Prediction of the Adhesion Strength of Coating in Plasma Spray Deposition

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ABSTRACT

The goal of this work is to validate the existing plasma spray mathematical models, using a calculation method and the comparison with experimental data, in order to determine their validity. A preliminary evaluation of the adhesion based on the velocity and temperature of the particles is useful to be calculated by using the mathematical model. Given the thermal-physical properties and chemical composition of a Fe-based amorphous X-5 powder, a modified model was suggested. For comparison, a series of experiments using plasma spraying of the X-5 powder were conducted. The significance of the current study consists of the model validation by using the data of the plasma spraying of the Fe-based amorphous material as a potential substitution for saving production costs by using ordinary air as the plasma generation gas. The findings show the discrepancy between the models and the experimental results. The prediction of adhesion using the mathematical models does not cover essential parameters such as the enthalpy of the particle stream. It is necessary to improve the mathematical models, including the modified one, based on the experiment results, with different pairs of particles and substrate materials. The proposed formula is applicable during the preliminary design of the spray process and the development of a new torch construction.

Keywords-adhesion; coating; particle velocity; particle temperature; validity; prediction

I. INTRODUCTION

Plasma spray deposition is an additive manufacturing technology whose application range extends beyond the traditional thermal spraying approach, including the plasma spraying approach, which relies heavily on melting. The plasma energy encourages the nitriding process to modify the structure and mechanical properties of the surface, but the temperature and chemical process prevail [1]. Many engineers and researchers have worked to produce thermal spray technology without melting, and the velocity of the particles is the primary driver in creating the deposition. Cold Spraying (CS) is the term for this procedure. Particles stick to and deposit onto a substrate in CS without melting, allowing functional coatings to be deposited. In recent years, technologies that use particle kinetic energy in addition to thermal energy, such as High-Velocity Oxy-Fuel (HVOF) flame spraying and thermal spraying, have been developed. The most common example of these new approaches is the CS method. The natural oxide film on the surface is damaged by repeated particle impacts during the activation phase, known as incubation, and an increase in chemical activity due to the creation of the nascent surface is critical to adhesion. Due to the quick impact of the solid particles, it was inferred that the contribution of atomic diffusion to adhesion is low. Plastic deformation occurs when the substrate material and particles undergo explosive bonding upon particle contact, undulating in a wavy pattern, and layers are created by mixing in tiny interfacial areas [2]. During this collision, however, severe plastic deformation caused by particle impact might dramatically affect the bonding process, causing shear instability. The natural oxide films on the particle and substrate are removed, and the surface is activated, resulting in the formation of a strong sticky area. The particle and substrate constituent parts are in intimate contact, as shown in TEM images, in the absence of inclusions such as the oxide at the adhering interface. As a result, surface activation is a critical element in particle adhesion mechanisms, and the CS technique's massive plastic deformation following impact significantly adds to this surface activation. Figure 1 depicts a method in which plastic deformation is triggered by the kinetic energy released by the collision of CS material particles. A material jet in the outer peripheral direction caused by thermal shear instability breaks and eliminates the particles and oxide coating on the substrate surface. A new chemically active surface is revealed, and the new surfaces are strongly connected to each another.

In recent publications, some mathematical models describing the plastic deformation have been introduced, including the thermal spraying when the heating particles deposit on the substrate. However, the main disadvantage of these models is the lack of technological parameters, which limits their application in the prediction of adhesion strength in coatings as a significant criterion for deposition evaluation. In this context, a new modified mathematical model dealing with the sufficient prediction of adhesion strength must be introduced and validated. In the event of a disagreement, the empirical formula should be considered the most important recommendation.

In this paper, the digital method for the calculation of the predicted adhesion by the digital own program PROGX100 is described and the amorphous powder X-5 is proposed for the plasma spraying, while in the experimental part the relationships between enthalpy and air flow rate and between adhesion, power of plasma, and velocity of particles were specified, while some significant recommendations and the direction of future studies are presented.

II. METHODOLOGY

Significant plastic deformation was found at the interface edge due to the presence or absence of the remaining oxide film, and fracture and strong bonding were observed due to the ease with which the oxide film was removed. On the other hand, the quantity of plastic deformation in the area of the particle center was tiny hence the oxide film persisted even after the impact, generating a very weak bond. According to [3], in addition to substrate hardness, particle velocity and spray angle can influence deformation behavior and the ultimate state of the oxide film. The equation in [1] justifying the role of the critical velocity (Vcr) as the condition governing the feasibility of bonding was impacted by a rise in the local temperature beyond the melting point.

$$V_{cr} = 667 - 14\times\rho_p + 0.08\times T_m + 0.1\times\sigma_u + 0.4\times T_{Pi} \quad (1)$$

where ρ_p is the density of the particle, T_m is the melting point of the particle, σ_u is the tensile strength of the particle, and T_{Pi} is the initial temperature of the particle.

Due to adiabatic deformation, when particles hit at high speeds, the particle temperature quickly rises to the melting point. During the collision, much of the kinetic energy of the particle will be transformed into heat, encouraging plastic deformation. Softening is expected to produce particles that constitute the material jets. The Johnson-Cook (JC) model [4] is widely used in manufacturing to simulate high-speed deformation and can be used for high-velocity particle spraying [5, 6]:

$$\sigma = \left[A + B\times\varepsilon_p^n \right]\times\left[1 + C \times ln \times \frac{\dot{\varepsilon}_p}{\dot{\varepsilon}_0}\right]\times \left[1 - \left(\frac{T - T_{ref}}{T_m - T_{ref}}\right)^m \right] \quad (2)$$

where σ is the flow stress, A, B, C, m, and n are the material constants, ε_p^n is the plastic strain, $\dot{\varepsilon}_p$ and $\dot{\varepsilon}_0$ are the actual strain and the reference strain rates, respectively, and T, T_m, and T_{ref} are the actual, melting, and reference temperatures, respectively. The restriction of models (1) and (2) is the practical reference because, in the thermal spray, the activation energy and temperature of the contacting surface at the time of the impact must prevail. In this regard, the Kudinov V.V. model should be examined since it simplifies the computation of adhesion in terms of its prediction for designing the thermal process [7]:

$$\frac{d\,N(t)}{dt} = [N_o - N(t)]\times\nu\times\exp\left[-\frac{E_a}{k\times T_k}\right]\times\exp\left[\frac{S}{k}\right] \quad (3)$$

where E_a is the activation energy in this case, ν is the frequency of the atoms own oscillation, T_k is the contact's absolute temperature, N_0 is the number of atoms on the surface of the substrate or particle that make up the mutual physical contact, $N(t)$ denotes the number of atoms energized during the activation time t, S denotes the activation entropy in the chemical reaction zone, and k is the Boltzmann constant. Since the predominant metal has a BCC or FCC crystal structure, (3) will be transferred to the new type:

$$\frac{d\,N(t)}{dt} = [N_o - N(t)]\times\nu\times\exp\left[-\frac{E_a}{k\times T_k}\right] \quad (4)$$

The rate of the chemical interaction on the phase boundary is defined by the relative adhesion of the particle with the substrate:

$$\frac{\sigma\,(t)}{\sigma_{max}} = \frac{N(t)}{N_o} \quad (5)$$

where $\sigma\,(t)$ is the adhesion resulting for duration t, σ_{max} is the maximum adhesion obtaining for the entire cycle of the spraying. The influence of the particle velocity on the adhesion can be calculated according to [8]: $\sigma = V^2\times\rho$, where V is the particle velocity and ρ is the density of the particle material. By the integration, (4) will be changed as follows:

$$\frac{\sigma\,(t)}{\sigma_{max}} = 1 - \exp\left[-\frac{\nu\times t}{\exp\frac{E_a}{k\times T_k}}\right] \quad (6)$$

In (6), it is accepted that [7]:

$$E_a \approx k\times T_k\times(\ln t_0 + 30), \qquad t_0 = \left(\frac{h}{2\alpha}\right)^2\times\frac{1}{a_1} \quad (7)$$

For most of the materials, $\nu = 10^{13}\times c^{-1}$; h = 0.1×$d$, where d is the diameter of the particle and $E_a \approx 1.35 - 1.65$ eV.

Equation (7) describes the change in adhesion depending on the activated temperature during the collision of the particle with the substrate. On the other side, the quantity T_k can be calculated with respect to the known equation [7]:

$$T_k(t) = T_0 + T_k^0;\ \ T_k^0 = \frac{(T_m - T_0)\times K\varepsilon}{K\varepsilon + F(\alpha)} \quad (8)$$

where h is the height of the solidified particle in the collision on the substrate, t_0 is the time during which the particle is solidified and the constant contact temperature T_k is activated, T_0 is the initial temperature of the substrate, and a_1 is the coefficient of the thermal conductivity of the particle. $\alpha = f(K\varepsilon, K_L)$ and α varies in 0 - 1 depending on $K\varepsilon$ and K_L.

$$K\varepsilon = \frac{\lambda_1}{\lambda_2}\times\sqrt{\frac{a_2}{a_1}};\ K_L = \frac{c\times T_m}{1.77\times L} \quad (9)$$

where $K\varepsilon$ is the criterion of the thermal activity of the particle in relation to the substrate, K_L is the criterion defining the latent heat of fusion of particle material, λ_1 and λ_2 are the coefficients of the thermal conductivity of the particle and the substrate, respectively, a_1 and a_2 are the coefficients of the thermal diffusivity of the particle and substrate, L is the latent heat of the fusion, c is the heat capacity of the particle, T_m is the melting point of the particle material, $F(\alpha)$ is the integral of the probability, and $\alpha = f(K\varepsilon, K_L)$, the root of the function.

$$K\varepsilon + F(\alpha) = K_L \times \frac{e^{-\alpha^2}}{\alpha} \quad (10)$$

α can be defined by using the φ-α diagram [7], where α is the abscissa of the intersection between the two curves [7]:

$$\varphi = K\varepsilon + F(\alpha) = \varphi(K\varepsilon;\ \alpha) \quad (11)$$

$$\psi = K_L \times \frac{e^{-\alpha^2}}{\alpha} = \psi\ (K_L;\ \alpha) \quad (12)$$

The temperature during the solidification of the particle is defined by [7], with T_I being the temperature of the particle:

$$T_k = \frac{T_1 \times K\varepsilon}{K\varepsilon + F(\alpha)} \quad (13)$$

Given the properties of the particle and the substrate materials, the prediction of the change in adhesion in thermal spraying can be calculated using (6)-(9). The second method is to use (10) and (11) to find the root of the contacting temperature given $K\varepsilon$ and K_L. Since the adhesion depends not only on the heating effect, but also on the plastic deformation during the collision of the melting particle on the substrate, it is useful to consider a modified model for the prediction of changes in the adhesion:

$$\frac{\sigma(t)}{\sigma_{max}} = 1 - \exp\left[-\frac{\nu \times t}{\exp\frac{3 \times (\xi - \chi \times \frac{\sigma}{E})}{q \times \alpha \times T_k}}\right] \quad (14)$$

where ξ is the strain in which the atomic bond in unstable, $\chi = \Sigma/\sigma$ is the coefficient of the local overload, Σ and σ are the local and average tensile strengths. In case of the collision of the melting particle on the substrate: $\chi = 1$; $q = \alpha_s/\alpha$, where α_s and α are the coefficients of thermal extension on the surface and in the volume of the stiff body.

The theoretical value of the predicted adhesion is calculated using (13) and (14) by the digital program PROGX100, given the thermal-physical properties of the materials as follows: The substrate is mild carbon steel, and the particle material is Fe-based amorphous powder X-5 [9]. The chemical composition of powder X-5 is given in Table I.

TABLE I. CHEMICAL COMPOSITION OF X-5 (%,WT)

C	Cr	B	Mo	Ni	Mn	Si	Nb	V	W	Fe
0.73	5.0	0.25	4.20	-	1.25	0.84	0.54	1.20	-	remain

III. EXPERIMENTAL PART

The general scheme of the plasma spraying system can be seen in [10]. G1 is the source of power, G2 is the plasma torch, R1 and R2 are the rotameters, V1 and V2 are the valves, N1, N2, N3, N4, and N5 are the nipples, T1 is the thermometer, and T2 is the throttle. The power source is a direct current source with a steep volt-ampere slope, an idle voltage of 300V, and a voltage adjustment limit of 50 to 600V. The plasma arc is conducted according to a two-step scheme. Water serves as the coolant, with inlet and outlet via valve V1 and rotameter R1. The thermometer T1 is used to measure the temperature and provide the data needed to calculate the enthalpy of the plasma jet. The accuracy degree of this rotameter is 2.5%. The water flow pressure at the inlet is 0.4-0.6MPa. The primary gas and secondary gas are fed through the V2 valve. The flow rate of gas is determined by the rotameter R2. The throttle T2 is used to smooth the current pulsation. The plasma spraying of powder X-5 experiment was carried out under the following conditions: Atmospheric plasma spraying was utilized (SG-100 TAFA-Praxair, USA). The primary gas was air, and the carrier gas was nitrogen. The chemical composition of Fe-based powder was analyzed by energy dispersive spectroscopy with the SM-6510LV. The experimental data are shown in Table II. To measure the velocity of spraying particles, the special high-speed camera Shimadzu HPV-1 was used [11]. It is useful to mention that the velocity of the particle stream was measured instead of that of separate particles. The measurement was conducted based on the principles of photogrammetry [12].

The average mass temperature of a plasma jet is evaluated indirectly by enthalpy. Because deposition is a continuous process of buildup from the particle stream, it is preferable to compare the average mass temperature to the temperature of the individual particle. The plasma stream containing the particles is a non-equilibrium system with temperature fluctuations over spatial and temporal scales [13]. The enthalpy is defined as the effective power of the plasma jet divided by the consumption of the primary gas [14]. The adhesion strength of the coatings on the steel substrate was determined by the Universal Compression Testing Machine (Model HT-2101A-300, Taiwan) according to the ASTM C633 standard. Experiments were conducted with a measurement error of about 2.5%. The empirical equation was derived using the least squares method. On one side, the theoretical calculation using the mathematical models derived with the assumption of separate particles during the collision on the substrate. But on the other side, the experiment was conducted in real conditions when the plasma stream included numerous particles with their own velocity and temperature. The significant value of the validation underlies this work, helping reduce the existing gap between the mathematical models and the experiments. The context stimulates the new modified model as it approaches the real condition and applicable empirical equations for the prediction of the adhesion of the coating.

A. *Case Study 1: Plasma Spray of Amorphous Powder X-5*

The influence of the plasma power and the flow rate of the air on the enthalpy will be investigated in this case. The interval of variation for the flow rate of the air is 0.5g/s in the range of 0.5–3.0g/s. Particle size ranges between 40 and 100μm. The spray distance is 120mm. Increased power of the plasma stream is expected when the flow rate of the air increases, resulting in the elevated enthalpy of plasma stream with particles (Figure 1) [9]. In Figure 1, x, ●, and ■ denote current of 120, 180, and 220A, respectively.

B. *Case Study 2: Investigation on Adhesion*

The material used was again X-5 powder, with particle size ranging in 40–100μm, with 9mm nozzle diameter. The current varied as: 120, 150, 180, 220, and 240A. The flow rate varied between 0.46 and 3.17g/s. The results of the measurement of adhesion bond and velocity are shown in Figure 2.

C. Case Study 3: Relation between Particle Velocity and Air Flow Rate

This series of experiments occurs when the gas flow rate and the power of the plasma stream (via the current) are changing simultaneously. The data from the measurement of the particle velocity will be used for the evaluation of the adhesion of the coating. The experiment parameters were: X-5 material, size of particles: 40–100μm, nozzle diameter: 9mm (Figure 3).

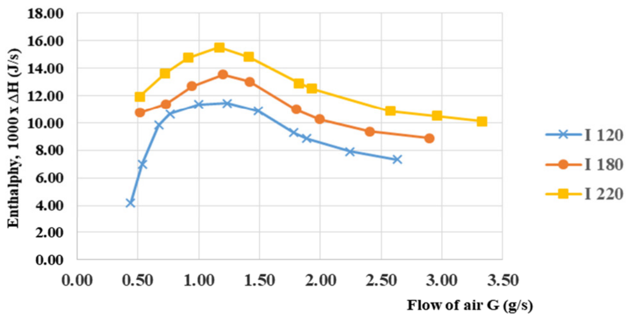

Fig. 1. Relation between enthalpy, plasma power, and air flow rate.

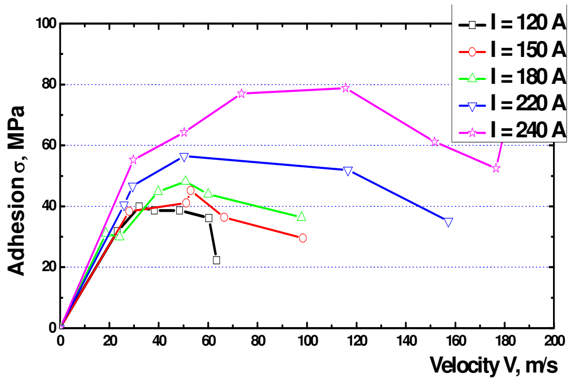

Fig. 2. Relation between adhesion, plasma power, and particle velocity.

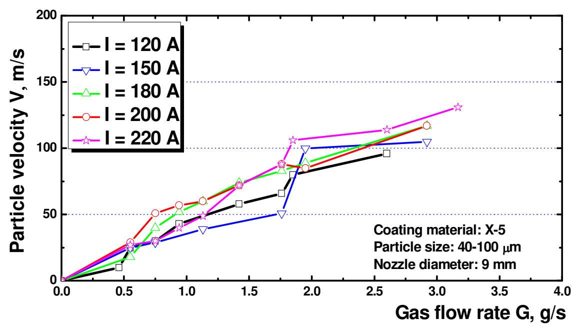

Fig. 3. Relation between particle velocity, plasma power, and air flow rate.

IV. RESULTS AND DISCUSSION

The theoretical result calculation according to (13) and (14) gave the values shown in Table II, given: $q = 5$, $\xi = 0.2$, $\chi = 1$, $\alpha = 0.33\times10^{-4}$°K^{-1}, $T_0 = 300$°K (without the preheating) for X-5 powder.

TABLE II. CALCULATION OF RELATIVE ADHESION

No	T_k, °K	$V_{particle}$, m/s	*d*, μm	$\sigma(t) / \sigma_{max}$, %
1	960	5	70	100
2	960	10	70	100
3	960	15	70	100
4	960	20	70	100
5	960	25	70	100
6	960	30	70	100
7	960	35	70	100
8	960	40	70	100
9	960	45	70	100
10	960	50	70	100
11	960	75	70	100
12	960	100	70	100
13	960	125	70	100
14	960	150	70	100
15	960	175	70	100
16	960	200	70	100

From Table II, the relative adhesion is monovalent (100%) when the particle velocity is changing, which is inconsistent over the course of the experiment in Case Study 2. When the particle's velocity is low, its kinetic energy is also low, resulting in low plastic deformation and a significantly lower level of dislocation in the structure. This effect probably caused the reduction of the adhesion between the coating and the substrate. This could be taken into account by the coefficient in (14). In Case Study 1 (see Figure 1), the increase in the flow rate had a positive influence on the particle velocity and its temperature (indirectly via the enthalpy of particle flow). However, there is a shroud surrounding this increase, in which the enthalpy of the particle decreases due to the particle's short in-flight time. To create a high density and a good adhesive bond in the coating, one should increase both the velocity and the enthalpy of the particle. However, it is difficult to assess their partial contribution theoretically. As the velocity is increasing, there is a small difference in adhesion bonds for small velocities (less than 40m/s), but in the intensive case, the adhesion bond is approaching the value of 80MPa, which exceeds the maximum limit reported in recent publications [15]. When changing adhesion bonds, the maximum number of them (the pick) tends to shift toward higher velocity as plasma power increases. This means that the adhesion bond depends on both velocity and enthalpy (plasma power). Except for very low air flow rates, the tendency of the wear resistance to change with the flow rate of the air almost coincides with the change in the adhesion bond. This can be understood, due to the fact that the low velocity impacts not only the good coherence bond, but also the poor adhesion bond. Based on the experimental results in Figures 1-3, an empirical formula demonstrating the relationship between adhesion bond, velocity, enthalpy, and current was proposed in [16]:

$$\sigma = \frac{1}{V^{x_1} \bullet \Delta H^{x_2} \bullet I^{x_3} \bullet x_4 + V^{x_5} \bullet \Delta H^{x_6} \bullet I^{x_7} \bullet x_8} \quad (15)$$

where: x_1 = -1.454, x_2 = 0.926, x_3= -0.398, x_4 = 0.003, x_5 = 0.836, x_6 = - 1.182, x_7 = - 0.9, and x_8 = 3598. V denotes the velocity [m/s], ΔH [J/g] the enthalpy, and I the adhesion bond [MPa].

V. CONCLUSIONS

In this work, the analysis of some mathematical models related to the plastic deformation applicable in the case of plasma spray deposition revealed their limitations for the prediction of the adhesion strength of the coating due to the lack of specific technological parameters such as the power of plasma, the velocity, and the enthalpy of the plasma stream, including particles. The newly modified model takes into account some specific physical and mechanical properties of the material and approaches more the real conditions of the collision of the heating particle onto the substrate. However, the discrepancy with the experiment results requires a significant improvement. Meanwhile, the plasma sprayring of Fe-based amorphous powder using ordinary air as a plasma generation gas once again proved the importance of the velocity and enthalpy of the particle stream for the strengthening of the adhesion since they positively encourage the plastic deformation according to the Johnson-Cook model. It is useful to note that as plasma power increases, the maximum values of adhesion shift to higher powers that can reach 80MPa when the power of plasma is approaching 80kW. To the best of the author's knowledge, the empirical formula (15) showing the relative correlation between qualitative parameters of the coating and spray process parameters, including the current, flow rate, plasma power, enthalpy of the torch, and velocity of the particles, had not been published before. This formula can be applied to the preliminary design of not only the plasma gun construction but also the technological process of the deposition. It is useful to continue the research in the future to cover the gap between the mathematical models and the empirical equations in the applicable prediction of the adhesion of the plasma-sprayed coatings. It is necessary to include the technological parameters of the spraying process into the model instead of the typical physical and mechanical properties of the given materials.

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