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## A comparative study on the performance of suspension plasma sprayed thermal barrier coatings with different bond coat systems



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#### ABSTRACT

The performance of suspension plasma sprayed (SPS) yttria stabilized zirconia (YSZ) thermal barrier coatings (TBCs) after isothermal treatment at 1150 °C was investigated. The NiCoCrAlY bond coats were applied by air plasma spray (APS) and high velocity oxygen fuel (HVOF) techniques. It was found that the microstructure of SPS TBCs depends on the surface morphology of the bond coat. The SPS TBCs with a rough APS bond coat exhibited a longer lifetime than those with a smooth HVOF bond coat. To understand this phenomenon, the evolution of the microstructure, mechanical properties and the residual stresses in the TBCs and TGO were systematically studied. Results showed that the surface roughness and oxidation behavior of the bond coat play dominant roles in the SPS TBC failure.

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#### 1. Introduction

Thermal barrier coatings (TBCs) are widely employed in the hot sections of gas turbines to protect the metallic components from high temperatures and to extend the life of the engines [1–5]. A TBC system consists of four layers: i) a ceramic top coat, typically made of 7–8 wt.% yttria-stabilized zirconia (YSZ), ii) a metallic bond coat, iii) a thermally grown oxide (TGO) that forms between the bond coat and the top coat at high temperature, and iv) a superalloy substrate [6,7]. The YSZ top coat is usually deposited by two standard techniques: air plasma spraying (APS) and electron-beam physical vapor deposition (EB-PVD). APS top coats exhibit a laminar structure with microstructural defects parallel to the top coat/bond coat interface. APS TBCs generally have a shorter thermal-cycling life than EB-PVD TBCs, therefore making APS TBCs suitable only for less demanding applications, e.g., industrial gas turbines. EB-PVD top coats by contrast are produced with a columnar structure which can better accommodate the thermal mismatch strain between the metallic substrate and the ceramic top coat, and thus leads to a longer thermal cycling life. However, the EB-PVD technique is much more complicated and expensive. Therefore, it is desirable to develop new techniques which could make coatings with a columnar structure at low cost.

Recently, suspension plasma spraying (SPS) has attracted much attention due to the ability to fabricate coatings with a columnar structure like EB-PVD while maintaining higher insulating porosity levels than APS [8–10]. The SPS technique uses a suspension of sub-micrometer or nano-particles as feedstock [11,12]. This type of microstructure significantly reduces the in-plane Young's modulus and thermal conductivity due to the existence of inter-columnar gaps and nano-sized intracolumnar pores. To date, research on SPS TBCs was mainly focused on the deposition mechanisms [13–16], indentation mechanical properties [17], processing diagnostics [18,19], the comparative study on thermal properties and sintering with APS coating [20] and the thermal cycle testing [9]. Few investigations have been carried out to study the failure behavior after thermal exposure. In addition, the question still remains as to which kind of bond coat is better for SPS TBCs.

The objective of this study is to investigate the failure behavior of SPS TBCs after isothermal treatment, and to understand the effect of different bond coats on the performance of SPS TBCs. The NiCoCrAlY bond coats were first deposited on a superalloy substrate using high-velocity oxygen fuel (HVOF) and APS. Then the YSZ top coat was applied on the bond coat surface using the SPS technique. The microstructure, mechanical properties, and the stress of the SPS TBCs after isothermal exposure were investigated. In addition, the effects of different bond

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coats on the microstructure of the top coat were studied and related to the lifetime of the SPS TBCs,

#### 2. Experiments

Two different TBCs systems were investigated in this work, i.e. 1) SPS TBCs with an APS bond coat, and 2) SPS TBCs with a HVOF bond coat. All the samples were sprayed on grit-blasted Hastelloy-X substrate with dimensions of 30 mm  $\times$  30 mm  $\times$  1.54 mm. The APS and HVOF bond coats were applied by F4-MB gun and D[2600 gun (Sulzer Metco, Switzerland), respectively. Bond coat were based on NiCoCrAlY alloy powder with the same chemical compositions but different size distributions, AMDRY 365-2 and 365-1 (Sultzer Metco, Wohlen, Switzerland) for APS and HVOF processing, respectively. The different size distributions of bond coat powder was used to control the bond coat roughness. The top coat was applied by SPS using the plasma torch 100HE (Progressive Surface, Michigan, USA) and a LiquifeederHE suspension feed system. The 8 wt.% YSZ powder with a diameter of  $d_{50} = 50$  nm was dispersed in an ethanol-based nanofeedstock suspension (solid loading 25 wt.%). The plasma gun consisted of a power of 105 kW, the stand-off distance was 70 mm, the raster step height was 2 mm and the coating deposition rate was about 10 µm/pass. The suspension was fed into the plasma jet at a flow rate of 45 ml/min, using a solid stream nozzle mounted orthogonal to the plasma flow. The target thicknesses of the bond coat and top coat were ~200 µm and ~350 μm, respectively.

The samples were cut into  $10~\text{mm} \times 10~\text{mm}$  plates using a Struers Accutom-5 cutting machine, and were subjected to isothermal heat treatment at 1150 °C with a holding time of 5, 25, 50 and 100 h. At least five samples were tested at each heat treatment condition. And five samples were used to establish the failure modes. The heating and cooling rate was 5 °C/min and 3 °C/min, respectively.

The polished cross-sections of the TBC samples were observed using a scanning electron microscope (Philips XL30) coupled with an energy dispersive spectrometer (EDS, Rontec). All the images were taken in backscattered electron (BSE) mode. The EDS spectrum analysis was carried out using software Esprit 1.8.

The elastic modulus and the hardness of the YSZ top coat were determined using a nano-indenter (MTS<sup>XP</sup>, USA) with a Berkovich tip. The measurements were performed on the polished cross-section of the TBC samples using continuous stiffness module. The indentation depth was assigned as 1000 nm. The strain rate was 0.05 s $^{-1}$ . At least 25 indents were made on each sample with 100  $\mu$ m intervals between adjacent indents in a horizontal direction parallel to the bond coat/top coat interface and at 50  $\mu$ m intervals perpendicular to the bond coat/top coat interface. The hardness and modulus were evaluated from the unloading curve using the Oliver and Pharr method [21]. The Poisson's ratio was set as 0.20 for the YSZ top coat.

The stresses in the YSZ top coat and in the TGO were evaluated using Raman spectroscopy and Photoluminescence piezospectroscopy (PLPS), respectively. They were collected using a Raman microprobe (LabRAM HR Evolution, Horiba) coupled with a 532 nm argon laser. The laser beam was focused on the top surface of the YSZ coating through a  $\times 50$  objective lens. The laser spot size was about 2  $\mu m$  in diameter and the laser power was kept at 25 mW. The PLPS measurement was also performed on the top surface of the YSZ top coat. The stress references for the TGO and YSZ top coat were a single crystal sapphire and free-standing top coat [22,23], respectively. After thermal exposure, the free-standing top coats were obtained by dissolving the substrate in aqua regia for about 10 h. The TGO on the backside of the YSZ top coat was carefully removed by grinding using SiC paper. For both measurements, at least 25 positions were randomly selected from the surface. All the spectra were fitted by a mixed Lorentzian-Gaussian function using the NGSLabspec software to obtain the peak position.

#### 3. Results

#### 3.1. Microstructure

Fig. 1 and Fig. 2 show the microstructure evolution of the SPS TBCs deposited on the APS bond coat and the HVOF bond coat, respectively. The most notable feature is the segmentation structure and the layered porosity bands. The dark layered porosity bands across the sample directly correspond to the number of spraying passes. The roughness of APS and HVOF bond coat have a range of Ra 11-12 µm and Ra 5-6 µm, respectively. There is a significant difference in the microstructure between the top coat deposited on the APS and on the HVOF bond coat. The rough APS bond coat surface leads to a fan-like structure appearing with regularity in the top coat, as shown in Fig. 1a. The layered bands inherited the curved surface profile, suggesting that SPS TBC sprayed onto the rough APS bond coats would accentuate the undulations of the bond coat. After 25 hours of thermal exposure, the cracks transverse the TGO and propagate mainly along the TGO/bond coat interface (insets of Fig. 1b). The TGO formed on the APS bond coat exhibits thickness heterogeneities (e.g., from 1.5 µm to 4.3 µm), and contains mixed oxides. The inter-splat pores and internal oxides were observed in the APS bond coat. The amount of internal oxides in the APS bond coat increases with the thermal exposure time. In addition, more internal oxides form in the APS bond coat at the root of the segmented crack, as shown in Fig. 1c (location A), suggesting that the inter-columnar gaps act as short-circuit for oxygen.

In contrast, the SPS YSZ deposited on the HVOF bond coat shows a relatively smooth surface. Most of the inter-columnar gaps are across the coating thickness and perpendicular to substrate. Compared with APS bond coat, the layered bands in the top coat sprayed on the HVOF bond coat are most parallel to the substrate (Fig. 2a). Some cracks occur at the edges of columns and propagate along the layered porosity, as shown in Fig. 2a. Due to the presence of layered porosity bands, the bonding strength among the spraying passes is low. Cracks could be initiated by the quenching stress during spraying [24]. For example, Karger et al. [24] reported horizontal crack density increases with the increase of thickness of the spraying pass. Such cracks propagate along the layered porosity bands (i.e. splat boundary), which might be caused by the tensile stress induced by the sintering of the YSZ top coat. These cracks, once initiated, could propagate easily and meet each other. In an extreme situation, e.g., near the interface, it would lead to coating spallation. Because the HVOF bond coat is denser than the APS bond coat, no internal oxidation occurs in the bond coat, and the TGO thickness is more uniform compared with the APS bond coat.

Fig. 3 shows the cross-sectional image of the top coat between the layered porosity bands. The column contains a significant amount of inter-splat and intra-splat pores in as-sprayed condition (Fig. 3a). Most intra-splat pores are closed pores, which become spherical after thermal exposure, but will not disappear (Fig. 3b). This is beneficial to maintain the low thermal conductivity of the TBCs. The width of the inter-columnar gap increases with an increase of annealing time, due to the densification of the YSZ columns (Fig. 1d and Fig. 2d). There is no obvious difference of sintering effect on microstructure between the two types of TBCs from the observation of SEM. Previous research reported that the SPS coating has faster sintering rate than conventional APS coating due to the much higher volume fraction of fine pores [20,25,26].

#### 3.2. Failure mode

As shown in Fig. 2c, the delamination of SPS TBC sprayed on HVOF bond coat occurred after 50 hours of isothermal treatments, while the TBC sprayed on APS bond coat is still intact (Fig. 1c). The spallation area of SPS TBC sprayed on HVOF bond coat is ~30%. For both types of samples, the interfacial cracks usually nucleate from the imperfection sites of the interface, e.g., the peaks or valley of the TGO, as indicated in insets of Fig. 1b and 2b. This is very similar to the typical failure

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