Effect of Bond Coat Materials on Thermal Fatigue Failure of EB-PVD Thermal Barrier Coatings*

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Abstract

Effect of MCrAlY bond coat alloy systems on thermal fatigue failure of thermal barrier coatings (TBCs) was investigated, where the TBC specimen consisted of Ni-based superalloy IN738LC substrate, bond coat, and 8 wt.% Y₂O₃-stabilized ZrO₂ (YSZ) top coat. The top coat was fabricated by EB-PVD method with 250 µm in thickness. Three kinds of MCrAlY alloys were studied as the bond coat material. Employing the originally developed test equipment, thermal fatigue tests were carried out, by applying thermal cycles between 400 and 950 °C in air. Special attention was paid not only to the failure life of the TBC specimen, but also the underlying failure mechanisms. The experimental results clearly demonstrated that the effect of MCrAlY bond coat alloys on the thermal fatigue life was very significant. Some discussions were made on the experimental results based on the measurements of mechanical and metallurgical properties of the bond coat alloys: i.e., elastic stiffness, thermal expansion coefficient and high temperature oxidation resistance.

Key words: Thermal Barrier Coatings (TBCs), Bond Coat, Thermal Fatigue, EB-PVD, Thermally Grown Oxide, Cracking, Spalling

1. Introduction

Industrial gas turbines have been used as electrical generators by both utilities and private industrial companies (1)-(7). Today, many simple and combined cycle gas turbine plants have been built on a worldwide basis to provide additional power as well as to replace aging power plants. In recent times, combined cycle gas turbines with peak temperature of 1500°C have been developed. These units can operate at thermal efficiencies greater than 55% (3)(7). The durability of a gas turbine is principally limited by the components operating at high temperatures in the turbine sections, in particular the first stage nozzle and blades. Although many efforts have been made to develop new heat resisting materials during the last few decades (e.g., ceramics and their composites), Ni-base superalloys have been still the most common structural materials used in advanced gas turbines (3). In order to develop advanced gas turbines which can operate with higher thermal efficiency, thermal barrier coatings (TBCs) on microstructurally controlled Ni-based superalloys, as well as serpentine cooling passages and film cooling (3)(5), are essential technologies for the control of the metal temperature. There is no exaggeration to say that...
next generation gas turbines cannot be developed without advanced TBC systems.

In general, a TBC system consists of a heat insulating ceramic outer layer (top coat) and a metallic inner layer (bond coat) between the ceramic and substrate. One of current efforts to improve the performance of TBCs has been directed at the control of microstructure of ceramic top coat. The top coat has been applied either by air, low-pressure plasma sprayings (APS, LPPS, respectively) for these decades. However, these traditional methods have some disadvantages such as difficulty of control the top coat microstructure and less strain tolerance. More recently, electron beam physical vapor deposition (EB-PVD) process has been developed. The microstructure of EB-PVD top coat is characterized by a columnar grain structure. The individual grains of the columnar structure are bonded at their base, but have a weak bond between grains. The major advantage of the columnar microstructure is that it can reduce thermal stress built-up in the coatings. As a matter of fact, the elastic stiffness of EB-PVD top coat was significantly lower than that produced by the APS process. Thermal strain within the coatings is accommodated by free expansion of the column into the gaps, resulting in lower internal stress. However, there is such a disadvantage of the columnar structure that thermal conductivity was twice as high as that of an APS coating.

Meanwhile, thermally grown oxide (TGO) is often formed at the bond/top coat interface during the deposition a little, and continues to grow in service. In general the TGO must be critical to failure life of the EB-PVD coating. Since the phenomenon is taken place by inward and outward diffusion between the bond/top coat, it is natural to deduce that bond coat material system may have an intrinsic role in the TBC failure.

It is a main object of this work to investigate the effect of MCrAlY alloy system on the lifetime of the EB-PVD TBC specimens under thermal cycles in air. The dependence will be discussed on the basis of the measurements of mechanical, physical and metallurgical properties of the bond coat alloys.

2. Experimental Procedures

2.1. TBC specimen preparation

Generally speaking, thermal stress in TBCs is resulting from a constraint of thermal free expansion of the three-dimensional geometry of components. In the other words, thermal stress is built-up in bi- and tri- axial stress state; thus a two-dimensional plate type of specimen that has been often used so far is not always preferable to evaluate the thermal fatigue life of TBCs; a main subject of this work. Accordingly, hollow cylindrical TBC specimens (illustrated in Fig. 1) were used for the thermal fatigue test in this work. The specimens were prepared as follows.

All the TBC specimens consist of three layers; IN738LC Ni-base superalloy substrate, MCrAlY alloy bond boat, and YSZ (: 8 wt.% \( Y_{2}O_{3}\)-stabilized \( ZrO_{2}\) ) ceramic top coat. The chemical compositions of IN738LC substrate, YSZ top coat, and bond coats are given in Tables 1 and 2, respectively. Three kinds of so-called MCrAlY alloy systems; CoNiCrAlY, CoNiCrAlY+B, and Haynes C22 alloys were selected as the bond coat alloy. Here, the CoNiCrAlY alloy is a traditional alloy that has been often used in the industrial
TBC systems for a few decades. On the other hand, Haynes C22 and CoNiCrAlY+B are specific alloys chosen in this work. The reason why the latter two alloys were selected is from the following reasons: whereas it is possible to get these MCrAlY alloys on commercial base, they are containing boron in significant amount in their chemical compositions (see Table 2). An addition of boron; one of the grain boundary strengthening elements, is expected to have a specific role to minimize the undesirable effects of the cellular microstructure formed in the superalloy substrate (18)(20). The cellular microstructure might be originated in some cases in new generation Ni-base superalloys during a grit blasting process; an important process to fabricate the TBCs, but it may reduce the high temperature strength of superalloy substrates (18).

Each bond coat alloy was overlaid on the IN738LC Ni-base superalloy by about 100 µm in thickness, by a low-pressure plasma spraying method. After the bond coating, the ceramic top coat was coated by electron beam physical vapor deposition (EB-PVD) technique by about 250 µm in thickness at Japan Fine Ceramic Center, Nagoya. The coating equipment used was designed and built-up by Von Ardenne Anlagentechnik TURBO150. The basic coating conditions were: rotation of the specimen during the deposition, 10 rpm; heating temperature during the deposition, 900°C; electron beam power, 45 kW; and chamber pressure, 1 Pa, respectively. The EB-PVD top coat was applied only on the cylindrical specimen surface, and not at the edge surface (i.e., on the axial plane); thus, the edge surface was still bare after the coating (see the illustration in Fig. 1). After the coating the edge surface was mechanically polished by 0.1µm diamond paste, through which a degree of environmental damage during the thermal cycle test will be investigated (see section 3.3)

Hereinafter, each TBC specimen will be represented in terms of its bond coat alloys; e.g., CoNiCrAlY TBC specimen etc.

2.2. Measurement of basic properties of bond coat alloys

In order to measure the basic properties of the bond coat alloys, self-standing specimens were extracted from the coating block prepared by the same conditions and procedures as those for the TBC specimen. Here, the specimens were machined by means of an electro-discharge machining equipment. The properties measured are; proof and tensile strengths, elongation, thermal expansion coefficient, and isothermal oxidation resistance. The former two properties were measured under 10⁻⁶/sec in tensile strain rate, by using the solid cylindrical specimen of which gage section diameter and length were 1.5 mm and 10 mm, respectively. The thermal expansion coefficient was measured according to Japan Industrial Standard (JIS) Z2285, by using the small block specimen. The isothermal oxidation resistance of the bond coat alloys was compared through the measurement of weight gain during the isothermal exposure test at high temperatures, which were carried out according to JIS Z2281.
2.3. Thermal Fatigue Tests

A new type of thermal fatigue test system was developed in this work (see Fig. 2). On developing this new test system, special consideration was paid so that the following two conflicting factors were satisfied. The one is such a requirement that thermal cycles can be applied to the TBC specimen with negligible temperature difference between the ceramic top coat and the superalloy substrate. Once this requirement is satisfied, it enables us to easily analyze the thermal fatigue test result from mechanical point of view, unlike the usual engineering test methods; e.g., in burner rig test in which it may be generally too difficult to know thermal stress built-up during the test. The other is such a requirement that the thermal cycle test can be performed under thermal cycle frequency as high as possible, which makes us evaluate the fatigue life of TBCs within a limited time. In this work, these two requirements have been achieved by combining two different types of heating systems; an induction heating system and an infrared radiation system. The respective systems can heat up the specimen predominantly from the metallic substrate part, and from the surface part (i.e., ceramic top coat), respectively. By balancing and controlling those two phenomena, it is possible to uniformly heat up the specimen. Employing this test system the thermal cycles were repeated to the TBC specimens between 400 and 950°C with a frequency of 5 min/cycle, accompanying with a temperature distribution within 5°C anywhere in the TBC specimens. All thermal fatigue tests were carried out in air.

3. Results and Discussions

3.1. Microstructure of EB-PVD top coat

Figure 3 shows the microstructure of the top coat by EB-PVD. As often the case, the EB-PVD top coat has a columnar microstructure that is predominantly aligned perpendicular to the bond coat surface. Fine intercolumnar pores also exist. The diameter of individual columnar grain was about 1 µm in average at the bond/top interface, which increased gradually with their growing, resulting in 5 µm in average on the top surface. According to the X-ray analysis, it was found that the columnar grains grew along <100> crystallographic direction, and aligned to <010> direction toward the substrate rotation axis in the deposit process (16) (see the illustration in Fig. 3). However, this type of texture has not adequately developed at the top/bond coat interface, or at the beginning of deposition.

3.2. Basic properties of MCrAlY bond coat alloys

(1) Microstructures

The microstructures of three kinds of the virgin bond coat alloys are compared in Fig. 4. Many types of precipitates are appearing, depending on the alloy systems. These precipitates were identified by CoAl in the CoNiCrAlY, CoAl and borides in the CoNiCrAlY+B, and NiAl and NiAl with a little borides in the HaynesC22 (20). Both the volume fraction and the size of the precipitates increase in order: CoNiCrAlY+B > CoNiCrAlY > Haynes C22.
(2) Mechanical and Physical properties

Mechanical properties of the virgin and aged bond coat alloys were measured by means of the self-standing MCrAlY alloy specimens, where the aging was conducted at 1000°C for 1000 hours in air. Figures 5(a) and (b) show the tensile strength and rupture elongation, as a function of test temperature, respectively. The tensile strengths of all the alloys gradually decreased with increasing temperature until up to 600°C. At over 600°C, while the strength decreased very rapidly, the elongation increased with temperature.

Thermal expansion coefficient of the bond coat alloys were also measured at R.T. and 950°C, and is compared with that of the substrate; Fig. 6. It is found from this figure that all the bond coat alloys have higher thermal expansion coefficient than the substrate. Especially, the value of the CoNiCrAlY+B was larger than the others. Note that thermal stress induced during the thermal cycle test is proportional to the above mismatch, which must be directly relating to the life of the TBC specimens.

(3) Oxidation resistance

Figure 7 expresses the results of oxidation test of the bond coat alloys after the isothermal exposure test at 1100°C. From this figure, it is seen that the weight gains of both the Haynes and the CoNiCrAlY+B coatings were higher than that of the CoNiCrAlY. This trend was more significant in the CoNiCrAlY+B. Such difference in the oxidation properties seems to be attributed to the addition of B. The oxidation resistance should be directly relevant to the development of TGO. This point will be discussed later.

3.3. Thermal Cycle Test

(1) Visual Damages

When the TBC specimens were subjected to thermal cycles in air, some visual damages were nucleated on the specimen surface. Figure 8 reveals typical damage morphology in the TBC specimen with the Haynes C22 bond coat, to which the thermal cycles were repeated between 400 and 950 °C by 1000 cycles. It is seen that many cracks were nucleated to both the vertical and horizontal directions on Figs. 8(a) and (b). The both
types of cracks should be originated by hoop and axial stress components induced by thermal cycles, respectively. However it was too hard to identify the crack nucleation sites, at least from the specimen surface observation. The failure seems to be undertaken by the coalescence of many small cracks.

These crackings propagated accompanying with the intercolumnar fracture mode, and then a part of top coat has spalled out (Fig. 8(b)). The spallation predominantly started from the specimen edge, suggesting that the thermal cycle damage must be evolved from the free surface. However, it is worthy to point out, the damages that were seen near the specimen edge were comparably built up as well inside the specimen, as shown later (see Fig. 12). This must be because the EB-PVD top coat was a very porous body; oxidation could attack the TBC specimen from anywhere. It seems from Figs. 8(b) and(c) that the cracking and spalling were taken place by a unit of small segments of the top coat, where each segment consisted of one or some “seed” columnar grain(s) and many minor grains. The former would be finally grown major columnar grains of the segment, and the latter

![Graph 1](image1.png)  
**Fig. 5** Mechanical properties of bond coat alloys

![Graph 2](image2.png)  
**Fig. 6** Comparison of thermal expansion coefficients of bond coat alloys and substrate.

![Graph 3](image3.png)  
**Fig. 7** Oxidation resistance of bond coat alloys at 1100°C.
disappeared and absorbed into the major grains during the deposition process. When this is the case, it must be important for the control of spalling behavior to control the nucleation process of seed grains. Those damages; cracking and spalling, are easy to be found from the specimen surface, however, it must not be beneficial for us to express the thermal fatigue damage in terms of these damages since there is a problem how to define the cracking level quantitatively. In addition, because the EB-PVD top coat contains many intercolumnar gaps between the grains from the as-deposited stage (9), it is too difficult to distinguish between these initial cracks and those nucleated by thermal cycles.

(2) Thermal Fatigue Failure Life

The thermal fatigue failure life of the TBC specimens was defined by the number of cycles at which the spalling-off of the top coat arrives at 10 % of the total top coat area in fraction in this work. The smaller, the better this fraction is, since from a nature of thermal stress the thermal fatigue damage in general evolves very rapidly at early stage. However, when this fraction is set up too small, it is too difficult to measure the life exactly. On the other hand, when it is too high, it dose not make sense. According to the above definition, the failure lives of the respective TBC specimens are compared in Fig.9. It is clear that the effect of MCrAlY bond coat alloys on the thermal fatigue life is very significant. In this work, the lives are ranging in order: CoNiCrAlY specimen > Haynes C22 specimen > CoNiCrAlY+B specimen.

In Fig. 9, the failure lives of the respective EB-PVD specimens are compared with that of the atmospheric plasma sprayed (APSed) TBC specimen with CoNiCrAlY bond coat. It found that the former is significantly longer than the latter. This means, the EB-PVD process should be more beneficial than the traditional process against thermal fatigue failure.
The difference of thermal expansion coefficient between the bond and top coats was larger in order: CoNiCrAlY+B specimen > CoNiCrAlY specimen > HaynesC22 specimen (see Fig. 6). This implies that the thermal stress must be higher in order: CoNiCrAlY+B specimen > CoNiCrAlY specimen > HaynesC22 specimen. Note that this order does not always correspond to the thermal fatigue life given in Fig. 9. Thus, not only the mechanical factors but also the other factors might be contributing to the thermal fatigue life in this work. The resistance against cyclic oxidation at high temperatures might be such a candidate factor, as discussed later.

(3) Thermally Grown Oxide during Thermal Fatigue Test

Many researchers have confirmed that the thermally grown oxide (TGO) formed at the top coat/bond coat interface played an important role in the TBC failure. This must be also the case in this work. The compositional analysis of the top/bond coat and substrate/bond coat interfaces was carried out using electron probe micro analyzer (EPMA); Fig. 10. While the characterization of the oxide at the interface has been still on the way of investigation, the main oxides at the both interfaces are briefly summarized in Table 3. For an example, the two TGO layers formed in the CoNiCrAlY+B specimen were consisting of Al2O3, Cr2O3 and their spinels at the top/bond coat interface, and Al2O3 at the substrate/bond coat interface, respectively. It is worthy to note in Table 3 that the characterization of TGO was different depending on the TBC system.

A degree of oxidation attack has been often represented by the TGO thickness. However, this parameter is not always preferable in this work. Because it is necessary to cut the TBC specimen to measure the TGO thickness, the thickness can not be measured continuously by means of one specimen. On the other hand, once the bond coat is oxidized to form TGO layer, the volumetric change should be taken place there. When the bond coat is metallic system, the change is in dilatation generally. This change should appear in a change of relative height of the bond coat area to the substrate. Thus, the relative height, \( h \), was measured by means of a confocal microscope in this work (see an illustration in Fig. 11). Here, the measurement was carried out at four positions in circumferential direction at every 90º on the edge surface of the TBC specimen where the top coat was not deposited.

The measurement of \( h \) is given in Fig. 11, where the positive and negative values mean a swelling and a chipping of the bond coat, respectively. These graphs show that in all the TBC specimens the value of \( h \) increased in an almost monotonic manner with increasing the number of thermal cycles. The value of \( h \) get larger in order: CoNiCrAlY+B specimen > HaynesC22 specimen > CoNiCrAlY specimen. It is important to note that this order agrees well with that in the thermal fatigue failure life (compare with Fig. 9), suggesting an important role of bond coat alloy system in the failure.
The TGO thickness inside the specimen was measured along the specimen axial direction on the longitudinal section of the specimen; Fig. 12. It is found from Fig. 12 that the TGO thickness at the middle position of the TBC specimen was almost comparable to that at the specimen edge in all the TBC specimens. This result indicates that the environmental damage progressed anywhere comparably, through the porous EB-PVD top coat, suggesting that the degree of oxidation damage can be represented by the observation from the specimen edge surface so far as the top coat has not spalled out.

Further investigations are necessary to make clear the effect of TGO on thermal fatigue failure life. However, it is important to point out at this moment that a dilatation strain of bond coat resulting from the formation of TGO may induce an additional internal stress near the interface, which promotes a spalling of top coat.
4. Conclusions

The effect of MCrAlY bond coat alloy system on thermal fatigue failure of EB-PVD TBC specimens was investigated in this work. The following conclusions have been introduced:

1. The effect of MCrAlY bond coat alloy system on the thermal fatigue life of the TBC specimens was very significant, in which the basic properties of the bond coat alloys played important roles. Some of them were physical and mechanical properties that dominated the level of thermal stress; e.g. thermal expansion coefficient and elastic modulus. The other was an environmental resistance of the bond coat to high temperature oxidation to form the TGO and to induce an additional internal stress due to the dilatation strain.

2. The thermal fatigue life of the EB-PVD TBC specimens was significantly longer than that of the APSed specimens.

Acknowledgements

Financial supports by Grant-in-Aid (# 15360046) by the Ministry of Education, and "Nano-Coating" project by NEDO are greatly acknowledged.

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