as a result of an extremely large number of separate short-term thermal cycles associated with the generation of heat by cooling and solidification of the particles. Generally, there is turbulent gas flow, and the usual particle size distribution of feedstock materials will give rise to a random-like quantity of enthalpy to each particle. Also, the coordinates and the time of impact against the substrate can be regarded as random quantities. Assuming to a first approximation that the particles are of the point type and that the process of generation of heat from the individual particle is almost instantaneous, then the equation for the random heat flow $q^{(r)}$ from n particles may be written as:

$$q^{(r)}(x,y,t) = \Sigma q_k \delta(x - x_k)\delta(y - y_k)\delta(t - t^{(k)}) \qquad [3]$$

where δ is Dirac's delta function;[8] and q_k, x_k, y_k, and $t^{(k)}$ are, respectively, the enthalpy, x and y coordinates, and the time corresponding to the k^{th} particle.

At the same time, the temperature field formed in heating the substrate with the gas or plasma jet can be unambiguously determined by means of an average surface distribution $q^{(d)}$ of the heat flow from this source. In contrast to the previous case, this source will be referred to as the determinate.

Experimental data[2,9,10] show that $q^{(d)}$, the particle temperature (T_p), and velocity (V_p) distributions as well as distribution of spray pattern are very well approximated by normal laws with standard deviations σ_q, σ_T, σ_V, and σ_P respectively. Therefore, in this article normal laws will be used for the approximation of these parameters. Particularly, for $q^{(d)}$, it will be assumed that:

$$q^{(d)}(x,y,t) = \eta_g Q_g g\left(x,x_0,\frac{\sigma_g^2}{2}\right) g\left(y,y_0,\frac{\sigma_g^2}{2}\right) \qquad [4]$$

Here

$$g(x,x_0,\sigma) = \exp\{-(x - x_0)^2/4\sigma\}/(2\sqrt{\pi\sigma}) \qquad [5]$$

where Q_g is the power of the heat source; η_g is the efficiency of the substrate surface heating with the jet; and $x_0(t)$ and $y_0(t)$ are the coordinates of the intersection point of the spraying surface and the axis of the jet.

To estimate integral thermal conditions under TSP, the case of spraying on a substrate with thickness H positioned in the region $0 \le z \le H$ has been examined. The heat sources are positioned in the region $z < 0$ (i.e., away from the substrate surface) and travel along the x axis with constant velocity W. It has been assumed that the surface temperature of the previously sprayed coating during its heating by the two-phase flow is almost identical to the temperature of the coating/substrate interface ($z = 0$), i.e., the distortions in the temperature field caused by the thermal resistance of the coating layer have been neglected. For the given assumptions, the heat conductivity equation and the initial boundary conditions on the sprayed surface in the stationary system of the coordinates may be written as:

$$\frac{\partial T}{\partial \tau} = \Delta T; 0 \le z \le H \qquad [6]$$

$$T|_{\tau=0} = 0; -\lambda\frac{\partial T}{\partial z}|_{z=0} = q^{(r)} + q^{(d)}$$

where $\tau = at$; $u = W/a$; in which a and λ are the coefficients of thermal diffusivity and heat conductivity.

On the rear side of the sheet ($z = H$), various cooling conditions and corresponding boundary conditions may exist. However, it is evident that in any practical case for the temperature $T(x,y,z,t)$ of any point of the substrate, the following relationship exists, $T_i \le T \le T_a$, where T_i and T_a are the temperatures of this point in isothermal and adiabatic conditions on the surface $z = H$, respectively. Therefore, in subsequent considerations, two modeling cases will be examined:

- The rear side of the sheet is thermally insulated; thus,

$$\frac{\partial T_a}{\partial z}|_{z=H} = 0 \qquad [7]$$

- During the entire process, the rear surface temperature is equal to the initial temperature, i.e.,

$$T_i|_{z=H} = 0 \qquad [8]$$

Using the solution of Eq 6 with boundary conditions 7 or 8 for $\langle T_a \rangle$ and $\langle T_i \rangle$ calculations, it is possible to write the following:

$$\langle T_{i,a} \rangle = A_g \langle T_{i,a}^o(W^o,H^o,S_1,t^o) \rangle + A_p \langle T_{i,a}^o(W^o,H^o,S_2,t^o) \rangle \qquad [9]$$

where

$$A_g = Q_g \eta_g/(\lambda\sigma_p \pi^{2/3}\sqrt{2});$$
$$A_p = G\langle q_p \rangle / (\lambda\sigma_p \pi^{2/3}\sqrt{2});$$
$$W^o = W\sigma_p/2a; H^o = \sqrt{2}H/\sigma_p;$$

and

$$t^o = \frac{2at}{\sigma_p^2}; S_1 = \sqrt{1 + S_{g,P}^2}; S_2 = \sqrt{1 + S_{PT,P}^2};$$
$$S_{g,P} = \sigma_g/\sigma_P; S_{PT,P} = \sigma_T/\sqrt{\sigma_P^2 + \sigma_T^2}$$

where G is deposition rate; $\langle q_p \rangle$ is mean enthalpy of particles; W^o is the nondimensional torch traverse velocity; H^o is the nondimensional substrate thickness; t^o is the nondimensional time; and $\langle T_a^o \rangle$ and $\langle T_i^o \rangle$ are nondimensional mean surface temperatures corresponding to the interface conditions in Eq 7 and 8.[2]

The typical results of calculations of $\langle T_i^o \rangle$ for the isothermal conditions on the surface $z = H$ presented in Fig. 2 show that, from the instant at which the heat sources start to operate and up to $\langle T^o \rangle \approx 0.8 \langle T_{\max}^o \rangle$, the temperature depends only on t^o and varies according to the following law:

$$\langle T^o \rangle = \sqrt{t^o}$$

After reaching the given level, the rate of temperature increase decreases. Starting from $\langle T^o \rangle \approx 0.9 \langle T_{\max}^o \rangle$, the rate of temperature variation is very low and $\langle T^o \rangle$ approaches asymptotically its limiting quasistationary value $\langle T_{\max}^o \rangle$, which primar-

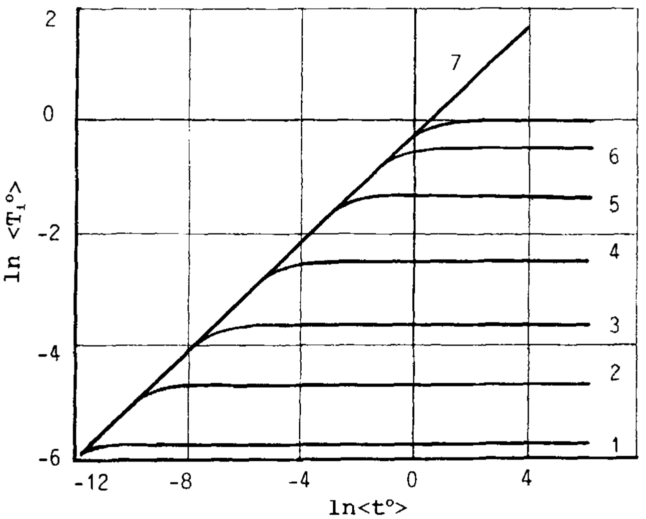

Fig. 2 Dependence of nondimensional mean temperature $\langle T_l^o \rangle$ on t^o. Curve 1: for $W^o = 185, 400$ for all H^o. Curve 2: for all W^o, $H^o = 0.87 \times 10^{-3}$. Curve 3: $W^o = 2870$, $H^o > 0.07$. Curve 4: $W^o < 45$, $H^o = 0.07$. Curve 5: $W^o < 5.6$, $H^o = 0.28$. Curve 6: $W^o = 0.07$, $H^o = 1.1$. The straight line (7) corresponds to $\langle T_l^o \rangle = \sqrt{t^o}$.

ily depends on W^o (nondimensional torch traverse velocity) and H^o (nondimensional substrate thickness).

It is necessary to note that, in some regions of W^o and H^o variations, it is possible to find very simple approximations for T^o_{max}. For example, for $W^o > 1.4$ and $(H^o)^2 W^o > 3.5$ (it was shown in Ref 2 that TSP parameters often correspond to this case), then $\langle T^o_{max} \rangle \approx 1.1/\sqrt{W^o}$; i.e., this parameter depends only on W and not on the sheet thickness and its rear cooling conditions. Such simple approximations are very convenient for temperature estimations and subsequent spraying parameter corrections.

4. Conditions of the First Monolayer Formation

Conditions of the first monolayer formations are primarily characterized by the mean particle velocity $\langle V_P^{(1)} \rangle$, temperature $\langle T_P^{(1)} \rangle$, as well as mean substrate surface temperature $\langle T^{(1)} \rangle$, to which the particles of the first monolayer are subjected.

The results of the analysis show that within the approximation mentioned $\langle T_P^{(1)} \rangle$, $\langle V_P^{(1)} \rangle$, and $\langle T^{(1)} \rangle$ can be expressed as:

$$\langle T_p^{(1)} \rangle = T_o \frac{\psi(B, S_{PT})}{(1 + S_{PT}^2)} \quad [10]$$

$$\langle V_p^{(1)} \rangle = V_o \frac{\psi(B, S_{PV})}{(1 + S_{PV}^2)} \quad [11]$$

$$\langle T^{(1)} \rangle = A_g f(B, S_{g,P}) + A_p f(B, S_{PT,P}) \quad [12]$$

where T_o and V_o are the particle temperature and velocity in the axis of the jet; f and ψ are nondimensional functions;[2] and B, S_{PT}, S_{PV}, $S_{PT,P}$, and K are nondimensional parameters defined as follows:

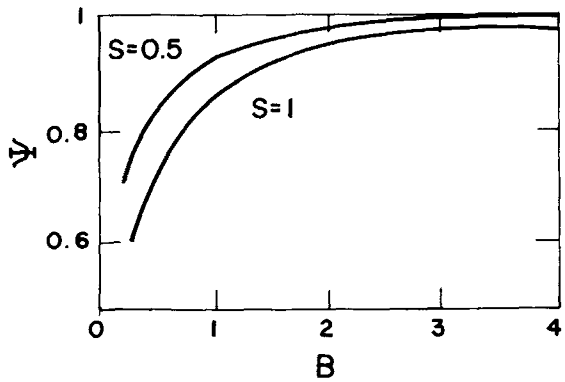

Fig. 3 Dependence of particle velocity with respect to the B parameter for the first monolayer of a deposit. The term B is a nondimensional parameter that is associated with the deformation of particles on impact against the substrate and thermal spray variables such as torch velocity, density of feedstock, spray pattern distribution, and coating deposition rate.

$$B = \frac{KW\rho_p d\sigma_p}{G}; \; S_{PT} = \frac{\sigma_p}{\sigma_T}; \; S_{PV} = \frac{\sigma_P}{\sigma_V};$$

$$S_{PT,P} = 1/\sqrt{1 + S_{PT}^2}; \; K = 5h/d$$

where d is the mean diameter of the spray particles; ρ_p is the density of the spraying material; and h is the mean particle thickness after solidification.

The results of calculations of $\psi(B,S)$ (Fig. 3) show that $\langle T_P^{(1)} \rangle$ and $\langle V_P^{(1)} \rangle$ are monotonically increasing functions of the B parameter. The growth rate of ψ depends on S; as S decreases, then the growth rate of ψ increases. In addition, it follows from the function behavior that for $B > 2.5$, the values of $\langle T_P^{(1)} \rangle$ and $\langle V_P^{(1)} \rangle$ flatten out at their limit values:

$$\langle T_p^{(1)} \rangle = T_o/(1 + S_{PT}^2) \text{ and } \langle V_p^{(1)} \rangle = V_o/(1 + S_{PV}^2)$$

The typical results of calculations of $f(B,S)$ presented in Fig. 4 demonstrate that $\langle T^{(1)} \rangle$ increases with B up to some critical value B^*. The mean temperature $\langle T^{(1)} \rangle$ reaches a maximum ($f = f_{max}$) at $B = B^*$ and then slowly decreases with a further increase in B. The parameter B^* depends on S in the following manner: the greater S, the less B^*.

It should be emphasized that the substrate surface temperature at $B = B^*$ is usually less than 70 to 100 °C, which is not high enough to activate a significant increase in its surface oxidation. Therefore, such behavior of $\langle T_P^{(1)} \rangle$, $\langle V_P^{(1)} \rangle$, and $\langle T^{(1)} \rangle$ suggests that the maximum values of the coating bond strength are in the range of $B^* < B_{opt} < 2.5$, i.e., in the range where the temperature and velocity of the particles, as well as the substrate surface temperature, have maximum values under the first coating monolayer formation.

5. Experimental Results and Discussion

The standard direct tensile pull-off adherence test[11] was used to determine the adhesion of coatings sprayed on a plane surface of 25-mm diameter steel cylindrical specimens. The surfaces to be coated were grit blasted. Standard plasma, arc, and

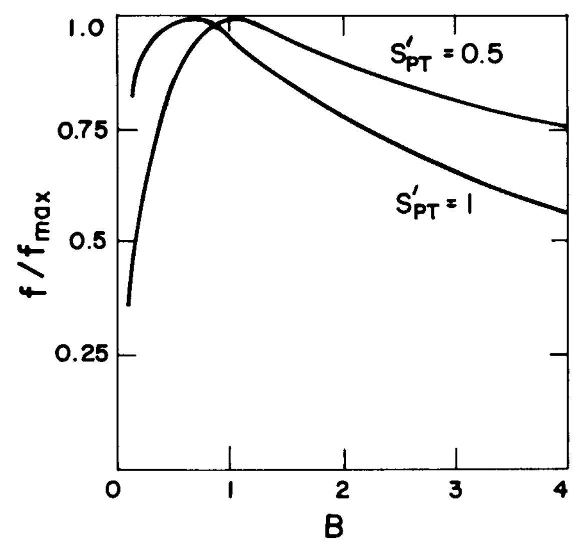

Fig. 4 Analysis of normalized (nondimensional) mean substrate temperature (f/f_{max}) corresponding to the first monolayer of the deposit. The term B is a nondimensional parameter that is associated with the deformation of particles on impact against the substrate and thermal spray variables such as torch velocity, density of feedstock, spray pattern distribution, and coating deposition rate.

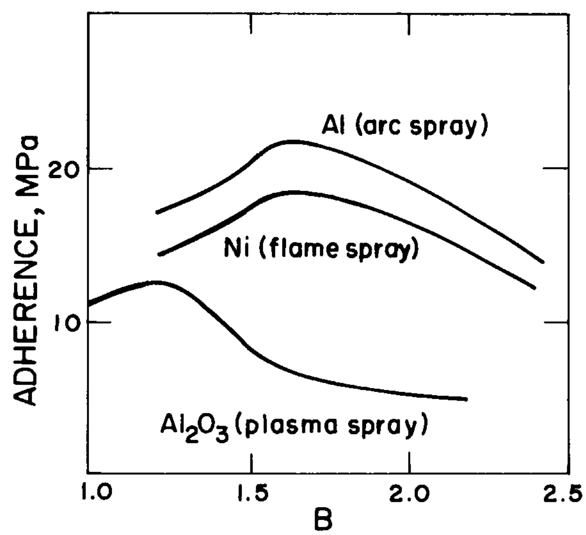

Fig. 5 Dependence of coating bond strength on the nondimensional parameter B. The term B is a nondimensional parameter that is associated with the deformation of particles on impact against the substrate and thermal spray variables such as torch velocity, density of feedstock, spray pattern distribution, and coating deposition rate.

flame equipment as well as automated spraying tables were used to spray coatings of 0.25 mm thickness with a precision of ±0.02 mm. The coatings investigated were aluminum and various types of steel (arc spray), alumina (plasma spray), and nickel alloy (flame spray). It should be noted that only the correlation between the parameter B and the coating bond strength was studied. Conditions of grit blasting, as well as powder size and properties of the gas/plasma jet, were fixed with their values being, perhaps, not optimal.

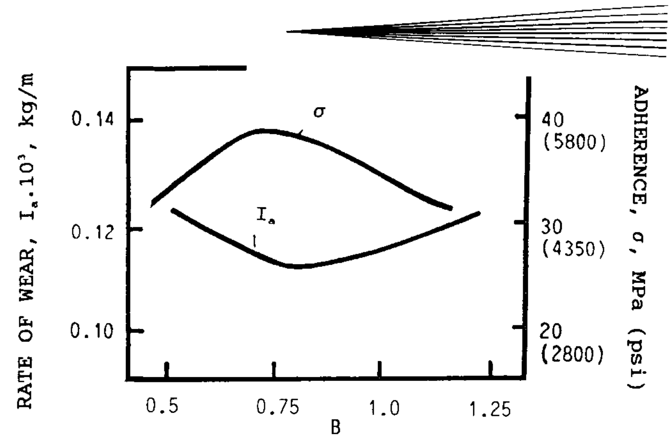

Fig. 6 Rate of wear and adherence versus B for the case of arc sprayed steel coatings.

The results of the bond strength measurements shown in Fig. 5 demonstrate that the coating bond strength reaches a maximum in the same theoretically predicted range of B, $B^* < B_{opt} < 2.5$. This phenomenon can be explained as follows.

At $B < B^*$ (low scanning velocity), the first monolayer is almost fully formed by the peripheral particles, which have comparatively low temperature and velocity. Moreover, as follows from the Fig. 4, when $B < B^*$, these particles collide with the cold substrate surface. As a result, the coating has low bond strength. Then, as the scanning velocity increases, the contribution of the peripheral particles begins to decrease, and increasingly more vacant spots on the substrate surface become available for the high-energy and high-temperature particles of the central jet zone to form the first monolayer. Over the range of $B^* < B_{opt} < 2.5$, the ratio between the central and peripheral particles in the first monolayer reaches its maximum. A further increase in B only leads to a decrease in T and an unjustified increase in the number of torch scans needed to ensure the same thickness of the coating.

Temperature conditions of TSP are more important for thermally sensitive coatings, i.e., for the case when coating quality strongly depends on the temperature conditions in the spray zone. A typical example of a thermally sensitive material is carbon steel. Therefore, arc spraying of carbon steel wire (0.95 to 1.05% C, 1.3 to 1.5% Cr, rem Fe) was used to verify the results of the integral mean temperature estimations.

Calculations of $\langle T \rangle$ show that, for $B = B^*$, $\sigma_P > 1.1 \times 10^{-2}\, m$ and $G < 3 \times 10^{-3}$ kg/s, the level of the integral temperature will be $\langle T \rangle \approx 50$ to 80 °C (during a single pass of the spray torch). Due to this low surface temperature, it is possible to expect at $B = B^*$ not only high coating strength, but also a high level of coating hardness. The effect of this superposition should be a high level of coating wear resistance, as demonstrated by bond strength and wear (conditions of the dry friction with abrasive) measurements shown in Fig. 6.

Of course, the spraying surface temperature estimates are approximate, and the optimal range of B obtained is wide enough to eliminate the necessity of experimental optimization. However, use of higher approximations for the actual distribution of the particle parameters can allow one, with the assistance of software, to narrow the range of B and to therefore predict the optimal relations between TSP parameters more accurately.

6. Conclusions

The approach proposed is a step to improve TSP modeling results. Even the simplest approximations for the main particle parameters have yielded reasonable agreement with bond strength data. The optimizing relationship between scanning velocity and other process parameters can be helpful for determining spray optimization, with further improvement in coating properties. Information about spray pattern distribution is a necessary supplement to any measurement of temperature, velocity, or other particle parameters that are needed to ensure the high predictability of TSP modeling.

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