Microstructural characteristics of plasma-sprayed MCrAlY/Al2O3-13 wt.%TiO2 coating prepared by induction heating-assisted laser remelting

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Abstract

The plasma-sprayed MCrAlY/Al2O3-13 wt.%TiO2 double-layer coating was treated using conventional laser remelting and induction heating-assisted laser remelting technologies to investigate the influence of laser remelting on the coating’s microstructure. The microstructural and interface characteristics of the different coatings were analyzed via scanning electron microscopy, energy dispersive spectroscopy, and x-ray diffractometry. Results showed that the Al2O3-13 wt.%TiO2 ceramic coating and MCrAlY bond coating of plasma-sprayed samples are typical lamellar structures with high porosity and that the nanostructural ceramic coating consists of fully melting and partially melting zones. A fine equiaxed grain remelting region is formed at the surface of the conventional laser-remelted ceramic coating. However, residual plasma-sprayed state is formed at the bottom of the ceramic coating. The microstructures at the bottom of the ceramic and bond coating are similar to the as-sprayed coating, but the compactness is somehow elevated. After induction heating-assisted laser remelting, the whole ceramic coating and bond coating are remelted, and compact structures are formed. Mechanical bonding occurs at the ceramic coating/bond coating interface and bond coating/substrate interface in the original as-sprayed coating. Through conventional laser remelting, mechanical bonding is the main bonding form on the specimen interface. Meanwhile, a certain degree of metallurgical bonding also exists. After induction heating-assisted laser remelting, the interface is completely transformed into metallurgical bonding. The conventional laser remelting can only strengthen the surface ceramic coating, but induction heating-assisted laser remelting can strengthen the surface ceramic layer and metal bond coating.

1. Introduction

Plasma spraying is a commonly used ceramic coating technology due to its high deposition velocity, high production efficiency, and wide range of application [1–6]. A metal bond coating is usually prepared between the surface ceramic coating and metal substrate to form a double-layer ceramic coating in order to reduce the internal stress caused by the difference in physical properties such as the linear expansion coefficient between the surface ceramic coating and the metal substrate; this process improves the bonding strength and other properties of the coating [7, 8]. However, plasma-sprayed coating will present typical lamellar structure with many pores and insufficient wear resistance [9, 10], thermal shock resistance [11–13], hot corrosion resistance [14–16], and thermal cyclic performance [17]. In addition, the coating and substrate are mechanically bonded, and the strain resistance is poor, thereby restricting its application range [8].

Laser remelting is an effective method to improve the performance of plasma-sprayed ceramic coating. During the laser remelting process, the heat source of the high-energy-density laser moves rapidly, and remelting materials are rapidly set to form uniform and compact microstructures, so as to remarkably improve
the coating’s quality and lengthen its service life [15, 18–32]. The common laser remelting technologies include conventional laser remelting [15, 18–32] and stepwise strengthened laser remelting [33].

In conventional laser remelting, a laser is applied to directly remelt the whole plasma-sprayed double-layer ceramic coating. Yang K et al [34] analyzed the effects of laser remelting on the microstructure and wear resistance of plasma-sprayed Al2O3–40%TiO2 coating. They found that laser remelting can reduce the pores and microcracks and removes the lamellar defect. The microstructure becomes homogenous and compact after laser remelting. Laser remelting can greatly improve the wear resistance of the plasma-sprayed coating. However, as cracks and peeling problems can be easily generated on the remelting coating under large melting depth, this method can only realize remelting strengthening of the surface ceramic coating. The bond coating/substrate interface and bond coating cannot be effectively strengthened and can be damaged during the working process of the whole double-layer ceramic coating [18–20, 34].

As for stepwise strengthened laser remelting, the metal bond coating is first sprayed with plasma, followed by strengthening treatment through laser remelting. After the surface treatment, the surface ceramic coating is sprayed, and the ceramic coating is strengthened via laser remelting [33]. Gao Y et al [33] fabricated a new type of thermal barrier coatings for gas turbine blades by laser remelting on Ni-based super-alloy substrates. They used a laser to prepare as-sprayed NiCoCrAlY bond coating and ZrO2 ceramic coating with double remelting. They found that an Al2O3 layer, which exists between the NiCoCrAlY bond coating and ZrO2 ceramic coating, provides for the adherence between them. Relative to conventional laser remelting, plasma spraying and laser remelting processes in the stepwise strengthening method need several cycles. The bond coating and ceramic coating need to be successively sprayed and remelted, complicating the process. Moreover, the preparation repeatability and stability of the coating quality are not high. Due to the stress superposition effect during secondary laser remelting, the coating can more easily go through cracking and peeling compared with conventional laser remelting. Therefore, the strengthening effect on the coating is superior to that of conventional laser remelting. However, this technology has not been extensively applied.

The laser cladding assisted with an induction heater represents a novel surface treatment technology [35], which combines the laser beam and induction heat source simultaneously. This technology not only can overcome the disadvantage of heat distribution by laser cladding with single heat source, but also has better metallurgical bond state and better processing stability. Moreover, it is easy to prepare free-cracking cladding coating with high performance. Inspired by the above research, in this study, a new induction heating-assisted composite laser remelting process was established, which can compensate for the disadvantages of conventional laser remelting, including energy shortage and unfavorable hot source distribution. It not only has the advantages of conventional laser remelting process (i.e., simple process and good stability) but can also realize synchronous strengthening of surface ceramic coating and metal bond coating. Thus, this process can be used to prepare a high-performance ceramic coating, which can be applied to the surface of critical hot-end parts and components such as aero-turbine engine blade. Conventional laser remelting and induction heating-assisted laser remelting were used to treat plasma-sprayed double-layer MCrAlY/Al2O3–13 wt.%TiO2 coating, and the effects of the two processes on the microstructure of the plasma-sprayed double-layer ceramic coating were compared.

2. Experimental method

2.1. Experimental materials

The substrate material was γ-TiAl-based alloy (TAC-2) smelted by the Research Institute of High-Temperature Materials, Central Iron and Steel Research Institute. The dimensions of the substrate were 100 mm × 100 mm × 8 mm, and its nominal chemical composition was Ti-46.5Al-2.5V-1Cr (atomic fraction, %). MCrAlY powders as bond coating were Y2O3-dispersed NiCoCrAl superalloy powders (KF-113A) produced by the Research Institute of Metal Materials, Beijing General Research Institute of Mining and Metallurgy. The size distribution of the MCrAlY powders ranged from 45 μm to 105 μm, and their nominal composition was Ni–20Co–18Cr–15Al–2Y2O3 (mass fraction, %). Ceramic powders were Nanox S2613P nano-aggregate Al2O3–13 wt.%TiO2 powders produced by the American Inframat Corporation, and their size distribution ranged from 10 μm to 50 μm. The scanning electron microscopy (SEM) morphologies of MCrAlY and nano-aggregate Al2O3–13 wt.%TiO2 powders are shown in figure 1.

2.2. Plasma spraying process

The MCrAlY/Al2O3–13 wt.%TiO2 double-layer coating was prepared using 3710 plasma spraying system (Chemplex Company, USA). The specimen was pretreated by grinding, degreasing, and sand blasting before spraying. The substrate was sand-blasted with 250 μm alumina sand to obtain a rough surface contour so as to strengthen the adherence of the coating. At present, gases such as argon (Ar), helium (He), hydrogen (H2), and nitrogen (N2) are used for the plasma. Ar and He are part of the monatomic gases, whereas H2 and N2 are part of
the diatomic gases. Although the enthalpy of diatomic gases is considerably higher compared with monatomic gases, the latter are preferred due to their easier transfer to plasma state, more stable electric arc, and lower working voltage. Moreover, the plasma temperature of monatomic gases is higher than that of diatomic gases. Thus, monatomic gases can be used for plasma spraying of high-melting-point ceramic materials. In this experiment, a primary Ar mixture with secondary He was used to increase the kinetic energy, velocity, and stability of the plasma beam. In addition, Ar was used as the powder carrier gas. In order to investigate the influence of process parameters of nanostructured Al$_2$O$_3$-13 wt.%TiO$_2$ coating during plasma spraying, an orthogonal array method was designed, and a series of fabrication experiments was conducted to identify the optimum process parameters [36]. Four main process parameters (i.e., spraying distance, spraying electric current, primary gas pressure, and secondary gas pressure) controlling the quality of coatings were optimized to produce a maximum bonding strength. Orthogonal design of plasma spraying process parameters of Al$_2$O$_3$-13 wt.%TiO$_2$ is listed in table 1. The plasma spraying process parameters, optimized through preliminary orthogonal experiments [36], are listed in table 2.

### 2.3. Laser remelting process

Laser remelting was implemented via the SLCF-X12 × 25 CO$_2$ laser machine. The schematic of induction heating-assisted laser remelting is shown in figure 2. As shown in the figure, the induction heating coil was fixed on the laser head. While the double-layer ceramic coating specimen was placed under laser remelting, the current was conducted through the induction heating coil using a high-frequency induction heater together with the laser head.
with induction heating to realize the combination of the laser and induction heat sources. After focusing, the laser beam acted upon the middle of the induction heating region of the specimen. The output powers of the laser and high-frequency induction heater were regulated by a computer numerical control (CNC) console. Meanwhile, the laser scanning speed was realized by the CNC console by controlling the laser head. Thus, the continuous multi-channel induction heating-assisted laser remelting of the specimen was realized.

Many parameters affect the quality of the laser-remelted coating, including laser power, spot size, defocus, laser beam mode, laser scanning speed, coating thickness, and amount of overlap. To reduce the cracking degree of the coating during laser remelting, a relatively low laser energy density was used. During laser remelting, the temperature distribution isoline on the cross-section shows a crescent shape as a result of heat conduction in the matrix [37]. To maintain the remelting thickness of the coating in the outer area of spot, 10%–30% of overlap is usually used. For induction heating process, the main parameters include power and frequency of the high-frequency induction heater, induction heating distance, and zone. The process parameters of the conventional laser remelting, optimized through preliminary experiments, were as follows: laser power, 500 W; size of rectangular spot, 5 × 3 mm; laser scanning direction, along the 3 mm side of the light spot; scanning speed, 700 mm·min\(^{-1}\); and amount of overlap, 20%. The laser processing parameters of induction heating-assisted laser remelting were similar to those of the conventional laser remelting, and the induction heating parameters were as follows: power and frequency of the high-frequency induction heater, 10 kW and 30 kHz, respectively; distance from the specimen surface to the induction coil, 5 mm; and size of the induction heating zone, 30 × 20 mm.

2.4. Characterization and analysis
The microstructures of the specimen after being subjected to plasma spraying, conventional laser remelting, and induction heating-assisted laser remelting were analyzed using a JSM-7100F (JEOL, Japan) field emission scanning electron microscope (FESEM) with an EDS (Oxford, UK) energy dispersive spectroscope (EDS) and a D/max2500 (Rigaku, Japan) x-ray diffractometer (XRD).

3. Results and discussion
3.1. Microstructure of plasma-sprayed coating
Figure 3 shows the cross-sectional SEM morphologies of the as-sprayed MCrAlY/Al\(_2\)O\(_3\)-13 wt.%TiO\(_2\) coating. As can be observed from figures 3(b) and (c), the coating presented typical lamellar structural characteristics of plasma spraying. A considerable proportion of pores existed at the ceramic coating and bond coating. Moreover, the ceramic coating consisted of two parts, namely, nanoparticle fully melting region and partially melting region, and presented a two-phase structure as previously described in many literature [38–40]. The pore formation was mainly caused by the mutual overlapping and stacking of sprayed particles, volume shrinkage of molten particles, and precipitation of gases in molten particles after the coating was cooled to room temperature. However, as the melting of metal powder particles was more sufficient during the plasma spraying process, the molten powder particles experienced deformation after contacting the substrate with good tiling property.
Therefore, compared with the ceramic coating, the bond coating was more uniform and compact and had fewer pores. The microstructure of the plasma-sprayed nanostructure $\text{Al}_2\text{O}_3-13$ wt.%$\text{TiO}_2$ ceramic coating and its formation mechanism are described in detail in our previous study [41].

### 3.2. Microstructure of the conventional laser-remelted coating

Figure 4 displays the cross-sectional SEM morphologies of the conventional laser-remelted $\text{MCrAlY}/\text{Al}_2\text{O}_3-13$ wt.%$\text{TiO}_2$ coating. Under the comprehensive actions of the laser power, energy density, temperature field distribution in the laser action zone, heat conductivity coefficient of the ceramic material, and coating thickness, the remelted ceramic coating presented lamellar characteristics and could be largely divided into the surface remelted zone and bottom residual plasma-sprayed zone according to the different organizational forms. During the laser remelting process, the surface $\text{Al}_2\text{O}_3-13$ wt.%$\text{TiO}_2$ ceramic coating is remelted and recrystallized. Compact and fine equiaxed grains were formed in the surface remelted zone as shown in figure 4(b), which were different from the conventional columnar crystal organization formed and grew along the direction of the heat flow [20, 42, 43]. The main reason behind the formation of the fine equiaxed grains was that the partially melted zone (residual nanoparticles) in the original plasma-sprayed coating significantly influenced the remelted ceramic coating structure [44]. As the dispersed nanoparticles repressed the grain growth in the remelted zone and slowed down the abnormal growth of the grains during the laser remelting process, microstructures with uniform and fine grains were formed; meanwhile, the quantity of internal defects of the grains was also reduced [45, 46].

The laser remelting temperature did not reach the melting point of the material at the bottom of the ceramic coating and at the bond coating. Thus, lamellar structures similar to the original plasma-sprayed coating were present at the bottom of the ceramic coating (figure 4(c)) and at the bond coating (figure 4(d)). However, as a certain sintering effect was reached due to the high-temperature action of laser remelting, their compactness was evidently improved. The influence of laser remelting on the plasma-sprayed nanostructure $\text{Al}_2\text{O}_3-13$ wt.%$\text{TiO}_2$ ceramic coating has been discussed in our previous study [44]. Thus, it will not be further considered in the present study.
3.3. Microstructure of induction heating-assisted laser-remelted coating

The cross-sectional SEM morphologies of the induction heating-assisted laser-remelted MCrAlY/Al$_2$O$_3$-13 wt.%TiO$_2$ coating are shown in figure 5. Through induction heating-assisted laser remelting, fine equiaxed grains (figure 5(b)), similar to those formed through conventional laser remelting, were formed on the surface of the ceramic coating, and the bottom of the coating was completely remelted (figure 5(c)). In addition, the bond coating was remelted, and completely compact structures were formed (figure 5(d)). This is mainly because (1) the induction heating-assisted laser remelting composite process could make up for the defect of the conventional laser remelting (i.e., energy shortage). Thus, the ceramic coating and bond coating in the double-layer coating could be simultaneously remelted through laser remelting just once. (2) In the induction heating-assisted laser remelting composite process, the laser beam acted in the middle of the induction heating zone, thereby making up for the unfavorable laser heat source distribution and reducing the temperature gradient between the remelted zone and the surrounding zone. Furthermore, the large induction heating zone exerted preheating and slow cooling effects on the molten pool, thereby effectively mitigating the cracking trend of the remelted coating. Therefore, relative to the conventional laser remelting, a larger melting depth was allowed in the induction heating-assisted laser remelting [47–49].

3.4. Phase composition

The XRD spectra of the original powder, the as-sprayed coating, the conventional laser-remelted coating, and the induction heating-assisted laser-remelted coating are shown in figure 6. The Al$_2$O$_3$-13 wt.%TiO$_2$ powder is composed of metastable $\gamma$-Al$_2$O$_3$ phase, $\theta$-Al$_2$O$_3$, rutile-TiO$_2$, and Al$_2$Ti$_3$O$_15$, and the primary phase is stable $\alpha$-Al$_2$O$_3$. The as-sprayed coating resulting from the prominent $\gamma$-Al$_2$O$_3$ is indexed, and some $\alpha$-Al$_2$O$_3$ and brookite-TiO$_2$ are also present. After plasma spraying, all $\theta$-Al$_2$O$_3$ and some $\alpha$-Al$_2$O$_3$ transferred into $\gamma$-Al$_2$O$_3$. This is mainly due to the rapid solidification of the liquid droplets during the plasma spraying process [50, 51]. Moreover, rutile-TiO$_2$ changed to brookite-TiO$_2$ after plasma spraying. The XRD pattern of the conventional laser-remelted coating is shown in figure 6(c), which proves that the metastable $\gamma$-Al$_2$O$_3$ changed to stable $\alpha$-Al$_2$O$_3$ due to the recrystallization of the plasma-sprayed surface ceramic coating during laser remelting [20]. In addition, most brookite-TiO$_2$ reacts with Al$_2$O$_3$ and forms Al$_2$Ti$_3$O$_15$ solid solution. There is a small amount of Ti$_6$O$_7$ and Ti$_2$O$_3$ generated from the oxygen reduction of TiO$_2$, and the rest of brookite-TiO$_2$ is converted into rutile-TiO$_2$ again. Similar to conventional laser remelting, $\gamma$-Al$_2$O$_3$ of the induction heating-assisted laser-remelted coating is converted to $\alpha$-Al$_2$O$_3$. However, TiO$_2$...
fully reacts with Al₂O₃ to form Al₂TiO₅ solid solution because of the larger size and longer life of pool during the induction heating-assisted laser remelting.

### 3.5. Discussion of coating interface bonding

The bonding formed at the coating interface is mainly divided into mechanical bonding and metallurgical bonding [8, 52]. For a double-layer MCrAlY/Al₂O₃-13 wt.%TiO₂ coating, the mechanical bonding mainly means bonding between the ceramic coating and the bond coating and between the bond coating and the substrate through mechanical riveting force. Moreover, the bonding strength of the coating is relatively low. A remarkable element counter-diffusion takes place between the ceramic coating and the bond coating and between the bond coating and the substrate under metallurgical bonding, so the bonding strength is improved by a large margin.

The line scanning results of specimen elements showed that there were mainly Al, O, and Ti elements in the ceramic coating; Ni and Co elements in the bond coating; and Al and Ti elements in the substrate. Almost no interdiffusion occurred at the original plasma-sprayed coating, ceramic coating/bond coating interface, or bond coating/substrate interface, indicating that the interfaces presented typical mechanical bonding. Through conventional laser remelting, element interdiffusion existed at the ceramic coating/bond coating interface and bond coating/substrate interface to a certain degree, suggesting that the mechanical bonding was the main interface bonding form, along with metallurgical bonding to a certain extent. Under induction heating-assisted laser remelting, the interface was already transformed from physical bonding of the original plasma-sprayed state into chemical bonding, being typical metallurgical bonding, as shown in figure 7. In addition, an enrichment phenomenon of Al element can be observed in the MCrAlY/Al₂O₃-13 wt.%TiO₂ interface. This is attributed to the melting of ceramic coating and bond coating during the induction heating-assisted laser remelting process. Thus, Al, which is less dense in the MCrAlY coating, floats to the top of the bond coating (interface of ceramic coating and bond coating). Al is redistributed in the bond coating and forms an Al enrichment region in the surface of the bond coating, which is similar to the result of [33].

The interface of the plasma-sprayed coating was under typical mechanical bonding. Sand blasting and roughing treatments were performed for the substrate before spraying so that the substrate presented microscopic unevenness. When passing through the plasma arc, the MCrAlY alloy powders at the bond coating...
were heated and melted and impacted the substrate surface at high speed. The liquid-stage metal was spread on the uneven surface of high-temperature alloy and formed favorable ‘anchor hooking effect’ with the substrate, which improved the bonding strength between the MCrAlY bond coating and the substrate.

The conventional laser remelting did not melt the bottom of the ceramic coating or bond coating. However, the high-temperature action of the laser remelting process still reached a certain sintering effect, which promoted the element interdiffusion between the bond coating and the ceramic coating and between the substrate and the bond coating to a certain extent. Although complete metallurgical bonding could not be

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**Figure 6.** XRD analysis results: (a) the original Al₂O₃-13 wt.%TiO₂ powder, (b) the as-sprayed coating, (c) the conventional laser-remelted coating, and (d) the induction heating-assisted laser-remelted coating.

**Figure 7.** EDS line scanning result of the induction heating-assisted laser remelted MCrAlY/Al₂O₃-13 wt.%TiO₂ interface.
realized, it did contribute to the improvement of interface bonding strength. Thus, an interface mainly presenting mechanical bonding with a certain degree of metallurgical bonding was formed.

However, the induction heating-assisted laser remelting composite process could simultaneously melt the ceramic bonding and bond coating in the double-layer coating structure. Thus, the ceramic coating/bond coating interface and bond coating/substrate interface realized metallurgical bonding simultaneously. Based on these findings, this process not only overcomes the deficiency of the traditional laser remelting, which can only strengthen the surface ceramic coating, but also avoids the problem of stepwise strengthening process, which is too complicated. In addition, the remelted coating will not be easily cracked. Thus, it can be applied to the surface of some critical hot-end parts and components as a very promising new laser remelting process.

4. Conclusions

The effects of conventional laser remelting and induction heating-assisted laser remelting on plasma-sprayed MCrAlY/Al2O3-13 wt.%TiO2 double-layer coating were comparatively analyzed in this study.

(1) The plasma-sprayed Al2O3-13 wt.%TiO2 ceramic coating and MCrAlY bond coating have typical lamellar structures and high porosity. Moreover, the nanostructure ceramic coating is composed of fully melted region and partially melted region. Fine equiaxed grains are generated at the upper part of the conventional laser-remelted ceramic coating. There is a residual plasma-sprayed zone at the bottom of the ceramic coating. The microstructures at the bottom of the ceramic coating and those at the bond coating still remain similar to the original plasma-sprayed coating, but only the compactness is improved. The whole ceramic coating is remelted due to higher energy input by the induction heating-assisted laser remelting. The bond coating is also remelted, and compact structures are formed.

(2) The metastable γ- Al2O3 phase in the plasma-sprayed coating is completely transformed into stable α-Al2O3 phase after laser remelting.

(3) The ceramic coating/bond coating interface and bond coating/substrate interface in the original plasma-sprayed double-layer coating present mechanical bonding. Through conventional laser remelting, mechanical bonding is mainly manifested at the specimen interface, together with certain metallurgical bonding. However, the interface of the induction heating-assisted laser-remelted coating is completely transformed into metallurgical bonding.

(4) For induction heating-assisted laser remelting, an enrichment phenomenon of Al element exists in the MCrAlY/Al2O3-13 wt.%TiO2 interface. Because Al is less dense in the MCrAlY bond coating, it is remelted and floats to the top of the interface of the ceramic coating and bond coating.

(5) The conventional laser remelting can only strengthen the surface ceramic coating, but the induction heating-assisted laser remelting is capable of further strengthening the surface of the ceramic layer and metal bond coating simultaneously. On this basis, the induction heating-assisted laser remelting is a very promising new laser remelting process.

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