The Effects of Temperature and Substrate Curvature on TBC Lifetime and Residual Stress in Alumina Scales beneath APS YSZ

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In order to assess the role of temperature on the lifetime of thermal barrier coatings (TBCs) and the development of residual stresses in Al_2O_3 scales grown under yttria-stabilized zirconia (YSZ) top coatings, two vacuum plasma sprayed (VPS) NiCoCrAlYHf bond coating (BC) compositions, with and without Si, were deposited on Hf-rich directionally-solidified (DS) 247 substrates and then coated with air plasma sprayed (APS) YSZ. Samples were thermally-cycled at temperatures ranging from 1075 to 1150 °C with 1-h cycles in air with 10% H_2O . Photo-stimulated luminescence spectroscopy (PSLS) was used to map residual stresses in the Al_2O_3 scale at the YSZ/BC interface from the same region at regular cycling intervals. All samples exhibited similar stress distributions after 100 1-h cycles with the exception of the YHfSi BC cycled at 1150 °C which had a lower average compressive stress with more delaminations. This specimen also had the shortest TBC lifetime which shows that the PSLS measurements correlate well to interfacial damage accumulation in TBCs. The presence of Si had no effect on lifetime or residual stress but was found to reduce the amount of internal oxidation. The effect of sample curvature on the interfacial stress and TBC lifetime was also assessed by comparing TBC-coated rod specimens to flat buttons. Rod specimens failed much earlier than flat specimens and had lower Al_2O_3 residual stress.

Keywords: photo-stimulated luminescence piezospectroscopy (PLPS); furnace cycle testing (FCT); bond coating; alumina scale; TBC; directionally-solidified superalloy

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1.Introduction

Land-based turbine engines used in natural gas combine cycle (NGCC) plants represent the most efficient and lowest emissions approach to power generation using fossil fuels. Higher turbine efficiency is achieved by increasing the turbine inlet gas temperature, which can decrease the durability of turbine engine materials. The current approach is to use thermal barrier coatings (TBC) on Ni-base superalloys that are internally cooled which reduces the alloy temperature below the level where creep and oxidation are problems [1-3]. The TBC consists of both a metallic bond coating for oxidation resistance and a low thermal conductivity yttria-stabilized zirconia (YSZ) top coating to reduce the metal surface temperature.

Laboratory evaluations of TBCs typically begins with furnace cycle testing (FCT) where a cycle consists of heating a coated coupon in a furnace and holding for a period time (e.g., 1 or 100 h) at a fixed temperature without a temperature gradient across the coating followed by cooling to near room temperature. Our previous work has utilized FCT to study the effect of bond coating composition[4], roughness [5], superalloy composition[6], gas composition[4-10] and cycle frequency [8, 9] on TBC lifetime. However, in some studies the FCT results were not statistically significant and, without a thermal gradient, FCT has limited value for evaluating multi-layer or low thermal conductivity top coatings.

In the present FCT study, bond coating composition, temperature and specimen geometry were varied. Bond coatings with Y, Hf and Si additions were introduced more than 30 years ago [1, 11]. It has been well-established that co-doping of reactive element (RE) additions such as Y and Hf produces better oxidation resistance than Y alone [12-15]. However, the role of Si is less established so coatings were sprayed with commercial powder made with and without Si additions. In addition to coupons, alloy 247 rod specimens were coated[16] in order to move

towards a specimen geometry that was more indicative of turbine hardware and to evaluate the FCT methodology. In addition to cycling to TBC failure, the residual stress in the thermally-grown alumina scale was measured on all samples using photo-stimulated luminescence piezospectroscopy (PLPS). Initial results on rod specimens showed a different residual stress level in the alumina scale as well as surprisingly shorter FCT lifetimes.

2. Material and methods

The superalloy substrate used for all samples was directionally-solidified alloy 247 with a composition shown in Table 1. Superalloy disks (1.59 cm diameter, 2mm thick) and rods (12.5mm diameter, ~30 mm long) were coated at Stony Brook University's Center for Thermal Spray Research (CTSR) and several similar rods were coated at the Jülich Research Center (FZJ). For this study, disks were coated on one side using vacuum plasma spraying (VPS) with two commercial NiCoCrAlYHf powders, one containing 0.61 wt% Si (YHfSi) and the other with almost no Si (YHf). Other than the difference in Si, the bond coating compositions were identical as shown in Table 1. Rod specimens were coated with the same YHfSi powder using the high-velocity oxy-fuel (HVOF) process and air plasma spraying (APS). The surface roughness (R_a) of the rods was measured using a Keyence model VR-3000 optical profilometer to be 8.3 μm which was the same as the flat disks. Following annealing at 1080 °C for 4 h in a 10^{-4} Pa vacuum, a ~200 μm thick APS YSZ top coating was deposited on the bond coatings. For the rod specimens, ~2 mm of bond coating was left exposed at each end of the rod. At FZJ, the bond coating was an oxide dispersion strengthened (ODS) CoNiCrAlYHf powder with 1% additions of HfO_2 and Y_2O_3 , Table 1.

For 1-h cycles, sets of three identical disk specimens were cycled in air with 10 ± 1 vol.% H_2O at 1075, 1100, 1125 or 1150°C. The cycles were performed in an automated vertical furnace rig with the specimens held on alumina rods using Pt-Rh wire and cooled in laboratory air for 10 min to $<30^\circ\text{C}$. For 100-h cycling of the rod specimens, only one rod of each composition was cycled at 1100°C in the same air+10% H_2O environment. The cycling was performed in a horizontal tube furnace with the specimens heated to temperature in flowing argon and the wet air introduced at 1100°C. The specimens were furnace cooled under argon.

As described in detail elsewhere[4, 6-8], residual stress in the Al_2O_3 scale was measured using PLPS through the YSZ coating using a Dilor model XY800 Raman microprobe (Horiba Scientific, Edison, NJ). Two-dimensional maps 670 X 500 μm in size with a step size of 10 μm were collected from the same region of the disks at regular thermal cycling intervals. Stress in the rod samples was collected after 100 h along a line 20 mm long with each measurement separated by 10 μm for a total of 2000 measurements. After failure, samples were mounted in epoxy, polished and imaged using optical microscopy and scanning electron microscopy (SEM).

3. Results and discussion

Fig. 1 shows the average lifetime of the VPS-coated disk samples with APS YSZ and the error bars show one standard deviation. At 1100°C, there was no statistical difference between the YHf and YHfSi bond coating lifetimes, and both were similar to the average lifetime previously obtained for HVOF YHfSi bond coatings [9]. Both VPS coatings also were evaluated at 1125°C but, while the average lifetimes were lower than at 1100°C, the effects of Si and temperature again were not statistically significant. To further study the effect of temperature, one group of YHf specimens was cycled at 1075°C and one group of YHfSi specimens was

cycled at 1150°C. Compared to the 1125°C results, these changes in temperature did produce statistically-significant differences and, as expected, the average TBC lifetime dropped with increasing temperature. It should be noted that only 3 specimens of each type were evaluated at each condition and larger groups might be better able to differentiate variables using FCT.

Fig. 2 shows PLPS stress histograms for both VPS bond coatings at each temperature following 100 1-h cycles. The stress in the scale on the YHf coating measured at room temperature increased following furnace cycling from 1075 to 1125 °C. This is likely due to greater thermal expansion mismatch stresses being generated between the substrate and the scale upon cooling to room temperature. A similar effect occurred with the scale on the YHfSi bond coating which was under a slightly higher stress at 1125 °C than 1100 °C. However, this trend did not continue with the scale on the YHfSi bond coating cycled at 1150 °C which decreased significantly in compressive stress after 100 cycles, which represented >50% of the average lifetime for this specimen, Fig. 1. Also, the stress distribution was much broader at 1150 °C. The stress relief in the scale was due to damage accumulation during thermal cycling as has been reported previously [17-20]. Only slight differences in the histograms were observed between the YHf and YHfSi coatings at 1100 and 1125°C.

To illustrate the effects of time and temperature on the alumina residual stress, Fig. 3 shows scale stress maps collected from the YHfSi bond coating cycled for 5, 100 and 300 1-h cycles at 1100 °C (Figs. 3A-C) and 100 1-h cycles at 1150 °C (Fig. 3D). All of the stress maps showed low-stress circular regions (blue in Fig. 3) scattered about a higher stress field. The low stress regions correspond to the peaks at the bond coating interface where out-of-plane tensile stresses reduce the local hydrostatic compressive stress in the scale and may lead to scale cracking and stress relief [6, 8]. The frequency and size of these regions appears stable with

thermal cycling duration and temperature which shows that the effect of roughness on the scale stress is localized. In contrast, the scale stress in the flatter regions gradually declines in stress as damage accumulates in the scale from thermal cycling. The scale stress after 100 cycles at 1150 °C (Fig. 3D) is lower than that at 1100 °C (Fig. 3B) showing that higher cycling temperatures increases the rate of damage accumulation in the scale thereby hastening TBC failure. The results of Fig. 3 show the importance of collecting stress maps from a single location to better characterize the interfacial microstructure, as well as the opportunity to use PLPS as a method for predicting remaining lifetime of TBCs.

The failure of the VPS-sprayed coatings also was characterized using polished cross-sections. Images after failure at 1125°C are shown in Figure 4. One difference between the YHf and YHfSi coatings is an increase in the amount of internal oxidation or “pegging” at the bond coating-alumina interface for the Si free coatings. A similar but less distinct difference was observed after failure at 1100°C. Figure 5 shows a back-scattered electron image taken at the scale/bond coating interface of the YHf sample after failure occurred at 440 cycles at 1125 °C. The oxide peg penetrated into the bond coating and contained lighter-contrast phases that were Hf-rich oxides. Pegging is typical with reactive element (RE) additions, especially with a soluble addition like Hf [21]. Recently Gleeson and co-workers suggested that Si may reduce the activity of Hf, thereby reducing the internal oxidation [22]. However, the difference in internal oxidation did not appear to affect lifetime and is removed from the cracking at failure. A smoother scale with less pegging may be expected to have a higher residual stress with less out-of-plane tensile stresses. The YHfSi specimen did show a slightly higher stress at 1100°C compared to the YHf specimen. However, very little difference in stress was observed at 1125°C.

Because the rod specimens were much larger than the disk specimens, they could not be suspended for 1-h FCT. Therefore, the rod specimens were evaluated in 100-h cycles. Figure 6 compares the lifetime of three different rod specimens (single specimens, not an average) to previous work on disk specimens with a HVOF YHfSi bond coating. While it was concluded that the disk specimens failed due to interdiffusion (i.e. Al depletion from the bond coating) after an average lifetime (5 specimens) of 2300 h, the rod specimens failed after only 4-6 cycles (400-600 h). The rod specimens included YHfSi bond coatings sprayed by APS and HVOF at CTSR. In addition, a rod specimen (alloy 247 rod from the same batch) with a HVOF ODS CoNiCrAlYHf bond coating and APS YSZ top coating was sprayed at FZJ. Fig. 7 shows the cross-section of the failed HVOF-coated rod from CTSR after 4 100-h cycles at 1100 °C, which exhibited a similar failure location to the flat VPS samples (see Fig. 4).

The reason for the early failure of the rod specimens is not clear and is being evaluated. Fig. 8a shows box plots of the compressive residual stress in the scale for a disk specimen measured during 100-h cycling at 1100 °C in air with 10% H₂O. The residual stress in the scale gradually decreased with thermal cycling, which mirrors the results shown in Fig. 3. The decline is due to damage accumulation in the scale which eventually leads to TBC failure which occurred after 2100 h for this sample.

Fig. 8b compares the alumina residual stress in the disk specimen after one 100-h cycle to the stress measured in the 3 rods after the same exposure at 1100°C. The residual stress in the scale grown on the HVOF and APS rods was significantly lower than the stress in the disk specimen. In contrast, the residual stress on the rod with the ODS CoNiCrAlYHf bond coating had a higher median value than the others. This difference could be due to the dispersion strengthening improving the creep strength of the coating, the CoNiCrAlYHf matrix compared to

NiCoCrAlYHfSi or different spray parameters used at FZJ. Future work will compare a conventional CoNiCrAlYHf bond coating to the ODS version. Nevertheless, the coating life of both HVOF rod specimens made at CTSR and FZJ was 400 h. Unfortunately, since the lifetimes of the rods were unexpectedly short, there was no opportunity to develop a stress vs. time plot as shown in Figure 8a.

4. Conclusion

Alloy 247 disk specimens with VPS NiCoCrAlYHf bond coatings with and without Si additions and APS YSZ top coatings were evaluated at different temperatures using 1-h cycles in air with 10% H₂O. At 1100 and 1125°C, there was no statistically-significant change in the average lifetime or difference in the alumina residual stress with and without Si in the bond coating. However, more oxide pegs were observed in the coating without Si, particularly at 1125°C. When the 1-h cycles were conducted at 1075°C or 1150°C, the average lifetimes were statistically different than 1125°C with the expected drop in lifetime with increasing cycle temperature. In order to begin exploring the effect of specimen geometry on coating performance and alumina residual stress, alloy 247 rod specimens also were evaluated using 100-h cycles in air with 10% H₂O at 1100°C. The rod specimens failed much more quickly than similar disk specimens and the reason for the early failure is currently being explored. An analysis of the stress state in the TBC on the rod and disk specimens is being conducted and different YSZ microstructures are being deposited to determine their effect on TBC lifetime on the rod specimens.

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Table 1. Chemical compositions (weight % or ppmw) determined by inductively coupled plasma analysis and combustion analysis.

Material	Ni	Cr	Al	Re	Co	W	Ta	Mo	Ti	C	Hf	Other (ppmw)
VPS YHf	44.7	17.9	12.8	<	23.4	<	<	<	<	<	0.39	6700 Y, 200 Si
VPS YHfSi	44.2	17.7	13.1	<	23.2	<	<	<	<	<	0.38	6900 Y, 6100 Si
HVOF	48.0	16.7	12.3	<	21.6	0.01	<	<	<	<	0.25	6820 Y, 3600 Si, 2 S
ODS	31.2	20.3	8.18	<	37.6	<	<	<	<	0.3	<	13080 Y, 7410 Hf, 2600 Fe
247	59.1	8.5	5.6	.02	10.2	10.0	3.2	0.6	1.0	0.2	1.32	<3 Y, 600 Si, 11 S

< Indicates below the detectability limit of <0.01%.

List of figure captions

Figure 1. TBC lifetimes following 1-h cycles in air + 10 vol% H₂O versus cycling temperature. The data points at 1100 and 1125 °C are offset for clarity. The error bars are the standard deviation of 3 measurements.

Figure 2. Histograms of the Al₂O₃ scale stress following 100 1-h cycles at different temperatures. Solid lines and dashed lines correspond to the YHf and YHfSi bond coatings, respectively. The 3468 stress measurements were binned in groups of 50 MPa.

Figure 3. Two-dimensional scale stress maps collected on the YHfSi bond coating after (a) 5 cycles, (b) 100 cycles and (c) 300 cycles at 1100 °C and (d) 100 cycles at 1150 °C.

Figure 4. Light optical microscope images collected on (a) a failed YHfSi TBC after 280 cycles at 1125 °C and (b) a failed YHf TBC after 300 cycles at 1125 °C.

Fig. 5. SEM image of a peg at the scale bond coating interface of the YHf coating after 300 cycles at 1125 °C.

Figure 6. Lifetimes of flat HVOF buttons and rods following 100-h cycles at 1100 °C in air + 10% H₂O. The error bars for the flat data is the average of 5 measurements. Only 1 sample was cycled for each of the three rod specimens.

Figure 7. Light optical microscope image collected on a failed HVOF TBC on a rod after 4100-h cycles at 1100 °C.

Figure 8. Box plots of compressive stress in the scale (a) versus hours at 1100 °C in 10% H₂O during 100-h cycles for the flat specimen with an HVOF bond coating, and of (b) the flat HVOF compared to the three rod specimens following 100 h at 1100 °C. The error bars correspond to the 99th and 1st percentiles of the stress measurements.

Figure 1

Figure 2

Figure 3

Figure 4a

Figure 4b

Figure 5

Figure 6

Figure 7

Figure 8