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Mechanism of pool separation and stratification in laser remelting and cladding[®]

Zeng Dawen(曾大文), Xie Changsheng(谢长生), Sheng Yaming(盛亚明)

College of Materials Science and Engineering,

Huazhong University of Science and Technology, Wuhan 430074, P. R. China

Abstract: The laser remelting with a two-layer material system (upper material was Al-30 % Ti-20 % Ni alloy, substrate was commercial aluminum alloy) and the laser cladding of a commercial 45 steel with copper powder (including 25 % SiC) were carried out using a 2 kW continuous CO₂ laser. For the case of laser remelting, a upper pool in the alloying layer and a lower pool in the substrate separated by the unmelted Al-Ti-Ni alloy were observed. For laser cladding, a stratified pool was observed, whose top layer was Cu alloy liquid and bottom was Fe alloy liquid. The mechanism of laser pool separation and stratification is illustrated by numerical calculation of heat transfer process of the two-layer system, combining with material physical properties (especially mixed enthalpy). A classification criterion for laser pool with the two-layer material system has been presented and four types of the laser pool are divided into unique pool, separated pool, mixed pool and stratified pool, which provides a theoretical basis for obtaining a excellent surface coating.

Key words: laser cladding; laser melting; pool separation; pool stratification; mechanism

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1 INTRODUCTION

In recent years, a great deal of attention has been focused on laser remelting and cladding technique, which have been used extensively in industries because of improving wear-resistance, corrosion-resistance and thermal-barrier of the material surface^[1,2]. In order to obtain a good surface coating, experimental and numerical investigations on characteristics of laser pool were carried on by many researchers. ZrO₂ + Ni composite coating was produced by laser cladding on 4Cr13 steel substrate and stratified structure in clad has been observed by Pei et al [3] (ZrO2 ceramic and Ni element separation). Laser cladding experiments were done by Wang et al [4] on an AISI 45 steel substrate with Cu + 5% Cr₂O₃ powder, and stratification phenomenon has been observed too (Cu rich laver, Fe rich layer and the space between them is $50 \mu m$). A

fully mixed, alloyed surface coating was produced by laser alloying on a surface of a 50 mm × 50 mm × 10 mm alumimum sample, on which a Ni element layer was prepared by plasma spray technique^[5]. Various types of laser pool were observed by different researchers, but few reports study on the mechanism of laser pool separation and stratification, and there is no criterion for classification of laser pool.

Therefore, in this paper, the mechanism of separation and stratification of laser pool is investigated by laser cladding and laser remelting with a two-layer material system; the classification criterion is presented and four types of the laser pool are divided into unique pool, separated pool, mixed pool and stratified pool. The author's purpose of this paper is to provide the theoretical bases for obtaining the excellent surface coating and to build the foundation for numerical research of the process of metallurgical

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kinetics of laser pool.

2 EXPERIMENTAL PROCEDURE

The substrate used in laser remelting experiment was a 15 mm thick plate of commercial aluminium alloy. The plate was cut into a rectangle sample with a gauge of 80 mm × 60 mm. The sample was grit-blasted on one surface and followed by cleaning with acetone in an ultrasonic cleaner. A 100 µm thick 50 % Al-30 % Ti-20 % Ni alloying coating was sprayed on the cleaned surface of the sample, using plasma spray technique. Then, laser remelting was carried out using a 2 kW continuous CO2 laser. The average power used for remelting was 1.4 kW, diameter of beam was 0.8 mm, and laser intensity was of Gaussian distribution. The sample was manipulated by mounting it on an X-Y table, whose the movement was controlled by a computer. The sample was moved at a speed of 250 mm/s. In order to make the alloying layer combine with the substrate more tightly, the low speed (60 mm/s) laser remelting was carried out before remelting (250 mm/s) and the overlapping of each laser track by the followed track was 45 %.

The substrate used in laser cladding was AISI 45 steel whose size was 80 mm × 30 mm × 10 mm. The adhering metal was LCu-03, whose chemical composition was Si 0.3%-Al 5.39%-Fe 4.73% and the ceramic powder was SiC. The surface of the AISI 45 steel was grit-blasted, got the rust off, degreased, cleaned by acetone and dried in a furnace. The fully mixed powder which consisted of the SiC particles and Cu powder according to the proportion of 1:3 (in mass) was coated on the cleaned surface of the substrate using the self-made adhesive, and the 1 mm thick alloying coating was formed. The sample was then put into the heat treatment furnace at 100 °C for drying in order to remove the gas in the alloying layer. The power of laser used in cladding was 2.0 kW, diameter of beam 5.0 mm and scanning speed 5 mm/s.

Small specimens were cut from the samples after cladding (or remelting). The cross-section (perpendicular to the alloying surface) of the alloying layers was mounted and polished metallographically. The specimens were etched in the aqua regia. These etched surfaces were observed in a JSM-35C scanning electron microscope. The compositional variations on the cross-section were measured using a JXA-8800R electron probe micro-analyzer (EPMA). In order to get the accurate compositions, the specimens were not etched for the EPMA analysis.

3 RESULTS

The pool micrograph of laser remelting with the two-layer material system is shown in Fig. 1. A bright zone and a black zone have been seen

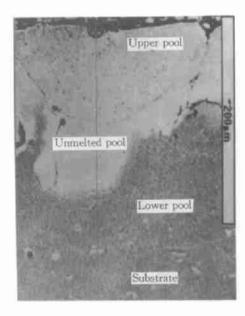


Fig. 1 Micrograph of pool of laser remelting

clearly in Fig. 1. The results of the line analysis along direction of the pool depth indicate that Ti and Ni elements are rich, and Al is poor in the bright zone and that Ti element is not contained, Al is rich and Ni is poor in the black zone (Fig. 2). Therefore, the bright zone is the alloying layer and the black zone the commercial aluminum substrate. It is clearly shown in Fig. 1 that there is a melting pool in the bright zone, and another pool also exists in the substrate, which are separated by unmelted alloy. The phenomenon of the pool separation demonstrates that bond between the alloying coating sprayed

and the substrate is mechanical and not metallurgical. So the bonding force is weak, and the alloying coating is easy to flake off, which is disadvataged to the properties of the alloying coating. At present, the effects of weak convection on the dendrite growth are vastly concerned and studied by many researchers, especially the effects of the weak convection on the morphological stability[6-8]. Concerning the lower pool, it is totally surrounded by the solid wall and the flow of the metal liquid, in which is only driven by the buoyancy, which is comparatively little. So the convection of the metal liquid is weak, and the lower pool offers a place for research of the crystal growth under the condition of the weak tonvection.

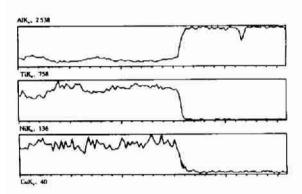


Fig. 2 Alloying element profile along direction of pool depth of laser remelting

Fig. 3 indicates the pool micrograph of laster cladding. The pool is divided into the bright layer and black layer and a distinct interface exists between the two layers. Table 1 show the composition analysis results of the two layers. From the table, it is obvious Cu is rich in the black layer which is easy to etched by aqua regia and Fe in the bright layer which is hard to be etched. If the phenomenon of the pool stratification were observed in the ceramic composite coating (for example ZrO₂ ceramic and Ni ele-

 Table 1
 Composition of stratified pool(%)

 Location
 Cu
 Fe
 Si
 O
 Al
 Ni
 Mn

 Black layer
 87.77
 5.75
 —
 0.4
 4.5
 —
 0.27

 Bright layer
 7.75
 78.10
 7.61
 0.65
 1.17
 4.41
 0.31

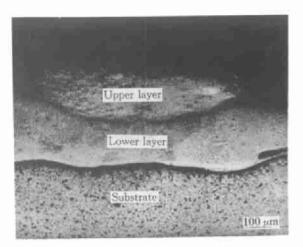


Fig. 3 Micrograph of laser cladding

ment stratification), the properties of the ceramic composite coating would have been extremely deteriorated. However, the stratification of Cu-Cr in situ metal composite coating could improve the strength and conductivity. Therefore, the effects of the pool stratification on the properties of the alloyed coating are decided by the material system.

4 POOL CLASSIFICATION

The previous experimental results showed that the characteristics of the laser pool are different as the material systems (or the processing parameters) are different. It is necessary to develop a classification criterion, which could rationally classify for the laser pool. The separated or unique pool is relative to the temperature distribution along the direction of the pool depth, and is decided by the incident energy, which laser offers. That is to say, it is decided by the processing parameters. Assuming that a unique pool exists and the alloying layer is penetrated, the stratified or mixed pool is decided by both the thermal dynamics and the metallurgical kinetics process. However, it is difficulty to develop a classification model considering the effects of the metallurgical kinetics process, because the process is very complex and the mechanism of mass transfer is issuable [9]. The thermal properties not only decide the thermal dynamics process, but also have an important influence on the metallurgical kinetics process. Therefore, the properties have an important effects on the pool characteristics. The metal liquids are easy to stratify for the positive mixed enthalpy ΔH , which indicates that mix of the metal liquids requires the external energy. On the contrary, the metal liquids are easy to mix and alloy for the negative mixed enthalpy.

Under the prerequisite for giving the materials and the processing parameters, the temperature distribution of the two-layer material system is decided by the following energy equation:

$$\rho \frac{\partial \theta}{\partial t} + \mathbf{V} \cdot \nabla (\rho H) = \nabla \cdot (\frac{k}{c} \nabla \theta)$$

where θ is temperature, t is time, k is conductivity coefficient, H is enthalpy, c is specific heat capacity, ρ is density, V is velocity vector. The material has an important effects on the thermal properties and the temperature distribution, so the thermal properties for numerical calculation are decided by the following method (Fig. 4):

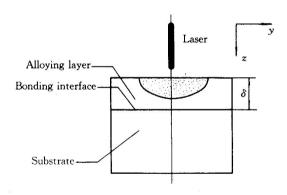


Fig. 4 Schematics of laser pool with a two-layer material system

- (1) Specific heat capacity c, conductivity coefficient k and density ρ are decided by the alloying layer, if z is equal to or less than δ (δ is the thickness of the alloying layer).
- (2) The specific heat capacity c, the conductivity coefficient k and the density ρ are decided by the substrate, if z is larger than δ .

The highest temperature θ_p of the bonding

interface between the substrate and the alloying layer may be gotten by solving the energy equation using the numerical technique. Assuming that the free surface of the alloying layer has been melted, the laser pool of the two-layer material system can be classified four types, according to the highest temperature θ_p of the bonding interface between the alloying layer and the substrate(Fig. 5):

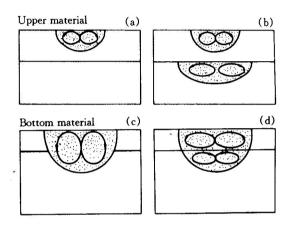


Fig. 5 Classification of laser pool with two-layer material system

- (a)—Unique pool; (b)—Separated pool;
- (c)-Mixed pool; (d)-Stratified pool
- (1) The unique pool: $\theta_{\rm p} < \theta_{\rm m_s} (\theta_{\rm m_s} \text{ is melting temperature of the substrate})$.
- (2) The separated pool: $\theta_{\rm m_a} > \theta_{\rm p} > \theta_{\rm m_s} (\theta_{\rm m_a})$ is melting temperature of the alloying layer), and there are upper and lower pool separeated by the unmelted alloying layer.
- (3) The mixed pool: $\theta_p > \theta_{m_s}$, $\theta_p > \theta_{m_s}$, and the mixed enthalpy ΔH is negative, which illustrates that the metal liquids are easy to mix.
- (4) The stratified pool: $\theta_{\rm p} > \theta_{\rm m_s}$, $\theta_{\rm p} > \theta_{\rm m_a}$, and the mixed enthalpy ΔH is positive, which illustrates that the metal liquids are easy to stratify.

When the thickness of the alloying layer is large yet it is not enough to be penetrated by the incident energy, the substrate is not melted and the only one pool exists in the alloying layer (type of the unique pool seen in Fig. 5 (a)). When melting point of the alloying layer is

rather larger than that of the substrate ($heta_{
m m}$ \gg $\theta_{\rm m}$) and energy aborsorbed by laser pool is not enough to penetrate the alloying layer, there are two separated pools in the alloving layer and the substrate (Fig. 5(b)). The flow phenomena of the metal liquid exist in both pools, but the causes inducing flow of the metal liquid are different. The surface tension gradient is the main driven force for the flow of the metal liquid in the upper pool, but no surface tension gradient exist in the lower pool and the main driven force is the buoyancy, because the lower pool is enclosed by the solid wall. When thickness of the alloying layer is thin and the energy absorbed by laser pool is enough to penetate the alloying layer, if the mixed enthalpy is negative, metal liquids fully mixed and the mixed pool can be seen from Fig. 5(c), which is a typical laser alloying process; if the mixed enthalpy is positive, the liquid in the pool stratified and the stratified pool can be seen from Fig. 3. Driven by the surface tension gradient, upper-layer metal liquid flows, and driven by the bonding interface tension gradient, under-layer metal liquid flows also. The flow of the metal liquids in the pool is the typical two phase flows^[9] and the process of convection and heat transfer in the pool is more complex (Fig. 5(d)).

The different flow characteristics of the metal liquid in the pool cause the different numerical model due to the different types of the laser pool with the two-layer material system. The development of classification criterion of the laser pool with the two-layer material system provides the foundation of the investigation on the metallurgical kinetics process.

5 DISCUSSION

According to the classification criterion of laser pool with the two-layer material system, the results of classification are good agreement with these of the experiments for the laser pools of the previous experiments (Table 2), which demonstrated that the classification criterion is reliable.

For the laser remelting with the two-layer

Table 2 Type of laser pool

Type of pool	θ _p */℃	$\theta_{\mathrm{m_a}}/\mathfrak{C}$	$\theta_{\mathrm{m_s}}/\mathrm{C}$	ΔH	Note
Unique pool	1 246	1 320	654	< 0	Remelting
Stratified pool	1 768	1083	1 536	>0	Cladding
Mixed pool	1657	1 455	654	< 0	Alloying

^{*} Calculated value

material system, the interaction time between laser and the alloying layer is short, the energy absorbed by the pool is small and the alloying layer is not penetrated (the highest temperature of the bonding interface between the substate and the alloying layer was 1246 °C, smaller than the melting temperature 1 320 °C of the alloying layer)[10] due to the higher scanning speed 250 mm/s. When the alloying layer melts, the substrate absorbs the numerous heat expelled from the alloying layer and melts to form a pool, because the melting point of the commercial aluminum (654°C) is much smaller than that of the alloying layer (1 320 $^{\circ}$ C). The separation phenomenon of pool do not exist for the laser remelting with only one material system.

Since conducitivity coefficent of Cu (398W. $m^{-1} \cdot C^{-1}$) is 4 times higher than that of Fe $(80.3 \mathrm{W \cdot m^{-1} \cdot C^{-1}})^{[11]}$, the absorbed heat of the Cu-base alloying layer transfers to the substrate rapidly, resulting in increasing of the substrate temperature, and the substrate is melted. Under the conditions of equilibrium, the melted Cu and Fe should be fully miscible each other according to the equilibrium phase, but the mixed enthalpy ΔH , mixed free energy ΔG and mixed entropy ΔS are positive^[12], and the mix requires external energy, so the mix of melted Fe and Cu is difficult on the view of thermal dynamics. The larger bonding interface tension between the melted Fe and Cu (compared with that of the solid and liquid of Fe or Cu, seen in Table. 3) restrained the mix of melted Fe and Cu. Moreover, the diffusive miscibility between the melted Fe and Cu is limited due to small lifetime of the laser pool (less than 1s), which illustrates that it was difficult to mix the melted Fe and Cu from the view of the kinetics. Therfore, the pool stratified and the clearly separated line between the melted Fe and Cu in the laser

Table 3 Surface tension γ of alloying system^[11]

5233 103000000000000000000000000000000000	Cu-Fe	Cu solid-liquid	Fe solid-liquid	Fe-air	Cu-air
θ/\mathfrak{C}	1 300	1 274	1 831	1 560	1 130
γ /(N·m ⁻¹	0.43	0.177	0.204	1.880	1.268

pool has been observed. The bonding interface tension gradient caused by the temperature gradient between the melted Fe and Cu drives the melted Fe flowing, thus the flow of the metal liquid in the stratified pool is the typical two-phase flow.

6 CONCLUSIONS

- (1) The phenomena of the pool separation and the pool stratification have been observed by the experiments of laser remelting with a twolayer material system and laser cladding.
- (2) The temperature distribution and the material thermal properties are very important factors causing the pool separation and stratification.
- (3) The classification criterion is presented and the laser pool with the two-layer material system is divided into four types: unique pool,

separated pool, mixed pool and stratified pool.

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