Latest advances in characterization of metallic materials for aerospace industry at European Synchrotron Radiation Facility (ESRF)

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Abstract
In 2021, the European Synchrotron Radiation Facility ESRF – EBS (Extremely Brilliant Source) was upgraded, becoming the first new generation high-energy synchrotron. The highlights of the research in metallic materials characterization for aerospace industry will be presented:
• Residual stresses and their relief have been measured in an additive manufactured 316L stainless steel arch structure;
• Creep samples of a serf-healing Fe-3Au-4W (wt.%) alloy have been investigated, using nano-Computed Tomography, observing the cavities and the precipitates;
• Ductile fracture is examined, through in-situ laminography in AA2024-T3 aluminium alloy, investigating the evolution of void and particle populations and the local strain field.

1. Introduction

The European Synchrotron Radiation Facility (ESRF) has enabled for more than 30 years major scientific breakthroughs in the understanding of living and condensed matter. In 2021 was completed the upgrade of ESRF – EBS (Extremely Brilliant Source), which is opening new vistas for X-ray science, providing a growing community of users with unique facilities to tackle the complex global challenges facing our society. ESRF – EBS is the first high-energy fourth-generation synchrotron light source, with X-ray performances increased by a factor 100 compared to the previous source and with a 30% reduction in electricity consumption. ESRF-EBS hails a new era for X-ray science in the exploration of matter at all scales and down to the atomic scale. As a result of its properties, synchrotron radiation is used increasingly as a response to industrial challenges related to the life cycle of materials, observing, characterizing and understanding the structure of matter are at the heart of these challenges for industry [1].

The aerospace industry is one of the most successful industries in Europe, but also one of the most challenging, and it is characterized by a high degree of research intensity and rapid developments. Due to this, aerospace industry has a high strategic importance in the development of innovative technologies and an important contribution to this leadership role is the innovative capacity of the suppliers with their production equipment and technology research [2].

The highlights of the research activities in characterization of metallic materials for aerospace industry made in these 2 years will be presented:
• Residual stresses and their relief after heat treatment have been measured in an additive manufactured 316L stainless steel arch structure, using synchrotron x-ray diffraction. The residual stresses, generated by the additive manufacturing production process, have been measured comparing the synchrotron x-ray diffraction spectra to the stress-free powder spectrum. Further, the additive manufactured part has been heat treated at 700 °C with a soaking time of 2 hours. Then, the residual stress relief has been evaluated. These experiments are part of EU-funded EASI-STRESS project [3];
• Creep specimens, which have experienced creep tests at constant 145 MPa σ at 550 °C and interrupted after 10h, 50h, 100h, 150h and 223h, performed on a self-healing Fe-3Au-4W (wt.%) alloy, have been investigated using synchrotron x-ray nano-Computed Tomography (CT). This technique allows the observation of 3D microstructure of the cavities and the precipitates, differentiating Au-rich and W-rich ones. Nano-CT measures also the orientation of cavities and precipitates compared to the stress direction [4];

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Ductile fracture under simple shear stress $\tau$ is examined, using synchrotron x-ray in-situ laminography in an aluminium alloy AA2024-T3. This analysis has been used to investigate the evolution of void and particle populations and perform Digital Image Correlation (DIC) to assess the local strain fields. Thus, the damage mechanisms leading to fracture have been highlighted [5].

**Stress relief of the residual stresses of an additive manufactured AISI 316L arch structure (part of EU-funded EASI-STRESS project)**

Laboratory-based residual stress measuring techniques often are not effective assessing the residual stresses on relative bulky components, due to the nature of residual stresses to be assessed and the given material system. However, synchrotron hard x-rays with their higher penetration depth can access more easily those areas, reducing the number of processing steps (e.g. tooling or machining) to investigate residual stress fields. Further, synchrotron hard x-rays may allow the possibility to perform this test non-destructively [6].

An arch concept was selected as a good candidate to measure residual stresses in additively manufactured parts due to its geometry, since it contains a void, which is one of the design advantages of employing additive manufacturing. Further, the chosen component lends itself to establishing a triaxial stress state, which has been largely absent from additive manufacturing benchmarks for residual stresses to date. The arch part, shown in Figure 1, has been produced by laser-based powder-bed fusion (L-PBF) and printed onto a build platform, leaving the specimens attached after building, such that the measurements could observe real stresses in components which have been generated during the additive manufacturing process. The base material selected was stainless steel 316L, due to its very stable austenitic microstructure throughout the continuous thermal cycling that material experiences during L-PBF processes. Further manufacturing parameters are listed in Table 1.

<table>
<thead>
<tr>
<th>Layer parameters</th>
<th>Spot size: 100 $\mu$m</th>
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<tbody>
<tr>
<td>Laser parameters</td>
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<td>Recoating parameter</td>
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<tr>
<td>Powder characteristics</td>
<td>AISI 316L, size distribution 15-45 $\mu$m, R10 recycling level</td>
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Table 1: Processing parameters for the additive manufacturing arch sample.

Figure 1: Additively manufactured arch geometry, primary L-PBF scan strategy and coordinate system.
The effectiveness of stress-relieving heat treatment performed on this specimen has also been examined. The specific treatment selected has the objective of maximizing stress relief, whilst preserving the underlying microstructure. Thus, the sample has been held at 700 °C for 2 hours and then cooled in an inert atmosphere passively, as prescribed by current best practice. Indeed, it has been shown by previous observations of L-PBF 316L that the underlying microstructure remains unchanged up to 600 °C. At 700 °C, the microstructure appears quite stable and only minor precipitation at long holding times (> 20 hours) has been observed. However, the microstructure undergoes strong modifications at 800 °C very quickly and in 30 minutes the solidification cells are completely destroyed [7].

The residual stress measurements have been performed at ESRF ID15A beamline in x-ray diffraction (XRD) full transmission geometry, using a synchrotron white beam, measuring on two planes XZ and YZ with a gauge volume of 0.2 x 3.4 x 0.05 mm. The raw powder spectrum has been used as stress-free reference, resulting in \( d\theta \) value equal to 1.0806. Finally, the residual stress values have been calculated comparing the Fe FCC(311) peaks and using a Young modulus \( (E) \) equals to 193 GPa and a Poisson’s ratio \( (\nu) \) of 0.28. The results are displayed in Figure 2.

Figure 2: Comparison of the residual stress \( \sigma_{xx} \), \( \sigma_{yy} \) and \( \sigma_{zz} \) measured in the as-built (AB) sample and in the same part after heat treatment (HT).

It has proven that residual stresses in L-PBF components in the as-built (as the AB sample) condition are typified by high tensile stresses at the periphery, balanced by compressive stresses at the centre, and the highest residual stress is typically proximate to the last layer deposited [8]. With the application of a stress-relief heat treatment (as experienced by the HT sample), those regions that have a high initial deviatoric stress state respond the most, on the other hand, regions having a more balanced hydrostatic stress state respond the least. The deposition strategy and geometry were chosen in order to roughly balance the stresses in the \( x \) and \( y \) direction through the top of the arch. Moreover, the centreline is expected to be in a state of plane stress in the \( x \) and \( y \) directions. This is shown in Figure 2. The stresses in the \( x \) and \( y \) direction are similar in terms of their positions along the ligament and magnitudes, starting from the top of the ligament at \( z=0 \) with extremely high residual tensile stresses. For the as-built component in \( z \) direction, the measurements demonstrate the plane stress state (e.g. \( \sigma_{zz}=0 \)) particularly near-surface, as it was expected, confirming the hypothesis of a stress state close to being plane stress.

The HT sample shows the same trends observed for the AB sample for the stresses acting in \( x \), \( y \) and \( z \), respectively. However, the peak in tensile stress has been strongly reduced by approximately 300 MPa, which is most occurring at
the top surface of the part. Further observations for the heat-treated results show that the regions with the highest residual stresses relaxation are located near surface and at the arch apex. While stresses acting in the $x$ and $y$ were approximately balanced in the as-built condition, there is a subtle difference of approximately 100 MPa at the apex of the arch between $x$ and $y$. The $x$ direction shows higher compression stresses than $y$, due to the level of constraint being higher in the $x$ direction, since more material is present by geometry in this orientation overall as opposed to $y$, which spans the arch. A slightly compressive stress, acting through thickness of the ligament along the $z$, balances the discrepancy between $x$ and $y$ directions.

Finally, residual stress measurements performed by synchrotron white beam in XRD full transmission geometry appear to be promising for measuring non-destructively triaxial residual stress fields in relative bulky components.

**Synchrotron x-ray nano-Computed Tomography of self-healing Fe-3Au-4W (wt.% ) alloy creep specimens**

Metals experience creep, a time-dependent plastic deformation, when exposed to a combination of a high temperature and constant stress levels even below the quasi-static yield stress, ultimately leading to failure due to accumulated damage. Creep damage often begins with the nucleation of grain boundary cavities, which further grow and coalesce into larger cracks.

To improve the creep resistance, the traditional mechanism is to tune the microstructure and composition, such that the initiation of the cavities is postponed as long as possible by precipitation strengthening, solid solution strengthening, dislocation hardening or the combination of these mechanisms. However, since the nucleation of the cavities is unavoidable, self-healing alloys have been proposed as an alternative strategy, in which the early-stage cavities can be filled autonomously by the precipitation of the supersaturated solute atoms pre-dissolved in the matrix. Further, the continuous growth and the coalesce of the cavities can be postponed [9].

The damage and healing behaviour in a Fe-3Au-4W (wt.%) ternary system, in which two healing agents can act simultaneously, has been investigated. In bcc-Fe, the solute Au and W atoms both diffuse faster than the host Fe atoms and this condition is essential for an effective healing of creep cavities by solute transport. Creep experiments at 550 °C with a constant stress of 145 MPa were applied and stopped after different times to investigate the microstructure development at different stages of creep. The results are reported in Figure 3.

![Creep curves for the Fe-3Au-4W (wt.%) alloy samples at a temperature of 550 °C with a constant applied load of 145 MPa.](image-url)
Synchrotron X-ray tomography has been used to visualise the evolution of both the grain-boundary cavities and the precipitates within the cavities at different stage of creep, and the interaction between the two healing agents with two voxel sizes of 100 (Figure 4) and 30 nm (Figure 5).

Figure 4: Segmented volume rendering of the cavities (orange) and precipitates (blue) in the Fe-3Au-4W (wt.%) alloy after creep at 550 °C with a constant applied stress of 145 MPa. The images are obtained with a voxel size of 100 nm. The applied stress is normal to the top view.
In the sample after 10 h of creep, some cavities and sporadic precipitates are observed. Most of the cavities are empty, indicating that the cavities are in their initial growth state just after nucleation. In the sample after 50 h of creep, more creep features, especially more precipitates, are observed; moreover, the cavities and precipitates start to show interaction: one or more precipitates are usually found in the vicinity of a cavity, indicating that the precipitates have multiple nucleation sites at the cavity surface. In 100 h sample, both the cavities and the precipitates are located at the grain boundaries, especially oriented perpendicular to the load direction, and experience growth. Cavities on some grain boundaries are significantly elongated, indicating a fast propagation of the cavities in that direction. In 150 h sample the competition between cavity growth and precipitate healing can be observed: the precipitates form around the cavity surface until the cavity is eventually wrapped by the precipitates. The morphology of the cavities and precipitates provides indirect evidence that the precipitates are formed after the occurrence of the cavities. In the sample after 223 h of creep, a late stage of cavities linkage can be observed, resulting in a cleavage of the grain boundaries (Figure 6).

Figure 5: Examples of segmented volume rendering for creep cavities and healing precipitates. The orange and blue volumes indicate cavities and precipitates, respectively. Precipitates were found at the notch and at the linking point of the cavities.
In previous studies, it was found that the isolated and linked cavities show a different healing behaviour. The isolated cavities can be healed continuously until a fully filling is achieved, while a linkage of the cavities can cause a sharp drop in filling ratio [10]. In the present work, linkage of the cavities is not frequent due to the limited creep strain; nevertheless, as shown in Figure 7, the average filling ratio of the partly filled cavities decreases with time, indicating an increasing cavity volume and a decreasing average filling ratio. This occurs since the partially-filled cavities and the precipitates therein both experience an increase in volume fraction, but the volume fraction of cavities increases faster, resulting in a decrease in the filling ratio.

Figure 6: Cavities partly filled by both Au-rich and W-rich precipitates in the 223 h sample.

Figure 7: Filling ratio of individual cavities as a function of the cavity volume for the Fe-3Au-4W (wt.%) alloy after different creep times.
The distribution in orientation of the cavities and the precipitates as a function of the angle between the long axis of the cavity/precipitate and the stress direction is reported in Figure 9. After 10 h of creep test, the freshly nucleated cavities show no preferred orientation. Increasing creep time to 50 h and beyond, most cavities are found to form with an angle close to 90°, indicating that the creep cavities prefer to nucleate at the grain boundaries oriented perpendicular to the stress direction and that they tend to propagate in the grain boundary surface. The precipitates also show a preference for a 90° angle with respect to the loading direction for creep times higher than 100 h. The time lag in the formation of the preferred orientation between the precipitates and the cavities provides strong evidence that the creep cavities initiate the formation of precipitates, since cavity nucleation has already started at the earliest creep, while the preferred orientation of the cavities only develops later. The precipitates nucleate after the occurrence of the cavities, and, thus, they grow in accordance with the shape of the cavities.

Thus, synchrotron X-ray nano-tomography have been performed on Fe-3Au-4W (wt.%) self-healing alloy samples being exposed to the same creep condition for different loading times. It showed the creep and the self-healing mechanisms: during creep, the grain-boundary cavities are continuously formed and subsequently healed by precipitation on the cavity surface. It allowed to highlight that the linkage of the cavities is rare, due to the limited creep strain and the low strain rate and cavities nucleated at an early creep stage can be fully healed, while due to a decrease in the diffusional solute flux and the inter-cavity spacing over time, a longer time is required to fully fill the late formed cavities. This results in an overall decrease in filling ratio of the partially filled but still growing cavities with time. X-ray nano-tomography has also determined that supersaturated Au solute diffuses significantly faster than the supersaturated W, resulting in a faster filling of creep cavities by Au-rich precipitates than W-rich precipitates. This difference in healing kinetics indicates that supersaturated Au and W may operate on different time scales, and combining both can significantly extend the time scale over which self-healing of creep damage can potentially be achieved [4].

### Synchrotron X-ray nano-Computed Tomography of self-healing Fe-3Au-4W (wt.%) alloy creep specimens

Ductile failure is the process in which plastic deformation promotes, through void nucleation, growth and coalescence, the progressive damage of polycrystalline metals until fracture initiation. Synchrotron laminography has proven to be particularly suitable for investigating specific regions of interest in sheet metal, using as “natural markers”, second phase particles or voids and allowing Digital Volume Correlation (DVC) [11]. The ductile fracture under simple shear is examined using X-ray laminography in an aluminium alloy AA2024-T3 1 mm thick sheet. Projection digital image correlation (DIC) is employed extensively to analyze the strain field and track individual voids through the loading steps. The laminography experiment is performed through a displacement controlled device, symmetric and monotonic stepwise loading via a dedicated electromechanical loading device is performed at 30 μm/min. The scanning region is positioned between the roots of the two notches on one side of the sample (Figure 10). Six loading steps are applied...
prior to fracture with the corresponding force levels (-300N, -600N, -700N, -790N, -865N and -920N) and a final scan is performed on the broken specimen. A pink beam from a single-line undulator with a 13 mm period and a peak X-ray energy around 26 keV is chosen, as it allows for a good compromise between X-ray transmission/penetration power and image contrast. The physical size of one cubic voxel is equal to 0.65 μm.

Figure 10: Sketch of the laminography specimen with highlighted area of interest.

The strain field has been measured by projection DIC and in Figure 11 the evolution of the effective strain field on the specimen gauge section is reported. Throughout the test, a localization of the deformation is observed, in between the two cutouts, close to the notches, two highly-strained regions form near the free top and bottom surface edges, while towards the middle of the gauge section the strain levels decrease slightly (~20%). A homogeneous strain field is observed in the highly stretched regions, with an effective strain of 0.3 in the last step prior to failure. The defects population consists of small and large intermetallic particles, as well as voids. The material features a high initial volume fraction of pre-existing voids in the matrix (around 0.7%). These defects and their own features have been tracked at each experimental step as shown in Figure 12.

Figure 11: Effective strain field evolution during the loading history measured by projection DIC.
Based on the experimental observations, the timeline from damage initiation to final fracture for the examined AA2024-T3 can now be based on the evolution of the two main features of the microstructure observed in the laminography experiments: intermetallic particles and pre-existing voids.
Figure 16a highlights the steps leading to the failure, which is depicted in Figure 16b. Indeed, between steps 2 and 4, early into the plastic loading, at low effective strain levels of the matrix material of less than 0.1, the strong but brittle intermetallic particles break along the axis normal to the direction of principal stress, thus, almost perpendicular to the direction of maximum principal strain, and at 45° to the orientation of the final crack. At the same time, the pre-existing porosities follow the matrix material deformation and rotation with no noticeable change in volume.

Between steps 4 and 6, the intermetallic particles continue to break, fragmenting multiple times normal to the direction of principal stress, while the previously formed gaps grow with the change in the matrix strain field. The orientation of the gaps in conjunction with the strain field should produce shrinkage normal to the maximum principal strain and extension along that axis; however, void contraction is impeded by the debris of the fragmented particles leading to void growth. At the same time, de-cohesion between the particle and the matrix is observed, induced by the mechanisms previously described. On the other hand, the deformation of pre-existing voids continues to be primarily dictated by the rotation of the matrix and is not prone to any sudden event, even under large deformations.

Figure 16: Crack mechanism as observed around an intermetallic: evolution of an intermetallic particle with loading (a) and final laminography and 3D reconstruction view of a crack formed at the tip of an intermetallic (b). Both scale bars represent 100 μm.
Between steps 6 and 7, formation of micro-cracks occurs (Fig. 16 b) and the voids between the fragmented particles continue to grow. At this stage, smaller particle debris seems to be able to move freely leading to wider gap opening (Figure 16b top and right arrow). The larger particle fragments are confined by the motion of the matrix material (Figure 16b left arrow), resulting in stress concentrations around them and subsequently micro-cracks are generated. The large extension of the void on the broken sample appears to be a sudden event, which could not only be related to the matrix deformation kinematics, but rather to the accommodation of the local stress field variations induced by the failure of the intermetallic particle and the formation of strain localization band. This feature’s existence can only be proven experimentally by the change in the shape and size of the pre-existing voids from step 6 to 7. Here, close to the location of the final crack, a significant rotation and straining is observed.

Thus, the chain of events leading to fracture of AA2024-T3 under shear begins with the failure of the brittle intermetallic particles normal to the maximum principal stress direction, followed by the stagnation of the particle fragments in the displacement field of the matrix, which induces a void growth mechanism.

This fracture mechanism bears similarities with the shear fracture mechanism of FB600 steel [12]. In the present study, in-situ X-ray synchrotron laminography is used to observe the failure mechanism of a 2024-T3 aluminium alloy under shear-dominated loading. Consistent with the kinematics of simple shear, an almost constant pre-existing void volume is observed throughout the experiments along with a strong elongation and flattening of the voids. Pre-existing voids deform by rotating, elongating and closing, thereby strictly following the homogenized strain field. The void volume fraction shows a significant growth of newly nucleated voids just towards the end of the tests. On the other hand, intermetallic particles fail in a two-stage manner. At low strain levels, a first crack forms inside the particles normal to the direction of the maximum principal stress. These nucleated voids open up during loading, thereby leading to large cavities. Finally, the broken intermetallic particles are subjected to large rigid-body motions upon failure. Away from the intermetallic particles, the fracture surface is covered by small ductile dimples containing dispersoids.

In addition to experiments, an RVE simulation with periodic boundary conditions is performed to model the failure mechanism of the intermetallic particles. The simulation results show that opening of the pre-cracked particles leads to high strain concentrations at the mesoscopic level that ultimately results in strain bands linking the particles.

References

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