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Tracking the Calcium–Magnesium-Alumino-Silicate (CMAS) Infiltration into an Air-Plasma Spray Thermal Barrier Coating using X-ray Imaging

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Abstract

The progressive infiltration of a free-standing air plasma spray (APS) thermal barrier coating (TBC) by Calcium-Magnesium-Alumino-Silicate (CMAS) at 1250°C is investigated followed non-destructively by time-lapse X-ray micro-computed tomography (μ -CT). The top ~50 μ m of the TBC expands rapidly (within the first 20 min) by as much as 70% due to the physical ingress of the CMAS along the splat boundaries and becomes exfoliated. ~~The remainder of the TBC swells at a slower rate (ultimately by 15%) but remains intact.~~ The ingress and resulting exfoliation are shown to be driven by capillary forces and with the exfoliated depth depends dependant on the CMAS loading. ~~The remainder of the TBC swells at a slower rate (ultimately by 15%) but remains intact.~~

Key words: aero engine; high temperature; failure; environment; volcano ash

Thermal barrier coatings (TBCs) are widely used in the gas-turbine engines to provide thermal protection for the metallic components [1-4]. The state of the art TBCs consist of a 7-8 wt.% Y₂O₃-stabilized ZrO₂ (7YSZ) topcoat, a NiCoCrAlY or Pt/Al diffusion bond coat and a Ni-based superalloy substrate [2, 5]. Although the application of TBCs has allowed an increase in gas temperatures, the demand for even higher operating temperatures is posing materials issues for the current TBCs [2, 4, 6, 7]. For example, silicate debris (e.g. sand, volcano ash etc.) ingested by the engine adheres to the surface of TBCs and forms glassy calcium–magnesium-alumino-silicate (CMAS) [8]. At high temperatures (~1230°C [9]) the CMAS is molten

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1 and can wet the TBC surface. Penetration of CMAS into the pores in the topcoat
2 makes it less strain-tolerant [6, 8, 10-14]. CMAS also induces depletion of Y^{3+} in the
3 YSZ as the solubility of Y^{3+} in CMAS is higher than that of Zr^{4+} [6, 8, 10-13]. As a
4 result, the $t \rightarrow m$ phase transformation may occur upon cooling accompanied by a 3-
5 5% volume expansion [6, 15] and a loss of coating toughness [15].
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10 Recent studies of TBC degradation by CMAS have resulted in an improved
11 understanding of the damage mechanisms, especially those related to CMAS
12 infiltration and the associated thermo-chemical reactions [16]. In general, the
13 infiltration of CMAS involves a series of complex physical and chemical processes.
14 The temperature gradient experienced by the TBC in service strongly affects the
15 CMAS infiltration by controlling its viscosity [17]. Detailed chemical and
16 microstructural analysis on the infiltrated TBCs has shown that different phase
17 transformations in the APS 7YSZ can arise depending on the local chemistry of the
18 CMAS [6, 13]. Recently, in-situ Raman spectroscopy was used at high temperature
19 to identify the chemical reactions associated with CMAS infiltration [10]. This
20 suggests that in order to better understand the failure of TBC by CMAS infiltration, it
21 is important not only to probe the infiltrated sample, but also to investigate the
22 infiltration process [10], especially with respect to microstructure evolution [16].
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36 | X-ray micro computed tomography (μ -CT) has already been used for TBC
37 characterisation, for example to track microstructure evolution as a function of
38 thermal exposure including TGO growth [18], the localised rumpling of TGO [19], the
39 displacement induced by thermal treatment [20], as well as cracking and changes in
40 porosity within the YSZ topcoat [21, 22]. More recently, the evolution of pores in the
41 TBCs under volcanic ash corrosion has been investigated by X-ray CT [23].
42 However, the evolution of TBC microstructure during CMAS infiltration has yet to be
43 observed.
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53 Here the infiltration of CMAS into an APS TBC is followed by time-lapse X-ray μ -CT.
54 The aims are (1) to build-up an overarching view of the CMAS infiltration process
55 and (2) to observe the microstructural evolution of the TBC during CMAS infiltration
56 in order to explore the kinetics of CMAS infiltration and the associated volume
57 changes in the TBC to underpin life-time prediction models.
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2 8 wt.% Y_2O_3 -stabilized ZrO_2 (8YSZ) was deposited onto a Hastelloy-X plate having a
3 NiCoCrAlY bond coat by APS. The thickness of the coating was about 280 μm . The
4 plate was cut into 7 x 5 mm^2 rectangles. To obtain free-standing TBCs, the
5 rectangles were immersed in aqua regia to remove the metallic substrate. CMAS
6 paste comprising 33CaO-9MgO-13AlO_{1.5}-45SiO₂ mol% was prepared by mixing the
7 powders of each oxide constituent and attrition milling the blend in isopropanol. The
8 as-prepared paste was applied to the middle of the free-standing TBC followed by a
9 5-min treatment at 1250 °C (10 °C/min heating and cooling rate) in order for the
10 CMAS paste to wet the TBC surface.
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20 In order to correlate contrast of features recorded by micro-CT with conventional
21 scanning electron microscopy (SEM) sections, a 500 μm diameter cylindrical sample
22 was cut from the CMAS-coated APS TBC plate using a precise laser cutting machine
23 [24]. An X-ray μ -CT scan was acquired prior to destructive sectioning to extract a
24 central section sample for SEM for side by side comparison.
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31 To follow the microstructural evolution at different stages of CMAS infiltration, a
32 matchstick specimen measuring 1 mm x 3 mm x 0.3 mm was cut from the centre of
33 the CMAS-coated freestanding TBC. A previous study on the same TBC sample has
34 shown that a large deformation can be induced by CMAS infiltration after heat
35 treatment at 1250 °C for ~98 min [9]. Once a tomograph of the specimen in the as-
36 received state had been collected, the specimen was heat treated in air to 1250 °C
37 for 20 min using a chamber furnace. During the heat treatment, the heating and
38 cooling rate was kept at 5 °C/min. After cooling to ambient, an X-ray tomograph of
39 the same region of interest was collected before the specimen was put back into the
40 furnace for the next heat treatment step to acquire a time-lapse sequence.
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51 A Zeiss Xradia Versa 520 X-ray microscope (XRM) was used to acquire the X-ray
52 tomographs. For all the CT scans, the X-ray tube was operated at 120 kV. Each
53 scan comprises 1601 projections equi-angularly distributed over 360° sample
54 rotation. The reconstructed virtual slices have a voxel size of 0.72 μm .
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1 The X-ray tomographs collected at different stages of thermal exposure were co-
2 registered to a single sample coordinate system. To enable correlative
3 characterisation [25], the datasets were pre-aligned manually and registered using
4 the 'Register Image' module in Avizo 9.4 (Thermo Scientific). The registered
5 datasets were re-sliced to pick out virtual slices showing the same region of interest
6 (RoI) of the sample at different stages of heat treatment.

11 After co-registration, Digital volume correlation (DVC) was undertaken for the intact
12 layer between successive tomographs using the DaVis 8 (LaVision) software. A
13 multi-pass correlation algorithm was chosen with the window size decreasing from
14 128 x 128 x 128 voxels to 24 x 24 x 24 voxels over 4 iterations and the window
15 overlap was kept at 50%. After each pass, the displacement vector is deleted if its
16 correlation value is less than 0.9. Moreover, the missing values on the grid points
17 where the displacement vectors have been deleted are added via bilinear
18 interpolation. The relative volumetric change of the sample can be calculated using
19 the divergence of the displacement field as [26]:

$$\theta = \nabla \cdot \bar{\mathbf{u}} = \frac{\partial u_x}{\partial x} + \frac{\partial u_y}{\partial y} + \frac{\partial u_z}{\partial z} \quad (1)$$

35 Prior to differentiation, a median filter (3 x 3 kernel size) is applied to the
36 displacement field in order to reduce noise.

40 Figure 1 compares a virtual slice taken from the X-ray tomograph of the free-
41 standing APS TBC after applying the CMAS with an SEM (BSE) micrograph
42 obtained destructively for the same slice (RoI). The CMAS is observed as a dark
43 region (almost as dark as the pores) in the CT image because of the low atomic
44 number of the constituents, whereas it is much lighter in the SEM image. The higher
45 spatial resolution SEM image confirms the spherical features showing the darkest
46 contrast on the μ -CT virtual slices (e.g. yellow arrow) are pores and the grey regions,
47 which tend to be aligned horizontally (e.g. yellow square), correspond to inter-splat
48 gaps filled by partially consolidated YSZ particles. The μ -CT virtual slice
49 demonstrates that the fragmented YSZ zones evident in the SEM sections have not
50 been introduced post mortem by the grinding and polishing process. Further, cross-
51 correlation confirms that the crack-shaped features (highlighted by the red ellipse)

are inter-splat cracks. Very fine cracks (less than 0.6 μm wide) cannot be resolved by X-ray μ -CT.

Figure 2 shows virtual cross-sections taken from the center of the thermally exposed coupon after different thermal exposure times. By following the microstructure evolution in this region of interest (RoI), it is possible to appreciate the kinetics of the CMAS infiltration process. Figure 2 (a) shows that the CMAS has completely covered the top surface of the TBC before infiltration. The CMAS loading on this specimen was estimated from the tomographs to be $22.3 \pm 2.6 \text{ mg/cm}^2$ (Appendix A).

The CT image sequence shows that the CMAS infiltration creates two distinct regions within the TBC. After 20mins (Figure 2 (b)), the CMAS has infiltrated the upper 20~50 μm of the TBC (i.e. the region above the red dash line in Figure 2 (b)) which as a result has become exfoliated from the underlying material. Exfoliation has followed the morphology of the splats, as shown by the yellow ellipse in Figure 2 (b). This suggests that the CMAS tends to attack the inter-splat gaps preferentially. Pores have developed in the exfoliated layer (e.g. red circles in Figure 2 (b)) during the first 20 min. The largely defect-free region of the coating near the very top of the coating (e.g. above the yellow dash line) has retained its morphology despite being in direct contact with the CMAS and has rafted upwards lifted by the exfoliation beneath it. In addition, pores are seen under the lifted splats. In some cases, cracks have developed within the lifted YSZ as indicated by the red arrow in Figure 2 (b).

At longer exposure times, the exfoliated surface region appears to 'deflate' somewhat while the CMAS infiltrates deeper into the TBC primarily via the boundaries of the splats. This is accompanied by a slight increase in the depth of the exfoliated layer which takes in more TBC splats (its extent delineated by the red dashed lines in Figure 2 (c) and (d)). Inside the exfoliated layer, which has developed in the first 20 min of heat treatment, a large number of small spherical pores (e.g. yellow circle in Figure 2 (c)) have formed after 40 min. Such pores

haven't agglomerated into larger pores. The larger spherical pores that have developed in the first 20 min (e.g. red circles in Figure 2 (b)) have deflated somewhat as the CMAS has penetrated deeper into the coating. The surface crack on the lifted YSZ has closed as the large pore beneath it deflates. The contrast for the CMAS in the exfoliated layer has become brighter as heat treatment proceeds. This could be a result of the dissolution of heavy element ions such as Y^{3+} and Zr^{4+} into the CMAS since the contrast in the CT virtual slices reflects the X-ray mass attenuation coefficient of the probed sample. This could have changed the viscosity of the remaining CMAS and led to different behavior of the pores at different stages of heat treatment [12].

It is noteworthy that the underlying intact (unexfoliated) region of the TBC appears to become infused with CMAS at longer exposure times (evident as a darker contrast compared with the YSZ in the X-ray μ -CT virtual slices). Comparison of Figure 2 (a) and (c) indicates that the CMAS has completely penetrated the free-standing coating as the splats at the bottom of the coating appear to have become separated (e.g. the region indicated by the black dashed line).

The temporal evolution of the average volume expansion is plotted in Figure 3. The volume expansion in the top 50 μ m of the coating surface is marked and can be estimated directly from the tomograms according to:

$$\Theta = \frac{V_t - V_0}{V_0} \quad (2)$$

where V_0 is the initial volume of the exfoliated and V_t is the volume of the same region after different treatment steps.

The average volume expansion in the exfoliated layer is estimated to be 70% after the first 20 min. This essentially matches the sum of the volume of the CMAS plus that of the TBC which became exfoliated. This indicates that the depth of the exfoliated layer could be related to the CMAS loading. The following 20 min of thermal exposure has led to a small decrease in the volume expansion (to about

60%). Combining with the microstructure observation in Figure 2, this decrease in volume expansion is largely due to the deflation of the pores and the infusion of the CMAS into the intact layer. Eventually, the volume expansion has stabilised at around 60%.

Regarding the intact **layerunderlayer**, no appreciable volume expansion occurs in the first 20 min. However, DVC reveals a volume expansion as the CMAS begins to infuse into the intact layer over the next 20 mins and that this continues, if somewhat more slowly, over the following 20 mins, as shown in Figure 3, being greatest nearest the exfoliated layer and decreasing towards the backface (the detailed DVC results are shown in Appendix C). The fact that a dilatational strain is recorded through this whole region corroborates the inference from Figure 2 that there is a gradual infusion of this layer.

Given that the CMAS melts at $\sim 1230^{\circ}\text{C}$ [9], at 1250°C the molten CMAS can flow freely into the porous YSZ driven by gravity. Meanwhile, the fine pores/cracks also create a capillary pressure, which draws the molten CMAS into the pores. In order to compare the contribution from these two factors, two test coupons of the same size (1 mm x 0.3 mm x 3 mm matchstick) were prepared and sandwiched together with CMAS between them as illustrated in Figure 4 (a). From a CT scan of the stack prior to thermal exposure, the equivalent CMAS loading (assuming the loading was shared by the TBCs at top and bottom) was estimated to be $10.6 \pm 1.0 \text{ mg/cm}^2$. The stack was exposed to 1250°C for 60 min and then rescanned by X-ray CT.

Figure 4 (b) and (c) compare the virtual cross-sections through the centre of the TBC stack. There is no appreciable difference in the infiltration depth between the top and bottom TBCs. **This suggests that capillary pressure drives the CMAS infiltration process.** Both TBCs have each developed a two-layer structure rather like that in Figure 2. In contrast to that case, there is no sign of infusion into the intact layers possibly due to a lower CMAS loading.

1 In summary, non-destructive time-lapse X-ray μ -CT has been used to follow the
2 infiltration of CMAS into a free-standing APS TBC. Its non-destructive nature
3 combined with DVC means that it can detect quite subtle movements of the CMAS
4 and the associated strain changes over time. The evolution of the TBC
5 microstructure by the CMAS infiltration is marked and has been elucidated. Based
6 on these microstructure observations, the following conclusions can be reached:
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- 10 • The CMAS quickly (certainly within 20 minutes at 1250°C) infiltrates the
11 CMAS loading driven by strong capillary driving forces along the inter-splat
12 boundaries. This causes significant (70%) expansion and exfoliation of the
13 region immediately below the surface. The depth of this exfoliated region
14 appears to be limited by the CMAS loading.
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- 16 • Over a longer timescale there is some loss of CMAS from this exfoliated
17 region causing it to deflate slightly. This is accompanied by a much lower level
18 of swelling of the intact region beneath (<15%).
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- 20 • The onset of exfoliation at the coating surface is not dependent on the CMAS
21 loading while the depth of the exfoliated region and the onset of the
22 observable infusion into the intact layer are dependent on the CMAS loading;
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32 Under real service conditions, the CMAS would be expected to accumulate over
33 time. Given that infiltration is rapid this may cause progressive infiltration and
34 exfoliation with increasing depth over time. Further in practice the temperature
35 gradient through the TBC thickness may lead to different infiltration behaviour, limiting
36 the penetration depth. These idealised experiments suggest that time-lapse X-ray
37 and DVC could be combined in future with in-situ investigations of CMAS infiltration
38 to examine the effect of realistic temperature gradients as well as the effect of
39 constraint from the substrate. Nevertheless, these experiments provide valuable
40 insights into CMAS infiltration and can underpin models of the process needed for
41 better lifetime prediction.
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Figure captions

Figure 1 Comparison of the same region of interest for the as-received APS TBC after CMAS deposition (a) a virtual X-ray μ -CT slice and (b) the corresponding polished section viewed by SEM (BSE mode). The inter-splat pores (yellow arrow), fragmented YSZ zones (yellow square) and inter-splat cracks (red ellipse) are visible under X-ray μ -CT and SEM.

Figure 2 Cross-sectional virtual slice through the centre of the tested coupon showing the same region of interest at different stages of CMAS infiltration: (a) as-deposited and after exposure at 1200 °C for (b) 20 min; (c) 40 min and (d) 60 min. Videos showing the CMAS infiltration process are included in Appendix B.

Figure 3 Volume expansion profiles through the thickness of the TBC after different heat treatment time. The error bar for the volume strain in the intact layer represents the standard deviation from the DVC results.

Figure 4 Sandwich sample arrangement for infiltration test: (a) schematic showing the sample geometry and X-ray μ -CT virtual cross-sections of the same region of interest (b) before infiltration and (c) after thermal exposure to 1250°C for 60mins. The yellow dash line highlights the interface between the exfoliated layer and the intact layer.

(a)

(b)

(c)

Appendix A

Quantitative image analysis was conducted to estimate the local CMAS loading for each specimen. This was achieved by segmenting the cross-sectional micrographs obtained by X-ray μ -CT as shown in Figure A1(a) and (b). The volume of the CMAS is estimated by counting the number of voxels corresponding to the CMAS phase and the mass of the CMAS is estimated by multiplying its volume with its density ($\sim 2.69 \text{ g/mm}^3$ [27]).

Figure A1 X-ray μ -CT micrographs for estimation the local CMAS loading: (a) raw virtual slice; (b) segmented image with the CMAS and YSZ coating coloured differently and (c) 3D volume rendering showing the CMAS on top of the YSZ coating.

Appendix B

Video B1. Video showing the same virtual cross section through the middle of the test coupon at different stages of CMAS infiltration.

Appendix C

The average volume expansion for the intact layer has been calculated according to Eq. 1 using the displacement field determined by DVC. The displacement fields determined by DVS are shown in Fig. C1. The top exfoliated layer has deformed too much for correlation to be achieved. The first 20 min has induced little swelling in the intact layer. The outward-pointing arrows as seen in the virtual cross sections for 40 min and 60 min suggest a volume expansion induced by the CMAS.

(a)

(b)

(c)

Figure C1 Displacement fields below the severely infiltrated zone induced by CMAS infiltration after heat treatment at 1200 °C for (a) 20 min; (b) 40 min and (c) 60 min. The arrows coloured using the magnitude of the displacement vector.