Rare Metal Materials and Engineering
Volume 51, Issue 5, May 2022
Available online at www.rmme.ac.cn

Science Press

Cite this article as: Cheng Bo, Li Jianjun, An Guosheng, et al. Durable Columnar Structure Design for Thermal Barrier Coatings[J]. Rare Metal Materials and Engineering, 2022, 51(05): 1660-1666.

REVIEW

Durable Columnar Structure Design for Thermal Barrier Coatings

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Abstract: The thermal barrier coatings (TBCs) are employed to protect metallic components from high temperature gas, and their microstructure and service performance are closely related to and have the mutual effect with the preparation process and service condition. This research reviews TBCs failure mechanisms caused by different stress inducements from the perspective of thermally grown oxide (TGO) thickening, phase transformations, and sintering. Subsequently, the preparation methods of TBCs with columnar structure are summarized, as well as their functions of releasing stress during TBC service. Although the preparation process and service conditions are different, the lifetimes of TBCs prepared by different methods are evaluated by comparing with that of the standard TBCs. The long lifetime service mechanism of TBCs with column structure is summarized. This research reviews the interaction effects among the stress source, microstructure, preparation process, and service performance of TBCs, providing guidance for the advanced and durable TBCs design.

Key words: thermal barrier coatings; durable structure design; columnar structure; lifetime

The thermal barrier coatings (TBCs) are mainly employed in the hot parts of gas turbines to protect the metallic components from hot gas[1-6]. The ceramics of high temperature resistance and good thermal insulation are deposited on the substrate surface to separate the high-temperature environments from substrate, therefore prolonging the TBCs service lifetime at extremely high temperature (even higher than the alloy melting point). The efficiency and thrust-weight ratio are hence improved greatly[7,8]. TBCs, as a complex multifunctional system of 100~500 μm in thickness, consist of ceramic top coat (TC), metallic bond coat (BC), and substrate, as shown in Fig. 1[9-12]. Traditionally, the low thermal conductive ceramic materials with layered structure are applied to TBCs TC, resulting in a good thermal insulation performance (100~300 °C) of TBCs. TC material mainly is 8wt% yttria-stabilized zirconia (8YSZ), due to its optimal properties for the application under 1200 °C[13-19]. The dense nearly-inner-oxidation-free metallic BC is usually prepared by cold spray (CS) or low-pressure plasma spray (LPPS) [20-23], which reduces the thermal mismatch between TC and substrate. The thermally grown oxide (TGO) inevitably grows on BC surface during preparation process[24,25]. During service, TBCs suffer extremely high temperature exposure and great temperature variation, which degrades the microstructure and performance of TBCs and introduces the stress, thereby influencing TBC service lifetime[26-30]. The introduced stress acts as the driving force for the crack propagation, and when the cracks coalesce to a large scale, TBCs fail. As a series system, the failure of any part will result in the failure of the whole TBCs system. Therefore, the longer service life of TBC system requires less stress. Since the durability and reliability of TBCs are the priority for application, over the past decades, a lot of efforts have been made to extend the lifetime of TBCs mainly through structure design and material system.

This research reviews the developments in the field of

Received date: May 17, 2021
Foundation item: Sponsored by State Key Laboratory for Mechanical Behavior of Materials; National Natural Science Foundation of China (51901093, 51674130); Hongliu Distinguished Young Talent Support Program Project of Lanzhou University of Technology; National High-End Foreign Experts Program of China (GDT20186200331); International Science and Technology Correspondent Program of Gansu Province (17JR7WA017); Sponsored by Science and Technology International Cooperation Demonstrative Base of Metal Surface Engineering Along the Silk Road
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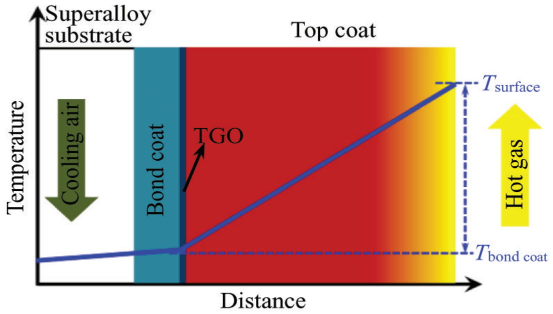

Fig.1 Schematic diagram of typical TBC system[12]

durable columnar structure design of TBCs. The degradation and failure inducements of TBCs are summarized. The columnar structure, corresponding lifetime, main challenges, and future development trends in TBC fields are discussed.

1 Degradation and Failure of TBCs

Microstructure and performance changes occur during exposure at extremely high temperature or during the temperature variation, which results in TBC failure, such as whole or partial TC spallation. It is believed that three main inducements are responsible for TBCs failure: TGO thickening, phase transformations, and sintering.

1.1 TGO thickening

During the high temperature exposure, the formation and growth of TGO are inevitable. The continuous uniform thin layered TGO is beneficial to the oxidation resistance of TBC system. However, the continuous thickening of TGO causes the great growth in stress, particularly after the generation of mixed oxides, which may result in TBC failure, especially at relatively low temperatures. Rabiei et al[31] reported that when TGO thickness is below the critical value h_c≈5.5 μm, the service behavior of TBCs is barely unaffected, i.e., the cracks introduced during the thermal spray process in TBCs are basically unchanged. Dong et al[20] prefabricated a series of TBCs with the TGO thickness of 1.3~7.7 μm by controlling the oxidation time, and found that the thermal cyclic lifetime is sharply decreased from 2000 cycles to 400 cycles with increasing the initial TGO thickness from 5 μm to 7.7 μm. Therefore, it is concluded that the critical TGO thickness is about 6.0 μm. If the thickening speed of TGO is reduced, TBC lifetime can be extended. Many efforts have been made to control the composition and growth of TGO. Zhang et al[21] developed a pre-diffusion method combined with pre-oxidation process, and found that the formation of continuous Al_2O_3 TGO on BC surface is promoted and the mixed oxides are effectively restrained, thereby prolonging TBC lifetime. Zhang et al[32] developed a TGO pretreatment method to form a dense continuous TGO layer, which largely delayed the growth rate of TGO and prolonged TBC service life.

1.2 Volume change of TC by phase transformation

Since TBCs serve under harsh operation conditions, TC material cannot meet the following basic requirements[33,34]: low thermal conductivity, high melting point, phase and composition stability at operation temperature, thermal expansion match with the metallic substrate, and low sintering rate under high temperature exposure. But it is quite difficult to find one material to satisfy all these requirements at the same time. The state-of-the-art TC of TBC material is 7wt%~8wt% yttria-stabilized zirconia (YSZ) because of its excellent comprehensive properties, such as high toughness, high coefficient of thermal expansion, and relatively low thermal conductivity. Actually, TC made from YSZ after air plasma spray (APS) mainly consists of nonequilibrium tetragonal phase (t') and a small amount of tetragonal phase (t) and monoclinic phase (m). The t' phase is characterized by excellent strength and fracture toughness, which is favorable for TBCs. However, the t' phase is unstable after high temperature exposure for long time due to the diffusion of Y_2O_3 stabilizer, which can be transformed into the cubic phase (c) and tetragonal phase (t) with high and low Y_2O_3 contents, respectively. The tetragonal phase is transformed into monoclinic phase (m) during cooling with volume expansion of 3%~5%[35-37]. The phase transformation of 8YSZ is one of the vital inducements for TBC failure, especially when the service temperature is above 1200 °C. Therefore, TC material without phase transformation between room temperature and operation temperature should be investigated.

1.3 TC sintering

After the high temperature exposure for long time, the ceramic surface will inevitably undergo the sintering, healing of lamellar structure, the increase in thermal conductivity and elastic modulus, and the degradation in strain tolerance. Cipitria et al[38,39] proposed a systematic mode to describe the sintering process of TBCs after APS, which quantitatively characterized the evolution of linear shrinkage and thermal properties. However, it is found that the sintering dynamics process presents a fast tendency at the initial stage and a slow one in the following stage. Besides, this mode cannot explain the fast sintering phenomenon during the initial stage of high temperature thermal exposure, because it is based on the hypothesis of equal gaps between the lamellar structures. For the real TBCs after APS, the cracks between the lamellar structures always show a wedge-shape, as shown in Fig. 2[40]. Li et al[40-42] developed a comprehensive sintering mechanism based on the real structure of TBCs after APS: the fast increase in mechanical and thermal properties in the initial thermal exposure duration (stage-I), and the slow increase in mechanical and thermal properties during the following duration (stage-II). Cheng et al[43] reported that during the thermal exposure under different temperature conditions, TBC failure happens when TC is sintered to a certain degree. It is believed that the sintering behavior of TBCs is associated with the evolution of elastic modulus of TC. During the thermal cycling, especially during the rapid cooling stage, the increased elastic modulus and temperature gradient result in the rapid shrinkage of coating surface region. The localized shrinkage produces large tensile stress and promotes the peeling of TC. Cheng et al[44] studied the effects of sintering

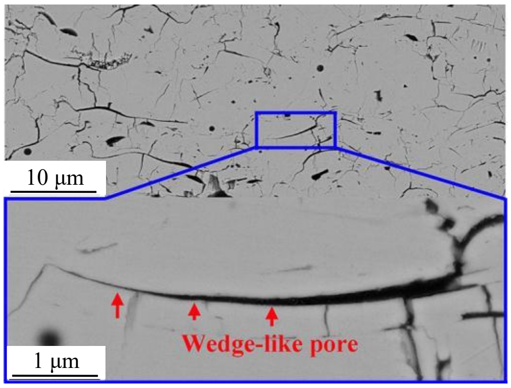

Fig.2 Wedge-shaped cracks of TBCs after APS[40]

temperature on the delamination of TBCs by gradient thermal cyclic tests, and found that the thermal cyclic lifetime is decreased with increasing the TC surface temperature from 1250 °C to 1350 °C. Thus, the high temperature sintering accelerates TC delamination, i. e., the higher the sintering temperature, the shorter the sintering duration (from the initial state to a specific modulus). It is also inferred that the lifetime of TBCs exposed at relatively low temperatures can be evaluated by that at high temperatures. In addition, all results exhibit the same failure mode, namely the multilayered splats delamination, under different conditions.

2 Columnar Structure Design of TBCs

For the traditional APS process, the continuous stacking of splats forms the lamellar-structured coating, and the average bonding ratio between the ceramic lamellae is only about 30% for the commonly used coating[45] with gaps, voids, and microcracks. Yang et al[46] proposed the critical deposition substrate temperature and found that the critical temperature of 8YSZ splats is about 600 °C. When the substrate temperature (or preheating temperature) is lower than the critical temperature, only 1/3 effective bonding can be formed between the splats. However, with increasing the temperature to 825 °C (over the critical temperature), the complete bonding interfaces are developed, which can regulate and control the microstructure of ceramic coatings. The lamellar structure is favorable for the improvement of TBC thermal insulation. However, it always shows a low fracture toughness and inferior thermal shock resistance. The columnar-structured TBCs show a high strain tolerance, and the stress introduced by extremely high temperature can be effectively reduced through the gap between the columns. The lifetime of columnar-structured TBCs is several times longer than that of the lamellar ones[47-49]. From the in-plane direction, the columnar size varies from several microns to hundreds of microns in alloys after different preparation methods, such as electron beam-physical vapor deposition (EB-PVD), suspension plasma spray (SPS)/solution precursor plasma spray (SPPS), and plasma spray-physical vapor deposition (PS-PVD).

2.1 TBCs after EB-PVD

EB-PVD method was firstly applied to the TBC working blades of turbines engine[50]. In 1990s, Ukrainian Paton Welding Institute successfully applied the electron beam melting technique to the physical vapor deposition processes, which improved the coating deposition efficiency and reduced the cost to a large extent[52]. Currently, EB-PVD technique is widely used for the preparation of TBCs for aero-engine working blades and guide vanes. During EB-PVD process, a vacuum atmosphere with pressure lower than 10^{-3} Pa is indispensable and the feedstocks are heated to a gas state by high density electron beam. The gaseous feedstocks form the column structure on the substrate surface under the deflection magnetic field. The typical microstructure of EB-PVD TBCs is shown in Fig. 3[53]. The nanoscale porosity in the columnar grains can reduce the stress (including the sintering stress and the mismatch stress between ceramic TC and alloy substrate) during TBCs service. In addition, compared with TBCs after APS, EB-PVD can achieve a denser TBC structure through chemical bonding, resulting in a good bonding strength perpendicular to the TC/BC interface. The dense structure can also hinder the attachment and penetration of low melting point pieces, such as $CaO-MgO-Al_2O_3-SiO_2$ (CMAS)[53]. However, the high cost and relatively low efficiency hinder the development of EB-PVD process. In addition, the thermal conductivity of TBCs after EB-PVD is about 1.2~2 times higher than that of TBCs after APS, which is adverse for TBCs application.

2.2 SPS/SPPS

The feedstock powder for APS of TBCs is 10~100 μm in size with the deposition splats of Φ40~200 μm×1~3 μm (thickness). It is reported that the mechanical properties of TBCs are improved by decreasing the splat size (corresponding to the feedstock powder at submicron or nanometer scale) which is restricted by the powders with suitable characteristics (size distribution, flowability). The

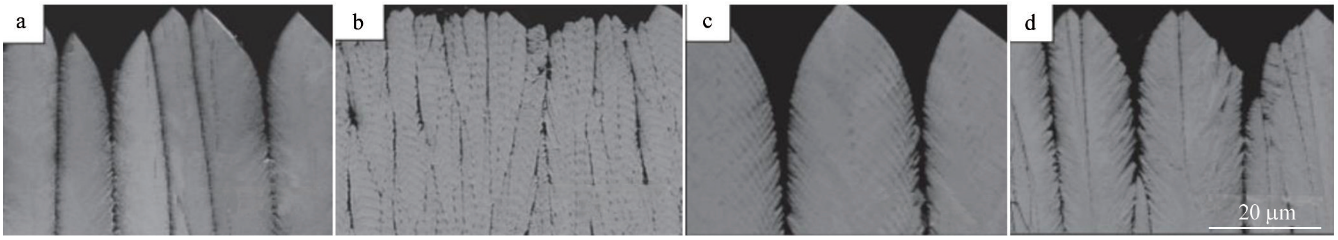

Fig.3 TBC morphologies with intermediate (a), fine (b), and coarse (c, d) microstructures after different EB-PVD processes[52]

above disadvantages can be ameliorated by the liquid carrier through SPS and SPPS processes. The submicron and nanometer particles are dispersed into the liquid medium (water or ethanol) or solutions of precursor salts. For SPS process[54-58], the fine feedstock powders are injected into the plasma jet with the assistance of the liquid medium, then the solvent vaporizes in the plasma, and the fine solute forms the individual particles or agglomerates in melting or partial-melting state. Finally, the particles are deposited on the substrate surface. It should be noted that in order to obtain a uniform coating microstructure, the fine powders should disperse into the solvent (water or ethanol) homogeneously. The deposition behavior depends on the size of solute and the agglomerates, thereby influencing the coating microstructure. Fig.4 shows the cross-section microstructures of coatings after different SPS processes[59]. Fig. 4a shows the microstructure with vertical cracks, and the periodic cracks can be observed in the whole cross section. Fig. 4b shows the feather-like columnar structure, which can improve the strain tolerance of TBC system.

For SPPS process, the solution precursor is employed for spraying stock[60-62]. During SPPS process, the solution precursor is injected into the plasma jet. A series of chemical reactions occur, and then the target coating material is synthesized. The coating microstructure depends on the fact whether the material is in the finished state or in the melting state before striking the substrate. The solution precursor employed in SPPS process always shows a high stability with even dispersion[63-65]. TBCs after SPPS show the porous microstructure with nano-grains and evenly spaced segmentation cracks, which improves the thermal insulation performance and prolongs the thermal cycling lifetime. Fig. 5 shows the typical microstructures of coatings after SPPS[66].

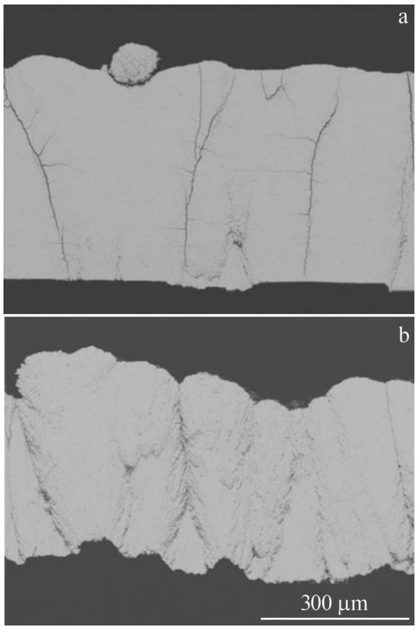

Fig.4 Microstructures of vertically cracked SPS coating (a) and columnar-structured SPS coating (b) [59]

2.3 TBCs after PS-PVD

PS-PVD is a novel plasma spraying technique based on the low pressure plasma spray to deposit the undetectable areas[67-70], which is beneficial for the surface coating with complex structures, such as double vanes. Besides, PS-PVD process can achieve a series of microstructures by adjusting the deposition parameters, such as the quasi-columnar structures and the columnar structures[71]. The microstructure depends on the ratio of gas phase to liquid phase in the plasma flame flow before deposition. The gas phase dominates the desirable columnar structure and the liquid phase tends to form the dense lamellar structure. Fig. 6 shows the cross-section microstructures of coatings after PS-PVD at different spray distances[72]. Fig.6a shows the quasi-columnar-structured coatings with broad inter-columnar gaps and some defects inside the columns after PS-PVD at distance of 600 mm. Fig.6b shows the symmetrical columnar structure with feather features surrounding the columns deposited at 1000 mm in

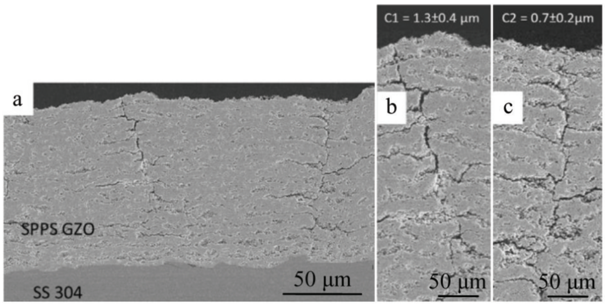

Fig.5 Cross section morphology of $Gd_2Zr_2O_7$ coating after SPPS with two vertical cracks of different thickness (a); magnified images of thick crack (b) and thin crack (c) in Fig.5a[66]

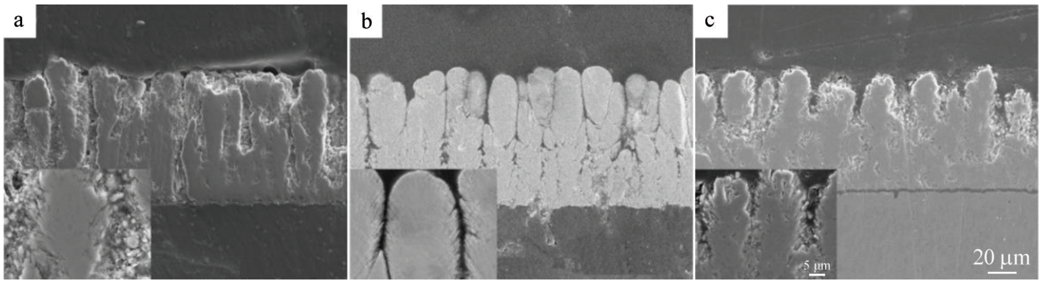

Fig.6 Cross-section microstructures of coatings after PS-PVD at different spray distances: (a) 600 mm, (b) 1000 mm, and (c) 1400 mm[72]

spray distance. With further increasing the deposition distance to 1400 mm, the coating exhibits the nonsymmetrical columns with obvious defects inside the columns.

3 Lifetime of Durable TBCs

Lifetime is also important to the service safety and stability of TBCs, but there is no unified method or standard for TBCs lifetime evaluation. Besides, the experimental lifetime highly depends on the test method and equipment. Nevertheless, the results can still be considered as the reference for the lifetime comparison between the columnar-structured TBCs and the standard TBCs after APS. The advanced test methods are required to conduct under the real service conditions of TBCs. TBCs are heated to a certain temperature, held for a while, and then cooled down to room temperature. However, there is no unified definition for TBC failure. In experiments, TBC lifetime is considered as the number of thermal cycles when the surface peels off to a certain percentage (1%, 10%, or 50%) during thermal cyclic test.

Schulz et al[73] investigated the thermal cyclic lifetime of TBCs prepared by EB-PVD and APS on different substrates, and found that the lifetime of TBCs after EB-PVD is twice longer than that after APS under the thermal exposure coupled with heating at 1100 °C for 50 min and forced air cooling for 10 min. It is proved that EB-PVD is an effective and promising way to acquire excellent strain tolerance and thermal shock capability to prolong the TBC lifetime due to the unique columnar microstructure. The failure mode of TBCs after EB-PVD is the buckling of long small TC strips, which is always initiated near TGO.

Seshadri et al[55] prepared different TBCs with unique mechanical and thermal properties by SPS, and these TBCs had various microstructures, including the porous feather-like columnar structure, narrow columnar structure, wide columnar structure, and segmented lamellar structure. These TBCs were subjected to thermal cyclic tests at 1100 °C, and the results indicated that TBCs with porous feather-like columnar structure show the longest lifetime of about 1000 cycles. Although the standard TBC after APS outperforms the optimal TBCs after SPS, SPS is still a promising process for materials and related applications.

Gell et al[74] reported the spallation mechanisms of TBCs after SPPS under the thermal cycle test at 1121 °C. The lifetime of TBCs after SPPS is 2.5 times longer than that after conventional APS, indicating the better durability even compared with that of TBCs after EB-PVD. The excellent durability of TBCs after SPPS is attributed to the improved strain tolerance caused by the segmentation cracks. The stress reduction of TBCs after SPPS is usually more than 50%.

Rezanka et al[75] reported the thermal cycling lifetime of TBCs prepared by PS-PVD. The thermal cycling tests were conducted in a burner rig at the surface temperature of 1250 °C. The lifetime of TBCs after PS-PVD is at least 2 times higher than that of conventionally sprayed TBCs, which can be attributed to the good stress release capacity of PS-PVD treatment. Rossmann et al[76] concluded that both PS-PVD and EB-PVD can release the stress with the thermal cycles proceeding, and TBCs after PS-PVD exhibit greater stress release capability and slightly lower stress values than those after EB-PVD do.

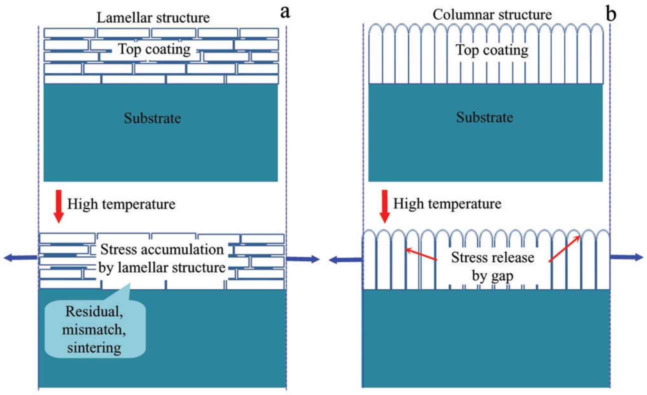

Fig.7 Schematic diagrams of TBCs with lamellar structure (a) and columnar structure (b)

It can be seen from the above analysis that TBCs are mainly employed to protect the key parts of gas turbines from high temperature hot gas. However, the high temperature exposure leads to the healing of pores and microcracks in TC and the performance degradation, especially the elastic modulus and thermal conductivity, i.e., the sintering process will inevitably weaken the strain tolerance of TBCs.

TBCs after APS show great advantage of thermal insulation property, mainly due to their unique lamellar structure combined with intrinsic low thermal conductivity. As shown in Fig. 7a, TC is built-up by the successive stacking of splats, and the introduced cracks and pores during deposition process contribute to the thermal insulation performance. Li et al[41] proposed a multi-splat layered delamination mode to describe the failure mechanism of TBCs after APS. Due to the shrinking-stress generated at TC surface, the lateral cracks connect to the coating surface. With the thermal cycling further proceeding, the stress accumulation promotes the crack propagation until the final multi-splat layered delamination. As for the columnar-structured TBCs (Fig. 7b), the typical characteristics are the gaps between columns. During the high temperature exposure, the heat flux is parallel to the column deposition direction, which generally leads to worse thermal insulation performance of alloys than that of TBCs after APS. However, it is also beneficial to the stress release of TBCs. Because TC is bonded to the substrate, during the thermal cycle tests, the mismatch stress between TC and the substrate can be effectively released by the gaps, resulting in improved thermal insulation performance and durability.

4 Summary and Outlook

The thermal barrier coatings (TBCs) are crucial in the development of advanced aviation and land-based gas turbines. The stress results from the thickening of thermally grown oxide (TGO), volume change of top coat by phase transformation, and the sintering of top coat which dominates the failure of TBCs. The columnar structure of ceramic top coat can improve the service durability of TBCs.

The demand of higher thrust-weight ratio and efficiency of material system should be considered. Although there is no standard for the evaluation of TBC lifetime, the reliability and repeatability of preparation process should also be considered. Therefore, the further study about TBCs should focus on the following aspects:

1) The development of new TBCs material for application at 1400 °C, and the study of related thermal stability, such as phase stability, thermal conductivity, coefficient of thermal expansion, and sintering behavior.

2) The optimization of columnar structure of TBCs by reducing the gaps or surface sealing treatment to improve the corrosion resistance against CaO-MgO-Al_2O_3-SiO_2 (CMAS) for longer service life of TBCs.

3) The combined utilization of coating preparation processes for better thermal insulation performance and longer lifetime at high temperatures.

4) The establishment of lifetime evaluation standard of TBCs for stable TBCs system under the real service conditions.

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耐久性柱状结构热障涂层设计

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摘　要：热障涂层（TBC）被广泛应用于热端金属部件表面，使其免受高温气体侵蚀，其微观组织结构及服役性能与制备工艺和服役状态密切相关并相互作用。本文从热生长氧化物（TGO）增厚、相变、烧结等方面讨论了各种应力诱因导致的TBC失效机理，综述了目前常用于陶瓷层柱状结构制备的方法及其在缓和TBC服役过程中应力的应用。由于TBC制备工艺及服役条件不尽相同，通过与标准TBC比较，评价了不同工艺下制备的TBC使用寿命，总结了柱状结构TBC的长寿命服役机理。本文综述了柱状热障涂层的应力源、微观结构、制备工艺和使用性能的相互作用关系，为高性能TBC设计提供了参考。

关键词：热障涂层；耐久结构设计；柱状结构；寿命

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