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Performance of Vacuum Plasma Spray and HVOF Bond Coatings at 900° and 1100°C

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Abstract

The effects of Ti and B additions to a vacuum plasma sprayed (VPS) NiCoCrAlYHfSi bond coating on thermal barrier coating (TBC) performance were studied at 1100 °C and 900 °C and compared to high-velocity oxy-fuel (HVOF) bond coatings. Using alloy 247 substrates and air plasma sprayed Y_2O_3-stabilized ZrO_2 top coatings, additions of B or Ti+B did not improve the average TBC lifetime in 1-h cycles at 1100 °C in air with 10% H_2O. The addition of Ti resulted in a decrease in lifetime. Photo-stimulated luminescence spectroscopy was used to map residual stresses in the thermally-grown Al_2O_3 scale. At 900 °C, closer to a typical land based turbine operating bond coating temperature, specimens were examined after ten 500-h cycles in laboratory air and air with 10%H_2O to study the effect of H_2O. The addition of water vapor had little effect on the measured parabolic rate constants at 900 °C and a comparison of the oxide microstructures in both environments is reported.

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

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1. Introduction

Natural gas combined cycle (NGCC) power plants are the largest current source of electricity in the U.S. and improving the efficiency of the gas turbine is the least expensive strategy to reduce CO_2 emissions (i.e. the "least regret" strategy [1]). Higher gas turbine efficiencies are achieved by increasing the inlet gas temperatures. However, this can lead to decreased lifetime of the thermal barrier coatings (TBCs) that protect the hottest section of the turbine [2-4]. For land-based gas turbines, these coatings consist of a metallic bond coating for oxidation resistance and a yttria-stabilized zirconia (YSZ) top coating with low thermal conductivity.

In searching for a more durable bond coating, a recent study looked at boron additions to cast NiCrAlYHf alloys and observed a potential benefit in cyclic oxidation resistance in wet air [5]. Given that, the current study looked at the effect of B and/or Ti additions to a base NiCoCrAlYHfSi [6-8] bond coating. Titanium could be incorporated into the bond coating due to interdiffusion with the superalloy substrate [9] but Ti prefers to form γ' rather than forming a boride (TiB or TiB_2) [5] as was observed in FeCrAl alloys [10]. Vacuum plasma spray (VPS) processing was used in order to minimize the oxidation of B during deposition. Following an established methodology [6, 9, 11] the average TBC lifetime was evaluated in 1-h furnace cycle testing (FCT) at 1100 °C in a gas mixture of air+10%H_2O to account for the presence of water vapor in the turbine combustion environment. In addition, the stress in the alumina scale formed during oxidation was characterized during cycling using photo-stimulated luminescence piezospectroscopy (PLPS). Several studies have examined the detrimental role of water vapor on TBC life at 1100 °C [6, 9, 11-13]. However, this test temperature is much higher than the operating metal temperature in a land-based turbine. Thus, a second set of experiments was conducted on the same alloys at 900 °C using 500-h cycles in laboratory air and a gas mixture of air and 10%H_2O in order to study the effect of water vapor closer to the actual operating temperature.

2. Material and methods

Directionally-solidified 247 superalloy[1] disks (1.59 cm diameter and 2 mm thick) were coated on one side at Stony Brook University's Center for Thermal Spray Research (CTSR) with six different bond coating compositions listed in Table 1. All VPS bond coatings contained between 0.59 and 0.70 wt% Y and between 0.38 and 0.40 wt% Hf as a starting composition. In addition to this YHf bond coating, bond coatings containing 0.61 wt% Si (YHfSi), 0.60 wt% Si and 0.31 wt% Ti (YHfSiTi), 0.62 wt% Si and 0.065 wt% B (YHfSiB), and 0.48 wt% Si, 0.33 wt% Ti and 0.077 wt% B (YHfSiTiB) were vacuum plasma sprayed. For comparison to the VPS version, the YHfSi powder also was deposited using the high-velocity oxy-fuel (HVOF) process. The VPS spraying occurred in two batches with a group of YHfSi bond coatings sprayed in each batch. Following annealing at 1080 °C for 4 h in a 10^{-4} Pa vacuum, a ~200 µm thick APS YSZ top coating was deposited using a Metco F4MB torch on the bond coatings. The YSZ porosity was 12.2 ± 1.30 %.

To determine average TBC lifetime, three identical disk specimens of each bond coating composition were cycled in air with 10 vol% H_2O at 1100 °C in 1-h cycles. The specimens were held on alumina rods using a Pt-Rh wire in an automated vertical furnace rig and the specimens were cooled in laboratory air for 10 min to <30 °C for each cycle. The failure criterion was

[1] Certain commercial materials or equipment or services are identified in this paper to adequately describe the experimental procedure. This does not imply their endorsement by the Oak Ridge National Laboratory, UT-Battelle LLC, and the US Department of Energy nor that these materials and equipment and services are necessarily the best choices for these purposes.

>20% coating loss. At 900 °C, specimens were exposed in 500-h cycles in laboratory air and air with 10 vol% H_2O and were not tested to coating failure. In this case, the HVOF YHfSi bond coating was applied to a single crystal PWA 1483 superalloy substrate; all the other specimens were on directionally-solidified (DS) Mar M 247.

A Raman microprobe (Dilor model XY800, Horiba Scientific, Edison, NJ) with a Ar^+ laser operating at 5145 Å was used to measure the mean hydrostatic stress in the Al_2O_3 scale non-destructively through the YSZ top coating [14, 15]. 3468 measurements were collected across a two-dimensional map 670 X 500 μm in size with a step size of 10 μm. The same area near the center of the specimen was measured on each specimen following subsequent thermal cycling which allowed the stress evolution to be measured. After coating failure or exposure at 900 °C, specimens were mounted in epoxy, polished and imaged using light microscopy and scanning electron microscopy.

3. Results and discussion

3.1 Furnace cycle testing at 1100 °C

Fig. 1 shows the average TBC lifetime, in cycles to failure, of seven groups of specimens cycled at 1100 °C in air + 10 vol% H_2O in 1-h cycles. There was no statistically-significant difference between the lifetimes of the three bond coatings from the first VPS batch. As reported previously [14], there was no difference in lifetime between the bond coatings with and without Si. In batch 2 of the VPS coatings, the bond coating that contained Ti had a lower lifetime than the other two coatings. The two VPS YHfSi bond coatings had similar average lifetimes that were both higher than the lifetime for HVOF YHfSi. The B and Ti+B additions did not improve the lifetime compared to the YHfSi baseline coatings. Subsequent experiments [14, 16] have used rod specimens [17] because of the FCT results on disk specimens where edge effects may be contributing to the TBC failure [6, 11].

Fig. 2 shows PLPS stress histograms at 20 and 200 cycles for one specimen of each of the bond coating batches. At 20 cycles, all the histograms peaked around 1 GPa of residual compressive stress in the thermally-grown α-Al_2O_3 scale but at 200 cycles the stress profiles diverged with the batch 1 samples (solid lines) shifting towards lower compressive stress and the batch 2 samples (dashed lines) holding at 1 GPa or increasing in compressive stress as seen with YHfSi. The two YHfSi bond coatings from the two batches showed the largest difference in the mode (or peak) stress of any two samples, 0.2 GPa, at 200 cycles despite being the same composition. The lower lifetime of the YHfSiTi sample shown in Fig. 1 did not correspond to a significant difference in the stress in Fig. 2. The stress relief in the scale observed in the batch 1 sample was likely due to damage accumulation during thermal cycling [18-21].

Fig. 3 shows stress maps collected from the first and second batch YHfSi specimens after 300 cycles at 1100 °C. Consistent with the histograms, the scale on the YHfSi coating from the second batch is under higher compressive stress than the coating from the first batch. It is speculated that this is due to more damage accumulation in the batch 1 sample during cycling leading to less compressive stress in the oxide scale. Both coatings contain regions of near-zero stress which show up as blue regions in the maps. Typically, these are caused by stress relief from cracking around asperities at the bond coating/YSZ interface caused by out of plane tensile stresses during thermal cycling [9].

Fig. 4 shows back-scatter electron images of the scale formed on YHfSi samples from batches 1 and 2 after failure at 1100 °C. The scale from the first batch looks similar to that formed on HVOF YHfSi specimens [22] in terms of oxide thickness. The second batch has a very thick alumina scale with Y- and Hf-rich oxide incorporation throughout the scale and with protrusions of oxide into the bond coating which is indicative of internal oxidation. This is consistent with a prior observation of a bond coating with higher Y and Hf contents that was combined with a low oxygen content in an as-deposited VPS coating [23]. With a high O content in an APS or HVOF coating, the Y and Hf are oxidized during deposition and are relatively immobile during thermal cycling and the formation of the oxide scale. With a lower O content, more Y and Hf remain unoxidized in the coating, presumably in solution or as Ni-rich yttrides [24-26]. During oxidation at 1100 °C, the Y and Hf diffuse toward the bond coating surface and become incorporated into the growing alumina scale, resulting in faster growth due to rapid transport through the RE-rich oxide precipitates or along the alumina-precipitate interface [27]. The differences between the VPS batches suggests that a better vacuum was achieved in batch 2 than in batch 1. An outcome of this comparison suggests that spray powders with higher Y and Hf contents will oxidize more rapidly when deposited by higher vacuum VPS applications. Further characterization is in progress to attempt to measure the O content in these two bond coatings. Whereas FCT did not reveal a difference between these two coatings in TBC lifetime, PLPS was able to detect a significant difference in the stress which further illustrates its usefulness as a non-destructive characterization tool.

Fig. 5 quantifies the alumina scale and β-NiAl depletion zone thickness of all seven bond coatings after failure at 1100 °C. As noted in Fig. 4, the scale formed on the batch 2 VPS coatings was clearly thicker than batch 1 and this led to a thicker β depletion zone since more Al was consumed from the coating to form Al_2O_3. As seen in Fig. 1, despite the batch differences, there was little effect on the TBC lifetime. However, note these measurements were not performed at a constant time but after failure. Thus, the YHfSiTi coating had similar β depletion after a shorter lifetime than the other batch 2 coatings.

3.2 Furnace cycle testing at 900 °C

For the set experiments at 900°C, Fig. 6 shows the specimen mass gain versus exposure time for specimens with four different bond coatings exposed to 500 h cycles in dry air and air with 10% H_2O. After an initial transient mass gain during the first cycle, the rates of subsequent mass gain were all very similar. The mass gains were slightly lower for the HVOF YHfSi coatings on the 1483 alloy. The slight mass loss for the specimen exposed in wet air at 4500 h could be caused by spallation of the uncoated coupon edge [6, 28]. The results suggest that there was no significant effect of water vapor on the scale growth rate at 900 °C for these coatings with all of the rate constants near 2-3x10^{-13} g^2/cm^4s.

Specimens were removed from testing after 5,000 h at 900 °C without coating failure and were metallographically prepared. Figs. 7a and 7b show the scale on the VPS YHfSiTiB specimen cycled in dry air and air + 10% H_2O, respectively. There is no noticeable difference in the scale thickness or morphology, Figure 7. Interestingly, the scale on this Ti-B containing batch 1 VPS coating at 900 °C appears similar in morphology to the batch 2 VPS coatings at 1100 °C, Fig. 5. The scale on the HVOF bond coating (Fig. 7c) after 5,000 h in wet air at 900 °C was much thinner and more uniform than the VPS coating (Fig. 7b) which is likely due to less internal oxidation during exposure for this sample. As part of an effort to develop an Al depletion model for these VPS coatings, several longer-term exposures were conducted at 900 °C in wet air. Fig. 7d shows the scale formed after 10,000 h. As expected, it is thicker than that formed after 5,000 h, Fig. 7b. Fig. 8 shows the thickness of the scale and depletion zone measured from images like those shown in Fig. 7. Only the YHfSi specimens showed a significant difference in the scale thickness between the two atmospheres with the wet sample being thicker although this did not show up as a change in the growth rate in Fig. 6.

4. Conclusion

TBCs on 247 alloy substrates with VPS NiCoCrAlYHf bond coatings with varying additions of Ti and B were exposed to 1-h cycles at 1100 °C in air with 10% H_2O and to 500-h cycles at 900°C in laboratory air and air with 10%H_2O. No statistically significant effect of bond coating composition on TBC lifetime was observed except for the Ti containing specimens which failed sooner than two other coatings sprayed in batch 2. Thus, no beneficial effect of B on TBC lifetime was found in these experiments. The largest difference in oxide scale microstructure occurred between the two VPS batches. It is speculated that the O content of the second VPS batch was lower, resulting in free Y and Hf becoming more heavily incorporated in the alumina scale and accelerating its growth. This difference between the two VPS batches did not strongly affect the average lifetime of the coatings, however, PLPS did non-destructively reveal a higher compressive scale stress in the batch 2 samples. Specimens cycled at 900°C for 5,000 h showed similar parabolic rate constants in both laboratory air and air with 10%H_2O indicating that the addition of water vapor did not affect the scale growth rate.

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

Material	Ni	Co	Cr	Al	Y	Hf	Si	Ti	B	W	Ta	Mo	C	Other (ppmw)
VPS YHf	44.7	23.4	17.9	12.8	0.67	0.39	0.02	0.004	0.001	<	<	<	<	<3 S, 300 Fe
VPS YHfSi	44.2	23.2	17.7	13.1	0.69	0.38	0.61	<	0.001	<	<	<	<	10 S, 300 Fe
VPS YHfSiTi	43.4	22.9	17.6	13.7	0.70	0.38	0.60	0.31	0.003	<	<	<	<	9 S
VPS YHfSiB	44.0	23.1	17.9	13.1	0.59	0.40	0.62	0.07	0.065	<	<	<	<	11 S
VPS YHfSiTiB	43.8	23.2	18.0	13.1	0.59	0.38	0.48	0.33	0.077	<	<	<	<	13 S
HVOF YHfSi	48.0	21.6	16.7	12.3	0.682	0.25	0.36	<	<	0.01	<	<	<	2 S
Alloy 247	59.1	10.2	8.5	5.6	<0.0003	1.32	0.06	1.0		10.0	3.2	0.6	0.2	200 Re, 11 S
Alloy 1483	60.6	8.8	12.0	3.4	<0.0003	0.002	0.004	4.0		4.1	5.2	1.9	0.07	<1 S

< indicates below the detectability limit of <0.01%.

List of figure captions

Figure 1. TBC lifetimes (cycles to failure) following 1-h cycles in air + 10 vol% H_2O versus cycling temperature. The error bars are one standard deviation of 3 measurements about the mean value.

Figure 2. Hydrostatic stress histograms of the Al_2O_3 scale stress following (a) 20 1-h cycles and (b) 200 1-h cycles. Solid lines and dashed lines correspond to specimens from batch 1 and batch 2, respectively. The 3468 stress measurements were binned in groups of 50 MPa.

Figure 3. Two-dimensional scale stress maps collected on the VPS YHfSi bond coating after 300 cycles at 1100 °C sprayed during (a) batch 1 and (b) batch 2.

Figure 4. Back-scatter scanning electron microscopy images collected on (a) a failed YHfSi TBC from batch 1 after 640 cycles at 1100 °C and (b) a failed YHfSi TBC from batch 2 after 600 cycles at 1100 °C.

Figure 5. Box plots of the (a) alumina scale thickness and (b) β depletion zone thickness after failure. Each box was determined by measuring 20 locations from three speciemsn each (total of 60 measurements).

Figure 6. Specimen mass gains versus exposure time at 900 °C for 4 bond coating compositions. The solid and open symbols represent cycling in dry and wet air, respectively. A second line for a VPS YHfSiTiB bond coating that was cycled for 20 500-h cycles (total of 10,000 h) is also shown.

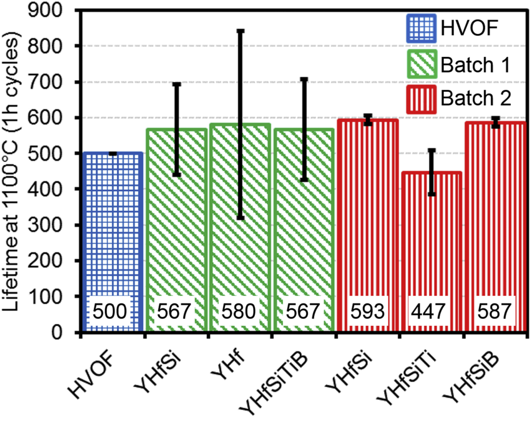
Figure 7. Light optical microscopy images of the YSZ/bond coating interafce following 10 500-h cycles at 900 °C of (a) the VPS YHfSiTiB bond coating on 247 in dry air, (b) the VPS

YHfSiTiB on 247 in wet air, and (c) the HVOF bond coating on 1483 in wet air. Also shown is (d) the VPS YHfSiTiB bond coating on 247 in wet air after 20 500-h cycles at 900 °C.

Figure 8. Box plots of the (a) alumina scale thickness and (b) β depletion zone thickness after 5000 h of exposure at 900 °C. The filled and open boxes correspond to heating in dry and wet air, respectively. An additional box for a VPS YHfSiTiB bond coating that was cycled for 20 500-h cycles (total of 10,000 h) is also shown. Each box was determined by measuring 20 locations from three speciemsn each (total of 60 measurements).

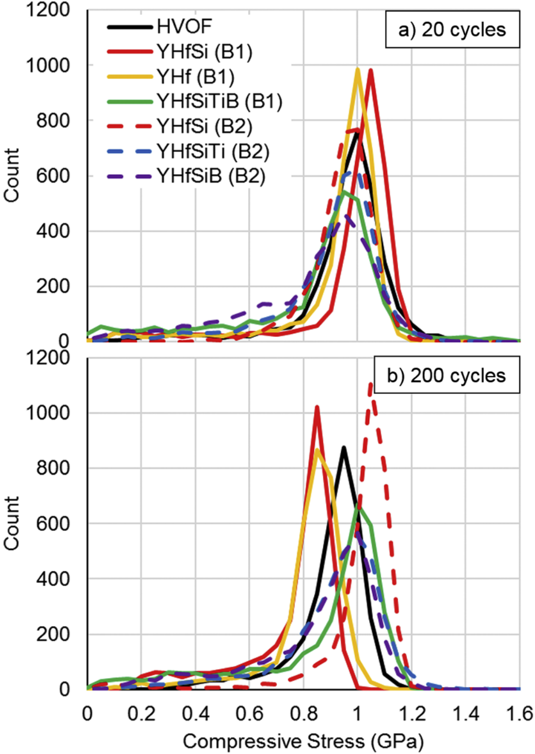
Highlights

- The effect of Ti+B in the BC on TBC lifetime was studied on DS247 in air + 10%H_2O.
- Cycling at 900°C in dry and wet air had the same scale growth rate and morphology.
- PLPS stress maps were collected on oxide scales beneath the YSZ top coating.



900
800
700
600
500
400
300
200
100
0
Lifetime at 1100°C (1h cycles)
HVOF
Batch 1
Batch 2
500
567
580
567
593
447
587
HVOF
YHfSi
YHf
YHfSiTiB
YHfSi
YHfSiTi
YHfSiB

Figure 1



HVOF
YHfSi (B1)
YHf (B1)
YHfSiTiB (B1)
YHfSi (B2)
YHfSiTi (B2)
YHfSiB (B2)
a) 20 cycles
b) 200 cycles
Count
Compressive Stress (GPa)
0
200
400
600
800
1000
1200
0
0.2
0.4
0.6
0.8
1
1.2
1.4
1.6

Figure 2

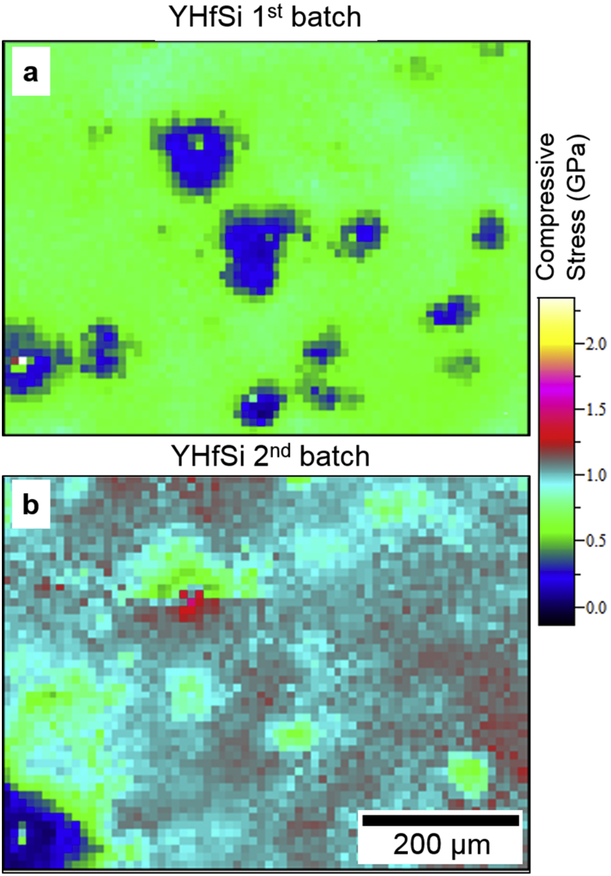

YHfSi 1st batch
a
YHfSi 2nd batch
b
200 µm
Compressive Stress (GPa)
2.0
1.5
1.0
0.5
0.0

Figure 3

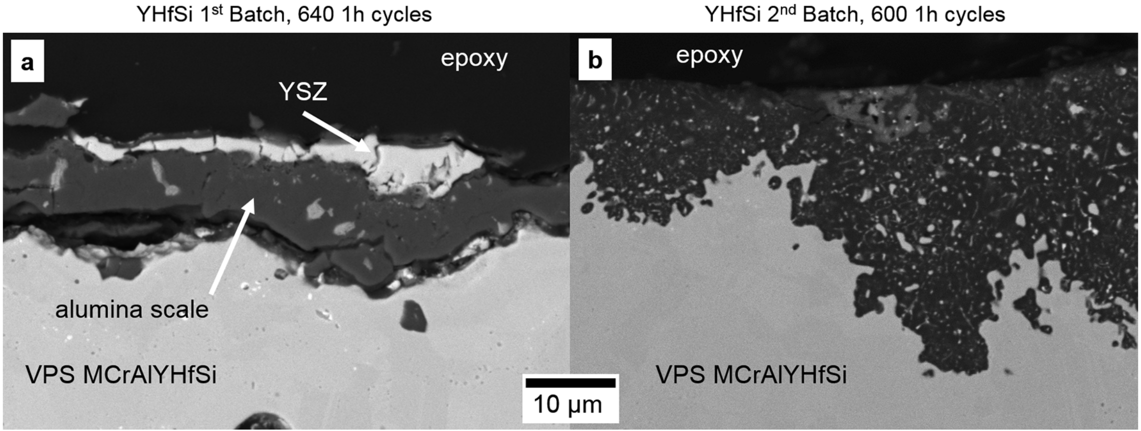

YHfSi 1st Batch, 640 1h cycles
YHfSi 2nd Batch, 600 1h cycles
a
epoxy
YSZ
alumina scale
VPS MCrAlYHfSi
b
epoxy
VPS MCrAlYHfSi
10 μm

Figure 4

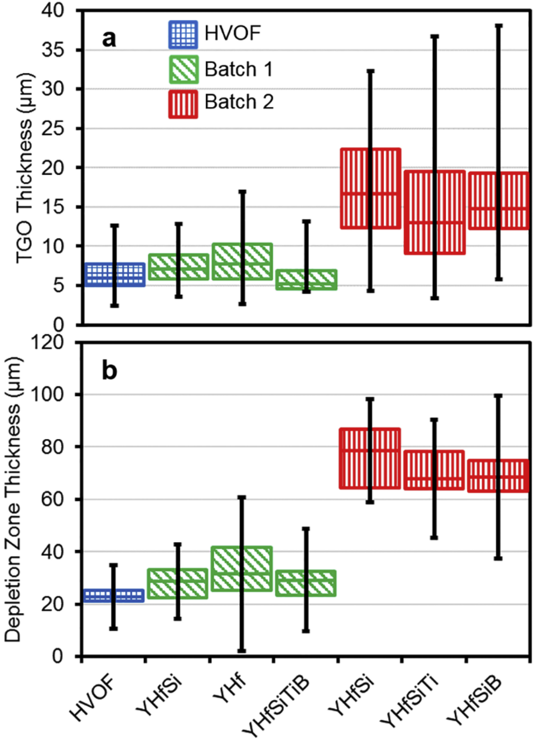
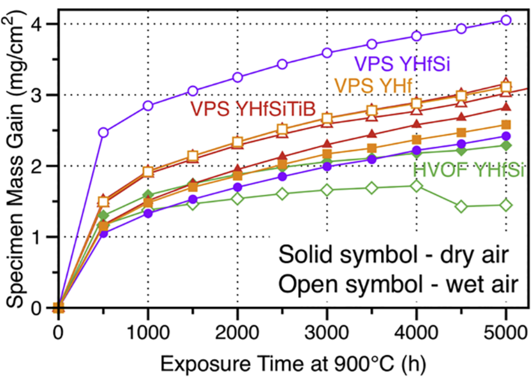

a
b
HVOF
Batch 1
Batch 2
TGO Thickness (µm)
Depletion Zone Thickness (µm)
0
5
10
15
20
25
30
35
40
0
20
40
60
80
100
120
HVOF
YHfSi
YHf
YHfSiTiB
YHfSi
YHfSiTi
YHfSiB

Figure 5



Specimen Mass Gain (mg/cm^2)
Exposure Time at 900°C (h)
VPS YHfSi
VPS YHf
VPS YHfSiTiB
HVOF YHfSi
Solid symbol - dry air
Open symbol - wet air
0
1
2
3
4
0
1000
2000
3000
4000
5000

Figure 6

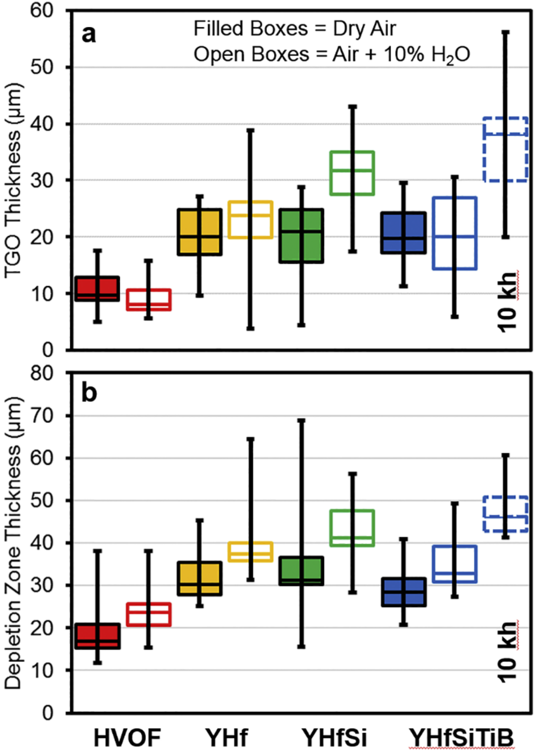

a
Filled Boxes = Dry Air
Open Boxes = Air + 10% H_2O
TGO Thickness (μm)
0
10
20
30
40
50
60
10 kh
b
Depletion Zone Thickness (μm)
0
10
20
30
40
50
60
70
80
10 kh
HVOF
YHf
YHfSi
YHfSiTiB

Figure 7

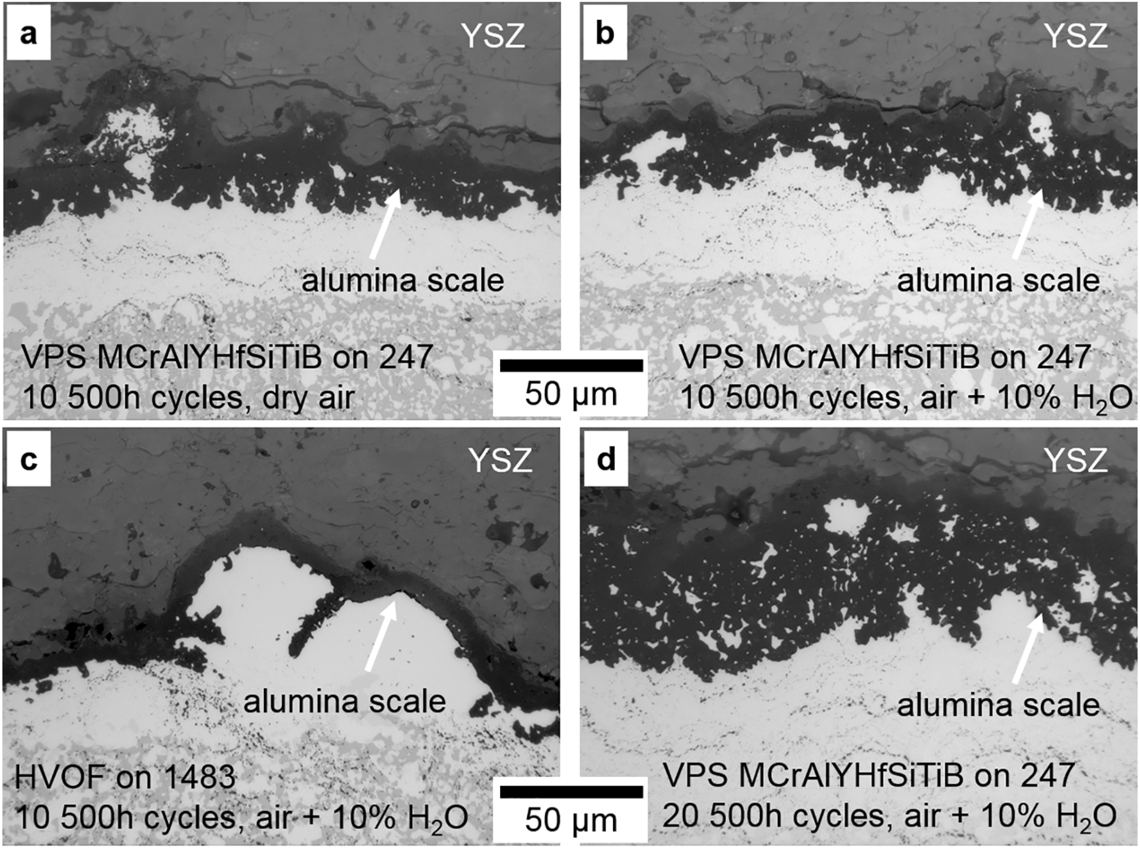

a
YSZ
alumina scale
VPS MCrAlYHfSiTiB on 247
10 500h cycles, dry air
b
YSZ
alumina scale
VPS MCrAlYHfSiTiB on 247
10 500h cycles, air + 10% H_2O
50 µm
c
YSZ
alumina scale
HVOF on 1483
10 500h cycles, air + 10% H_2O
d
YSZ
alumina scale
VPS MCrAlYHfSiTiB on 247
20 500h cycles, air + 10% H_2O
50 µm

Figure 8