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Microstructure and mechanical properties of transition zone in laser additive manufacturing of TC4/AlSi12 bimetal structure

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Abstract

Ti/Al bimetallic structure (BS) has a good development prospect and broader application potential in aerospace engineering. Considering the limitation that dissimilar welding is only applicable to the thin plate, it is necessary to explore a new manufacturing process for Ti/Al BS. In this study, a TC4/AlSi12 BS was prepared by laser additive manufacturing (LAM). TC4 zone, AlSi12 zone and transition zone were formed in the LAM process. Due to the sufficient diffusion reaction, the transition zone with a width of about 0.8 mm was obtained. At the same time, a few micro-cracks were found in the transition zone. The microstructure and phase composition of the transition zone had been emphatically studied. Research results showed that the presence of Si element made the phase composition of the transition zone more complicated. The structure evolution from TC4 to AlSi12 was: \( \alpha - Ti \rightarrow Ti_3Al \rightarrow TiAl+(TiAl+Si) \rightarrow Ti_5Si_3 \rightarrow TiAl_3+(\alpha-Al+Si) \rightarrow \alpha-Al+Si+(\alpha-Al+Si) \). The hardness distribution of BS was uneven, with the highest value reaching 524 HV. The tensile strength of the TC4/AlSi12 BS was about 110Mpa, and the fracture location was located in the transition zone.

1. Introduction

Lightweight is a vital assessment index in the selection of materials in the aerospace field [1–3]. Titanium alloys and aluminum alloys have become the primary structural materials in the aerospace field due to their characteristics of lightweight and high strength [4, 5]. Their composite components combine the respective advantages of the two materials. While meeting the application requirements, it can also achieve lightweight, improve the thrust-to-weight ratio of the aircraft and reduce the cost of structural parts [6, 7], widely application prospect in the field of aerospace. Until now, titanium alloy and aluminum alloy dissimilar metal composite components have been applied in aircraft cabin heat sinks, wing honeycomb sandwiches, Airbus aircraft seat rails, high-speed train carriages, and other structures [8–10]. However, how to realize the effective connection of titanium alloy and aluminum alloy is a current problem and still faces serious challenges [11]. At present, dissimilar welding is the main manufacturing method of Ti-Al alloy joint, but it has more or less limitations [12–16]. For example, fusion-brazing and non-fusion welding can obtain a thinner reaction interface due to lower heat input and limited atomic diffusion distance. But it is generally suitable for thin plates below 1 mm. Although thicker plates can be welded by laser and electron beam, it does not exceed 3 mm. Moreover, Ti-Al intermetallic compounds (IMC) such as \( Ti_3Al, Ti_2Al, TiAl \), are easy to form during the welding process, which leads to increased crack sensitivity of welded joints and becomes the weak link of the structures [17–20].

Therefore, it is necessary to explore a new Ti/Al connection method. LAM technology combines laser multilayer cladding technology with traditional rapid prototyping technology. The powder material is melted by a high-power laser, and the three-dimensional solid parts with complex structures are formed layer by layer under the synchronous control of the computer. Compared with dissimilar welding, LAM technology has special advantages for direct forming and manufacturing of dissimilar materials, especially large-sized structural...
parts. It can be more flexible and changeable to accurately control the deposition process and composition of each layer to achieve complex structural parts without metallurgical defects and high-quality manufacturing. LAM technology can realize the direct deposition manufacture of dissimilar materials, add transition layer connection and gradient transition connection, etc [21], which broadens the connection method of dissimilar materials and provides new possibilities for the connection of dissimilar materials. Up to now, there are many researches on the LAM technology of individual aluminum alloys or individual titanium alloys [22–27], but there are few research reports on the connection of titanium alloys and aluminum alloy dissimilar materials through the LAM technology.

In this study, the LAM technology was used to directly deposit TC4 titanium alloy on the TC4 titanium alloy substrate, and then AlSi12 alloy was deposited on it, which realized the forming connection of the TC4/AlSi12 BS. The microstructure, phase composition, element distribution, hardness, and tensile properties of the transition zone were studied, which provide a new technological method and metallurgical theoretical basis for the manufacturing of Ti/Al BS.

2. Experimental procedures

In this experiment, the hot-rolled TC4 titanium alloy was used as the substrate. Chemical treatment was carried out on the substrate surface before the deposition to remove oxide film and oil stains. The spherical powders of TC4 (mass fraction of Al 6.05%, V 3.81%, balanced Ti) and AlSi12 (mass fraction of Si 12.062%, Fe 0.096%, balanced Al) with a particle size range of 50–120 μm were prepared, as shown in figure 1. Before the LAM experiment, all the powders were vacuum-dried at 100 °C for 2 h to remove the moisture that might be absorbed on the surface of the powder. The TC4/AlSi12 BS was prepared by the LDM8060 system (Raycham, China). At the same time, in order to prevent the generation of pores and ensure the chemical composition and mechanical properties of the sample, the water and oxygen content in the sealed cabin were strictly controlled below 20 ppm.

The metallographic samples including TC4, AlSi12 and reaction region were cut. Keller (HNO3: HCL: HF: H2O = 2.5: 1.5: 1: 95) reagent was used for etching, after grinding and polishing. In order to reveal the internal structure, the scanning electron microscope (SEM SU8010) was used to observe the microstructure of the BS sample. The chemical element composition and distribution were performed by the energy dispersive spectrometer (EDS). The phase composition was identified by an x-ray 7000-type diffraction (XRD) and the scanning angle was 20°–90° at a scanning speed of 4° min⁻¹. The hardness distribution of the sample was tested with a Vickers hardness tester, with the loading force was 9.8 N and the retention time was 10 s. The tensile properties of the sample were tested by an electronic universal testing machine at a tensile rate of 0.2 mm min⁻¹.

3. Results and discussion

3.1. Process parameter optimization and general observation

The manufacturing process of TC4/AlSi12 BS consisted of two steps: the deposition of TC4 on the substrate and the deposition of AlSi12 on the deposited TC4. The deposited TC4 was equivalent to the substrate of AlSi12. The intensity of Ti-Al reaction determined the bonding strength of the BS, not the deposition parameters of TC4. Therefore, this paper focused on the deposition process of AlSi12 on TC4 surface, while TC4 adopted a mature process (as shown in table 1). First, AlSi12 alloy was deposited directly on the TC4 substrate under six groups of different laser powers with other parameters unchanged to simulate the addition process of AlSi12 on the TC4...
Penetration inspection results showed that the cracking was serious at 1800 w and 2000 w, and almost no deposition at 1000 w with a thickness of only 0.1 mm (figure 2). Considering the difference in cumulative heat of single-layer cladding and additive manufacturing of structural parts. Smaller size verification samples were prepared at 1600 w, 1400 w, and 1200 w, as shown in figures 3(a)–(c). Cracks appeared in the Ti/Al deposition layer. Penetration inspection results showed that the cracking was serious at 1800 w and 2000 w, and almost no deposition at 1000 w with a thickness of only 0.1 mm (figure 2). Considering the difference in cumulative heat of single-layer cladding and additive manufacturing of structural parts. Smaller size verification samples were prepared at 1600 w, 1400 w, and 1200 w, as shown in figures 3(a)–(c). Cracks appeared in the Ti/Al

| Number | Laser power (w) | Inspection results |
|--------|-----------------|--------------------|
| 1      | 2000            | ![Image](image1.png) |
| 2      | 1800            | ![Image](image2.png) |
| 3      | 1600            | ![Image](image3.png) |
| 4      | 1400            | ![Image](image4.png) |
| 5      | 1200            | ![Image](image5.png) |
| 6      | 1000            | ![Image](image6.png) |

**Figure 2.** Penetration inspection results of TC4 substrate deposited directly by AlSi12 under different laser powers.

**Figure 3.** (a)–(c) TC4/AlSi12 BS small size verification samples at 1600 w, 1400 w and 1200 w laser power, (d)–(e) Large size schematic diagram and physical specimen.
joint at 1600 W and 1400 W, while no cracks appeared below 1200 W. Therefore, optimized process parameters were determined according to the experiment, as shown in Table 1. Finally, a 40 × 20 × 40 mm TC4/AlSi12 BS sample block was prepared under the optimized parameters, and tensile and metallographic samples were cut from it. The TC4 and AlSi12 were deposited at a height of 20 mm respectively. The schematic and physical specimen are shown in Figures 3(d), (e). From the outside of the specimen, the two parts composed of silver-white aluminum alloy and brown titanium alloy were excessively smooth, and no metallurgical defects were observed.

### 3.2. Microstructure and phase composition

SEM morphology of cross section of the TC4/AlSi12 BS is shown in Figure 4. Three regions were formed in the LAM process, namely TC4 zone, AlSi12 zone and transition zone, respectively. The width of the transition zone was about 0.8 mm, which was thicker than reported in the literature [3, 11, 18].

Figure 5 shows the SEM images and the corresponding EDS mapping of area A-D in Figure 4. It can be seen that Ti element diffused to area C. This indicated that metallurgical bonds have completely formed of two components. XRD was applied to determine the phase composition of the BS with different regions. According to the x-ray diffraction results of Figure 6, we can find that both areas B and C contained TiAl₃, α-Al and Si phase. TiAl₃ grew toward the AlSi12 zone in the form of dendrites, and the primary α-Al grew as equiaxed crystals. In addition to α-Al and TiAl₃, layered (α-Al + Si) eutectic structures and locally precipitated Si were also found in area B. Compared with area B, the size and quantity of α-Al and TiAl₃ in area C were reduced, while the eutectic structure (α-Al + Si) increased. The strongest peak in the XRD diffraction pattern (Figure 6) also changed from TiAl₃ to Al phase. Area D was wholly composed of AlSi12 alloy. The dendritic TiAl₃ and bulk Si phases completely disappeared, resulting in a more uniform eutectic structure.

The two components of TC4 and AlSi12 react directly in the A zone, resulting in the most complex microstructure in the cross section. In this case, further magnified images of area A at different multiples are shown in Figures 7(a)–(c). According to the different microstructures of this area, it can be further divided into five regions I–V (Figure 7(b)). Based on the EDS analysis results in Table 2 and the XRD pattern in Figure 6, the microstructure and phase composition in five regions were analyzed in detail. Region I was TC4 deposition layer, which was mainly composed of lath-like α-Ti according to previous research results [28]. Due to laser action, the deposited TC4 locally melted when the first layer of AlSi12 was deposited. Ti and Al atoms aggregated and reacted at the solid-liquid interface. At this time, Ti atoms were sufficient, and Ti₃Al compound was attached to the surface of the solid TC4 for non-spontaneous nucleation. Finally, Ti₃Al phase transition region with a width of about 3 μm was formed in region II, as shown in Figure 7(c).

Compared to TiAl₃, Si has a lower solubility in Ti₃Al and TiAl [29]. Therefore, Si atoms were continuously pushed toward the solid-liquid interface during the formation of Ti₃Al. As the diffusion continued, TiAl

| Table 1. LAM process parameters for TC4/AlSi12 BS. |
|-----------------|-----------------|-----------------|-----------------|-----------------|
| Material       | Laser power (W) | Laser scan speed (mm min⁻¹) | Powder feed rate (g h⁻¹) | Layer thickness (mm) |
| TC4            | 2400            | 12               | 310             | 0.7             |
| AlSi12         | 1200            | 8                | 170             | 0.4             |
Figure 5. SEM images and the corresponding EDS mapping of area (A)–(D).

Figure 6. XRD pattern results of the BS with different regions.
compounds were formed along the Ti₃Al phase transition region when the content of Ti atoms was equal to that of Al atoms. Si atoms can only exist in the form of eutectic at the grain boundary between TiAl. Finally, a hypereutectic region, with a width of about 10–20 μm, composed of primary TiAl phase and (TiAl+Si) eutectic structure was formed in the region III.

The Study of CHEN et al. shows that Si element is most likely to segregate when the mole fraction of Ti is about 0.5, which can be attributed to the lowest chemical potential, as presented in figure 8[30]. With the formation of TiAl compound, the mole fraction of Ti at this time was exactly about 0.5. When Ti and segregated Si just meet the ratio of 5:3, the strip Ti₅Si₃ phase was formed in region IV (figure 7(a)), and a similar result was reported by Gupta [29]. The DES mapping in figure 5 shows that an Al-poor strip was formed in area A, and the Al-poor strip was rich in Ti and Si. It was consistent with the inference of Ti₅Si₃ compounds.

The formation of Ti₃Al, TiAl and Ti₅Si₃ consumed a large amount of Ti, making Al atoms occupy the absolute content. Accompanied by lower formation free energy [31], a large number of TiAl₃-type phases were formed in the form of columnar crystals, as shown in figure 7(a). Between the columnar crystals was (α-Al+Si) eutectic structure. This was similar to the area-scan results of the columnar crystals rich in Ti and the inter-columnar crystals rich in Al and Si (figure 5). The atomic radius and electronegativity of Al and Si are similar, a small amount of Si atoms will replace a part of Al atoms in TiAl₃ lattice and eventually forming columnar Ti(Al, Si)₃ compound phase [32, 33]. In the LAM process, the cooling rate of the upper part of the molten pool was larger under the combined action of heat conduction and heat radiation. TiAl₃ is an ordered structure with preferential growth orientation along the direction of the maximum cooling rate [34]. Therefore, it grows into liquid AlSi12 alloy in the form of columnar crystals. The formation rate of TiAl₃ is higher than the consumption

| Location | Ti fraction (%) | Al fraction (%) | Si fraction (%) | V fraction (%) | Possible phases   |
|----------|----------------|----------------|----------------|--------------|------------------|
| 1        | 81.81          | 15.14          | 0              | 3.055        | Ti               |
| 2        | 64.32          | 27.93          | 5.63           | 2.13         | Ti₃Al            |
| 3        | 39.44          | 49.47          | 9.27           | 1.82         | TiAl+Si          |
| 4        | 43.50          | 51.26          | 3.41           | 1.84         | TiAl             |
| 5        | 58.44          | 6.07           | 32.52          | 2.96         | Ti₅Si₃           |
| 6        | 24.52          | 62.86          | 11.68          | 0.93         | Ti(Al, Si)₃      |
rate of Ti atoms [34]. When it was far away from TC4 zone, the content of Ti atoms was less, which forms strong internal stress. In addition, the violent flowed of the liquid metal under the action of the laser resulted in fragmentation of columnar TiAl3 into blocks, as shown in figure 5.

From the above analysis, it can be seen during the LAM process of the TC4/AlSi12 BS, the microstructure evolution from TC4 zone transit to AlSi12 zone was $\alpha$-Ti $\rightarrow$ Ti$_3$Al $\rightarrow$ TiAl+(TiAl+Si) $\rightarrow$ Ti$_2$Si$_3$ $\rightarrow$ TiAl$_3$+(α-Al+Si) $\rightarrow$ α-Al+Si+TiAl$_3$+(α-Al+Si) $\rightarrow$ α-Al+Si+(α-Al+Si). Microstructure formation was mainly determined by the diffusion behavior of alloying elements and the free energy of formation of the phase itself. Xu et al [31] confirmed by MEHF model that the formation sequence of intermetallic compounds in Ti-Al alloys above 500 °C is TiAl$_3$, TiAl and Ti$_3$Al. It indicated that elemental diffusion was the main factor in the formation of these compounds in this study.

In addition, microcracks were found locally in the transition zone, as shown in figure 9. Different from the research result of Tian et al [17], the crack expanded in a direction approximately perpendicular to the Ti$_3$Al phase transition region, rather than the direction parallel to the Ti/Al interface layer commonly found in Ti/Al bimetallic joints. The crack was originally located at the intersection of the Ti$_3$Al, TiAl and Ti$_2$Si$_3$ compounds. First, the three compounds are relatively hard and brittle, and there are differences in shrinkage during solidification. Secondly, continuous layer-by-layer deposition was required during the LAM processing, and repeated heating and cooling led to residual stress in the component. Thirdly, the Ti$_2$Si$_3$ compound extended directly to the phase transition region of Ti$_3$Al, and the hypereutectic region (TiAl+(TiAl+Si)) in region III was narrow, making the interfacial energy to prevent the crack growth relatively small. Three reasons together lead to the initiation of the crack and the expansion along the edge of the Ti$_2$Si$_3$ compound, extending to region V.

Figure 8. Influence of Ti mole fraction on Si chemical potential.

Figure 9. SEM of crack morphology in the transition zone: (a) low magnification, (b) high magnification.
3.3. Micro-hardness and tensile properties

The hardness distribution of TC4/AlSi12 BS is shown in figure 10. TC4 zone hardness was about 330 HV. High hardness compounds such as Ti₃Al, TiAl and Ti₅Si₃ were formed in the transition zone due to the metallurgical process.

![Figure 10. Hardness distribution of LAM TC4/AlSi12 BS.](image)

![Figure 11. (a) Schematic diagram of tensile test pieces, (b) stress-strain curve, (c) SEM of fracture morphology, (d) EDS analysis results at points A and B.](image)

3.3. Micro-hardness and tensile properties

The hardness distribution of TC4/AlSi12 BS is shown in figure 10. TC4 zone hardness was about 330 HV. High hardness compounds such as Ti₃Al, TiAl and Ti₅Si₃ were formed in the transition zone due to the metallurgical process.
reaction between Ti/Al/Si. Therefore, the hardness increased to a maximum of about 524 HV at the position where the distance from TC4 zone fusion line was about 50 μm. With the increase of the distance from TC4 zone, the transition zone was mainly composed of α-Al, TiAl₃, and (α-Al+Si) eutectic structure, which reduced its hardness to about 90 HV. The hardness was almost constant when completely transferred to AlSi12 zone, about 62 HV.

The sample of the TC4/AlSi12 BS sample was cut for tensile tests. The size of the tensile test pieces is shown in figure 11(a). Figure 11(b) shows engineering stress-strain curves with an average tensile strength of approximately 106 MPa. It was comparable to the research results obtained by Tomashchuk et al. They used laser welding 2 mm thick aluminum alloy AA5754 to titanium alloy Ti6Al4V [3]. The surface of the tensile fracture had many cleavage steps showing the characteristics of quasi-cleavage fracture (figure 11(c)). The result of the fracture spot scanning analysis (figure 11(d)) shows that the ratio of Ti to (Al+Si) was about 1:3, which was very consistent with the previously reported Ti(Al, Si)₃ phase. And it has the same structure as TiAl₃ except that some of the Al atoms are replaced by Si atoms [33]. The specific fracture location should be located in region V near the Ti₅Si₃ side in the transition zone. Compared with the TC4 and TiAl₃ compounds, the difference in thermal expansion coefficient between TiAl₃ and aluminum alloy is larger, resulting in the TC4/AlSi12 BS being easy to produce stress concentration in TiAl₃. The AlSi12 alloy has good plasticity, so under load, TiAl₃ phase in brittle and lamellar was easy to initiate cracks until fracture failure occurs.

In this study, the reaction between Ti/Al/Si elements was sufficient. This is attributed to the longer diffusion distance and reaction time due to the higher heat input and heat accumulation in LAM process. As a result, a thicker transition zone with local cracks was obtained. The role of Si element made the internal organization of the transition zone more complicated. Therefore, further exploration and research were needed in LAM process to reduce the thickness of the transition zone and obtain Ti/Al composite structure without cracks.

4. Conclusions

In the paper, the feasibility of LAM technology in Ti/Al BS manufacturing was explored, which provides a new possibility for Ti/Al dissimilar material connection manufacturing. The main conclusions were summarized as follows:

1. TC4/AlSi12 BS was prepared under the optimum process parameters of the LAM, in which the macro forming was smooth and good metallurgical bonding was realized.
2. TC4/AlSi12 BS formed a transition zone with a width of about 0.8 mm during the LAM manufacturing process. From TC4 zone to AlSi12 zone, Ti₃Al, TiAl, Ti₅Si₃, Ti(Al, Si)₃ compounds were precipitated successively in the transition zone.
3. The hardness of the TC4/AlSi12 BS transition zone fluctuated greatly, with the maximum value of about 524 Hv and the minimum value of about 90 Hv due to the distribution of different compounds.
4. The tensile strength of the TC4/AlSi12 BS was about 110 Mpa. The fracture surface mainly contained TiAl₃ phase, and the fracture location was located in region V near the Ti₅Si₃ side in the transition zone.

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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

Declaration of competing interest

We declare that we do not have any commercial or associative interest that represents a conflict of interest in connection with the work submitted.
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