Thermal durability of thermal barrier coatings with layered bond coat in cyclic thermal exposure

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The effects of the bond coat structure on the microstructure evolution and lifetime performance of thermal barrier coatings (TBCs) were investigated through the cyclic thermal fatigue (CTF) and thermal-shock (TS) tests. The single layer and the first layer in the layered bond coat were prepared by high velocity oxy-fuel (HVOF) spray process using nickel-based feedstock. The second layer in the layered bond coat and the top coat were formed by air plasma spray (APS) process using nickel-based metallic feedstock and 8 wt% yttria-stabilized zirconia, respectively. The CTF tests were performed till 872 cycles with a dwell time of 60 min at a surface temperature of 1100°C and a bottom temperature of 950°C. Also, the TS tests were conducted until 300 cycles with a dwell time of 60 min at 1100°C. After the CTF and TS tests, the TBC system with the layered bond coat showed a better thermal durability than that with the single layer. The hardness value of the bond coat by HVOF process was dramatically decreased after the both tests, without much change in the bond coat by APS process. The diffusion trends of elements were similar with each other, less dependent on the bond coat structure, increasing amounts of cobalt and aluminum and decreasing those of nickel and tungsten. The microstructure evolution of the top coat, the growth behavior of thermally grown oxide layer, and the thermal durability were strongly affected by the thermal exposure condition and the bond coat structure.

Key-words: Thermal barrier coating, Bond coat, Layered structure, Air plasma spray, High velocity oxy-fuel spray, Thermal durability

1. Introduction

The thermal barrier coatings (TBCs) are widely employed in high temperature components of gas turbines, to increase the fuel efficiency and to improve the engine performance.1)–3) TBCs consist of three layers with a heat-resistant ceramic top coat, an intermetallic bond coat, and a superalloy substrate. The top coat which provides thermal barrier function to TBC system should have a low thermal conductivity and be able to sustain the thermal expansion mismatch with the substrate.4)–6) The bond coat provides a rough surface for mechanical bonding of the top coat, protects the underlying substrate against the high temperature oxidation and corrosion, and reduces the thermal expansion mismatch between the substrate and the top coat.7)–9) With increasing turbine inlet temperature for improving the fuel efficiency, the role of TBC is emphasized to protect the substrate and the bond coat from oxidation and corrosion, resulting in the increase of top coat thickness to above 600 μm.7,9,10) It was reported that the increase in thickness of the top coat can reduce the surface temperature of components at the rate of 4–9°C per 25.4 μm.9,10) However, if the top coat thickness is reached to above 1500 μm, the top coat will spontaneously fail during preparation or in service due to the thermal and residual stresses.9,11) Many technologies have been suggested to provide higher thermal durability to TBC system, such as dense vertical cracked (DVC) TBC, high porosity coating, and suspension plasmas spray (SPS) process, even though there are disadvantages of the higher thermal conductivity of DVC-TBC, the low adhesive strength and damage resistance of high porosity coating, and the less practical use of SPS.12)–16)

Many researches have been focused on the thermo-mechanical properties of the top coat related to the lifetime performance. However, major problems of TBCs in a field application are the spallation and wear of top coat under the thermal and corrosion fatigue in working environments. These are more related to the interface stability of TBC system, especially at the interface between the top and bond coats. Therefore, new strategy for improving the interface stability must be considered and proposed to enhance the thermal durability and lengthen the lifetime performance of TBC system. The bond coat plays an important role in ensuring structural effectiveness and affording extra adhesion of the top coat to the substrate. Many techniques have been proposed and applied to form the bond coat, such as vacuum plasma spray (VPS), low-pressure plasma spray (LPSS), air plasma spray (APS), high-frequency pulse detonation, and high velocity oxy-fuel (HVOF) spray.17)–26) The APS process is widely used to prepare the bond coat in TBC system, because of its economic benefits. However, in order to meet requirements, such as better thermal durability and longer lifetime performance, the HVOF spray process is employing for preparing the bond coat, showing a dense microstructure with low oxide contents.

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The LPPS process is used to prepare the bond coat, even though the LPPS process is limited application, due to the production cost and low production rates. In this study, the microstructure of bond coat was controlled to improve the interface stability, and the effects of the bond coat structure on the thermal durability of TBC systems were investigated through the cyclic thermal fatigue (CTF) and thermal shock (TS) tests, including the lifetime performance of each TBC system. Two types of TBC system were prepared with different bond coat structures of the single and double layers. The microstructure evolution, mechanical properties, and fracture behavior of the TBC systems were compared before and after the tests. The relationship between the delamination or fracture behavior and the bond coat structure has been discussed, based on the elemental analysis and microstructure before and after cyclic thermal exposure tests.

2. Experimental procedure

2.1 Materials and specimen preparation

A nickel-based superalloy (GTD-111, a nominal composition of Ni–14Cr–9.5Co–1.5Mo–3Al– 4.9Ti–3.8W–2.8Ta–0.1C–0.01B, in wt %) with a coin shape was used as a substrate. The dimension of specimen was 20 and 5 mm in diameter and thickness, respectively. The substrate was sand-blasted using an alumina (Al₂O₃) powder with particle size of 50 mesh. The bond coat was designed as the single and double layers. The first layer in the layered bond coat and the single layer were coated on the substrate by HVOF spray process using Diamond Jet®-2600 DMJ (Plasma Technik). The second layer was deposited onto the first layer by APS process using 9 MB (Sulzer Metco, Switzerland). AMDRY 962 powder (Sulzer Metco Holding AG, Switzerland, composition of Ni–22Cr–10Al–1.0Y in wt %, particle size of 56–106 μm) was used as a feedstock powder to prepare the bond coat in APS process, and AMDRY 997 powder (Sulzer Metco Holding AG, Switzerland, composition of Ni–23Co–20Cr–8.5Al–4Ta–0.6 Y in wt %, particle size of 5–37 μm) was used to prepare the bond coat in HVOF spray process. The thicknesses for the first, second, and single layers were approximately 120, 120, and 250 μm, respectively. Finally, the top coat was coated on the both single and double layers by the APS using METCO 204C-NS (Sulzer Metco Holding AG, Switzerland, 8 wt % Y₂O₃ doped in ZrO₂, particle size of 45–125 μm). The thickness of the top coat was approximately 450 μm. All coating processes were done within 2 h. Detailed spray parameters for the HVOF spray and APS processes are shown in Tables 1 and 2 with modifications to the manufacturer’s specifications.

2.2 Cyclic thermal fatigue and thermal shock tests

A bottom-loading programmable cyclic furnace was used for determining life performance of the TBC systems. The CTF tests were performed until 872 cycles at a surface temperature of 1100°C with a temperature difference of 150°C between the top and bottom surfaces of specimen with a dwell time of 60 min, and then natural air cooling was allowed for 20 min at room temperature. The failure criterion was defined as 25% buckling or spallation of the top coat in the CTF. The TS tests were carried out until 300 cycles in a muffle furnace. When the temperature of the furnace reached to 1100°C, the specimens were pushed into the furnace. In the TS, the specimens were held about 60 min in the furnace and then directly quenched into water. Throughout the TS, the temperature of water was between 20 and 35°C. More than 50% of the region spalled in the top coat or cracked at the interface between the top and bond coat was adopted as the criterion for failure in water quenched specimens. At least four specimens were tested for each condition.

2.3 Characterization

The selected TBC specimens before and after the CTF and TS tests were preprocessed to observe cross-sectional microstructure and to measure mechanical properties. The mounted samples into a fluid epoxy resin were polished using silicon carbide paper and polished using the 3 and 1 μm diamond pastes. The cross-sectional microstructure of TBC specimens was observed using a scanning electron microscope (SEM, JEOL Model JSM-5610, Japan). The thickness of the thermally grown oxide (TGO) layer formed at the interface between the bond and top coats were measured using a drawing software. The hardness values of the bond coat and top coats were determined using a micro indenter (HM-114, Mitutoyo Corp., Japan) with a Vickers tip for a load of 3 N. To obtain reliable values, more than 10 points were tested for each result. The size of indentation impression was measured by the SEM and all experiments were performed at room temperature, and standard test method for "Vickers Indentation Hardness of Advanced Ceramics (ASTM C1327-03)" was employed for the hardness measurement of this study. The chemical composition of each TBC system after cyclic thermal exposure tests was determined using an energy-dispersive X-ray spectrometer (EDX, Oxford Instruments, Oxford, UK).

Table 1. Feedstock powders and coating parameters for preparing the bond and top coats

| Parameters          | First and single layer bond coats (HVOF) | Second bond coat (APS) | Top coat (APS) |
|---------------------|-----------------------------------------|------------------------|----------------|
| Feedstock           | AMDRY 9951                              | AMDRY 962              | METCO 204C-NS  |
| Feedstock           | AMDRY 997                               | AMDRY 962              |                |
| Feedstock           | AMDRY 962                               |                        |                |
| Feed rate           | 40 g/min                                | 60 g/min               | 45 g/min       |
| Gun distance         | 200 mm                                  | 110 mm                 | 110 mm         |
| Gun speed            | 700 mm/s                                | 500 mm/s               | 500 mm/s       |
| Step distance        | 4 mm                                    | 4 mm                   | 5 mm           |
| Voltage              | 70 V                                    | 70 V                   | 75 V           |
| Current              | 500 A                                   | 500 A                  | 500 A          |

Table 2. The flow rate of carrier gas in preparing the bond and top coats

| Carrier gas | First and single layer bond coats (HVOF) | Second bond coat (APS) | Top coat (APS) |
|-------------|-----------------------------------------|------------------------|----------------|
| Air         | 42 NLPM                                 |                        |                |
| O₂          | 12 NLPM                                 |                        |                |
| H₂          | 62 NLPM                                 | 23 NLPM                | 27 NLPM        |
| Ar          | —                                       | 96 NLPM                | 96 NLPM        |
3. Results and discussion

3.1 Microstructure of as-prepared TBCs

The cross-sectional microstructures of as-prepared TBCs with different structures in the bond coat are shown in Fig. 1 for the TBC with the ordinary bond coat (single layer) and Fig. 2 for the TBC with the layered bond coat (double layer). In all cases, the top coat of 450 ± 40 μm was prepared by the APS, showing typical defects, such as splat boundaries, pores, and un-melted powders. In the TBC with the single layer bond coat, the thickness of the bond coat was 250 ± 20 μm. The double layer was well deposited without any delamination or cracking at the interface between the APS and HVOF bond coats [Fig. 2(D)], and the thickness of each bond coat layer was well controlled as 120 ± 15 μm for the first layer and 120 ± 10 μm for the second layer. The bond coat prepared by the HVOF showed a dense microstructure and a less oxide content, whereas the amount of visible defects and oxide contents for the APS bond coat were in a high level. There was not any visible crack, which caused a failure or delamination, in the microstructures of the interface between the top and bond coats [Figs. 1(C) and 2(C)]. Usually the HVOF spray process creates the more smooth interface between the bond and top coats than APS process, because the particle velocity in HVOF spray process can be produced up to about 700 m/s and that in the conventional APS system (9 MB) is 350 m/s. For this reason, the surface prepared by APS process is more rough than that by HVOF spray process, which would improve the structural effectiveness in TBC system.

3.2 Lifetime performance of TBCs

The effects of the bond coat structure on the thermal durability were investigated through the CTF and TS tests. The cross-sectional microstructures of the TBCs with the single and double layers in the bond coat after the both tests are shown in the Fig. 3. The TBC with the single bond coat was fully delaminated in the range of 661–685 cycles [Fig. 3(A-1)]. In the TS, the top coat was fully delaminated in the range of 58–62 cycles [Fig. 3(A-2)]. The thermal and residual stresses at the interface of the bond and top coats caused by temperature difference and the mismatch in coefficient of thermal expansions (CTEs) between the top and bond coats lead to delamination and failure. The less lifetime cycle in the TS may be due to the large temperature difference between the substrate and the top coat during the quenching and annealing processes. Larger temperature difference causes higher thermal and residual stresses at the interface of the bond and top coats. The TBCs with the double bond coat had no delamination until 872 and 300 cycles in the CTF and TS tests, respectively [Fig. 3(B)]. Even though the number of cycle in the TS is smaller than that in the CTF, the thermal degradation, such as oxidation in the bond coat and cracking in the top coat was more severe in the TS, indicating that the thermal fatigue condition employed in evaluating the thermal durability is more important than the actual testing time. The improved thermal durability in the TBCs with the double bond coat would be due to the better structural effectiveness of the second bond coat to the top coat. It was verified that the thermal durability could be improved by controlling the microstructure in the bond coat. In our previous study,38) the TBC with APS bond coat showed no delamination in the CTF, indicating that the bond coat prepared by APS process is more efficient in the thermal durability than those by HVOF spray process. The TBCs with the bond coats by the HVOF spray and LPPS processes showed the Al depletion region between the bond and
top coats. Usually the depletion of Al in the VPS bond coat is more serious than that in the APS bond coat in oxidation environment, showing about 6 and 4 wt% for the VPS and the APS, respectively. The Al depletion is very important in the TGO growth and the thermal durability. Therefore, the lifetime of TBCs with the APS bond coat was superior to that of TBCs with the VPS bond coat. The result of previous study is in agreement with our results of the CTF and TF tests.

The magnified cross-sectional microstructures of the TBC with the single bond coat before and after the CTF and TS tests are shown in Fig. 4, showing less oxidation in the bond coat after the both tests. The TGO layer was newly developed in the range of 7–9 μm, independent of the thermal fatigue condition employed. If the thickness of TGO layer is greater than 10 μm, the interface between TGO layer and top coat normally starts to be delaminated and shows a failure phenomenon, and the formation and composition of TGO layer have been well documented. The magnified cross-sectional microstructures of the TBC with the double bond coat before and after the CTF and TS tests are shown in Fig. 5. The small horizontal cracks were integrated into the long and thick horizontal cracks in the top coat after the TS, while the thick and imperfect defects of the “splat” boundaries/cracks, interlamellar cracks, and pores were newly developed and extended. These thermal degradation phenomena were well described in Fig. 6, including the TGO growth and the “rumpling” phenomena of TGO layer. The duplex oxide scale was formed in the TGO layer, especially after the CTF; black color is formed early and gray one is formed late. The duplex oxide scale are composed of the inner (black color: Al₂O₃) and outer (gray color: Cr₂O₃, NiAl₂O₄, and other spinel structures) layers. The thickness of TGO layer in the TBC with the double bond coat was thicker than that with the single bond coat, because of relatively long cycles in the CTF and TS tests. In the CTF, the TGO layer was generated over 10 μm thickness, which more thicken after the TS, even though the number of cycle in the TS was less than that in the CTF. This was verified again that the thermal durability is dependent on the thermal fatigue condition.

The oxidation phenomena were progressed in the first bond coat, showing the less oxidation in the second bond coat and at the interface of the first and second bond coats. The oxidation in the bond coat will lead to a change in sign of stresses due to the smaller CTEs of TGO layer. The failure or fracture behavior in TBCs is associated with the stresses arising from the mismatch of CTEs between top coat and TGO layer since the residual stress is a source of strain energy that can promote spallation of TBCs. Cracks were shown at the interface between the top and bond coats of the TBC with the double layer bond coat after the both tests (Fig. 6), but no fully delamination, meaning that the adhesive strength at the interface between the top and bond coats could be improved by the layered structure in the bond coat.
3.3 Mechanical property and element analysis

The hardness values of the top and bond coats before and after the CTF and TS tests were measured using a Vickers indentation method, which are shown in Fig. 7. The indentation tests were conducted on the sectional plane with a load of 3 N at room temperature. The hardness value of the top coat in the as-prepared TBC was determined to be 2.9 ± 0.1 GPa (mean ± standard deviation). The bond coats prepared by the APS and HVOF processes showed the hardness values of 2.8 ± 0.3 and 4.5 ± 0.2 GPa, respectively. The hardness value of the bond coat prepared by HVOF spray process was higher than that by APS process, which was in good agreement with the microstructure observed in Figs. 1 and 2. The hardness values were much similar with our previous studies, indicating that the microstructure was well controlled. The hardness values of the single bond coat were dramatically decreased as to 2.7 ± 0.1 GPa after the CTF and 2.3 ± 0.2 GPa after the TS. The decrease of hardness value in the HVOF bond coat (single layer and the second layer in the double layer bond coat) is due to the phase transformation by element diffusion, while oxidation in the APS bond coat compensates the decrease of hardness value by element diffusion. The TBC with the single bond coat was delaminated after the both tests and hence there was no hardness value for the top coat. In the TBC with the double bond coat, the hardness values of the top coats were increased and decreased in error range for the CTF and TS tests, respectively. The bond coat prepared by APS process (the second layer) showed the slightly increased hardness values of 3.1 ± 0.4 GPa after the CTF and 3.2 ± 0.4 GPa after the TS, and the first bond coat prepared by HVOF spray process represented the similar trend with the single bond coats. The hardness values were also more severely affected by the TS rather than the CTF, independent of the bond coat structure.

The elemental analysis of each TBC with the single and double bond coats was performed after the TS, which are shown in Fig. 8, including the cross-sectional microstructures. Also, the compositions of the substrate and the feedstock powders (for the first and second layers) are shown in the graphs. The elements of nickel (Ni), tungsten (W), cobalt (Co), aluminum (Al), and chromium (Cr) were analyzed in this study. The amounts of the Ni and W elements were decreased by passing the boundary of the substrate and the bond coat, while those of the Co and Al were increased, showing less variation in the Cr. The diffusion trends of elements were similar with each other, less dependent on the bond coat structure. The double bond coat showed the relatively higher amounts of the Ni and Al and smaller amounts of the Co than the single bond coat. These results are due to the formation of TGO layer and well reflected in the hardness variation. As a result, the elements of Co, Al, and Ni are mainly diffused during the test, and the double bond coat indicated less diffusion in the main elements, indicating that the hardness values of the second layer were less decreased than those of the first layer in the double bond coat.

4. Conclusions

The influences of the bond coat structure on the thermal durability of TBC system with the top coat prepared by the APS were investigated through the CTF and TS tests. The bond coat was designed as a layered structure and controlled with different coating processes and feedstock powders, without visible cracks or delamination at the interface of the both TBCs with the single and double bond coats. After the CTF, the TBC with the single bond coat was delaminated in 661–685 cycles. However, the TBC with the double bond coat was survived after 872 cycles without delamination or cracking at the interface between the bond and top coats. After the TS, the top coat of the TBC with the single bond coat was fully delaminated in 58–62 cycles, but the TBC with the double bond coat was not delaminated until 300 cycles, indicating that the bond structural effectiveness is one of important factors for improving the thermal durability of TBC system. The hardness values were more severely affected by the TS rather than the CTF, independent of the bond coat structure. The diffusion trends of elements were similar with each other, less dependent on the bond coat structure. These observations allows us to design a certain layered structure in TBCs, especially in the bond coat, to propose an efficient coating to ensure the thermal durability and mechanical properties in the cyclic thermal exposure and thermal shock environments.
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Fig. 8. The cross-sectional microstructures and results of elemental analysis after TS tests for TBCs with the single and double bond coats: (A) TBC with single bond coat and (B) the TBC with double bond coat.
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