

The laser deposition $\text{CoCrFeNiAl}_x\text{Nb}_y\text{Ti}_z$ high-entropy alloy coating is optimized based on the comprehensive evaluation method

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Abstract

This study is based on the CoCrFeNi alloy system. By adding alloying elements such as Al, Nb and Ti and using laser cladding technology for preparation, it aims to optimize the composition design of the alloy powder and the cladding process parameters. This experiment conducted an optimization study on the process parameters of laser cladding. The experimental results show that the appropriate addition of Al, Nb and Ti elements can effectively enhance its mechanical properties. This research provides important theoretical basis and practical guidance for the design and development of high-performance CoCrFeNi series alloys and the engineering application of laser cladding technology.

Keywords

$\text{CoCrFeNiAl}_x\text{Nb}_y\text{Ti}_z$ high-entropy alloy; laser cladding; mechanical property

1. Introduction

Titanium alloys, magnesium alloys and other alloys possess excellent properties such as high specific strength, good toughness and low density [1,2]. Meanwhile, due to its abundant reserves on Earth, it is widely used in fields such as aerospace and the automotive industry [3-4]. However, with the development of industry, these materials will be increasingly used in high-temperature, high-pressure and abrasive environments. The disadvantages of poor wear resistance and poor high-temperature stability limit their application [5]. To address these issues, many surface strengthening techniques have been adopted to enhance the wear resistance and corrosion resistance of these alloy surfaces, such as plasma spraying, physical vapor deposition (PVD), chemical vapor deposition (CVD), surfacing, carburizing, nitriding, etc.

Laser has been widely applied in material surface treatment due to its advantages

such as high energy density, good coherence and good directionality [6,7]. Laser surface treatment technologies include laser surface alloying, laser strengthening, laser cladding (LC), laser remelting, etc. For a long time, the design of most traditional alloys has aimed to enhance coating performance by adding trace elements to the main components. In 2004, Yeh et al. [8] proposed a new alloy design concept, namely high-entropy alloy (HEA). High-entropy alloys have four main effects: high-entropy effect, slow diffusion effect, lattice distortion effect, and "cocktail" effect. The high-entropy effect is a representative effect of high-entropy alloys and is mainly used to explain multi-principal component solid solutions. The inert diffusion effect is used to describe the formation of nanoscale pre-precipitated phases; The lattice distortion effect is used to explain the high-strength characteristics of high-entropy alloys [9]; The "cocktail" effect was first proposed by the Indian scholar Ranganatha[10]. This effect emphasizes the unique properties of each element. The material combination will produce a special synergy effect while significantly enhancing the design flexibility of HEAs. The CoCrFeNi series of high-entropy alloys is currently a popular system in the research of high-entropy alloys [11]. By adding elements such as Al, Ti, C, and Mo to CoCrFeNi, changes in crystal structure and improvements in performance can be achieved. High-entropy alloy coatings prepared by laser cladding typically possess excellent wear resistance, corrosion resistance, high-temperature oxidation resistance, soft magnetic properties, and outstanding radiation resistance. Due to the large temperature gradient, fine-grained and tough coatings will form on the substrate surface. Laser cladding coatings have the advantages of being able to form a good metallurgical bond with the substrate, having a small dilution rate and heat-affected zone, and being able to form a fine microstructure. They are environmentally friendly, simple, flexible and material-saving.

2. Experimental procedure

In this study, 9 single-layer coating layers (CoCrFeNi, CoCrFeNiNb_{0.3}Ti_{0.8}, CoCrFeNiNb_{0.3}Ti_{0.5}, CoCrFeNiAl_{0.7}Ti_{0.8}, CoCrFeNiAl_{0.7}Nb_{0.3}Ti_{0.5}, CoCrFeNiAl_{0.7}Nb_{0.5}, CoCrFeNiAlTi_{0.5}, CoCrFeNiAlNb_{0.3}, CoCrFeNiAlNb_{0.5}Ti_{0.8}) were fabricated on the surface of 45# steel substrate by laser cladding technology. The experimental data of the cross-sectional morphology ratio, dilution rate and deposition angle of the coatings were quantitatively scored using the comprehensive scoring method. Finally, the appropriate powder ratio and laser cladding process parameters were determined through hardness testing of the coatings.

The mechanical properties were evaluated using a Vickers microhardness tester (HV-1000) with a load of 1000f and a loading time of 10 seconds. Hardness measurements were conducted on the upper part of the coating, with three points measured at each height position. The average value was taken to reduce experimental errors. The horizontal and vertical spacing of the measurement points was 200 μ m.

3. Conclusions and Discussion

3.1. Cross-sectional morphology of a single-coat layer

Figure 2 presents the cross-sectional metallographic micrographs (OM) of all single-pass laser cladding coatings, which are used to evaluate the geometric morphology and forming characteristics of the coatings under different parameter conditions. Figure 1 defines the typical geometric parameters adopted in the analysis of the coating cross-section, including deposition width (W_d), deposition height (H_d), deposition area (A_d), and melted substrate area (A_m). Based on these basic geometric features, three key evaluation parameters that reflect the forming quality of the coating can be further calculated: aspect ratio (R), dilution rate (η), and deposition angle (θ). These three parameters are of great significance in the research of laser cladding processes and can effectively reflect the metallurgical bonding degree between the coating and the substrate, the matching relationship between energy input and material response, and the comprehensive effect of melt pool flow and solidification behavior. Among them, a higher aspect ratio usually indicates better height control in forming and lower material fluidity, which is suitable for multi-pass overlapping; an appropriate dilution rate reflects good base material bonding and alloy composition control; and the deposition angle is closely related to the continuity of the coating boundary transition zone. A larger deposition angle often indicates better edge forming and metallurgical transition quality. Therefore, these three parameters are widely regarded as the most representative quantitative indicators for evaluating the forming quality of single-pass laser cladding coatings, and have important guiding significance for optimizing process parameters and regulating microstructure properties [12, 13].

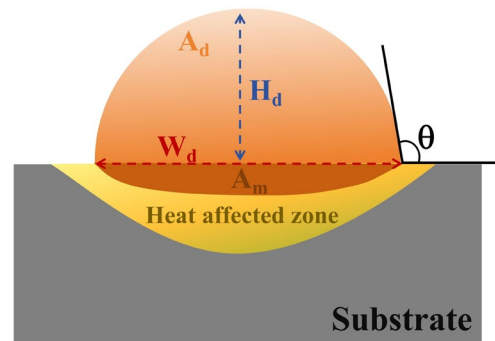


Figure 1. Schematic diagram of the cross-section of the high-entropy alloy coating

$$R = \frac{W_d}{H_d} \quad (1)$$

Here, R represents the aspect ratio, W_d is the deposition width, and H_d is the deposition height.

$$= \frac{A_m}{A_d + A_m} \times 100\% \quad (2)$$

Here, η represents the dilution rate, A_d denotes the area of the deposition region, and it is equal to the area of the substrate that A_m melts.

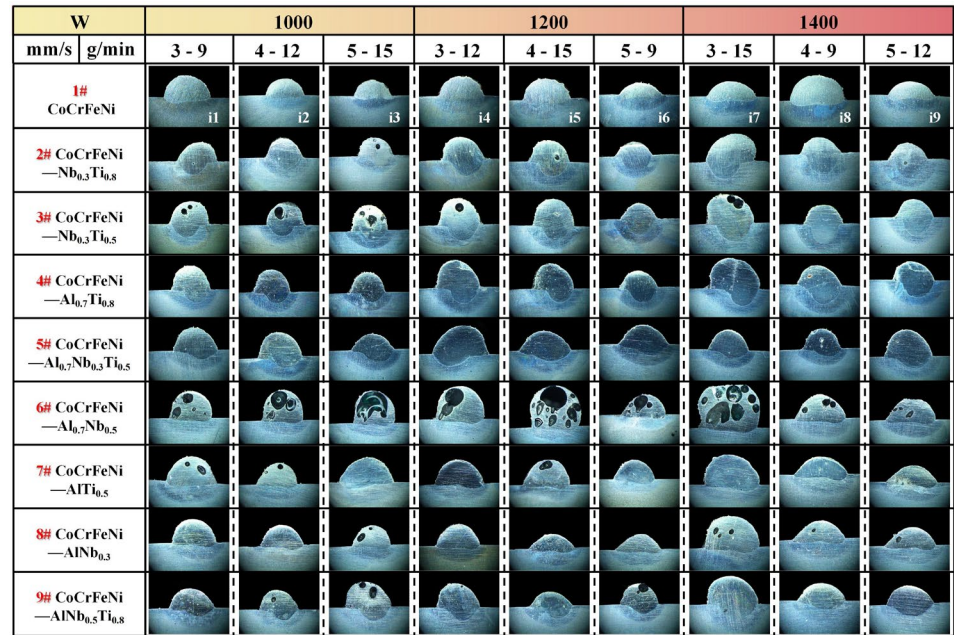


Figure 2. Cross-sectional morphology of high-entropy alloy coating

From the cross-sectional morphology of the single-layer coating shown in Figure 2, it can be observed that the fusion coatings of alloys 1#, 4#, and 5# all exhibited obvious cracks or pore defects under different process parameters, but the forming quality was good. Additionally, the specimens 2# (i1, i2, i4, i6-i8), 3# (i5, i6, i8, i9), 7# (i3, i4, i6-i9), 8# (i4, i6), and 9# (i1, i5, i7-i9) did not show any macroscopic cracks or pores. In contrast, alloy 6# exhibited obvious pore defects under multiple sets of parameters, which no longer met the requirements for subsequent performance testing and process optimization. Therefore, it will not be considered in the subsequent analysis. After undergoing metallographic etching, the base material, the heat-affected zone and the coating layer show distinct color differences, which is helpful for clearly identifying the interface position and the penetration depth characteristics. The size of the penetration depth is jointly influenced by the alloy composition and process parameters. The results show that the coatings formed by 1#, 5#, and 7#-9# alloy powders have smaller penetration depths, while the coatings corresponding to 2#-4# alloy powders have larger penetration depths. As shown in Figure 3, the aspect ratio (R), dilution rate (η), and deposition angle (θ) of each coating layer exhibit significant differences under different process parameters. Among them, the η of 1#, 2#(i3), 3#(i1-i5, i7), 4#(i1-i5, i7), 5#(i1, i2, i4, i7-i9), 7#, 8#, and 9#(i1-i4, i6, i7) samples are all within 10%-30%, while the η of the remaining samples is over 30%, which may cause unnecessary consumption of the base material. η is one of the important parameters for measuring the metallurgical bonding degree between the coating and the substrate during cladding, and its size not only affects the diffusion of interface elements and the bonding strength,

but also determines the degree of refinement of the final microstructure and the stability of the performance [14, 15]. A larger η indicates that more matrix is melted, which may result in coarse structures and higher internal stress, thereby reducing the performance of the deposited layer. On the other hand, a smaller η may lead to poor bonding between the coating and the substrate, increasing crack sensitivity, and even causing macroscopic cracks or delamination failure [16]. At the same time, it can be seen that θ increases with the increase of laser power and scanning rate. A larger θ is conducive to forming a more complete and smoothly transitioning coating geometry, thereby reducing the porosity and increasing the density and structural integrity of the deposited layer. This, in turn, leads to a decrease in the quality of the deposited coating. It is worth noting that in samples 2# - 9#, when the laser power is 1400 W, the scanning rate is 3 mm/s, and the powder feeding rate is 15 g/min, the R, η , and θ of the obtained coating show significant decreases. This is mainly due to the fact that a larger laser energy is preferentially used for the melting of the powder, resulting in a reduction in the effective energy density acting on the substrate surface, and thereby reducing the penetration depth and the degree of substrate dilution. It should be emphasized that the differences between the studies are expected. This is because, in addition to the main process parameters such as laser power, scanning rate, and powder feeding rate, factors such as the size and morphology of the powder particles, the design of the scanning trajectory, the equipment manufacturer, and the distribution of the laser spot also have a significant impact on the geometric shape and forming quality of the coating.

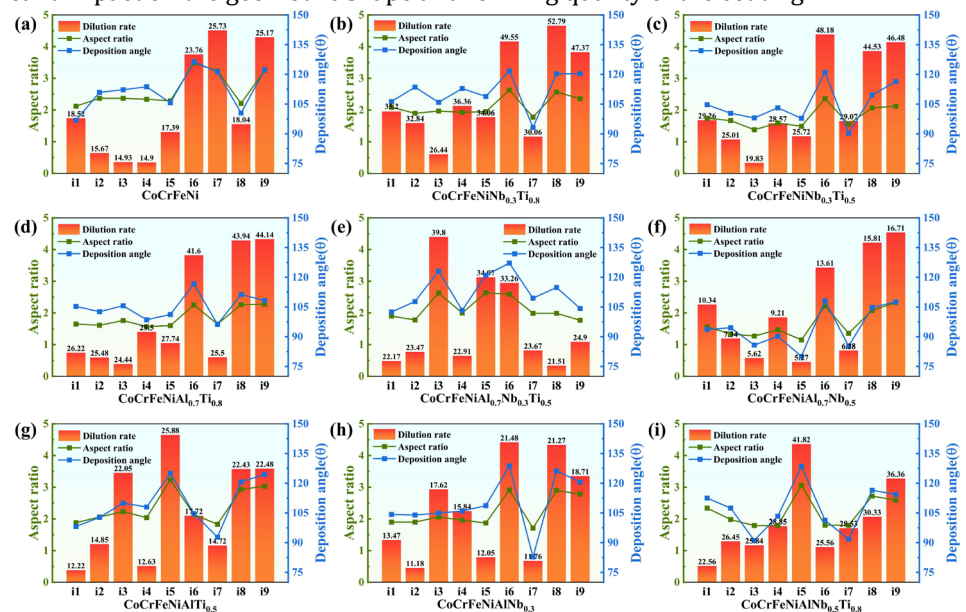


Figure 3. Forming parameters (dilution rate, aspect ratio and deposition Angle) of high-entropy alloy coating

3.2. Comprehensive Evaluation of Single-Coat Forming Quality

Generally speaking, a higher aspect ratio (1.5-2.5), a lower dilution rate (10%- 20%), and a larger deposition angle (100° - 120°) are required to fabricate a high-performance single-layer coating. However, in actual processes, it is often difficult to achieve the optimal match simply by adjusting a single process parameter. Therefore, it is urgent to introduce multi-objective optimization methods to seek the global optimal solution for coating performance. In multi-response optimization problems, when the number of target responses is small, the comprehensive scoring method is a commonly used and effective analytical approach. This method standardizes each performance indicator, assigns weights to the scores, and calculates the weighted average of each score value, thereby obtaining a comprehensive score reflecting the overall performance of the sample. Specifically, different indicators set weight coefficients according to their importance, and the final score can be used for ranking the advantages and disadvantages of different coating samples or parameter combinations [17, 18]. The performance of each single-layer coating under different process parameters is evaluated based on the comprehensive scoring method, and the calculated comprehensive score is shown in Figure 4. The calculation formula for the comprehensive score is as follows (i is the aspect ratio, j is the dilution rate, and k is the deposition angle)

$$\delta_i = \left| \frac{x_i - x_i^*}{x_i^*} \right| \quad (3)$$

$$P_i = 100 \times (1 - \delta_i) \quad (4)$$

$$P_{total} = \omega P_i + \omega P_j + \omega P_k \quad (5)$$

Among them, x_i is the relative deviation, x_i^* is the actual measured value, x_i^* is the ideal target value, P_i is the index score (if the relative deviation is too large, it is given a score of 0), P_{total} is the total score of the comprehensive evaluation, and ω is the weight coefficient ($\omega = 1/3$).

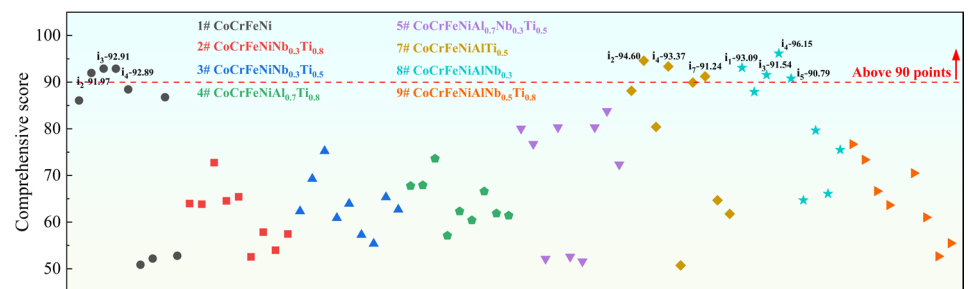


Figure 4. Comprehensive scoring results of high-entropy alloy coatings

From the comprehensive scoring results in Figure 4, it can be seen that a total of 9 groups of samples scored above 90 points, indicating that they performed exceptionally well in multiple evaluation indicators such as dilution rate, microhardness, and forming quality. Among them, the 8#(i4) alloy powder (with the composition of CoCrFeNiAlNb_{0.3} HEA) achieved the highest comprehensive score under the param-

eter combination of laser power of 1200 W, scanning speed of 3 mm/s, and powder feeding rate of 12 g/min. This shows the optimal comprehensive forming quality. Single-pass deposition is the basis for subsequent multi-layer and multi-track deposition. Reasonable process parameters not only directly affect the surface morphology of the deposition layer but also significantly impact the microstructure and properties within the deposition layer [19]. Therefore, in the pre-processing of multi-layer construction, in-depth research and optimization of the single-pass deposition behavior is an important prerequisite for ensuring the excellent macro and micro performance of the sample in the end.

3.3. Microhardness testing analysis

To select the coatings with superior performance, Vickers hardness tests were conducted on each sample, and the resulting hardness distribution curves are shown in Figure 5. Generally speaking, the higher the microhardness of the coating, the better its wear resistance and service life [20]. However, in this study, considering the macroscopic morphology of the coating surface and section as well as the forming quality of the single-coat layer, a comparative analysis was carried out on the three coating samples 7# (i4, i7) and 8# (i4). From the test results, the microhardness of 7# (i4, i7) samples was all higher than that of 8# (i4), but 7# (i4) had poorer surface forming quality than 7# (i7) and 8# (i4). Moreover, although the dilution rate of 7# (i7) sample performed well, its aspect ratio and deposition angle were smaller (Figure 5g), which was not conducive to forming a coating with good structural integrity. In contrast, the 8# (i4) sample was more excellent in terms of aspect ratio, deposition angle and dilution rate control, showing better forming performance and higher hardness.

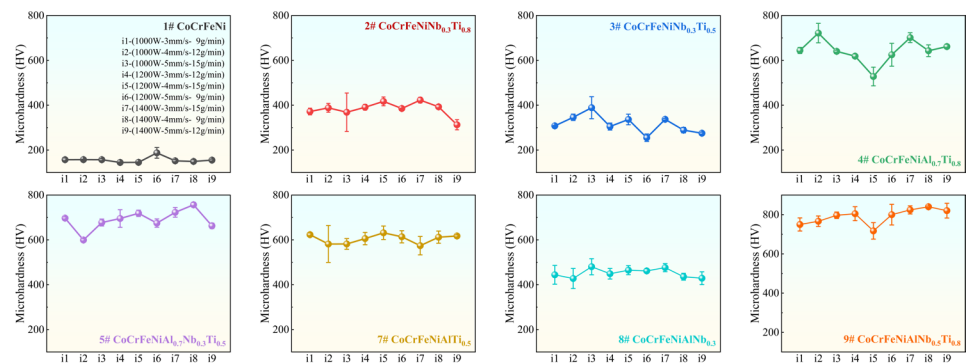


Figure 5. Microhardness curve of cross section of high entropy alloy coating

4. Summarize

In this study, a CoCrFeNiAl_xNb_yTi_z multi-component high-entropy alloy system was constructed through orthogonal experimental design, and the laser cladding process parameters were optimized. The optimal process parameter combination for the

CoCrFeNiAlNb_{0.3} high-entropy alloy (HEA) laser cladding coating with the best forming quality was obtained, and microhardness tests were conducted on the samples. The main conclusions are as follows:

- (1) Through the comprehensive scoring method, multiple forming quality indicators were quantitatively evaluated to determine the optimal process parameter combination for the laser cladding CoCrFeNiAlNb_{0.3} HEA coating as 8# (i4), that is, the laser power is 1200 W, the scanning speed is 3 mm/s, and the powder feeding rate is 12 g/min. This can result in a coating with higher forming quality.
- (2) Under the same combination of laser cladding process parameters, the microhardness of sample 8# (i4) (449.0 HV) was significantly higher than that of the CoCrFeNi alloy sample (1# (i4), with a hardness of 144.9 HV), with a hardness increase of approximately 2.1 times. Additionally, compared to the 45# steel substrate (259.6 HV), its hardness increased by approximately 73%, verifying the effectiveness of the alloy composition design and the optimization of the cladding process in improving the micro-mechanical properties.

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