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High-temperature protective coatings on superalloys①

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[Abstract] Protective coatings are essential for superalloys to serve as blades of gas turbines at high temperatures, and they primarily include aluminide coating, MCrAlY overlay coating, thermal barrier coating and microcrystalline coating. In this paper, all these high-temperature coatings are reviewed as well as their preparing techniques. Based on the most application and the main failure way, the importance is then presented for further deepgoing study on the high-temperature oxidation law of aluminide coatings.

[Key words] high-temperature coating; casting; superalloy; oxidation
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1 INTRODUCTION

Superalloys and high-temperature protective coatings are playing the essential and important role in the fields of modern energy-source industries and aviation-astronautics industries. Nowadays, they have been broadly applied to many areas such as various gas turbines as well as different aero-spacecrafts, rocket engines, nuclear reactors, submarines, heat power plants, petroleum chemical-industry devices. Among these uses, those served as thermal components of aviation-astronautics are particularly interesting.

When they are running, thermal components for the above uses will be bound to encounter the serious problem of high-temperature oxidation and heat corrosion (as shown in Fig. 1)[1,2]. In order to ensure a certain using life of these components, they have to simultaneously meet the enough mechanical strength and the good resistance to both high-temperature oxidation and heat corrosion. The general design rule is thus making the thermal components from superalloys with enough permanent strength, and developing the high-temperature protective coating on these components.

To make the thermal components work well, the importance of protecting the superalloys from the catastrophic oxidation and corrosion in corrosive atmospheres is identical to obtain and utilize their high strength. While, gas turbines are developing with an obvious feature of raising the entry temperature of burning gas as high as possible to increase the efficiency and the power, and of using the cheap fuel to decrease the oil-exhausting cost. These bring the increased protruding high-temperature oxidation and corrosion for the thermal component of engines, then high-temperature protective coatings are more and more taken the matter seriously.

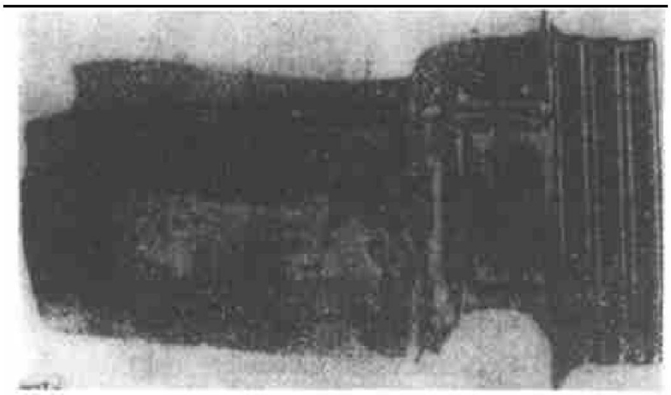

Fig. 1 Surface morphology of a blade of gas turbine after high-temperature corrosion[1]

2 HIGH-TEMPERATURE PROTECTIVE COATINGS

2.1 Aluminide coating

Aluminide coating includes two sorts of simple aluminide coating and modified aluminide coating. The former forms on superalloys by deposition of single element of Al, and it has the excellent resistance to oxidation, but not to heat corrosion[3]. Adding a little Cr, Si, Ti and Pt into the simple aluminide coating, the resistance of the coating to heat corrosion can be then obviously increased, so it is called the modified aluminide coating.

2.1.1 Cr-modified aluminide coating

Adding Cr into the coating, not only the resis-

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tance to oxidation and heat-corrosion but also the stability will be increased[4~6]. Al-Cr coating has the structure similar to that of simple aluminide coating. This coating consists mainly of NiAl for Ni-base alloys and of CoAl for Co-base alloys.

2.1.2 Si-modified aluminide coating

During the deposition of Al, adding some Si or Si alloy powder into the aluminisor, a little of Si can permeate into the coating. The research results show that, just the right content of Si will obviously improve the resistance to oxidation and heat-corrosion for the aluminide coating[7~9]. But the Si content in the coating should not be too high, otherwise the harmful phase of low melting-point will form between Si and Ni in the substrate at high-temperatures, meanwhile the coating turns brittle and easily peels off during oxidation[10].

2.1.3 Ti-modified aluminide coating

Al-Ti coating is a commercial aluminide coating developed by American General Electric Company (G. E. Co.)[11,12]. As for the advantages and disadvantages of using Ti to modify the aluminide coating, there are quite many divergences. For example, some patents[11,12] suggest that, the addition of Ti during the deposition of Al would benefit the oxidation performance of the coating. Besides, Al-Ti coating gains an advantage of the heat-corrosion resistance over the simple aluminide coating. Nevertheless, some other references[13~19] show that the addition of Ti into the coating would always be unfavourable to oxidation.

2.1.4 Pt modified aluminide coating

It is a tremendous breakthrough in the study on aluminide coatings for adding Pt into the coating to form Pt-modified aluminide coating[20]. About the mechanism that Pt increases the resistance of the aluminide coating to high-temperature oxidation and heat corrosion, the primary viewpoints are presented as follows. 1) Pt increases the bond strength of the α-Al_2O_3 scale[21]; 2) Pt increases the spontaneous coalescence ability of the α-Al_2O_3 scale[22]; 3) Pt increases the structural stability of the aluminide coating, and decreases the inter-diffusion between the coating and the substrate[23].

Compared with the simple aluminide coating, Pt-modified aluminide coating gets the more resistance to high-temperature oxidation and heat-corrosion for superalloys. The effect of Pt on the coating has been proved to be mainly improving the protectivity of α-Al_2O_3 scale, but the mechanism of this effect has not yet been thoroughly cleared up[24].

Because Pt is expensive, the preparation of Pt-Al coating costs very high. In the recent years, cheaper Pd has been tried to take the place of Pt and Pd-Al coating[25,26] arrives at a good result.

2.2 Overlay coating

Overlay coating is also called alloy coating, namely MCrAlY coating. It had been developed before the occurrence of Pt-Al coating. In MCrAlY coating, M is Fe, Co or Ni; Al is used to form protective Al_2O_3 scale; Cr is used to promote the formation of oxide scale and improve the ability of resisting heat corrosion; and Y is used to increase the adhesion power of oxide scale.

This coating has good performance of high-temperature oxidation and heat corrosion, as well as very good plasticity. There is only a little reaction between coating and substrate, but there is a large selectivity of the coating composition, and a wide range of the coating thickness[27]. The suitable composition can be selected based on various working environments and different substrate materials.

Adding Al into this coating, the oxidation resistance can be increased. However, if the Al content in the coating is too high, the plasticity of the coating will then be decreased. When a little of active element Y is added into the coating, the oxidation performance of the coating is obviously improved[28~30]. But active elements such as Y have the relatively low solubility in alloys, and they will locally gather when their contents beyond the limit of the solid solubility, resulting in the decrease of the resistance to oxidation and to heat-corrosion. Usually, the Y content in MCrAlY coating is appropriate below 1% (mass fraction).

2.3 Thermal barrier coating (TBC)

Thermal barrier coating is also called the composite coating. It was ceramics with low thermal conductivity that blocks the transfer of heat from the surface into the substrate, and lowers the substrate temperature, then prolongs the serving period of workpieces with this coating. ZrO_2 has the relatively low thermal-conductivity among all ceramics, and is more suitable for preparing the thermal barrier coating. Adding some amount of CaO, CeO_2, MgO or Y_2O_3 into ZrO_2, the cubic system of crystal ZrO_2 can be stabilized within a very wide range of temperatures.

Thermal barrier coating often consists of two layers. The outer is the ceramic layer of ZrO_2 stabilized by Y_2O_3, and the inner is the combining layer of MCrAlY. The ZrO_2 layer mainly plays a role of heat-insulation, and the MCrAlY layer is used to relax the unmatch of thermal expansion coefficients and elastic modulus between the ceramics and the substrate, and to increase the bond strength of coating/substrate.

The interface of this coating is vulnerable, and the coating always fails from peeling off[31~36]. The TBC with gradient structure, which was developed in recent years, can reduce the scaling of coating caused by the unmatch of thermal expansion coefficients between metals and ceramicses[37~41]. The typical structure of TBC is shown in Fig. 2[42].

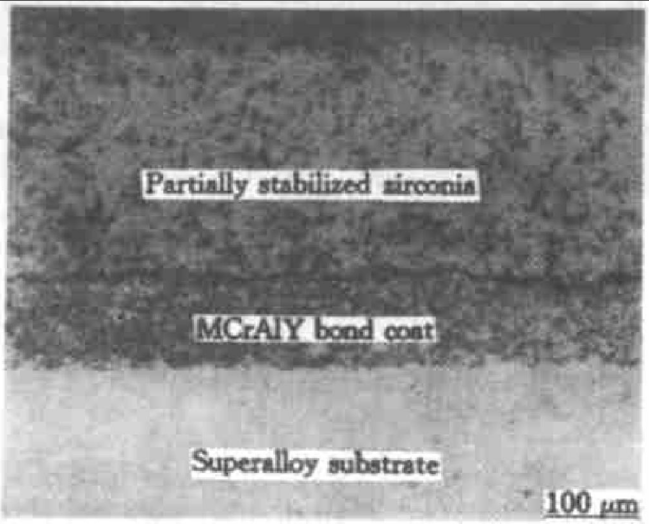

Fig. 2 Cross-sectional view of typical thermal barrier coating[42]

2.4 Microcrystalline coating

Microcrystalline coating may belong to a fully new type of high-temperature coatings (as shown in Fig. 3), which was developed in 1990s[41]. By the magnetic-control sputtering technology, the coating can be obtained with the same composition as that of the substrate alloy. This brings the coating the good compatibility with the substrate and the high bond strength, without the inner-diffusion. The oxidation behaviour of this coating is obviously superior to the substrate alloy.

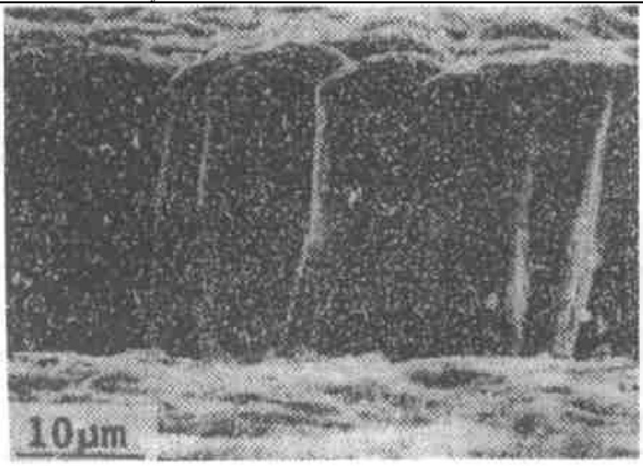

Fig. 3 Sectional SEM of sputtering microcrystalline coating on K38G alloy[41]

2.5 Comments

At present, there are many different sorts of high-temperature coatings. Although the new systems of coatings are broadly developed domestically and abroad, aluminide coatings still occupy the dominant position in the applied market all over the world. The reason is that this coating not only can meet the general property demands, but also has the features of low cost and stable performance.

The protective effect of coatings depends upon the ability of their forming the protective scale of alumina. Thus there should be enough Al content in the coating with suitable thickness. Giving consideration to both oxidation resistance and mechanical properties, Al content will be generally controlled at about 30% (mass fraction), and the coating thickness will in the range of 30~ 35 μm[42, 43].

3 PRODUCING TECHNIQUES OF HIGH-TEMPERATURE COATINGS

The main techniques for high-temperature protective coatings may be roughly divided into five kinds[1, 2, 42~ 44]: thermal permeation, physical technique, hot spraying, melting-condensation and sintering, and electrochemistry.

3.1 Thermal permeation

Thermal permeation technique includes pack cementation, gas-phase cementation (e.g. CVD), brei cementation and melting cementation.

1) Pack cementation. This method is one of the simplest technologies that can be used to manufacture aluminide coatings. Pack the workpiece into the cementation powder, air inert gas or reductive gas, heat up to some temperature and heat-shield for a period of time, then the coating with some thickness will form on the workpiece. The cementation agent is made form Al-containing metal powder and activating agent (with modifying material) which are in the light of certain proportion.

2) Gas-phase cementation. This method has a feature of no contacting between the workpiece and the cementation powder. The gas phase resulting from the reaction of cementation agents carries out both the chemical deposition and reactive diffusion on the surface of alloys at high temperatures, and then the diffusion coating forms.

3) Brei cementation. Some proportion of the permeation source, activating agent, bonding agent and solvent are mixed to prepare the suspension solution of brei. Coat it on the surface of workpieces, place the brei-coated workpiece in a vacuum device, air the protective gas under the vacuum state, then run the high-temperature diffusion, and the coating will form on the surface.

4) Melting cementation. This method is to coat the workpieces with the metal to permeate without filling and activating agents, which is then heated to melt and diffuse into the substrate alloy to form the coating.

3.2 Physical technique

This technique includes electron-beam physical vapour deposition (EB-PVD)[45~ 48] and sputtering deposition[49]. Using EB-PVD, the workpiece needs to be heated up to 800~ 1 100 ℃ before forming the coating, so that the inter-diffusion could take place between the obtained coating and the substrate to increase the bond strength of the coating.

When the coating material is high melting-point metals or complex alloys, using the sputtering method to produce the coating has its obvious advantage. During the deposition of complex alloys, the composition of the coating is easy to control, and it does not obviously differ from the composition of the target materials.

3.3 Hot spraying

The heat-resistant coating made by ordinary-pressure plasma spraying has high porosity, low bond strength, much oxide inclusion and bad corrosion resistance, so it can not be used as the high-temperature corrosion resistant coating of blades of gas turbines. Low-pressure plasma spraying (LPPS) is carried out in the vacuum with a little inert gas, by the effective heating method, metal particles finish melting and rapidly spray to the surface of the workpiece. Then the resulting coating has its compactness greatly increased and the oxide inclusion obviously decreased, which remarkably improves both the high-temperature corrosion resistance and the mechanical properties of the coating[46].

3.4 Melting-condensation and sintering

These techniques are usually used to prepare overlay coatings. Based on the composition of alloy coatings, the melting-condensation method is to make up the brei from various elemental powders in the demanded proportion to coat the workpiece, which is then treated at some temperature to form the coating by melting and condensation of the brei. Because of the limitation of the temperature that the substrate alloy can stand, this method make the composition of the coating very limited.

As a contrast, the sintering method may prepare the overlay coating at lower temperatures, so the composition of the coating is adjustable within a wider range. But the resulting coating is not compact, and need to be treated with thermal isostatic pressing.

3.5 Electrochemistry

This technique includes electroplating and electrophoresis. Together with other techniques, it can produce the modified aluminide coating or the alloy coating.

3.6 Technical application

The thermal cementation is the most and main method used to manufacture high-temperature corrosion resist coatings. Of this, the pack cementation is a traditional method which is the earliest and the commonest. Now this method still broadly comes into practice for intermetallic compound coatings.

As for aluminide coatings, both thermal cementation and physical technique are available, but the main coating technique is pack cementation, brei cementation, vacuum evaporating-plating diffusion or chemical vapor deposition. While both overlay coatings and thermal barrier coatings are produced mainly by vacuum electron-beam evaporating deposition (EB-PVD) or low-pressure plasma spraying (LPPS)[1,50], assisting with electroplating or cathode sputtering.

Owing to the so called "line-of-sight" (as shown in Fig. 4), the physical and hot-spraying techniques are very difficult to form the well-distributed coating on workpieces with complex geometrical shape. In addition, these techniques have a great effect on the geometrical size of workpieces. Under this situation, gas-phase cementation is particularly favorable.

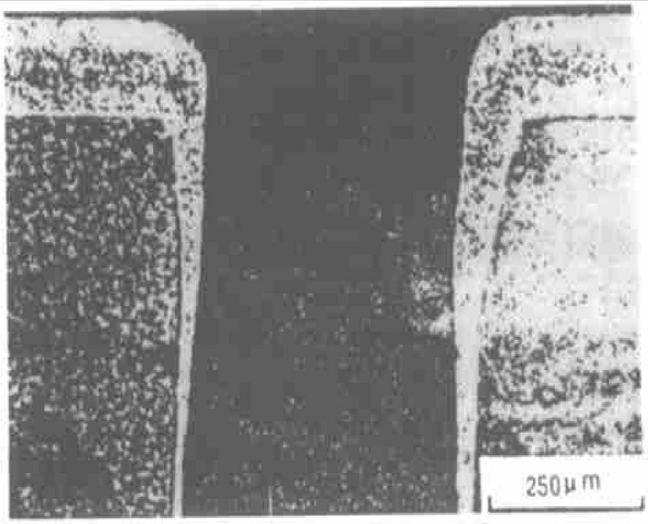

Fig. 4 Cross-section of cooling hole coated by EB-PVD suffering "line-of-sight"[50]

4 FAILURE OF THERMAL-COMPONENTS OF SUPERALLOYS AND HIGH TEMPERATURE OXIDATION

4.1 Concept of high temperature oxidation

High temperature oxidation means in narrow sense that the material reacts with oxygen to form the oxide at high temperatures, and in broad sense that the atoms, atomic groups or ions composed of the material lose their electrons[51]. Generally speaking, it refers to that of narrow-sense.

4.2 Failure of high-temperature components

4.2.1 Failure reasons

The factors, on which the serving period of thermal components of superalloys for gas turbines depends, mainly include four aspects as follows. 1) high-temperature corrosion caused by the medium of burning gas, including high-temperature oxidation and heat corrosion; 2) creep deformation caused by high-temperature permanent stresses; 3) mechanical fatigue caused by vibrating loads; 4) thermal fatigue caused by thermal periodic alternations.

4.2.2 Failure ways

As to gas turbines used for aviation, mechanical fatigue is often the main failure way of components when working temperature of turbines is below 800 ℃. Creep deformation, oxidation or thermal fatigue always results in the failure of components when the working temperature is above 1 000 ℃, and any above factor may become the main failure way at 800 ~ 1 000 ℃.

It can be seen that, high-temperature oxidation is the basic and significant failure way of thermal components within a wide range of the working temperature of gas turbines. While the metal temperature goes over 950~ 1 000 ℃ or the used fuel and the entering air are both relatively "clean" (without the pollution of S or/and V), high-temperature oxidation turns up to be the main way of the thermal component failure of aviation engines[1]. Especially with the gradual increase of the working temperature of modern aviation engines, high-temperature oxidation increasingly plays a key role in the failure of thermal components.

Usually, oxidation is very prominent for plane engines, and heat corrosion still exists but not serious relatively.

5 SUMMARY

Superalloys and high-temperature coatings develop closely linked with the power industry, especially with the aviation industry[42]. Some new metal-base high-temperature materials, such as ordered intermetallic compounds of Ni_3Al, NiAl, Ti_3Al and TiAl, have been developed in recent years, but all of them have some unsatisfying properties, which restrain them to come into practice or restrict their application[52]. Thereby, Fe-base, Ni-base or Co-base superalloys have been still mainly using as metallic high-temperature materials.

As the modern power device of various gas turbines and especially of different aviation engines, high-temperature oxidation of their thermal components is always their basic and main failure way. High-temperature coatings manifest an effective measure to resist it, of them the aluminide coating occupies the fundamental position used the most widely in this field.

The oxidation behaviors have been deepgoingly and systemically studied in all their aspects for metals or alloys. However, comprehensive quantization studies of oxidation laws have not yet been found for the aluminide coating, which is a special metallic material adhering to the superalloy, besides the general kinetic curves. It is well known that, isothermal oxidation performance, namely the static oxidation in air, is a most fundamental and important property for a high-temperature coating. Thus, the deepgoing study of quantization analysis is practically meaningful on the high-temperature oxidation law for the above aluminide coating.

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