第 49 卷第 1 期　　红外与激光工程　　2020 年 1 月
Vol.49 No.1　　Infrared and Laser Engineering　　Jan. 2020

A toughening method of the interface in double-ceramic-layer thermal barrier coating based on selected laser modification

Gao Lei[1*], Li Huiyun[2]

(1. General Standard (Beijing) Institute of Science and Technology, Beijing 100070, China;
2. Shaanxi Preschool Normal University, Xi´an 710100, China)

Abstract: In this study an innovative peg-nail structured laser modification was carried out double-ceramic-layer thermal barrier coating (DCL-TBCs) by a pulsed Nd:YAG laser which were fabricated by a NiCoCrAlYTa bond coat and two ceramic top coat (YSZ and $La_2Ce_2O_7$, LC) sprayed by air plasma spraying. Results indicate that the roughness of the laser modified surface has significantly improved, compared with as-sprayed coatings. The completely recrystallization can be found in the peg-nail structured laser modified unit, including a fully dense columnar microstructure. Due to the peg-nail structured laser modification which generated re-melting and resolidificating at the whole LC coat and partial YSZ coat, can greatly improve the interface bonding property and the bonding strength, the peg-nail structured laser modified samples have better spallation or delamination resistance than the normal as-sprayed DCL-TBCs.

Key words: thermal barrier coating; ceramic interface; laser modify; fracture; toughen

CLC number: TN929.1　　**Document code:** A　　**DOI:** 10.3788/IRLA202049.0105005

基于选择性激光改性的双陶瓷层热障涂层界面增韧方法

高　磊[1*],李慧芸[2]

(1. 环宇通标(北京)科学技术研究院,北京 100070;
2. 陕西学前师范学院,陕西 西安 710100)

摘　要: 该研究通过大气等离子喷涂法制备了粘接层为 NiCoCrAlYTa 合金、陶瓷层为 YSZ 和 $La_2Ce_2O_7$ (LC)的双陶瓷层热障涂层(DCL-TBCs),并提出了一种采用脉冲 Nd:YAG 激光的新型桩钉结构激光改性方法。结果表明,激光改性后,双陶瓷层热障涂层的表面粗糙度较喷涂前有明显提高;在激光改性的桩钉结构单元中可以发现陶瓷层的完全再结晶,以及致密的柱状微结构;由于激光改性构建的桩钉结构使得整个 LC 层和部分 YSZ 层产生了再熔化与再溶解,极大地提高了界面结合性能和结合强度,因此激光改性后的双陶瓷层热障涂层比常规的双陶瓷层热障涂层具有更好的抗脱粘性能。

关键词: 热障涂层;　陶瓷层界面;　激光改性;　断裂;　增韧

收稿日期: 2019-10-11;　**修订日期:** 2019-11-21

基金项目: 陕西省科技计划项目(2017NY-199)

作者简介: 高磊(1971-),男,高级工程师,博士,主要从事分析测试技术及仪器方面的研究。Email:13991160201@139.com

0 Introduction

Thermal barrier coatings (TBCs) are widely applied in gas turbines for aero engines and electricity generators, which can reduce the surface temperature of the substrate and slow down the oxidation, thus improving the durability of metal components and prolonging their service life[1-3]. Typical TBCs consist of superalloy substrates, metal bond coat (BC) and ceramic top coat (TC). Generally, the ceramic top coat uses 7-8 wt.% yttria partially stabilized zirconia (YSZ) which has excellent thermal insulation and high durability[4]. However, due to sintering and phase transformation at high temperature lead to decrease of strain tolerance, increase of Young´s modulus and change of volume during cooling, it cannot be used for a long time above 1 200 ℃[5-6]. Double-Ceramic-Layer Thermal Barrier Coating (DCL-TBC) is one of the most promising thermal protection systems for the next-generation gas turbine to achieve higher operating temperature[6-7]. The difference between them is that there are two ceramic layers deposited on the BC layer above the substrate in DCL-TBCs. The material of ceramic layer at the top should have low thermal conductivity and high phase stability, such as $La_2Zr_2O_7$(LZ) or $La_2Zr_2O_7$ (LC), which act as thermal insulator to protect the inside ceramic layer. The inside ceramic layer is usually made by the traditional TBC materials, i.e. YSZ, as a stress buffer layer to reduce the stress level in the top ceramic layer. This DCL-TBCs system can not only achieve high thermal barrier effect, but also alleviate thermal mismatch stress among components. It is an important development direction of high temperature stability, sintering resistance, high thermal barrier and long life thermal barrier coating system. Many researchers [8-10] have reported that DCL-TBCs have better thermal cycling performance than traditional single ceramic layer TBCs.

Nevertheless, the problem of early delamination failure of DCL-TBCs is still very serious, and its service life needs to be further improved. The design of double-layer ceramic coatings inevitably introduces additional interfaces into the coating system, making it more vulnerable to residual stress. Xu et al.[11] conducted thermal shock tests on DCL-TBCs fabricated by EB-PVD. It was found that interface debonding was the main failure mode. Cheng et al.[12] indicated that horizontal cracks in DCL coatings are the essential reason of coating failure. Because of the existence of temperature gradient in thermal cycling test, the porosity and modulus of elasticity which show different distribution in the thickness of the coating cause the horizontal cracking of the coating.

Some researchers[12-13] have found that the cycle life of coatings can be effectively prolonged by changing the thickness of double ceramic layers. The inside YSZ layer can reduce the stress level in the top ceramic layer by the stress buffer effect which is related to the thickness of the YSZ layer. Chen et al.[14] alleviated the thermal mismatch by adding a mixed layer to the LZ/YSZ system, which improved the life of TBC system. However, none of these methods can solve the problem of interface cracking easily, so the effect of increasing service life is very limited. And in practical engineering applications, the procedures of these methods are somewhat cumbersome and complicated.

Laser glazing is currently recognized as a potential way in improving the life of TBC coating due to the local heating resulting in small heat affected zone, precise operation and fast processing time [15-16]. However, laser glazing will increase the thermal conductivity of the coating, thus weakening the thermal insulation performance. In addition, the surface re-melting of the ceramic layer will introduce more new interfaces that separates the remelted coating from the original coating in the interior, resulting in greater residual stress. And it cannot improve bonding strength between two ceramic layers. Chang [17] designed a peg-nail structured laser modification implemented on traditional single-ceramic-layer TBCs, which improved the thermal shock lifetime around 3-5 times.

In this work, a peg-nail structured laser modification was created on DCL-TBCs for enhancing the thermal shock resistance. The effect of the peg-nail structured laser modification on DCL-TBCs was described.

1 Experiment procedure

1.1 Material preparation and spraying processes

A nickel based superalloy Hastealloy-X was used as substrate, and the dimension is 25.4 mm in diameter and 3 mm in thickness. The coating materials were NiCoCrAlYTa powder (Sulzer Metco Inc., USA, 30-60 μm). Before to spraying, the substrate were cleaned with acetone, and then grit-blasted with 100-mesh alumina grit. After grit blasting, a bond coating was sprayed on the substrate, with a thickness of about 100 μm, by the air plasma spraying (APS) method. Then, a YSZ ceramic layer of about 200 μm was sprayed by APS method. The detailed thermal spray parameters show in Tab.1.

Tab.1 Detailed thermal spray parameters of DCL-TBCs

Parameters	Bond-coat	LC (TC1)	YSZ (TC2)
Gun	Praxair JP-8000	METCO F4	METCO F4
Spraying method	HVOF	APS	APS
Power/kW	-	55	47
Distance/mm	400	120	160
Ar/H_2 (SLPM)	-	40/8	35/20
Feed rate/$g\cdot min^{-1}$	80	45	50
Gun velocity/$mm\cdot s^{-1}$	720	450	500

1.2 Laser modification procedures

A pulsed Nd:YAG laser was used to treat the selected areas of the surfaces of the as-sprayed DCL-TBCs samples. The power and wavelength of the laser are 220 W and 1 064 nm, respectively. During the laser treatment, the input current is selected as 100 A, the pulse width of 3.0 ms, the frequency is 10 Hz, and the laser spot diameter of 1 mm. The peg-nail structured units are distributed on the edges of the sample, as shown in Fig.1.

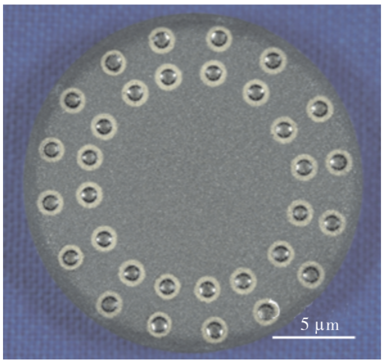


Fig.1 Laser modified plasma sprayed DCL-TBCs sample

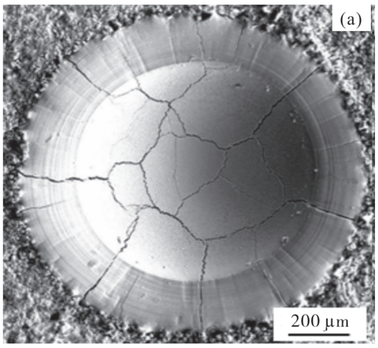
1.3 Thermal shock tests and characterization method

The thermal shock tests were conducted by heating and water quenching method. Both the as-sprayed and the peg-nail structured laser modified samples were treated. The heating time at 1 000 ℃ is 5 min, and the water-cooling time is 1 min. After that, the samples were picked up, dried and checked. The tests were repeated until TBCs failure which was defined as 20% spalling on surface of the coating. The surface morphology of the laser modified samples were observed by a microscope (BX53M, Olympus, Japan). The cross-sectional microstructure were characterized by a scanning electronic microscope (SEM) (Quanta 400, FEI, America).

2 Results and discussion

2.1 Morphological analysis

The surface morphologies of the peg-nail structured laser modified DCL–TBCs are shown in Fig.2. There are significant differences between the morphologies of the as–sprayed and the peg–nail structured laser modified DCL–TBCs. A rough surface with many visible protrusions can be seen in the as–sprayed coating, which is because partially molten particles are less prone to deformation than fully molten particles in plasma spraying process. While, the roughness of the laser modified surface has significantly improved. All the characteristics of the as–sprayed coating are removed after laser modification. A completely re–solidified surface, dense microstructure and a network of continuous cracks can be observed on the peg–nail structured laser modified units. The formation of cracks is attributed to the volume shrinkage and the relaxation of residual stresses during the rapid and non–uniform cooling down of molten zirconia to room temperature, and probably the large and localized temperature gradient which generates residual stresses after the laser passing[18]. According to the studies reported by Raheleh and Chang[15,17], the cracks in laser modified sections helps to enhance the thermal shock life times of the as–sprayed TBCs significantly by the formation of a continuous network of segmented cracks perpendicular to the surface and the increase in strain accommodation. In addition, a slightly flat annular transition region can be seen outside the peg–nail structured laser modified units, which is due to that the subsidence of the central area in the process of laser modification causes the surrounding surface area to deform.



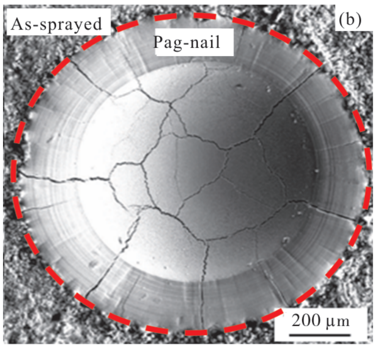


Fig.2 Surface SEM morphology of laser modified plasma sprayed DCL–TBCs sample (The area inside the red dotted line is the peg–nail structure, and the outside is as–sprayed coating)

2.2 Microstructural analysis

Figure 3 shows the cross–sectional microstructure of the peg–nail structured laser modified unit. It can be seen clearly that different layers of DCL–TBC which include bond coat above the substrate, two ceramic layers deposited on the bond coat, and the interface between them. Thickness of two ceramic layers and bond coat is about 200 and 70, respectively. Inside as–sprayed part of coating, many micro–cracks and voids distribute uniformly and randomly. However, the area affected by laser modification is very dense and compact, in which there are no holes or micro–cracks, but only some large segmented cracks perpendicular to the surface. This is because the laser remelts and compacts the as–sprayed coating, eliminating pores and cracks in it, creating what is known as a peg–nail structure. And the cause of crack formation was mentioned in the previous section.

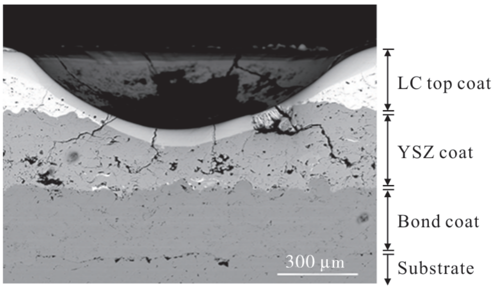


Fig.3 Cross–sectional SEM image of the peg–nail structured laser modified unit

As can be seen, the laser modified region penetrated the whole LC top coat and the partial YSZ coat. These sections occurred remelting and then re-solidification during laser modification, forming the dense peg-nail structure. In this way, the original relative interface between LC and YSZ becomes indistinguishably, and only the interface between YSZ and the peg-nail structured laser modified units is clearly visible.

Figure 4 shows the top surface microstructure of the peg-nail structured laser modified unit. As we can see, the completely recrystallization can be found in the peg-nail structured laser modified unit, including a fully dense columnar microstructure and some segmented cracks. Figure 5 shows the contrast of the cross-section microstructure before and after laser modification. The microstructure orientation from lamellar to columnar is the result of a typical the directed solidification and implies the presence of a thermal temperature gradient across direction of the heat flowing during laser treatment. In addition, the microstructure of the peg-nail structured laser modified units is modified to a combination of a thin discontinuous coarse equiaxed grains on the surface and columnar grains in the cross-section.

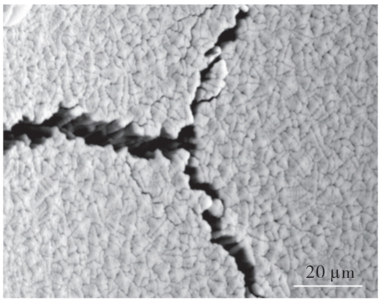


Fig.4 Top surface microstructure of the peg-nail structured laser modified unit

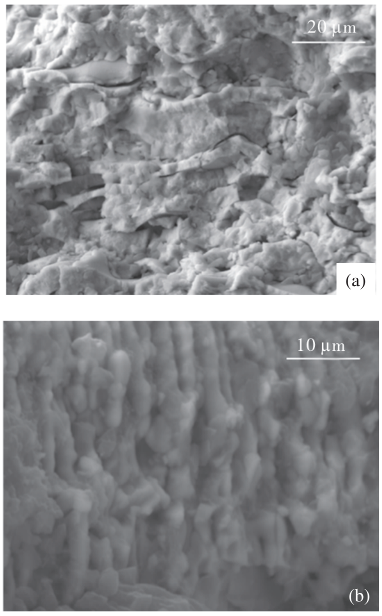
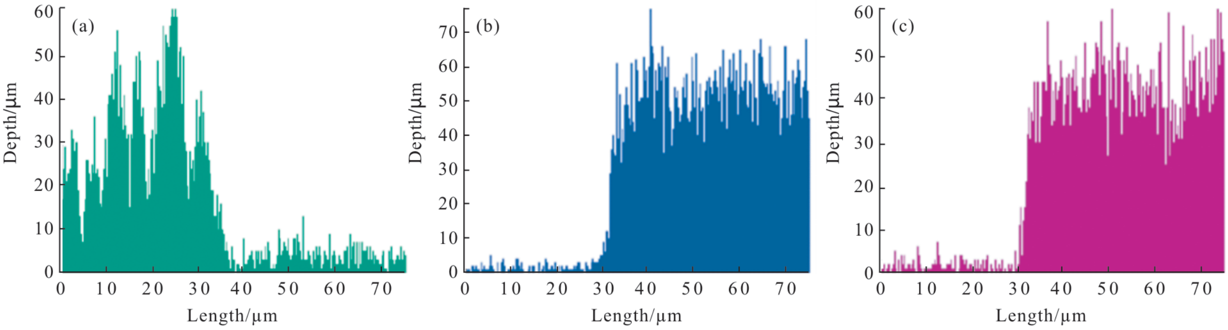


Fig.5 Cross-section microstructure before and after laser modification

Elements analysis is carried out on the cross-section of the peg-nail structured laser modified unit, and the result is shown in Fig.6. The elements analysis shows that the elements have diffused to some degree which is due to the rapid remelting and solidification during laser modification, indicating that the interface between the LC and YSZ coating in the peg-nail structured laser modified unit becomes a partial metallurgical bond.



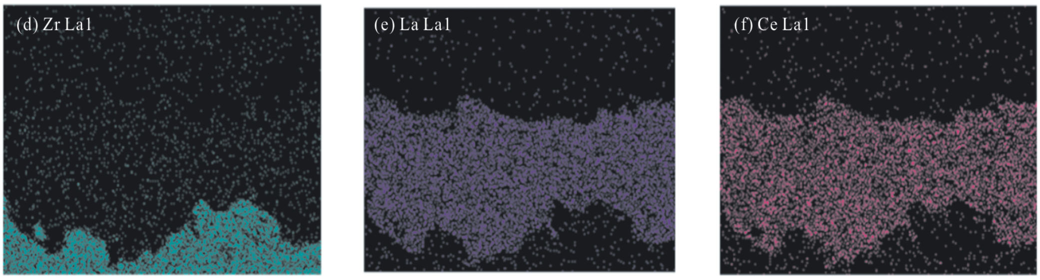


Fig.6 EDS results of the remelted peg-nail structure

2.3 Thermal shock behavior

In order to show the influences of the peg-nail structure laser modification on the thermal shock resistance of DCL-TBCs, the spallation area of two kinds of samples (with and without the peg-nail structure) are recorded in Fig.7. It further demonstrates that the thermal shock resistance of the peg-nail structured laser modified DCL-TBCs is much better than that of the as-sprayed DCL-TBCs: the laser modified sample exhibits an improvement of two times lifetime compared with the as-sprayed samples. It is worth noting that the result of two kinds of samples after 50 times thermal shock was very close. This is because the top coat peel off gradually from the boundary during thermal shock, and the outermost peg-nail structure still has a certain distance from the boundary of the sample. In the short-term thermal shock experiment, the spallation area was small and did not reach the peg-nail structure, which made the peg-nail structure unable to play its role. Therefore, after the 50-times thermal shock, the spallation area of two kinds of samples was very close. And the effect of laser modification was worse than that without the peg-nail structure after the 50-times thermal shock, which was considered to be within the error range.

Figure 8 shows optical microscope picture of the as-sprayed sample after thermal shock, and Fig.9 shows the optical microscope pictures of the peg-nail structured laser modified sample before and after thermal shock. It can be seen from Fig.8 that for the as-sprayed DCL-TBCs sample fracture occurred partly inside the LC top coat while partly at the interface between LC and YSZ

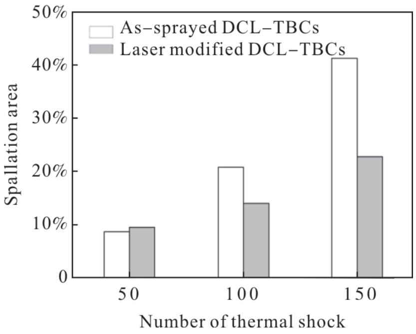


Fig.7 Spallation area ratio of as-sprayed and laser modified samples after different thermal shock cycles

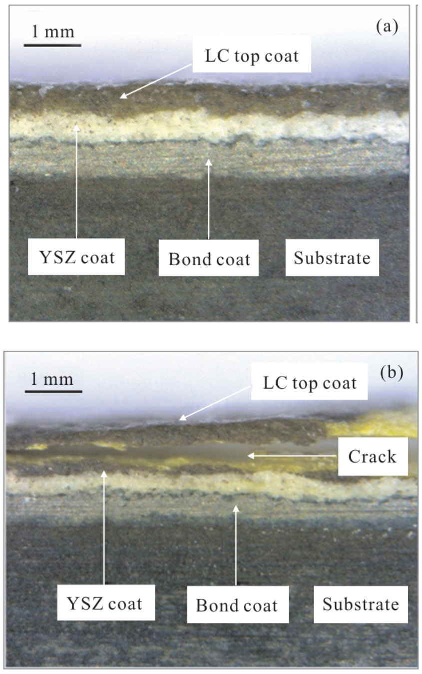


Fig.8 Optical microscope picture of the as-sprayed DCL-TBCs sample after thermal shock

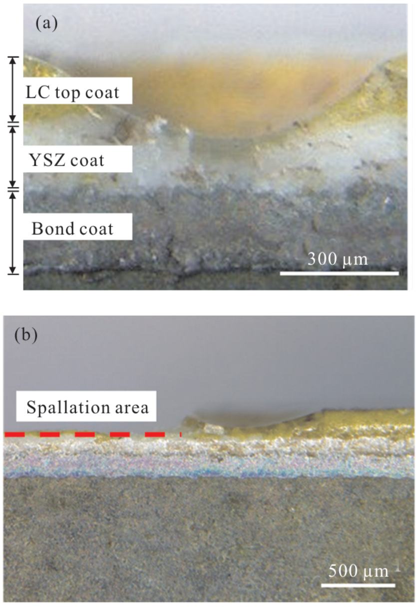


Fig.9 Optical microscope picture of the peg–nail structured laser modified DCL–TBCs sample before and after thermal shock

layer, causing large area spallation and delamination of top coat as a whole, which indicates the relative short thermal shock life. However, in Fig.9, for the laser modified DCL–TBCs sample, the area of fracture at the interface is significantly less than the as–sprayed DCL–TBCs sample, which means the peg–nail structured laser modified units have a marked restraining effect on the growth of interfacial crack. It can be found that the interfacial crack continues to grow until it reaches the peg–nail structured laser modified units. Laser modification can make the interface between the LC top coating and the YSZ coating become a partial metallurgical bond, which can greatly improve the interface bonding property and the bonding strength. The peg–nail structured laser modified samples have better spallation or delamination resistance, which is obvious advantage compared with the normal as–sprayed DCL–TBCs, because the delamination of two ceramic layer is one of the main reasons for the failure of the DCL–TBCs.

3 Conclusions

In this study, microstructure and thermal shock behavior of the plasma sprayed and the peg–nail structured laser modified DCL–TBCs were investigated. A peg–nail structured modification was carried out on double–ceramic–layer thermal barrier coating (DCL–TBCs) by a pulsed Nd:YAG laser. The surface roughness of the coating after laser modification is obviously improved compared with as–sprayed coatings. A fully dense columnar microstructure was obtained in the laser modified ceramic region. A special peg–nail structure was created which can greatly improve the bond strength at the interface and inhibit the expansion of interfacial cracks, so the peg–nail structured laser modified DCL–TBCs had better spallation resistance.

References:

[1] Zhao C, Zhao M, Shahid M, et al. Restrained TGO growth in YSZ/NiCrAlY thermal barrier coatings by modified laser remelting [J]. ***Surface and Coatings Technology***, 2017, 309: 1119–1125.

[2] Wei L, Tong X Y, Li J. Research status of self repairing thermal barrier coating and laser preparation method [J]. ***Information of Surface Engineering***, 2016, 16(1): 17–21.

[3] Zhang P, Li F, Zhang X, et al. Thermal shock resistance of thermal barrier coatings with different surface shapes modified by laser remelting [J]. ***Journal of Thermal Spray Technology***, 2019, 28(3): 417–432.

[4] Batista C, Portinha A, Ribeiro R M, et al. Morphological and microstructural characterization of laser–glazed plasma–sprayed thermal barrier coatings [J]. ***Surface & Coatings Technology***, 2006, 200 (9): 2929–2937.

[5] Basu D, Funke C, Steinbrech R W. Effect of heat treatment on elastic properties of separated thermal barrier coatings [J]. ***Journal of Materials Research***, 1999, 14(12): 4643–4650

[6] Cao X Q, Vassen R, Stoever D. Ceramic materials for

thermal barrier coatings [J]. ***Journal of the European Ceramic Society***, 2004, 24(1): 1–10.

[7] Cao X Q, Vassen R, Tietz F, et al. New double–ceramic–layer thermal barrier coatings based on zirconia–rare earth composite oxides[J]. ***Journal of the European Ceramic Society***, 2006, 26(3): 247–251.

[8] Xu Z, He L, Mu R, et al. Double–ceramic–layer thermal barrier coatings of $La_2Zr_2O_7$/YSZ deposited by electron beam–physical vapor deposition [J]. ***Journal of Alloys and Compounds***, 2009, 473(1–2): 509–515.

[9] Wang L, Wang Y, Sun X G, et al. Finite element simulation of residual stress of double–ceramic–layer $La_2Zr_2O_7$/8YSZ thermal barrier coatings using birth and death element technique [J]. ***Computational Materials Science***, 2012, 53(1): 117–127.

[10] Wang L, Wang Y, Sun X G, et al. Thermal shock behavior of 8YSZ and double–ceramic–layer $La_2Zr_2O_7$/8YSZ thermal barrier coatings fabricated by atmospheric plasma spraying [J]. ***Ceramics International***, 2012, 38 (5): 3595–3606.

[11] Zhenhua X U, Limin H E, Rende M U, et al. Double–ceramic–layer thermal barrier coatings of $La_2Zr_2O_7$/YSZ deposited by electron beam–physical vapor deposition [J]. ***Journal of Alloys & Compounds***, 2009, 473 (1): 509–515.

[12] Cheng B, Yang G J, Zhang Q, et al. Gradient thermal cyclic behavior of $La_2Zr_2O_7$/YSZ DCL–TBCs with equivalent thermal insulation performance [J]. ***Journal of the European Ceramic Society***, 2018, 38(4): 1888–1896.

[13] Han M, Zhou G, Huang J, et al. A parametric study of the double–ceramic–layer thermal barrier coatings part I: Optimization design of the ceramic layer thickness ratio based on the finite element analysis of thermal insulation (take LZ7C3/8YSZ/NiCoAlY DCL–TBC for an example) [J]. ***Surface and Coatings Technology***, 2013, 236: 500–509.

[14] Chen H, Liu Y, Gao Y, et al. Design, preparation, and characterization of graded YSZ/$La_2Zr_2O_7$ thermal barrier coatings [J]. ***Journal of the American Ceramic Society***, 2010, 93(6): 1732–1740.

[15] Ahmadi–Pidani R, Shoja–Razavi R, Mozafarinia R, et al. Improving the thermal shock resistance of plasma sprayed CYSZ thermal barrier coatings by laser surface modification[J]. ***Optics & Lasers in Engineering***, 2012, 50(5): 780–786.

[16] Zhou Y C, Hashida T. Thermal fatigue failure induced by delamination in thermal barrier coating [J]. ***International Journal of Fatigue***, 2002, 24(2–4): 407–417.

[17] Chang F, Zhou K, Tong X, et al. Microstructure and thermal shock resistance of the peg–nail structured TBCs treated by selective laser modification[J]. ***Applied Surface Science***, 2014, 317: 598–606.

[18] Zhu C, Li P, Javed A, et al. An investigation on the microstructure and oxidation behavior of laser remelted air plasma sprayed thermal barrier coatings [J]. ***Surface & Coatings Technology***, 2012, 206(18): 3739–3746.