Optimizing of hardness, microstructure and adhesive strength of plasma-sprayed FeSiAl coatings using orthogonal analysis

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Abstract. The traditional high-efficiency absorber FeSiAl has gradually achieved the attention at high temperature. The paper focuses mainly on the preparation and characterization of plasma-sprayed FeSiAl coatings. The plasma-sprayed FeSiAl coatings were prepared through the Praxair 100HE high enthalpy plasma spray gun on stainless steel 316 substrate by orthogonal experiment. The cross-section and properties of sprayed coatings were analyzed. The results reveal that the best parameters of sprayed coatings can be represented by C:Bi:A:D = 30 SCFH, 140 mm, 50 KW, 3 PRM. The optimal parameters can be expressed as C:D:B:Fe:Si:Al for adhesive strength, i.e., 50 SCFH, 2 PRM, 200 mm, 50KW. The cross-section of sprayed coatings is composed of fattening FeSiAl particles (pale gray) and oxidation phase of FeSiAl (dark). The percentage of disorder structure reduces in FeSiAl coatings.

1. Introduction

An electromagnetic shielding and microwave absorbing materials have been urgently needed to reduce electromagnetic radiation and radar reflection in some extreme environment, like high temperature [1]. FeSiAl plays a vital role among high temperature absorbing coating due to high permeability and a saturation magnetization in recent years [2-3]. As of now, the focus of attention is on the preparation process of excellent absorbing coatings. The preparation technics for absorbing coatings includes mainly chemical vapor deposition (CVD) [4], sol-gel method [5], plasma spraying method [2] and so on. Plasma spraying process gradually gains the public’ favor because of numerous spraying materials and high deposition efficiency. However, seldom systematic research concerning for FeSiAl coating is reported using the plasma spraying process.

The paper explored the influence the parameter of plasma spraying on hardness and adhesive strength of plasma-sprayed FeSiAl coatings using orthogonal analysis. Meanwhile, the orthogonal analysis was employed to calculate each control factors of spraying parameters on hardness and adhesive Strength of FeSiAl coatings. The similar investigation conclusions can be considered as guidance in industrial production for a mixture of FeSiAl and any other materials (for example, ceramic and alloy).
2. Experimental procedure

FeSiAl powder (9.6 wt. % Si, 5.4wt. % Al, and balance Fe) was used as raw material with an average particle size of 45 μm. Stainless steel 316 was regarded as the base material.

2.1. Preparation of coatings

The Praxair 100HE high enthalpy plasma spray gun of Progressive Technologies Inc was applied to produce FeSiAl coatings. The moving speed and step distance of the plasma gun was 150mm/s and 3mm respectively. Argon was used as the plasma carrier gas. The substrates were sandblasted with corundum sand before plasma-sprayed. FeSiAl coatings were deposited on Stainless steel 316 with respective serial numbers and different processing parameters in an L9 (3^4) orthogonal experiment. The as-sprayed coatings were controlled with a thickness of 0.15-0.30 mm.

2.2. Microstructure and properties

The size distribution of the FeSiAl powder was measured using a Malvern MS3000. SEM images were collected on a JEOL SEM 7600F electron microscope, with the accelerating voltage of 15 kV. Microhardness of these samples was obtained by using Digital Microhardness tester. Bonding strength was measured according to the national standard of “Thermal Spraying Determination of Tensile Adhesive Strength [6]”.

3. Results and Discussion

Various factors affect the plasma spraying coatings prepared by the different equipment, such as spraying power, spraying distance, carrier gas flow, power feeding rate, cooling gas flow, sandblasting distance, sandblasting pressure, sandblasting angle [7] and so on. Based on our preliminary investigation and survey for the Praxair 100HE high enthalpy plasma spray gun, the microstructure and properties of FeSiAl coatings can be controlled by spraying power, spraying distance, carrier gas flow, power feeding rate with the same sandblasting conditions and the others. To further explore the effect of these four significant factors on the microstructure and properties of FeSiAl coatings, three levels of each factor were selected to establish the orthogonal test, that is, L9 (3^4) four factors and three levels orthogonal arrangement test, as can be seen in tables 1 and 2.

To analyze the effect of these four significant factors on the microstructure and properties of FeSiAl coatings, range factor R was calculated, as shown in table 3. Figure 1 shows the response curve of control factors for microhardness and adhesive strength of P1-P9. As shown in table 3, four factors including carrier gas flow, spraying distance, spraying power, power feeding rate when microhardness was considered as the primary index. The optimal parameters for microhardness can be expressed as C3B1 A1D2 which is 30 SCFH, 140 mm, 50 KW, 3 PRM. Meanwhile, it can be concluded that primary and secondary sequences of spraying parameters for adhesive strength of coatings are carrier gas flow, power feeding rate, spraying distance, spraying power (see chart 3). The best parameters of spraying coatings can be represented by C3D1B2A3, in the case, 30 SCFH, 2 PRM, 200 mm, 50KW.

| Table 1. Control factors and levels of L9 (3^4) orthogonal. |
|------------------|------------------|------------------|------------------|
| Level | A [Spraying Power (KW) ] | B [Spraying Distance (mm) ] | C [Carrier gas flow (SCFH) ] | D [Power feeding rate (PRM) ] |
| 1 | 40 | 80 | 10 | 2 |
| 2 | 45 | 140 | 20 | 3 |
| 3 | 50 | 200 | 30 | 4 |

| Table 2. L9 (3^4) orthogonal experimental layout with microhardness and adhesive strength. |
|------------------|------------------|------------------|------------------|------------------|------------------|
| Experiment number | A | B | C | D | Microhardness (HV) | Adhesive strength (MPa) |
|------------------|------------------|------------------|------------------|------------------|------------------|------------------|
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| $P_1$ | 40 | 80 | 30 | 3 | 737.2 | 22.395 |
|------|----|----|----|---|-------|-------|
| $P_2$ | 40 | 140 | 10 | 4 | 500.8 | 7.037 |
| $P_3$ | 40 | 200 | 20 | 2 | 492.2 | 25.956 |
| $P_4$ | 45 | 80 | 10 | 2 | 467.9 | 13.044 |
| $P_5$ | 45 | 140 | 20 | 3 | 738.1 | 13.812 |
| $P_6$ | 45 | 200 | 30 | 4 | 589.9 | 29.442 |
| $P_7$ | 50 | 80 | 20 | 4 | 746.5 | 10.140 |
| $P_8$ | 50 | 140 | 30 | 2 | 825.8 | 56.576 |
| $P_9$ | 50 | 200 | 10 | 3 | 413.0 | 15.091 |

**Table 3.** Range analysis of $L_9(3^4)$ orthogonal experimental.

| Control factors | A          | B          | C          | D          |
|-----------------|------------|------------|------------|------------|
| $K_1'$          | 576.73     | 650.53     | 460.57     | 593.30     |
| $K_2'$          | 598.63     | 688.23     | 658.93     | 629.43     |
| $K_3'$          | 661.77     | 498.37     | 717.63     | 612.4      |
| $R'$            | 85.03      | 189.87     | 257.07     | 34.13      |

The sequence of the factors: $C>B>A>D$

The optimum scheme: $C_3B_2A_3D_2$

| $K_1''$         | 18.46      | 15.19      | 11.72      | 25.19      |
| $K_2''$         | 18.77      | 19.14      | 16.64      | 17.10      |
| $K_3''$         | 20.60      | 23.50      | 29.47      | 15.54      |
| $R''$           | 2.14       | 8.30       | 17.75      | 9.65       |

The sequence of the factors: $C>D>B>A$

The optimum scheme: $C_3D_1B_3A_3$

As shown in figure 1a, the value of microhardness increases with spraying power increasing to 50KW, the reason lies in the higher-power accelerates the melting degree of FeSiAl powder, which improving the density of coatings. The microhardness values firstly increase and then declines as the spraying distance changing. According to the gas cooling theory, the spraying powder has longer flights under the cooling gas, the containing lower heat energy decreases the melting degree of FeSiAl powder, finally, reducing the compactness of coatings. The phenomenon can be explained by the spraying powder has the best melting condition during the spray process. The heat energy of spraying powder for 200 mm keeps a lower value comparing with 140 mm and 80 mm spraying distance. The increasing carrier gas flow upgrades the values the microhardness, possible because the increasing carrier gas flow reduces the porosity and microcracks of coatings and the increasing the amplitude of carrier gas flow greater than the decrease of temperature. The microhardness values increase and then decrease upon the power and feeding rate increasing. It is probably that the added spraying powder cannot be timely incomplete fusion reducing the compactness of the coatings. As shown in figure 1b, the adhesive strength of coatings appears to be rising as the spraying power increase due to the higher-power promoting the compactness of coating. The adhesive strength of coatings gradually increases with the increasing spraying distance. It is highly possible that thermal failure has little influence on the coating. Meanwhile, a larger distance gives plenty of time to release the residual stress of the coating. The increasing carrier gas flow upgrades the adhesive strength because of the increasing carrier gas flow reducing the porosity and micro cracks of coatings. Interestingly, the adhesive strength of coatings is progressively lowering as the rising power feeding rate. It might be due to that the larger power feeding rate, the greater residual stress. The larger power feeding rate leads to the number of unmelted particles, which improving the residual stress of coatings.
Figure 1. Response curve of control factors: (a) microhardness, (b) adhesive strength.

SEM and EDS analysis for the $P_1$-$P_9$ coatings are shown in figure 2 and table 4. As can be seen from figure 2, it can be found that the coatings are composed of fattening FeSiAl particles (pale gray) and oxidation phase of FeSiAl (dark color). Flattening indicates FeSiAl particles fully melt in the spray process. The spray powder hit the surface of the substrate and lamellar cambium structure experiencing full deformation and stacking during deposition. Furthermore, the cross-sectional morphology emerges the feature of uniform, compact, less porosity. Selected areas A and C are the highly oxidized phases of FeSiAl. Selected areas B and D are flatting FeSiAl. Selected area E located in the substrate is found to contain Mn, Ni and Cr element besides iron. The highly oxidized phase formed due to taking place the inflight oxidation and the post-impact oxidation.

Table 4. EDS results corresponding to figure 2 (i) and (k) (normalized, in wt. %).

| Figure Number | Selected area | Fe  | Si   | Al   | O    | Cr    | Mn   | Ni   | Total |
|---------------|---------------|-----|------|------|------|-------|------|------|-------|
| 3 (i)         | A             | 70.86 | 12.00 | 1.98 | 15.16 |       |      |      | 100.00 |
|               | B             | 86.62 | 9.07  | 4.31 |       |       |      |      | 100.00 |
|               | C             | 73.57 | 8.31  | 2.81 | 15.32 |       |      |      | 100.00 |
| 3 (k)         | D             | 85.39 | 8.97  | 5.64 |       |       |      |      | 100.00 |
|               | E             | 50.16 | 1.20  | 3.02 | 25.44 | 1.57  | 18.60 |      | 100.00 |
Figure 3 shows the SEM-mapping photograph of Fe, Si, Al, O, Cr, Mn, and Ni in P9. Fe is in all zones of P9, and its content is the highest. Si and Al are mainly distributed in the coating region. Cr, Mn, and Ni are primarily distributed in the substrate region. O is randomly dispersed in the matrix.

![Figure 3. SEM Mapping of Fe, Si, Al, O, Cr, Mn, and Ni in P9.](image)

Figure 4 exhibits the X-ray diffraction (XRD) spectra of P1-P9 coatings deposited by plasma spraying and as-received. The as-received XRD pattern presents seven peaks at about 27.3, 31.2, 44.8, 53.2, 55.7, 65.4, and 82.9°, corresponding to (111), (200), (220), (311), (222), (400) and (422) of FeSiAl, respectively. It indicates that the as-received is nearly single phase and structure of the major phase includes A2, B2, and D03 [8-10]. According to the results of calculation and simulation by using Materials Studio, the disordered A2 structure consists of (200), (400) and (422) diffraction peak, corresponding to 44.8, 65.1, and 82.5°, respectively. B2 is defined as the intermediate state between A2 and D03. D03 contains a (111) diffraction peak at 27.3° and a (200) diffraction peak at 31.2°. (200) and (111) diffraction peak appear with the increasing degree of order for FeSiAl phase structure, also known as characteristic peaks of hyperlattice. For the P1-P9 coatings, the XRD patterns show the major diffraction peaks contain (200), (220), (400) and (422). Comparing with the XRD pattern of as-received, the intensity of diffraction peak (111) and (222) decrease, which means the percentage of the disordered structure reduces in FeSiAl coating. Maybe because the phase structure of FeSiAl powder gradually become disordered in the process of striking forward the substrate after melting.

![Figure 4. XRD patterns of P1-P9 samples.](image)

4. Conclusion
The plasma-sprayed FeSiAl coatings were prepared through the Praxair 100HE high enthalpy plasma spray gun on stainless steel 316 substrate by orthogonal experiment. The main conclusions in this paper are as follows:

(i) The best parameters of sprayed coatings can be represented by C\(_3\)D\(_1\)B\(_3\)A\(_3\) for adhesive strength, i.e., 30 SCFH, 2 PRM, 200 mm, 50KW. The optimal parameters can be expressed as C\(_3\)B\(_2\)A\(_3\)D\(_2\) for microhardness, i.e., 30 SCFH, 140 mm, 50 KW, 3 PRM.

(ii) The cross-sectional of sprayed coatings are composed of fattening FeSiAl particles (pale gray) and oxidation phase of FeSiAl (dark).

(iii) Comparing the original FeSiAl particles, the percentage of disorder structure is reduced in FeSiAl coatings.

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