Process optimization and coating properties of aluminum coating prepared by supersonic plasma powder feeding based on response surface

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Abstract. This paper adopts High Efficiency Supersonic Plasma Spraying (HEPJet) to produce aluminum coating under the condition of large powder delivery. Based on the Box-Behnken-Design (BBD) design in Response Surface Method (RSM), the interaction of Ar flow, power and spraying distance with the deposition efficiency and microhardness of aluminum coating was analyzed, and the spraying parameters of aluminum coating prepared by supersonic plasma powder feeding were optimized. The coating performance was analyzed by means of electron microscopy (SEM) and X-ray diffraction (XRD). The results show that the established quadratic equation model is reliable and the influence of spraying distance coating on deposition efficiency and microhardness is greatest. The parameters of spraying with large powder delivery were as follows: argon 200 L/min, power 50 kW, spraying distance 40 mm, and powder delivery amount 150 g/min. Moreover, the aluminum coating prepared by HEPJet has poor density, large porosity and severe oxidation, which provides reference value for the subsequent research on the preparation of aluminum coating by HEPJet.

1. Introduction

The corrosion of steel materials has always caused huge losses to China's national economy. According to statistics, the annual economic losses caused by corrosion in China account for about 2%-4% of the gross national product. Corrosion of steel materials causes a lot of waste of resources and energy. Therefore, it is very important to study the corrosion protection technology of steel materials [1-2].

Aluminum is an active metal that is easily oxidized in the atmosphere to form a chemically stable alumina. It has a good function of isolating corrosive media. At the same time, aluminum is a good anode material, and it has a cathodic protection effect on the steel material [3]. At present, methods for preparing aluminum coating mainly include thermal infiltration methods (vapor deposition method, vacuum evaporation method, hot dip plating method), physical methods (sputter deposition method, electron beam evaporation deposition method, electric pulse deposition method), and thermal spray method (flame spray, arc spray). These conventional methods have problems such as high cost, low
deposition efficiency, easy oxidation in the spraying process, and poor coating quality. HEPJet has the characteristics of low energy consumption, small gas flow, inert heat source, high temperature, high speed, small spraying distance, etc., which is beneficial to produce low-cost, high-performance coatings [4].

Response Surface Methodology (RSM) is a statistical method that uses the multivariate regression equation to fit the functional relationship between the factor and the response value by analyzing the contour of the response surface to find the optimal process parameters[5]. It is a function of one or more factors (such as power, main gas flow, spray distance, etc.) as a function of the system's response (such as coating deposition rate and microhardness hardness), using graphical techniques to display this functional relationship for the tester to directly observe the sensitive relationship between each factor and the response value.

Therefore, in this paper, based on the RSM and the quadratic equation response model of power, main gas flow, spray distance and deposition efficiency and microhardness of aluminum coating by HEPJet large powder feeding aluminum coating, gets the optimal spraying parameters and analysis the structural characteristics and coating properties of the aluminum coating to provide a reference for the follow-up work of HEPJet to prepare aluminum coatings.

2. experiment method
2.1 Sample preparation method
In this test, a high efficiency supersonic plasma spraying system (HEPJet, as shown in Figure 1) independently developed by the State Key Laboratory was used to prepare an aluminum coating. The spray material was aluminum powder produced by Beijing Mulberry Co., Ltd. (particle size ranged from 15 to 45 μm), and the powder morphology is shown in Figure 2.

The sample used for coating characterization was prepared from 10mm×50mm×5mm tempered 45 steel as the substrate. First, the surface of the substrate was polished by sand paper from small to large size, and the surface impurities and oxide film were removed. Use an ultrasonic cleaner to clean and remove oil on the surface. The surface of the matrix was roughened with 24 eyed emery before spraying.

2.2 Coating performance testing method
Powder and coating morphology were analyzed using Quanta 200 scanning electron microscopy (SEM). The chemical composition of the cross section of the coating was analyzed using a Genesis X-ray energy spectrometer (EDS). The phase composition of the coating was analyzed using an Advance type polycrystalline X-ray diffractometer (XRD). The microhardness tester was used to measure the microhardness of the coating. During the measurement, the loading load was 100g and the loading time was 10s.

2.3 Test design
2.3.1 Response factor selection
The factors affecting the HEPJet spray aluminum coating mainly include: main gas (Ar) flow rate, secondary gas (H2, N2) ratio of main gas flow rate, working current and voltage, powder feeding amount, powder feeding gas pressure and flow rate, spraying Distance and so on. For HEPJet, when
the main gas flow is constant, the secondary air flow is proportional to the working voltage, and the voltage is usually adjusted by adjusting the flow rate of the secondary gas. The maximum value of the voltage is limited by the main gas flow rate, because the maximum proportion of the secondary gas flow in the main gas is generally 25%. If it exceeds 40%, the effect on the jet enthalpy and voltage change is small, and at the same time it also reduces the life of the nozzle. The working voltage and working current mainly determine the spraying power. According to the previous spray experience, when the jet state is good, straight, non-forked, and unbiased, the powder feeding parameters such as powder feeding amount, powder feeding pressure and flow rate have relatively little influence on the coating performance[6], so this test uses HEPJet spray aluminum coating mainly considering three factors: main gas (Ar) flow rate, spray power (P) and spray distance (D).

2.3.2 Selection of parameter range

Due to particle velocity and equipment requirements, the gas flow rate is not less than 75L/min, so the Ar flow rate is at least 70L/min. As N2 increases, the voltage increases, but the device limit must not exceed 155V. The total power is directly affected by the voltage and current. The maximum allowable current is 600A. The power of the Al powder fully melted is from 30 kW to 55 kW. If the spraying distance D is too small, the coating is easily ablated by the high temperature flame flow. And when the spraying distance is too large, the oxidation degree of the Al powder increases. The single factor test method was used to optimize the parameter range, and the final spray parameters was determined as: Ar flow 70-340 L/min, power P 30-55 kW, and spray distance D 40-150 mm. The single factor optimization test parameters are shown in Table 1, and the test factor levels are determined as shown in Table 2.

| Tab.1 Single factor optimization test parameter table |
|---------------------------------------------|
| Num| Ar flow (L/min) | N2 flow (L/min) | P (kw) | D (mm) | Powder feeding amount (g/min) | number of times | Deposition rate (mm) |
|----|---------------|-----------------|--------|-------|-----------------------------|----------------|---------------------|
| 1  | 70            | 14              | 47.5   | 80    | 7                           | 20             | 0.17                |
| 2  | 95            | 19              | 47.5   | 80    | 7                           | 20             | 0.16                |
| 3  | 120           | 24              | 47.5   | 80    | 7                           | 20             | 0.18                |
| 4  | 95            | 19              | 47.5   | 60    | 7                           | 20             | 0.18                |
| 5  | 95            | 19              | 47.5   | 80    | 7                           | 20             | 0.15                |
| 6  | 95            | 19              | 47.5   | 100   | 7                           | 20             | 0.14                |
| 7  | 95            | 19              | 32     | 40    | 7                           | 40             | 0.47                |
| 8  | 95            | 19              | 32     | 150   | 7                           | 40             | 0.18                |
| 9  | 95            | 19              | 55     | 40    | 7                           | 40             | 0.7                 |
| 10 | 95            | 19              | 55     | 150   | 7                           | 40             | 0.3                 |
| 11 | 70            | 14              | 55     | 40    | 7                           | 40             | 0.62                |
| 12 | 200           | 30              | 55     | 40    | 7                           | 40             | 0.53                |
| 13 | 340           | 30              | 55     | 40    | 7                           | 40             | 0.68                |

| Tab.2 Factors and levels |
|--------------------------|
| Factor | Coded values and levels |
|--------|--------------------------|
| P (kW) | 30 42.5 55              |
| Ar (L/min) | 70 205 340          |
| D (mm) | 40 95 150               |

3. Results and discussion
3.1 Response surface test result

The experimental parameters were determined according to the conclusion of the optimization parameters of the single-factor experiment at the earlier stage. The Design test was conducted by
box-behnken Design using design-expert software, and the following 17 groups of response surface test results were generated, as shown in table 3. It can be seen from the table that the deposition rate of aluminum coating within the selected spraying parameter range is 0.1-0.52 mm/20 time, and the microhardness range is 26.98-52.26 HV.

| number | Ar flow(L/min) | Power P(kw) | Spray distance D (mm) | Deposition rate (mm/20 times) | Microhardness (HV) |
|--------|----------------|-------------|-----------------------|-----------------------------|-------------------|
| 2      | 205            | 55          | 150                   | 0.35                        | 26.98             |
| 3      | 70             | 55          | 95                    | 0.31                        | 45.86             |
| 4      | 205            | 30          | 40                    | 0.48                        | 38.52             |
| 5      | 205            | 42.5        | 95                    | 0.35                        | 40.5              |
| 6      | 70             | 42.5        | 40                    | 0.36                        | 52.26             |
| 7      | 340            | 42.5        | 150                   | 0.1                         | 42.74             |
| 8      | 205            | 42.5        | 95                    | 0.34                        | 42.86             |
| 9      | 340            | 55          | 95                    | 0.31                        | 44.44             |
| 10     | 205            | 30          | 150                   | 0.13                        | 31.82             |
| 11     | 205            | 42.5        | 95                    | 0.36                        | 41.09             |
| 12     | 340            | 42.5        | 40                    | 0.52                        | 48.16             |
| 13     | 205            | 42.5        | 95                    | 0.34                        | 43.26             |
| 14     | 205            | 42.5        | 95                    | 0.36                        | 43.55             |
| 15     | 70             | 42.5        | 150                   | 0.29                        | 38.02             |
| 16     | 70             | 30          | 95                    | 0.22                        | 45.26             |
| 17     | 340            | 30          | 95                    | 0.19                        | 44.25             |

3.2 Fitting model analysis

According to the data processing and analysis of the experimental results by design-expert software, the relation equation of three factors affecting the deposition rate (Dep) and microhardness (HV) of aluminum coating was obtained as follows:

\[
\text{Dep} = 0.35 + 0.056 \times P - 0.12 \times D - 0.088 \times Ar \times P + 0.05 \times P \times D - 0.07 \times Ar^2 - 0.023 \times P^2 - 0.037 \times D^2. (1)
\]

\[
HV = 42.25 - 0.23 \times Ar + 0.014 \times P - 5.26 \times D - 0.10 \times Ar \times P + 2.21 \times Ar \times D - 2.25 \times P \times D + 6.50 \times Ar^2 - 3.80 \times P^2 - 3.45 \times D^2. (2)
\]

It can be seen from the equation coefficients that the order of the three factors affecting the deposition rate of aluminum coating is D>P>Ar, and Ar has the least influence on the deposition rate. The order of the three factors affecting the microhardness of the aluminum coating is D>Ar>P, and the influence of P is the smallest.

The variance analysis was performed on the deposition rate model and the microhardness model. The results are shown in Table 4 and Table 5, respectively.
It can be seen from Table 4 that the F value of the deposition rate model is 321.61, and the P value is \( P<0.0001 \), which indicates that the relationship between the deposition rate and the main gas flow, power and spray distance regression equation is significant. The F value of the Degree of misfit is 0.42 and the P value is 0.751>0.05, and the influence relationship is not significant, indicating that the ratio of the obtained equation to the abnormal error in the actual fitting is small, and the regression equation is reliable. The P value of B and C in the model is \( P<0.0001 \), indicating that the influence of power and spraying distance on the deposition rate is very significant. The P value of the main gas flow is 0.0441<0.05, indicating that the main gas flow has a significant influence on the deposition rate, which is consistent with the coefficient analysis conclusion of the regression equation.

It can be seen from Table 5 that the F value of the microhardness model is 42.25, and the P value is \( P<0.0001 \), indicating that the relationship between the microhardness of the coating and the...
regression equation of the main gas flow, power and spray distance is significant, and the degree of mismatch The F value of the Degree of misfit is 0.42 and the P value is 0.7477>0.05. The influence relationship is not significant, indicating that the ratio of the obtained equation to the abnormal error in the actual fitting is small, and the regression equation is reliable. The P value of the spraying distance is P<0.0001, which indicates that the influence on the microhardness is most significant. The P values of the main gas flow and power are 0.6065 and 0.9748, respectively, which are less significant. This conclusion is consistent with the analytical conclusion of the regression equation.

According to the relation equations of main gas flow, power and spraying distance, deposition rate and microhardness, as well as the analysis of variance of deposition rate model and microhardness model, it can be concluded that spraying distance has the greatest influence on deposition rate and microhardness.

3.3 Response surface analysis

The response surface analysis of deposition rate and microhardness was performed by software, and the analysis results are shown in Figure 3 and Figure 4, respectively.

![Response surface analysis](image)

(a) Effect of Ar and P on deposition rate         (b) Effect of Ar and D on deposition rate

(c) Effect of P and D on deposition rate

**Fig.3** deposition rate model response surface map

It can be seen from Figure 3 (b) (c) that D has the greatest influence on the deposition rate, and the largest deposition rate is the smaller distance. According to the contour map, when the spraying distance is 40 mm, the deposition rate reaches the maximum. In Figure 3 (a), the deposition rate always decreases with the increase of Ar. This is consistent with the conclusion that the coefficient before Ar is negative in the deposition rate equation. It can be seen from the sedimentation rate equation that the influence of D on the deposition rate is negatively correlated. The deposition rate is decreasing as D increasing. This conclusion is verified in Figure (b). It can be observed from the curved surface diagram that when Ar flow remains unchanged, the change trend of deposition rate increases with the decrease of D. When D is 150 mm, changes in Ar flow have little impact on deposition rate, and the change trend of surface is not obvious. It is concluded in the sedimentary rate equation that the deposition rate is more affected by D than P, which is further verified in Figure 3 (c).

When the power is increased, the particles are more heated, which facilitates rapid deposition on the surface of the substrate. The smaller D, the less time it takes for molten aluminum particles to fly through the air, reducing the degree of oxidation and facilitating rapid particle deposition. When P and D are constant, Ar flow is too small, particles fly slowly, and the degree of oxidation increases, which is not conducive to deposition. Ar flow rate is too high, and the heating time obtained by the particles
is too short to melt into the best state, which affects the deposition rate. Therefore, an appropriate Ar flow rate is 200 L/min.

(a) Effect of Ar and D on hardness

(b) Effect of P and D on hardness

Figure 4 shows the comprehensive influence of Ar flow rate and D on microhardness. When the spraying distance is 40mm and the main gas flow rate is 70 L/min, the coating has the maximum hardness. When the main gas flow rate is the same, the hardness of the coating decreases with the increase of D. At the same spraying distance, the microhardness shows a trend of increasing first and then decreasing as the main gas flow increases. When the flow rate of Ar is small and the spraying distance is small, the Al particles have a good melting state, a short exposure time in the air and a small degree of oxidation. So a dense coating formed after the particles are deposited, which has the small pores and the largest hardness. When the spraying distance is constant, the hardness of the coating decreases as the main gas flow increases. This is because the increase of particle's flight speed leads to uneven heating and insufficient melting. Some particles are oxidized and more pores are formed, which reduces the density of the coating. In Figure 4 (a), when the Ar flow is the maximum and the spraying distance is the minimum, the surface appears the second highest point, because the particles have high flying speed and small spraying distance, the particles have a short flight time in the air and a small degree of oxidation. Therefore, when D is 40mm and Ar flow rate is 70 L/min, the microhardness of the coating obtained is relatively large.

Figure 4 (b) is a comprehensive effect surface diagram of P and D on microhardness. When the spraying distance is minimum 40 mm and the power is 55 kW, the coating hardness is the largest. The effect of D on the microhardness of the coating is still greater the distance and the lower the hardness. When the spraying distance is certain, the microhardness first increases and then decreases with the increase of P. When P is too small, the flame flow temperature is low, and the Al particles are not heated evenly, which fails to achieve the optimal melting state, the deposition effect is not good. The particles are not tightly bound together and the coating density is poor with many gaps. As the P increases, the temperature rises, the melting state of Al particles are good, the coating pores are reduced more densely, and the hardness increases. When the power is further increased, the microhardness is rather lowered because the jet temperature is too high, and the melting point of Al is lower at 660 ℃, resulting in over-melting of the particles. Therefore, when the choice of a small spraying distance is 40 mm, and the power can not be too small and not too large, preferably 40 kW, the coating is more dense and microhardness is larger.

3.4 Calculation of deposition efficiency under large powder feed rate

Deposition efficiency is one of the most important performance indicators, which provides a basis for the cost calculation of aluminum coating preparation under engineering practice. Weighing 50g of Al powder to spray on matrix until the powder is exhausted, and weigh the matrix after spraying. The mass increase of the test piece was measured, and the deposition efficiency was calculated by the formula (3).

\[ \eta = \frac{m}{50} \]  

(3)
Where $\eta$ is the deposition efficiency and $m$ is the weight gain of the test piece after spraying.

The response surface method was used to analyze the spraying factors affecting the coating. The spraying parameters of optimizing the HEPJet large powder feeding amount were as follows: powder feeding amount 150 g/min, main gas flow rate 250 L/min, power 45 kW, spraying distance 40 mm. The deposition efficiency test was carried out 6 times (see Table 6), and the results were averaged to obtain a deposition efficiency of 56%.

### Tab.6 Deposition efficiency test record table

| Test number | 1  | 2  | 3  | 4  | 5  | 6  |
|-------------|----|----|----|----|----|----|
| Weight gain/g | 25 | 28 | 31 | 32 | 26 | 27 |
| Deposition efficiency/% | 50 | 56 | 62 | 64 | 52 | 54 |

3.5 Performance analysis of aluminum coating prepared under large powder feed rate

After spraying, the surface and cross section of the coating were observed by scanning electron microscopy to analyze the morphology of the microscopic undercoat.

![Fig.5 Microscopic topography of the coating surface](image)

It can be found from Figure 5 (a) that the coating has a layered structure, and the thickness of the coating is different at uneven surface, which affects the density and service life of the coating. In Figure 5 (b), there are a small amount of large white particles, which is caused by uneven heating of the particles. The temperature of the particles have a large influence on layer-to-layer bond, and the dark region in Figure 5 (c) is pores, because the particles are easily oxidized.

![Fig.6 Microscopic topography of the coating](image)

The coating in Figure 6 (a) is relatively dense overall and is well bonded to the substrate. After enlarging the layer, a small amount of voids can be seen in Figure 6 (b), and a small amount of white spots may be unmelted particles. Due to the high-speed flame flow cannot melt all the particles into the optimal state, there are two micro-pits in Figure 6 (c). Due to the presence of unmelted particles and over-melted particles, micro-pits or voids are easily formed in the coating.

As shown in Figure 7, the EDS analysis of the coating revealed that the coating was mainly composed of Al element, and the content of O element was 11%. XRD analysis of the coating showed...
that the main component of the coating was Al and the coating contained a small amount of Al2O3, which oxidation was more serious.

Fig.7 XRD analysis images

4. Conclusion

(1) Among the various spraying factors of high efficiency supersonic plasma sprayed aluminum coating, the order of influence on the deposition rate of aluminum coating is spraying distance > power > main gas flow. And the order of influence on the microhardness of aluminum coating is spraying distance > main gas flow > power, where the spray distance has the greatest impact on both.

(2) The response parameters were optimized by the response surface method to obtain the powder feeding parameters: argon gas 200 L/min, power 50 kW, spraying distance 40 mm, and powder feeding amount 150 g/min.

(3) The aluminum coating is prepared by using a large amount of powder, and the microstructure of the aluminum coating is analyzed. The density of the coating is not good, the particles are not heated uniformly, and the temperature difference between the particles during the deposition process leads to uneven bonding, and the coating pores rate is large and the oxidation is serious.

Acknowledgements

This work was financially supported by the National Natural Science Foundation of China (51535011), National Key Basic Research Development Program (61328304).

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