Characteristics and performance of suspension plasma sprayed thermal barrier coatings on additively manufactured superalloy substrates

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ARTICLE INFO

Keywords:
Additive manufacturing
Thermal barrier coatings
Ni-based superalloys
Microstructural characterization
Thermal cycling
Thermal shock

ABSTRACT

The complex-shaped hot-section parts of new-generation turbine engines demand unique design solutions. Additive Manufacturing (AM) is an emergent production method that can produce metallic parts with complex geometries and minimal material wastage. In this work, the characteristics and performance behavior of Thermal Barrier Coatings (TBCs) deposited on forged and AM-built HAYNES®®282® superalloy substrates were studied and compared. The bond coats were produced by High-Velocity Air-Fuel (HVAF) spraying using NiCoCrAlY powder feedstock and TBC top-coats by Suspension Plasma Spraying (SPS) using water- and ethanol-based suspensions of Yttria-Stabilized Zirconia (YSZ). The microstructural features, adhesion, Thermal Cycling Fatigue (TCF) lifetime, and thermal shock lifetimes of the TBCs were comprehensively investigated. The results showed that the deposition of bond coats reduced the roughness and asperities of the AM-built substrates. Depending on the type of suspension used and the spray parameters employed, the TBCs exhibited vertically cracked and columnar microstructures. However, no significant differences in TCF and thermal shock lifetimes of TBCs on AM and forged substrates were observed. It is demonstrated that TBC systems can be produced on AM-built metallic substrates, and the resulting TBCs can have similar microstructures and properties as TBCs deposited on conventional substrates.

1. Introduction

The aviation and power-generating industries’ rising demand to increase gas turbine engine efficiency consistently drives researchers to seek novel approaches to accomplish the above. The purpose of increasing efficiency is not only to meet the energy demand but also to reduce fossil fuel consumption during operation and thus lower CO2 emissions [1,2]. Increasing turbine operating temperature is intimately related to efficiency enhancement. Consequently, Ni-based super-alloys with attractive high-temperature strength have been widely used in the hot section parts (combustor, turbine blades) of engines with temperatures exceeding 1000 °C to increase the efficiency of gas turbine engines.

To further increase the operating temperatures of superalloys, thermal barrier coatings (TBCs) have been developed to protect the substrates, which enable superalloys to operate above their usual upper temperature limit by about 150–200 °C [3]. These coatings are inherently required to accommodate the difference in coefficient of thermal expansion between topcoat and superalloy substrate and to grow a thin, compact, and adherent protective oxide scale promoting oxidation/hot corrosion resistance in order to ensure their long-term durability [4]. Manufacturing complex-shaped cooling channels in hot-section parts, especially in the combustion chamber, and improvements in TBCs have allowed the industry to further increase the gas inlet temperatures up to 1500 °C with excellent thermal efficiency, thrust-to-weight ratio and reduce the emission of noxious by-products. Nevertheless, this strategy is approaching the intrinsic limit imposed by traditional substrate manufacturing methods (subtractive manufacturing), since the nickel-based superalloys are difficult to form and conventionally machine because of their high hardness and toughness. To further optimize the design of critical components including the incorporation of complex-shaped cooling channels in the hot-section parts, especially in the combustion chamber, one of the promising
solutions is Additive Manufacturing (AM). The main reason for the increased interest in AM for gas turbine applications is the possibility to produce complexly designed components on-demand with requisite customization, accompanied by significant mass and cost reductions [5–7].

The most widely used hot section parts which are confronted with the highest temperatures are manufactured using Ni-based superalloys, such as HAYNES®282® (H282), which can withstand temperatures above 1000 °C while maintaining excellent thermo-physical properties and thermal stability [8,9]. Several AM processes can be used to manufacture complex lightweight components required for gas turbine applications using superalloys such as those above. In Metal Additive Manufacturing (MAM), the components are built by melting metallic powder, wire, or sheet using a heat source, which is often a laser beam or an electron beam [10]. One of the most relevant MAM processes for manufacturing gas turbine components with the Ni-based superalloy H282 is the Electron Beam – Powder Bed Fusion (EB-PBF). In this process, the feedstock material is a powder, the heat source is an electron beam, and the components are manufactured under a controlled atmosphere [11,12]. Despite the well-documented benefits of AM processing, their suitability to replace standard manufacturing processes is still being studied in detail; for instance, the capability to produce defect-free, consistent quality components with the desired surface texture.

The irregularities on the surface of the AM-built parts necessitate exploring the use of various post-treatments and surface engineering techniques that can be implemented on the manufactured parts to improve the surface characteristics as demanded to achieve the targeted functional performance. The requirement for TBCs to improve their temperature capability is obvious if the components are manufactured using AM methods. In this context, the performance and lifespan of TBC can be greatly influenced by the surface characteristics of as-built AM parts. As a result, this feature necessitates a thorough examination by contrasting the characteristics and performance of TBCs applied on parts produced conventionally and through additive manufacturing. Recent studies have shown that coating deposition by thermal spraying processes on AM components could be an attractive technological solution to protect and improve the functional properties of the AM components [17]. The metallic bond-coat of the TBC system is often made using the High-Velocity Air-Fuel (HVAF) process with powder feedstock, as this produces dense and adherent coatings which contain few oxides, which is required for this layer. The two most prevalent processes for producing the top layer of a TBC are Atmospheric Plasma Spray (APS) and Electron Beam – Physical Vapour Deposition process (EB-PVD). The EB-PVD process is used to achieve coatings with a characteristic columnar microstructure that has good strain tolerance. In recent years, the Suspension Plasma Spraying (SPS) process, which uses a liquid-based feedstock and allows spraying of finer powder particles, has been extensively used for TBC applications [14–16]. The SPS process has many advantages for coating production including the possibility of producing coatings with tailored microstructure types that are challenging to obtain by the APS process e.g., columnar, cracked-columnar, vertically cracked, and featherly of improved performance. TBC systems have been developed and used for several decades for high-temperature gas turbine applications, but no investigations on TBCs deposited by thermal spraying on AM components have been reported. As the development of coatings in general and SPS TBCs in particular on AM components is an area of immense technological relevance to future gas turbine engines, research is indispensable to evaluate the feasibility of applying TBCs on AM-built components in either as-built conditions or with suitable pre-treatment. Thus, the present work investigates the surface, microstructure, and functional properties of the SPS TBCs deposited on EB-PBF AM substrates as compared to similar TBC systems deposited on forged substrates.

2. Experimental details

2.1. Preparation of additive manufactured substrates

The AM substrate specimens made of HAYNES®282® superalloy were produced by the EB-PBF process using gas atomized powder feedstock supplied by Höganas AB (Höganas AB, Sweden). The nominal particle range of the powder was 45–90 (±5 %) μm and its chemical composition is given in Table 1. Square plates of dimensions 60 mm × 60 mm × 6 mm were manufactured using an Arcam A2X system (Arcam AB, Mölnlycke, Sweden), and coupons of 25.4 mm diameter were cut out from them by water jet cutting. The obtained coupons were subsequently used as AM substrates along with coupons of identical size fabricated from forged material. Fig. 1 depicts a plate produced by the EB-PBF process and a coupon obtained after water jet cutting.

2.2. Deposition of HVAF bond-coat

The bond coat deposition on the AM and forged substrates (in tables and figures it is referred as standard substrates hereafter) was carried out with an HVAF M3™ supersonic spray torch (Uniteqcoat Technologies, USA) using gas atomized NiCoCrAlY feedstock powder (AMPERIT405.001-Höganas AB, Sweden). Identical HVAF spray parameters were employed to deposit the bond coats on the AM and as well as on the forged H282 substrates used as a reference in this study. Prior to spraying, alumina powder grit blasting was performed on the forged substrates. The surface preparation by alumina grit blasting was not attempted on AM substrates as they seemed to already possess sufficient roughness in the as-built condition. The chemical composition of the NiCoCrAlY powder feedstock and the HVAF bond-coat spray parameters are tabulated in Table 2 and Table 3, respectively.

2.3. Deposition of SPS topcoat

The SPS method was used to deposit the top coats, with seven spray runs using various parameters and two distinct YSZ suspension feedstocks. The first was with 30 wt% solid-load in water using a powder with size d50 ~ 0.8 μm (Höganas AB, Sweden); the second was with 20 wt% solid-load in ethanol utilizing a powder with size d50 ~ 0.5 μm (Treibacher Industrie AG, Austria). The topcoats were sprayed with an SPS Axial III spray gun (Northwest Mettech Corp., Canada). Prior to spraying, the sample surfaces were cleaned with ethanol and compressed air. The spray parameters are presented in Table 4:

The spray parameters of runs M4, M5, and S were the same but carried out either with different suspensions or substrates as indicated in the table. The S samples (with forged substrate) were considered as the reference samples. As the deposition of SPS TBCs on AM substrates was being performed for the first time, 7 different spray parameters were used to deposit topcoats to investigate the resulting microstructures.

2.4. Heat-treatment of sprayed samples

Before carrying out other functional characterization, the coatings were subjected to heat treatment in a vacuum furnace to induce age-hardening of the H282 material. A standard age-hardening treatment, recommended for H282 superalloy [20], was carried out in two steps:

1st step: 1010 °C for 2 h with subsequent air-cooling.
2nd step: 788 °C for 8 h with subsequent air-cooling.

Table 1

<table>
<thead>
<tr>
<th>Element</th>
<th>Cr</th>
<th>Co</th>
<th>Mo</th>
<th>Ti</th>
<th>Al</th>
<th>C</th>
<th>B</th>
<th>Ni</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wt%</td>
<td>19.5</td>
<td>10</td>
<td>8.5</td>
<td>2.1</td>
<td>1.5</td>
<td>0.05</td>
<td>0.005</td>
<td>Balance</td>
</tr>
</tbody>
</table>
2.5. Surface roughness measurements

The roughness measurements were made to assess the arithmetic average roughness (Ra) of the substrates, bond coats, and topcoats of each of the 8 TBC samples. The arithmetic average roughness of the substrates, bond coats, and topcoats were measured on the 3D surface topography images of the samples obtained by WLI. The use of this method provides more accurate measurements than those obtained with a stylus profilometer, as it allows for deeper measurement into the asperities. The 3D surface topography images of the substrates, bond coats, and top coats were obtained using the 3D-profilometer Profilm3D (Filmetrics, USA). This 3D profilometer uses the WLI technique based on the principle of Michelson interferometry to generate topographic images of a surface. The measuring device consists of a broad-band light source and a detector (a charge-coupled device camera). For measurements using the WLI method, the device specifications were as follows: system 19 K004, objective Nikon 10× DI, zoom 4×, and normal speed. Table 5 shows the scan configuration, stitch parameters, and post-processing steps performed to obtain the 3D surface topography images. For each substrate, bond coat, and topcoat, 10 Ra measurements were made, in accordance with ISO4287 Amplitude, and the average was calculated with the associated standard deviation.

2.6. Microstructural characterizations

A detailed microstructural characterization of metallographically prepared surfaces and cross-sectioned samples was carried out in case of each of the 8 TBC-coated specimens (see Table 4) using a Hitachi TM3000 (Tokyo, Japan) table-top Scanning Electron Microscope (SEM). Thickness measurements of the bond coat and topcoat were carried out by capturing coating cross-sections of 20 SEM images at a magnification of 200×, and the thickness measurements were made on each image using the Image Analysis (IA) software (ImageJ).

The vertical crack/column density of the top coat was determined by capturing a total of 10 SEM images across the sample’s cross-section at a magnification of 200×. Using ImageJ software, a straight line of fixed and known length was then drawn at half of the thickness of the top coat. Then, all the vertical cracks or inter-columnar spacings intercepting the straight line were counted. From each SEM image, the vertical crack/column density was calculated using the following formula:

\[
\text{Vertical crack/column density (cracks/mm)} = \frac{\text{No. of vertical cracks/inter – columnar spacings intercepting the line}}{\text{True length of the line}}
\]
For each TBC sample, the resulting vertical crack/column density was calculated as the average of 10 vertical crack/column density measurements and specified with its standard deviation.

The porosity measurements in the top coat of each TBC sample was also performed by IA using the ImageJ software. To consider coarse and fine porosity, magnifications of 500× and 5000× were used to capture the SEM images, and 20 SEM images were taken for each magnification. For each type of porosity, an average was calculated, along with the associated standard deviation, from the 20 measurements made. Finally, for each TBC sample, the total average porosity of the top coat was calculated as the sum of the coarse and fine porosities.

### 2.7. Erosion tests

The room-temperature erosion tests on the TBC samples were performed using an air-jet erosion tester TR-470 (Ducom Instruments, Groningen, Netherlands), in accordance with ASTM G76-07. The erosive medium used was alumina powder (average particle size 50 ± 5 μm), dried at 150 °C in an oven for 12 h before the erosion test. For each of the different TBC systems, three erosion tests were carried out at three different orientations on the top coat. The weight of the sample before and after the erosion test was measured with a sensitive balance (PCE-AB 100 Instruments, Southampton, UK) to calculate the weight loss. Finally, for each of the TBC samples, an average weight loss per minute was calculated with the associated standard deviation. The parameters used for erosion testing are shown in Table 6.

### 2.8. Adhesion tests

Adhesion testing of the TBC samples was performed using a Universal Testing Machine (UTM) in accordance with ASTM C633 at GKN Aerospace Engine Systems AB (Trollhättan, Sweden). As per this standard, a test specimen of 25 mm diameter was joined with the top and bottom counter parts using an adhesive agent (FM 1000 epoxy glue, Cytec Industries Inc. NJ, USA). The whole glued specimen along with the counter parts were cured for about 90 min about 170 °C. The tensile load was, then, gradually applied using a universal MTS Criterion Model 45 tensile testing machine at a cross - head speed of 1.27 mm/min. The adhesion strength was calculated as the ratio between the load at which the rupture occurred and the coating surface area. For each of the top coats, the adhesion testing was carried out on 3 specimens in order to calculate an average value of the adhesive strength in MPa, with the associated standard deviation.

### 2.9. Thermal cyclic fatigue testing

To evaluate the lifetime of the TBC systems, a standard lifetime assessment test, Thermal Cyclic Fatigue (TCF) testing, was performed. The TCF testing was performed in an automated furnace under a normal atmosphere environment (Entech ECF 14/16, Sweden). The TBC samples were heated to 1100 °C for 1 h, and then rapidly cooled with compressed air for 10 min to approximately 100 °C. To assess the number of cycles to failure, digital image recordings of the sample surfaces were made at the end of each cycle. For each type of TBC, TCF tests were performed on three samples. The failure criterion was 20 % coating spallation and the number of cycles to failure was averaged, with the associated standard deviation. After the TCF tests, each of the TBC samples was metallographically prepared and post-failure analysis of the failed samples was performed under SEM. Energy Dispersive Spectroscopy (EDS) was used to determine the chemical composition of the thermally grown oxides.

### 2.10. Thermal shock lifetime

To simulate the actual conditions encountered in service, the TBCs samples were subjected to a propane gas combustion burner rig test at GKN Aerospace Engine Systems AB (Trollhättan, Sweden). In burner rig testing, samples were heated to a set topcoat surface temperature (about 1300 °C) using combustion burners for a set duration (75 s) and then forced to cool by compressed air to a set top coat surface temperature (about 600 °C) over a set duration (75 s). Prior to the start of the cycling, the samples were preheated by hot air guns from the back side (from the substrate side) to a set substrate surface temperature (about 700 °C) and maintained at that temperature throughout the cycling. The failure criteria for the burner rig test sample was 10 % visible spallation on the top coat surface. For each coating, eight samples were tested under thermal shock (Burner rig test) to determine the statistical variance.

### 3. Results and discussion

#### 3.1. Surface roughness measurements

The surface roughness measurements were done on pristine forged substrates, AM substrates, HVAF bond coats on forged/ AM substrates, and SPS topcoats for the 8 different spray runs. The surface analysis of the TBC samples is essential because the roughness of the substrate and bond coat can influence the microstructures of the bond coat and topcoat and the adhesion between the substrate and bond coat similarly between the bond coat and top coat and hence the functional performance of TBC. Fig. 2 shows the Ra results obtained by WLI. As expected, the surface roughness of the uncoated forged substrate is very low (1.8 ± 0.3 μm) while the surface roughness of the uncoated AM substrate is very high (46.9 ± 3.8 μm). This high value is directly linked to the AM process (that uses powder feedstock) so that: the surface roughness is related to the presence of peaks and valleys caused by unmelted powder particles, that generate high peaks, and sharp and deep cavities. After the HVAF bond-coat deposition on the AM substrates, the Ra value decreases from 46.9 ± 3.8 μm to 13.4 ± 1.0 μm. It can be assumed that the profile of the HVAF bond coat will not look like the profile of the AM substrate: in fact, the thickness of the bond coat probably masks the surface irregularities of the substrate and thus reduces the roughness. The roughness value (8.21 ± 0.43 μm) measured for the HVAF bond coat on the forged substrate is typical for the HVAF process on a forged substrate and is line with previous works [27]. Thus, it can be concluded that the deposition of the HVAF bond coat increases the surface roughness of the forged substrate, however decreases the high surface.

### Table 5

Scan configuration, stitch parameters, and post-processing steps (Profilm3D software).

<table>
<thead>
<tr>
<th>Scan</th>
<th>Back scan/Scan length</th>
<th>Analysis type</th>
<th>Grid total area</th>
<th>Stitch Details</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>400 μm/800 μm</td>
<td>Envelope peak/1.8 %</td>
<td>1200 μm × 1000 μm/9 pictures</td>
<td>3 × 3/Overlap: 0.5 %/Cropped to inter rectangle</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>N 1 – Fill invalids Method interpolate invalids from neighbour pixels</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>N 2 – Spatial filter Type smoothed/Method median/5 × 5 pixels</td>
</tr>
</tbody>
</table>

### Table 6

Parameters used for erosion testing.

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Values</th>
</tr>
</thead>
<tbody>
<tr>
<td>Impingement angle</td>
<td>90°</td>
</tr>
<tr>
<td>Erodent feed rate</td>
<td>2 g/min</td>
</tr>
<tr>
<td>Pressure</td>
<td>0.25 bar</td>
</tr>
<tr>
<td>Time</td>
<td>4 min</td>
</tr>
<tr>
<td>Erodent velocity</td>
<td>30 m/s</td>
</tr>
<tr>
<td>Stand-off distance (SoD)</td>
<td>10.0 mm</td>
</tr>
<tr>
<td>Nozzle diameter</td>
<td>1.5 mm</td>
</tr>
<tr>
<td>Impact angle</td>
<td>90°</td>
</tr>
<tr>
<td>Carrier media</td>
<td>Air</td>
</tr>
</tbody>
</table>
As predicted, the SPS topcoat of the forged sample has a lower surface roughness ($Ra_S = 10.5 \pm 0.5 \ \mu m$) than the AM samples. This is probably because the surface roughness of the bond coat has an impact on the microstructure of the topcoat. Consequently, the bond coat on the forged sample exhibited surface roughness of about 8 \(\mu m\) (lower), and thus a lower Ra value for the topcoat. Moreover, as explained in Section 3.1, this could also be due to the fact that the microstructure of sample S has more uniform columns compared to the AM samples, resulting in a smoother surface. For the SPS topcoats of AM samples sprayed under different conditions, the surface roughness varies from 14 \(\mu m\) to 25 \(\mu m\), but no trend seems to appear depending on the type of suspensions, water-based or ethanol-based. Indeed, the top coat sprayed with water-based suspension has a lower surface roughness ($Ra_{M1} = 14.8 \pm 0.4 \ \mu m$, $Ra_{M4} = 16.10 \pm 2.1 \ \mu m$, and $Ra_{M6} = 16.8 \pm 1.00 \ \mu m$) than the samples with top coats sprayed with ethanol-based suspension ($Ra_{M2} = 22.5 \pm 2.9 \ \mu m$, $Ra_{M3} = 19.80 \pm 1.9 \ \mu m$, and $Ra_{M5} = 24.21 \pm 3.2 \ \mu m$), except compared to the top coat of sample M7 ($Ra_{M7} = 14.9 \pm 1.8 \ \mu m$). A link can be made between the results of the surface roughness of the topcoats presented in Fig. 2, and the results of the vertical crack/column density presented in Fig. 5 (Section 3.2). Indeed, a trend seems to emerge as the higher the vertical crack/column density of the topcoat, the higher the surface roughness of the topcoat. This may be related to the fact that a higher column density indicates a higher number of columns through the topcoat, and therefore a greater alternation of columns with inter-column gaps which ultimately increases the surface roughness.

### 3.2. Microstructural Characterisation

The SEM images of the TBC samples composed of HVAF-sprayed bond coats and SPS-sprayed topcoats on AM and forged substrates are shown in Fig. 3 and Fig. 4, respectively. Table 7 shows the SPS top-coat thicknesses measured on SEM images for each of the spray runs. The top-coat thicknesses are observed to range from $378.0 \pm 17.7 \ \mu m$ (sample M2) to $603.3 \pm 33.4 \ \mu m$ (sample M6).

It is noticed that the surface roughness of the AM substrates is much higher than that of the forged substrate. Furthermore, for the TBCs sprayed on AM substrates, it is also possible to observe the presence of defects at the substrate/bond coat interface, such as porosities created by the deep and sharp cavities of the rough surface and un-melted H282 powder particles resulting from the AM. It is observed that the highly dense microstructures of the bond coats of the 8 samples are similar because they were sprayed with the same feedstock powder, HVAF spray parameters, and under the same conditions. Finally, it can be noted that despite the use of two different substrates, the difference in roughness values at the bond coat-top coat interface seems to be reduced when comparing TBCs on the forged substrate and on AM substrate. Thus, the top-coat profiles appear to be rather different due to the fact that 7 different spray runs were performed with different spray parameters and two different 8YSZ suspensions. However, significant change in topcoat microstructure has not been observed due to the impact of AM substrate finish (Fig. 4).

Fig. 3 shows the vertically cracked microstructures of the TBCs with topcoats sprayed with a water-based suspension, while Fig. 4 shows the columnar microstructures of the TBCs with top coats sprayed with an...
ethanol-based suspension. For the topcoat parameters, sample M1 was sprayed with a higher power, sample M4 was sprayed with reference parameters [22], and sample M6 was sprayed with lower SoD. The three microstructures obtained are dense vertically cracked (DVC) and on all three samples it is possible to observe the vertical cracks typical of this type of coating, and also horizontal cracks, called branching cracks, which appear to originate from the vertical cracks. Vaßen et al. [21] have shown that the growth of vertical cracks in SPS coatings is driven by a high number of tensile stresses in the coating. Between the vertical cracks, presence of fine porosity is also noted in all the three samples (M1, M4 and M6), as shown in the SEM image of the sample M6 (Fig. 3).

The vertical crack/column crack density for the SPS top coats are shown in Fig. 5. In Fig. 5, it can be noted that the vertical crack density is higher for sample M6 (6.4 ± 0.5 cracks/mm) than for samples M4 (5.1 ± 0.8 cracks/mm) and M1 (4.7 ± 0.7 cracks/mm), and these results correlate with what can be observed in the SEM images. In TBCs, vertically cracked microstructures are recognized to improve the ability of the coatings to tolerate the thermal stress created during temperature cycling conditions [22].

Fig. 4 shows SEM images of cross-sections of the TBCs on forged substrate as well as AM substrates with HVAF bond-coats and ethanol-based SPS top-coats. For the topcoat parameters, sample S on the forged substrate and sample M5 on AM substrate was sprayed with standard parameters [22] typically used to achieve columnar microstructures. Whereas sample M2 was sprayed with a higher power, sample M3 was sprayed with a higher suspension feed rate, and sample M7 was sprayed with higher power and lower SoD. The topcoat micrographs obtained for these 5 samples are quite similar to columnar microstructures, except for sample M7 which shows some columnar features but with a more vertically cracked microstructure compared to sample M5. For all samples, it is possible to observe inter-column porosity at the interfaces between columns, and fine intra-column porosity. The inter-column gaps are wider for samples M5 and S, and from the results shown in Fig. 5, the column density is also higher for these two samples sprayed...
with standard parameters (12.1 ± 0.9 columns/mm for M5, and 10.2 ± 0.9 columns/mm for S). It has been shown that larger inter-columnar gaps can improve the strain tolerance of the whole TBC system as well as its thermal cyclic lifetime, but that these gaps can also decrease the thermal conductivity of the top coat [21]. Although sample M7 looks more like a vertically cracked-columnar coating compared to the columnar coatings M2 and M3, its column density (or vertical crack density) is higher (9.0 ± 0.8 cracks/mm for M7 compared to 8.5 ± 0.8 columns/mm for M2 and 7.8 ± 1.0 columns/mm for M3). One comment can be made on the difference between sample M5 and sample S (the spray parameters are the same, but the substrates are different): both exhibit a columnar microstructure, but due to the higher roughness and slightly wavy surface of the HVAF bond-coat on the AM substrate, the columns in the top coat of sample M5 are highly non-uniform compared to the more homogeneous columns of the reference sample [22]. In general column size (density) will be influenced by the surface topology of the substrate or bond coat surface [14] and that on sample M5, the columns above the fairly high peaks are wider and larger than the other samples. The two main different types of microstructures obtained for the 8 samples, vertically cracked-type and columnar type, are most certainly due to the use of 8 wt% YSZ suspensions with different solvents, water, and ethanol. Ganvir et al., [22] identified the suspension surface tension, viscosity, and density, as the main parameters that affect droplets’ size and momentum during the SPS spraying. As a result, using a water-based suspension produces bigger droplets with greater momentum than using an ethanol-based suspension, which has a lower surface tension, lower density, and higher viscosity. It has been shown that droplets with higher momentum and larger size leading to vertically cracked microstructures, while droplets with lower momentum and smaller size leading to columnar microstructures. This was explained as follows: droplets with smaller diameters and low momentum are affected by the plasma flow drag in the border layer near the substrate, so these droplets will be entrained by the plasma flow along the substrate surface and deposit at shallow angles on the surface asperities, resulting in the formation of columns. Whereas larger droplets with higher momentum are less vulnerable to the plasma flow drag, resulting in a perpendicular impact on the substrate and close-to-normal deposition, resulting in the formation of vertically cracked structures [23]. This phenomenon explains why TBC samples M1, M4 and M6 whose top coats were deposited with a water-based suspension have vertically cracked microstructures, whereas TBC samples S, M5, M2, M3, and M7 whose top coats were deposited with an ethanol-based suspension have columnar microstructures.

3.3. Porosity evaluation

The porosity measurement results of the SPS top coats are presented in Fig. 5. Fig. 6. Fig. 7 shows the SEM images of the SPS top coats of the 8 samples at higher magnification (500×), which confirms porosity features. It is observed that sample M5 and sample S, have columnar microstructures, with the highest porosity (M5 = 17.74 ± 1.2 % and S = 15.4 ± 1.4 %). It is confirmed that the highest porosity was caused by the presence of substantial fine-scale pores within columns and wide gaps between columns (Fig. 7). However, it does not seem that the use of AM substrate has an impact on the porosity of the SPS top-coat. In fact, the top coats of samples M5 (AM) and S (standard), sprayed with the same suspension with identical parameters, have very close or even similar porosity rates if the standard deviations are considered. However, because the columns are less uniform, as described in Section 3.1, the inter-column gaps are wider at some places, which could account for the slightly higher porosity rate of sample M5.

Thus, comparing the porosity percentage of the columnar top coats sprayed with ethanol-based suspension, the highest are those of sample M5 and sample S which have wide inter-column gaps as explained above, and those of samples M2, M3, and M7 are much lower (6.3 ± 0.6 % for M2, 6.5 ± 1.4 % for M3 and 5.9 ± 1.4 % for M7). Based on Fig. 7, this difference could be explained by the fact that the top coats of samples M2 and M3 have almost no inter-column spaces and thus only the fine intra-column porosity comes into consideration. For sample M7, this lower porosity rate can be explained by the fact that the top coat microstructure is more vertically cracked-type with some columnar features and very low intra-column porosity. Moreover, the results might be consistent with studies by Ganvir et al., [16], they have shown about the relationship between porosity rate and spray parameters: lower power results in highly porous columnar structures, while using
higher power results in less porous columnar structures. And also reducing the SoD results in more flatter structures that crack during spraying and lead to vertically cracked microstructures. Here, the results obtained agree with this remark: compared to the top-coat M5 sprayed with standard parameters, the top-coat M2 sprayed with a higher power is less porous, and the top-coat M7 sprayed with higher power and lower SoD is less porous and more vertically cracked-type.

Then, comparing the porosity rates of the vertically cracked top coats sprayed with water-based suspension, the top coat of sample M4 is more porous than the other samples (11.08 ± 0.74 % for M4). In Fig. 7, it can be observed that despite the fact that all three samples, M4, M1, and M6, show vertical cracks and branching cracks within their microstructures, the top-coat M4 shows a much higher number of pores within the columns between the vertical cracks. It can also be assumed that the top-coat M6 has a higher porosity rate (7.7 ± 2 % for M6) than the top-coat M1 (5.9 ± 0.9 % for M1) because of the interpass porosity bands (Fig. 7).

The results obtained show that the AM substrates do not seem to have an impact on the porosity percentage of the topcoats, with the observed values being in a range similar to that observed for vertically cracked-type and columnar-type SPS topcoats on forged substrates. In addition, some samples have fine-scale porosity that could contribute to reducing the thermal conductivity, and some have inter-column gaps/vertical cracks that could improve the strain tolerance of the TBC systems [14].

3.4. Erosion testing

The erosion tests were carried out on the top coat of each of the 8 TBC systems. The erosion rate representing the weight loss per minute (mg/min), of all samples is depicted in Fig. 8. When selecting the duration of the erosion tests, particular attention was paid to the fact that the top-coats are not be completely eroded to enable valid comparison of results obtained with different TBC specimens. The erosion rate results of the 8 samples show almost equal values when considering standard deviations and no significant statistical differences are observed. The erosion rate of the top coat generally depends on its porosity, surface roughness, and coating cohesion. It has been shown that the more porous the coating, the higher the erosion rate: i.e. it is easier for the erosive media to remove material from the top coat [25]. Similarly, the higher the surface roughness of the coating, the higher the erosion rate:
surface asperities will be fractured and removed more easily [26]. However, neither of these two trends is clearly observed for the 8 different top coats, probably because their microstructures are very different. However, comparing the erosion rates of the vertically cracked top coats sprayed with water-based suspension, it can be seen that sample M4 has the highest erosion rate compared to samples M1 and M6 which are less porous. Comparing the erosion rates of the columnar top coats sprayed with ethanol-based suspension, it can be seen that sample S has the highest erosion rate compared to samples M2, M3 and M7 which are less porous. However, the differences between the values are minor, and sample M5 which has the most porous microstructure and the highest surface roughness does not have the highest erosion rate. It can also be inferred that in general higher the column density, higher the hindrance for the crack propagation (due to the impact of the erosive media) and thus lower the erosion rate.

3.5. Adhesion testing

The measured average adhesion strength values of the 8 different TBCs on AM substrates are presented in Fig. 9 and the digital images of TBC samples after adhesion testing are shown in Fig. 10. Comparing the adhesion strength of the vertically cracked-type TBCs, the samples M1
and M6 show the highest adhesion strength (42.45 ± 1.86 MPa for M1 and 37.43 ± 1.80 MPa for M6). The adhesion strength of the sample M4 is slightly lower (24.66 ± 2.64 MPa for M4). Based on Fig. 10, it can be seen for samples M1 and M4 that the failures occurred within the top coat, so the strength measurements obtained correspond to the cohesive strength of the top coats. For sample M6, the failure appears to have occurred at the bond coat - top coat interface. Also, from the porosity analysis it was found that M4 (Increasing order of porosity-M1 (5.97 %) < M6 (7.79 %) < M4 (11.08 %)) has the highest porosity, which may give raise to weak cohesive strength to the coating. Moreover, for the water based SPS coating sample M4 is deposited with least spray power raising to weak cohesive strength to the coating. Furthermore, for the water based SPS coating sample M4 is deposited with least spray power raising to weak cohesive strength to the coating. Moreover, for the water based SPS coating sample M4 is deposited with least spray power raising to weak cohesive strength to the coating. Moreover, for the water based SPS coating sample M4 is deposited with least spray power raising to weak cohesive strength to the coating.

Comparing the adhesion strength of the columnar-type TBCs, the samples M3 and M5 show the highest adhesion strength (46.72 ± 3.48 MPa for M3 and 46.24 ± 5.00 MPa for M5). The adhesion strength of sample M2 is slightly lower (38.27 ± 0.90 MPa for M2) and the lowest adhesion strength is that of sample M7 (21.06 ± 2.37 MPa for M7). This may be inferred from considering spray power employed to deposit ethanol based SPS coatings (Increasing order of plasma spray power M5 (122 kW) < M2&M3 (138 kW) < M7 (147 kW)), as higher the plasma spray power higher the residual stress and thus lower the adhesion.

Based on Fig. 10, it can be seen that the failures occurred at the bond coat-top coat interface in the outer perimeter of the samples. The AM samples are assumed to have the same surface roughness for their HVAF-sprayed bond coats, but the mechanical interlocking at the bond coat-top coat interface may be different depending on the presence of defects. Thus, a possible explanation for the different adhesion strength values would be differences in mechanical anchoring at the bond coat-top coat interface. A potential reason for the high adhesion strength of samples M3 and M5 could be an optimized combination of spray parameters that lead to more efficient bonding mechanisms at the bond coat-top coat interface.

Moreover, the columnar microstructures of ethanol based SPS coatings (M2, M3, M5 & M7) can possess higher adhesion strength due to greater surface area provided by the coating to the bond coat also due their superior interlocking columnar structures. Whereas the vertically cracked microstructures of water based SPS coatings (M1, M4 & M6) with branching cracks (high stress concentration) which could further promote fracture during the adhesion test.

3.6. Thermal cyclic fatigue analysis

For each different TBC system, two samples were tested for TCF lifetime estimation, and the normalized value of TCF lifetime results obtained by TCF testing are presented in Fig. 11. As the samples were exposed in a furnace and then cooled with compressed air, it can be deduced that there was no thermal gradient across the TBCs and underlying substrates, and therefore the oxidation of the bond coats was the main source of failure in TCF tests. Fig. 11 shows that samples M6 and M7 have the highest lifetimes, while the lowest lifetimes are observed for samples M1, M4, and S. The samples M2, M3, and M5 show intermediate lifetimes compared to the other samples. For TBCs with YSZ top coats, the common failure mode is the spallation of the top coat due to the thermal growth of the oxide layer (TGO) at the bond coat-top coat interface and due to the mismatch in CTE values between layers of the TBC system [25]. Firstly, it is observed that the use of AM substrates, which have higher surface roughness at the substrate-bond coat and bond coat-top coat interfaces, does not seem to have an impact on TCF lifetimes. Indeed, only sample M4 has a lower life than the standard sample S. The same applies to the impact of using ethanol-based or water-based suspensions: no trend seems to emerge.

As the HVAF bond coat is the same for all TBC samples, which only have different top coats, it can be assumed that the oxidation behavior of the bond coats is the same for all the samples. Therefore, it can be deduced that different microstructures for the top-coats have led to different TCF lifetimes. In fact, due to the columnar or vertically cracked microstructures, these TBCs have different strain tolerances, which can lead to more or less rapid failures [27]. It is noted that the two samples M6 and M7, with the highest lifetimes, were those sprayed with lower SoD. The slightly lower lifetimes of samples M1 and M4 could be due to the presence of numerous branching cracks in the vertically cracked top coats, compared to the columnar-type top coats. These horizontal cracks create a low energy path for crack propagation during TCF testing, leading to rapid crack propagation through the coating, which will result in faster coating failure [27]. After TCF failure, each TBC sample was observed under SEM, and the same observation was made: in the case of all samples, it was observed that the failure occurred at the thermally grown oxide (TGO)/top coat interface and that cracks start to grow near the TGO layer. EDS analysis of the TGO layers was also carried out for all samples and showed that TGOs are composed of aluminium oxides. The cross-sectional SEM image of the failed sample M5 is shown in Fig. 12(a) and (c) shows the EDS analysis of the TGO which reveals the presence of aluminium oxide. In Fig. 12(a) and (b), it is possible to observe the TGO layer that grew between bond coat and top coat. In the bond coat, it is possible to observe the β(NiAl)-phase (darker grey area), and above the β-depleted zone (lighter grey area). The β(NiAl)-phase acts as a reservoir of aluminium for the formation of the alumina TGO and the depleted zone corresponds to the path that the β-phase has to migrate to contribute to

![Fig. 10. Pictures of the TBC samples after adhesion testing: a) M1, b) M2, c) M3, d) M4, e) M5, f) M6, and g) M7.](image-url)
the growth of the TGO [25]. This mechanism leads to the depletion of the β-phase in the bond coat and thus to an increase in the thickness of the depleted zone. It is also possible that the β-phase of aluminium diffuses to the substrate, increasing the β-phase depletion [14].

The β-depletion zone thickness can be used as a criterion to assess the end of the coating lifetime. Indeed, a TGO layer formed of alumina oxides acts as a protective layer that prevents bond coat oxidation. When the β-phase is depleted, other oxides migrate more slowly forming the TGO layer. These spinel, nickel, and cobalt oxides have larger expansion volumes and are growing faster than aluminium oxides, and thus will cause rapid coating failure by spallation. For all samples, this β-phase was still present in the bond coat with a similar thickness as confirmed by Fig. 12(b). The β-depletion zone thickness was measured by Image Analysis on SEM images, at 10 locations, for each TBC sample and then an average was calculated from these values. The values obtained for the β-depleted zone thickness, for the different TBC samples, are quite close, in the range of 33 μm to 49 μm.

The formation of the TGO layer at the bond coat/top coat interface, which was observed on all 8 TBC samples, is slow but continuous, so the thickness of this layer may represent the lifetime under TCF testing. The TGO layer thickness was measured by Image Analysis on SEM images, at 20 locations, for each TBC sample and then the average and standard deviation were calculated from these values. The values obtained are shown in Fig. 13. As expected, it can be observed that the samples with the longest lifetimes have larger TGO thicknesses (7.9 μm ± 0.9 μm for sample M7) and the samples with lowest lifetimes have smaller TGO thicknesses (5.7 μm ± 0.9 μm for sample M4). Therefore, in line with what has been seen in the literature, for all 8 samples, the thickness of the TGO layer, which depends on the growth of the aluminium oxides, defines the TCF lifetimes of the TBC systems.

3.7. Thermal shock lifetime

Fig. 14 shows the results from burner rig test where the thermal shock lifetime data is normalized with respect to SPS forged sample coated with ethanol suspension. In Fig. 14 it can be seen that the forged sample showed the highest and M5 showed the lowest average thermal shock lifetime. The columnar microstructures (M2, M3, M7, and standard) showed a better thermal shock lifetime than the vertically cracked microstructures of M1, M4, and M6, which may be due to their higher strain tolerance capability [28]. This is also evident in Fig. 5 as higher the segmentation density in ethanol-based columnar coatings than in water, higher the strain tolerance and hence higher the thermal shock lifetime. Under thermal shock test, the coating experiences a temperature gradient primarily through the coating thickness introducing thermal stresses in the coating and thus coating spallation. The life of the coating during a thermal shock test mainly depends on porosity distribution, microstructure, thermo physical properties, etc. [28]. Thus, it is also important to note that, use of AM substrates which have rougher surfaces at the substrate-bond coat and bond coat-top coat interfaces, does not seem to have an effect on the thermal shock life of the coating. The minimum thermal shock exhibited by the samples M4 and M5 may be attributed to their higher porosity (Fig. 6) as compared to the other samples. In general, higher the porosity lower the toughness and thus lower the thermal shock life.

4. Conclusions

TBCs systems were applied on Additive Manufactured substrates made by the EB-PBF process using HAYNES®#282® superalloy powder.
The NiCoCrAlY bond coats were deposited by the HVAF process and the top coat was deposited using two different 8wt%YSZ suspensions using SPS at different spray parameters. The surfaces, microstructures, properties, TCF and thermal shock lifetimes of the TBCs on AM and forged substrates have been investigated. The following are the major conclusions drawn from this study:

- The bond-coat deposition masks the surface asperities of the AM substrates, despite the surface roughness of the AM substrates being higher than 40 μm.
- The TBCs with top coats deposited with a water-based suspension show vertically cracked microstructures with porosity ranging from 6 % to 11 %. The TBCs with top coats deposited with an ethanol-based suspension show columnar microstructures with porosity rates around 17 % for those with large inter-columnar gaps and high

![Thermally Grown Oxide thicknesses after Thermal Cyclic Fatigue testing.](image1)

![Thermal shock life time results.](image2)
intra-column porosity and with porosity rates around 7% for those that are denser.

- Furthermore, the surface roughness values of the top coats and the erosion rates correlate with the properties provided by each of the microstructure types. The same types of coatings have been obtained previously on forged substrates, using both types of suspensions and the same parameters, so the AM substrates do not seem to have an impact.

- The TCF lifetimes of the TBCs deposited on AM substrates show that failures occurred for the highest surviving samples. Thus, TBCs on AM substrates do not have shorter lifetimes than those deposited on forged substrates. However, it can be assumed that these short lifetimes come from HAYNES®282® substrates regardless of the manufacturing method - because using the same spraying parameters, for TBCs deposited on forged material substrates (Hastelloy).

- It is found that the thermal shock lifetime of the TBCs is completely independent of substrate material instead it primarily depends on microstructural characteristics and the coating toughness.

From the above observations, it can be concluded that the use of additively manufactured substrates does not have a major impact on the microstructures and properties of TBCs. In conclusion, it is possible to develop TBC systems on metallic substrates produced by an AM process, and the resulting TBCs can have the same microstructures as TBCs deposited on forged substrates.

**Declaration of competing interest**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

**Data availability**

Data will be made available on request.

**Acknowledgement**

Financial support from the Knowledge Foundation, Sweden for the project EcoGATE (Dnr. 2020007), as a part of which the present work was carried out, is gratefully acknowledged. The authors would like to thank their colleagues at University West, Jonas Olsson for producing the AM substrates and Magnus Sandberg for assistance during spraying of coatings.

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