Correlative Nano-Computed Tomography and Focused Ion-Beam Sectioning: A Case Study on a Co-Base Superalloy Oxide Scale

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Herein, the capabilities of a correlative tomography approach combining laboratory nano X-ray computed tomography, focused ion-beam sectioning, and energy-dispersive X-ray spectroscopy are utilized for the characterization of multilayered oxide scales of Co-base superalloys regarding their 3D morphology and chemical composition. The combination of complementary 3D imaging techniques allows for a precise and reliable segmentation of the pore space, oxide precipitates, and different phases of the initial material. Such information is instrumental to a microscopic understanding of oxidation in this new class of superalloys and key to improvement of oxidation resistance in the next-generation high-temperature materials.

1. Introduction

In recent years, the applicability of Co-base superalloys as high-temperature material has been a research topic of high interest. This new class of superalloys was first reported by Lee and rediscovered in 2006 by Sato et al. So far, the mechanical properties of Co-base alloys have been studied extensively and they are deemed promising for applications in the intermediate-temperature regime. Phase stability and high-temperature mechanical properties were significantly improved in the last years by addition of a wide range of alloying elements. However, to assure long-term stability of these new superalloys in application, not only their mechanical properties but also their resistivity against oxidation have to be studied and improved. For the understanding of elementary (transport) mechanisms during scale growth on complex Co-base superalloys under oxidizing atmospheres, it is crucial to gain insight into the porosity, pore connectivity, as well as phase formation and further evolution upon exposure at elevated temperatures. Recently, research on oxidation of Co-base superalloys has attracted a lot of interest and considerable progress has been achieved by dedicated alloy design.

Since pores and phases of these complex multilayered oxide scales extend into 3D with both nanometer- and micrometer-sized features, a site-specific, scale-bridging, and 3D characterization approach is required. One commonly used technique is focused ion beam/scanning electron microscopy tomography (FIB/SEM tomography or, shortly, FIB tomography). Utilizing the so-called “slice & view” technique, a region of interest (ROI) is serially sectioned by ion milling and every newly generated surface is imaged with the electron beam using either secondary electrons (SEs) or back-scattered electrons (BSE) or both. This technique provides stacks of high-quality SEM images for 3D reconstructions. Additionally, most FIB/SEM instruments offer the possibility to complement imaging with energy-dispersive X-ray spectroscopy (EDXS) for chemical analysis. The major drawback of FIB tomography, however, is its destructive nature. Since the ROI is successively milled away, the same sample volume cannot be further analyzed by complementary methods or tested (e.g., mechanically) once the 3D data have been acquired. In addition, FIB tomography can lead to difficulties when analyzing highly porous structures without structural integrity, nonconductive samples, or beam-sensitive materials. There are partial solutions and work-arounds for these problems. However, the destructiveness of the method is unavoidable.

In recent years, new developments in X-ray optics have made synchrotron technology with optical resolutions down to 50 nm accessible to lab instruments. These lab-scale X-ray nano-computed tomography (nano-CT) instruments offer a nondestructive 3D characterization on comparable length scales to FIB tomography. Equipped with capillary condenser, Fresnel zone plate objective, and Zernike phase ring, instruments such as the ZEISS Xradia 810 Ultra are capable of...
capturing 3D microstructures down to a spatial resolution of (50 nm)$^3$ with a quasimonochromatic X-ray beam.[32] While current nano-CT instruments are able to supply viable quantitative data on 3D sample morphologies, phase identification and image segmentation remain major challenges of this method.[37] However, due to its nondestructive nature, nano-CT facilitates in situ studies as well as subsequent investigations by complementary characterization techniques.[38]

Here, we present a characterization workflow that overcomes these problems by combining high-resolution nano-CT with FIB tomography. Simultaneous SEM imaging and EDXS mapping of same areas allow for a precise image segmentation and chemical phase identification and, beyond that, ultimately allows for the SEM/EDXS-informed reconstruction of complex 3D microstructures.

2. Results

2.1. Sample Preparation Routine

A cross section of the oxidized ERBOCo-1 sample is shown in Figure 1a. After exposure to synthetic air at 900°C for 12 h, the surface is covered by a 6–8μm multilayer oxide scale. In the following, the distinguished regions in the scale are named according to the nomenclature introduced in a previous study by Weiser et al.[3] We define the surface normal to be the z-direction; x and y are chosen arbitrarily. The Ni coating at the top is followed by a polycrystalline outer oxide layer ($d_1$) of about 3–3.5μm thickness. Two distinct layers make up the inner oxidation zone, the fully oxidized inner oxide layer $d_2$ and the internal precipitation region $d_3$. Below the internal oxidation front (IOF), the unoxidized superalloy single crystal with the distinct $γ/γ'_0$ microstructure is present. The layers will be referred to as $d_{1-3}$ and SX (single crystal). After cross sectioning and SEM pre-characterization, ROI was defined and covered with a protective carbon layer to avoid FIB damage during preparation. As shown in Figure 1b, a rectangular block with edge lengths of about 40 μm x 40 μm x 60 μm was cut out with the Ga$^+$ ion beam and attached to a micromanipulator. The block was transferred to a stainless steel pin and was fixed to it using carbon deposition. To avoid shadowing effects toward the sample by the support pin during the 180° rotation, the sample was stacked on top of a previously deposited carbon pad. Afterward, the sample was tilted toward the ion beam and was milled down to a final diameter of 15μm in annular fashion (see Figure 1c,d). Figure 2a shows an SEM image of the nano-CT pillar on top of the support pin after the final annular milling step.

2.2. Nano-CT

After the preparation routine described earlier, the pillar was transferred to the ZEISS Xradia 810 Ultra. A versatile feature of this particular X-ray microscope is the possibility to operate it in either absorption contrast mode (AC) or phase contrast mode (PC). The former mode is well suited for studying materials with medium or large densities showing considerable X-ray absorption in the size range of nano-CT samples. The latter mode can also be used for imaging materials with lower densities as long as phase shifts between adjacent regions of the sample are introduced.[32,39] The PC mode generates contrast by inserting a Zernike phase ring into the beam path which introduces a +3π/2 phase shift of the nonscattered X-rays with respect to the scattered ones, thus transforming phase shifts into amplitude contrast.[40] Particularly,
strong phase shifts occur at sample boundaries and pores and give rise to pronounced edge contrast. Thus, PC is well suited for revealing pore structures or light inclusions in a matrix of higher mass density. On the downside, the more complex contrast formation in PC mode results in apparent image artifacts, such as halos (at sample boundaries or pores) and shade-offs (in regions away from edges), which can make reconstruction and correct segmentation of 3D data challenging. Figure 2b,c shows transmission images of AC and PC, respectively. For this particular study, the latter was chosen for 3D analysis because of the higher contrast between the internal pore/precipitate structures and the surrounding matrix. In Figure S12, Supporting Information, we present an example of topologically closed-packed (TCP) phases in an oxidized ternary Co9Al9W. In this case, AC imaging is the method of choice because of the contrast generated by plate-like phases enriched in W. Depending on the material system to investigate, either contrast mode (or both in conjunction) can be utilized for a detailed analysis.

In our setup, the acquisition of AC and PC image tilt series can be performed in direct succession guaranteeing that the identical sample volume and orientation are studied in both modes. The pillar geometry of the sample allows for a full 360° tomography scan without missing-wedge artifacts. In a first step, both tilt series, taken in AC and PC mode, respectively, are automatically reconstructed, segmented, and visualized using standard analysis. For the present sample, the reconstruction of AC data mainly reveals the mass difference between the Ni coating, the two layers d2 and d1 containing light metal oxides, and the subjacent unoxidized superalloy (see Figure 2a and Figure 1a). The inner oxide layer d3 is analyzed using PC data because the edge enhancement of the Zernike PC improves the visibility of pores and light metal oxides.

One immediately apparent problem of standard analysis is that the porosity–distance plot derived from the PC data set shows a pronounced peak directly above the unoxidized superalloy (see gray graph in Figure 3b) where no pores are revealed in the SEM image of the sample cross section. Rather, the position coincides with the internal precipitation region d1 in Figure 1a, suggesting that the alumina particles in the referred layer were falsely attributed to the porosity. Indeed, a detailed examination of virtual slices through the nano-CT reconstruction reveals that both in PC and in AC mode, it is hardly possible to differentiate between pores and light metal oxides (e.g., alumina). A threshold-based image segmentation (manually or automated) is not applicable and renders a quantitative analysis of the reconstructed volume impossible. Moreover, for the case of PC imaging, the virtual slices through the reconstructed 3D data set exhibit a halo-like contrast at the transition from pore to surrounding material. This is a major drawback of the method and cannot be evaded in a routine measurement to the current date. The segmentation error caused by gray value thresholding images with such halo features complicates a quantification of the pore volume and can lead to artificially inflated porosity values.

In the following section, we describe a method of using manually segmented SEM images of relevant layers to calibrate and validate the thresholding parameters of the virtual slices of nano-CT data sets.

### 2.3. Correlative FIB/SEM Tomography

To overcome the problems of image segmentation and phase identification mentioned earlier, we developed the correlative tomography approach shown in Figure 4. The first step is the acquisition of AC and/or PC nano-CT tilt series. The tilt series are used to reconstruct the sample volume, which is then screened for specific features, e.g., structured pores, precipitates, channels, or cracks (see Figure 2). Once these features are identified and all nondestructive measurements are performed, the nano-CT pillar is transferred back to the FIB/SEM instrument for a correlative SEM–nano-CT analysis. To enable cutting of the pillar perpendicular to the pillar axis and simultaneous SEM imaging of the newly created pillar cross section, the sample is mounted onto a 52° pretilted holder. As shown in Figure 4b,c, only selected FIB cuts are performed to uncover the beforehand identified ROI, enabling simultaneous SEM imaging and EDXS mapping of the newly created surfaces. As nano-CT already provides a high-resolution 3D data set, a full FIB tomography is not required anymore. In comparison with a complete “slice and view” series, only a few specific SEM images (typically 1–10, depending on the sample complexity) need to be acquired and segmented. Due to the small number of single SEM images, their segmentation can even be performed (or proven) manually with great precision and reasonable time and effort. Additionally, EDXS elemental distribution maps of the same slices are acquired simultaneously to gather...
and identify relevant phases. The information obtained from the correlative FIB tomography measurements is then fed into the segmentation and quantitative analysis of the nano-CT data, yielding SEM/EDXS-informed nano-CT reconstruction, as shown in Figure 4d.

2.4. SEM/EDXS-Informed Nano-CT Reconstruction

In the case study presented in this work, after the nano-CT experiments, we performed ten FIB cuts and acquired SEM images and EDXS maps from the resulting cross sections. The correct position of the high-resolution SEM images of the horizontal cuts is then traced back and identified inside the nano-CT 3D reconstruction. The tilt and rotation angle of the virtual slices are adjusted until the best congruence between the virtual slices through the nano-CT data set and the SEM images is obtained. To facilitate the alignment procedure, distinctly shaped pores and particles were used for orientation and shape matching.

One representative example of this procedure is shown in Figure 5. The pore marked by the red circle in the SEM image, Figure 5a (top), was traced back and identified in the corresponding nano-CT virtual slice (bottom). A comparison of the two images shows excellent agreement of the pore structures across the whole image area. Even though the spatial resolution of the nano-CT virtual slice is significantly lower, almost all pores identified in the SEM image are also revealed in the nano-CT virtual slice.

However, both images show contrast phenomena which render reliable determination of 2D porosity by automatic gray value thresholding (e.g., in the study of Ridler and Calvard[43]) impossible. In the case of the SEM image, the SE contrast leads to increased intensities at pore edges (edge contrast) and steep pore walls (tilt contrast). Additionally, surface curtaining from FIB cutting gives rise to pronounced topography contrast even in regions without pores. In the case of the virtual nano-CT slice, the halo contrast of the PC mode already discussed earlier leads to an edge enhancement similar to the edge contrast in SEM. However, in

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Figure 3. Quantitative analysis of the SEM/EDXS-based reconstruction. a) 3D rendering of the analyzed volume (closed porosity (blue), interconnected porosity (green), Al₂O₃ precipitates (yellow) and cast porosity (red)), and corresponding distance to the single crystal (SX) over porosity plot b) showing five distinct peaks in regions with higher porosity. c) Histogram of the maximum Feret diameter of the alumina precipitates in side view and top view.

Figure 4. Sequence of the correlative tomography workflow: a) Qualitative 3D reconstruction using nano-CT to identify ROI for further analysis. Selected FIB cuts are performed to lay bare a ROI and enable simultaneous b) SEM and c) EDXS imaging. d) SEM image-based segmentation together with additional chemical information from EDXS allows for a reliable interpretation and quantitative analysis of the nano-CT reconstruction.
contrast to SEM, the significantly lower resolution of nano-CT makes it impossible, even by careful visual inspection and manual segmentation, to pinpoint the real position of pore edges with sufficient precision to extract reliable values for 2D porosity. Thus, nano-CT alone is not suitable for determining absolute porosities, at least for the sample and pore sizes studied here.

The fact that pore edges can be reliably determined from manual evaluation of SEM images (with precision in the range of nanometers), even in the presence of the contrast phenomena mentioned earlier, gives us a handle to improve the quantitative evaluation of porosity from nano-CT data by SEM-informed image segmentation. This correlative SEM/nano-CT approach is exemplarily illustrated in Figure 5b for the image areas already shown in Figure 5a. By careful manual segmentation of the SEM image, a good representation of the pore structure (marked in blue) is achieved, resulting in a 2D porosity of 10.4% for this particular cross section. The porosity value is subsequently used as a calibration for thresholding of the nano-CT virtual slice meaning that the threshold of the latter is adjusted until the same 2D porosity value is obtained (Figure 5b, bottom). By comparing the two images, it is shown that most of the pores identified in the SEM image are also present in the segmentation of the nano-CT virtual slice, even though the shape and interconnectivity of pores slightly deviate. This is to be expected as those details are at the limit of the resolution of nano-CT where halo contrast also comes into play. However, since the porosity analysis is based on high-resolution SEM data, we expect that the overall 2D porosity and pore arrangement are well represented with the present threshold of the nano-CT data. After this threshold calibration of the selected virtual slices, the same threshold is applied to the whole stack of the nano-CT 3D reconstruction, assuming that the limited resolution and image artefacts of nano-CT have the same effect on the apparent 2D porosity across the whole sample volume. Owing to the homogeneous and isotropic contrast throughout the slices of the nano-CT data set, this is a reasonable assumption, and one calibration step each, to determine the threshold values for the pore space and for the oxide particles, is assumed to be sufficient.

To discriminate between pores and light oxide precipitates in the 3D tomogram derived from nano-CT, a correlative approach involving SEM analysis is again required, as illustrated in Figure 6. Figure 6a shows an SEM image of a pillar cross-section which was prepared by FIB cutting after the nano-CT analysis was completed. The cross section is slightly inclined with respect to the layers of the oxide scale, thus revealing different depths in the sample across the imaged area. On the left side, the unoxidized superalloy with the characteristic $\gamma/\gamma'$ microstructure can be seen followed by a layer with large pores, a layer with oxide precipitates, and further layers with smaller pores (right). The arrows mark a representative large pore and alumina precipitate which appear with different contrast. The corresponding virtual slice through the nano-CT data set is shown in Figure 6b. Again, as in Figure 5, the arrangement of features revealed in the virtual slice is in excellent agreement with SEM. However, in contrast to SEM, the pores and alumina precipitates show almost identical contrast. From this comparison,
it is quite clear that pores and light oxide phases cannot be reliably discriminated based on nano-CT contrast alone but require additional information from SEM or EDXS.

In particular when using BSE for imaging (cf. Figure 6a), the different contrasts of pores and oxide phases can be used to differentiate between these constituents of the oxide scale. Additional chemical information can be obtained by acquiring EDXS mappings from cross sections cut by FIB. Figure 6c exemplarily shows elemental distribution maps of a cross section containing alumina precipitates and a single large pore. The enrichment of Al and O and respective depletion of Co clearly identify the dark contrast features in the SEM image as alumina precipitates. Based on SEM contrast alone, this interpretation would rely on presumptions or require prior knowledge of the oxidation process. In particular, EDXS becomes important when different oxides phases coexist in the oxide scales which is the common scenario.[2,3,20,21,25,44] The large pore can already be identified from the shadow contrast in the SEM image. Accordingly, the EDXS mappings do not show Al (area marked by the red circle) and O in this area but only slight depletion of Co which might result from partial absorption of X-rays generated in the material below the pore on their path to the detector. In summary, the EDXS analysis in Figure 6c corroborates the interpretation that the dark precipitates in the internal precipitation region d₁ are alumina precipitates. This knowledge of the precipitate nature is fed into the 3D segmentation process which we refer to as SEM/EDXS-informed nano-CT reconstruction as sketched in Figure 4d.

The key contribution of EDXS is the unique chemical identification of different phases (here only alumina) and pores, whereas SEM provides high-resolution images with material and pore contrast, enabling precise identification of phase boundaries and pore edges for a highly accurate segmentation.

2.5. Quantitative Analysis and Interpretation of Microstructure Data Set

Figure 3a shows the ROI extracted from the nano-CT data with the SEM/EDXS-informed segmentation (side view). For 3D segmentation, including the applied thresholding calibration, the whole SEM-informed 3D data set was first cropped into rectangular areas, and the different features are then segmented using gray value thresholding combined with a particle volume filtering algorithm (not considering volumes smaller than 0.065 μm³) to remove artifacts. Subsequently, as shown in Figure 3a, the segments are divided into four different species, namely, closed porosity (blue), interconnected porosity (green), alumina precipitates (yellow), and cast porosity (red). The 2D porosity (pore area over total area) is averaged over four adjacent virtual slices perpendicular to the long axis of the tomography tip (z-direction) and is used to calculate the 3D porosity (given in percentage). The porosity as a function of distance from the unoxidized single-crystal superalloy is shown in Figure 3b. Apart from the result of the SEM/EDXS-informed nano-CT reconstruction (black curve), the result of the preliminary analysis based on nano-CT alone is shown for comparison. Here, the alumina particles in the internal precipitation region d₁ were falsely interpreted as pores due to the almost identical contrast in the PC images (cf. Figure 6b).

In the following, we discuss and interpret the different features of the oxide scale. Starting from the top of the reconstructed volume in Figure 3a, we see a maximum porosity of 20.7% at a distance of 8.8 μm from the unoxidized superalloy (cf. Figure 3b). This extremely high value can be almost exclusively attributed to the three large pores in between the cobalt oxide grains and a highly porous spatially confined layer directly beneath the outer oxide layer d₂. These pores are interconnected and are located directly below grain boundaries in the outer oxide layer. Formation of microchannels that allows the access of gaseous oxygen to the inner oxide layer d₂ was repeatedly demonstrated for pure Co and Ni.[45,46] The insufficient resistance against internal oxidation of γ’-strengthened Co-base model alloy at 900 °C was reported in a recent publication.[47] Therefore, high oxygen levels can be assumed at the former sample surface of ERBOCo-1 during isothermal oxidation. Because of insufficient statistics due to the limited sample volume, the maximum porosity of about 20% strongly depends on the number of large pores contained within the pillar. The inner oxide layer d₁ exhibits two distinct peaks at positions 7.5 and 4.7 μm with porosities of 12.2% and 6.5%, respectively. Since the pores in the inner oxide layer are not connected, penetration of O₂ through d₁ can be ruled out. Nevertheless, the formation of voids that ultimately combines to pores is a consequence of extensive Co and Ni transport to the scale/gas interface. In between the two maxima, the porosity decreases to 0.6%. This experimental finding indicates another change in the oxidation mechanism. The diffusion of metal cations that is opposed to the transport of oxygen might originate from different regions of the sample. At this stage, the underlying processes cannot be explained completely by the elucidation of only one sample. The first nano-CT results suggest a local porosity maximum at a distance of 2.5 μm of roughly 15%. The falsity of this value is caused by the almost identical contrast of porosity and alumina in the PC images. The porosity plot for the SEM/EDXS-informed segmentation allows attributing this peak to the alumina precipitates and the porosity value was corrected to zero in this region. Other EDXS results (not presented here) suggest that in this region, the formation of mixed oxides (e.g., spinels) and chromia might have led to a volume expansion and closing-up of the porosity. The formation of spinels in inner oxide layers of comparable alloys was also experimentally observed and predicted by simulations.[2,3] FIB tomography was utilized to elucidate the pore distribution in the Cr-containing spinel layer of an Fe–Cr alloy after exposure to steam at 650 °C.[48] The publication demonstrated a higher degree of interconnectivity compared with the present study. When comparing the standard nano-CT analysis with the SEM/EDXS-informed nano-CT reconstruction, it is noteworthy that the calibration down scales the porosity for all peaks consistently.

Based on our correlative SEM/EDXS analysis, we were able to discriminate between pores and alumina particles in the virtual slices of the nano-CT reconstruction, ultimately allowing a 3D reconstruction of the internal precipitation region d₁. Figure 6c shows a histogram of the maximum Feret diameters of the precipitates[49] measured from two representative slices of this region (see Figure S1.1, Supporting Information). The precipitates are elongated perpendicular to the oxidized surface (i.e., along [001] direction). Most precipitates show a larger...
lateral diameter toward the unoxidized superalloy, which could hint at the direction of material transport during formation. The predominant growth direction being the z-direction however hinders a coalescence of the precipitates and thereby the formation of a continuous alumina passivation layer. Within the unoxidized superalloy at the bottom of the sample, we can identify a last small porosity peak with a porosity of 2.7% at a distance of 1.8 μm from the unoxidized superalloy, which is caused by small casting porosity in the bulk material. These pores are mostly round in shape and likely stem from the casting porosity in the bulk material.

3. Discussion

The multinary Co-base superalloy ERBOCo-1 forms a multilayered oxide scale with spatially separated porous layers and an alumina precipitate layer when subjected to synthetic air at 900 °C for 12 h. Two pore layers form within the oxide scale separated by a dense region likely containing small mixed metal oxides and chromia. Above a γ’-depleted zone, alumina precipitates form but do not coalesce into a continuous passivation layer. The alumina precipitates are elongated along the growth direction of the oxide scale. For the characterization of the oxide scale, we established a correlative tomography approach combining laboratory high-resolution nano-CT, FIB/SEM tomography, and EDXS, which allows the structural and chemical characterization of complex 3D microstructures. Combining information from such complementary methods is key to verify and corroborate gathered data. In our approach, we used high-resolution morphological information and material contrast from SEM images and chemical information from EDXs to facilitate the segmentation of tomograms derived from nano-CT. In contrast to conventional FIB tomography, nano-CT is applicable to nonconductive and highly porous materials. Moreover, the non-destructive nature of nano-CT enables subsequent complementary measurements on the same sample volume or even in situ studies, as long as segmentation calibration by FIB/SEM can be applied post-mortem or on another representative sample volume.

The presented approach demonstrated its suitability to reveal mechanistic insights into distinct processes that are crucial for a more complete understanding of oxidation behavior at high temperatures. Especially, the successful design of new materials that can operate in harsh environments of, for example, a gas turbine can benefit from such insights. These alloys rely on the formation of protective surface oxides to ensure the required resistance against corrosion. Still today, the formation of continuous and protective Cr₂O₃ and Al₂O₃ layers in Co- and Ni-base superalloys is not fully understood. The formation of voids below externally grown chromia scales unambiguously reduces the stability of the layer and ultimately results in scale spallation. Furthermore, voids or cavities in the internal oxidation zone might directly influence the morphology of alumina precipitates and therefore also affect the growth of protective Al₂O₃ layers. For example, controversies concerning the oxidation behavior of diluted Ni-Al systems at 800 °C were explained by the growth of cavities below the original alloy surface during the very early stages of scale growth. The presented combination of reduced FIB tomography with nano-CT can be used to characterize these cavities and simultaneously visualize the morphology of alumina precipitates in the internal oxidation zone. For short-term experiments and consequently thin oxide scales, no conventional metallographic preparation is needed. Therefore, it is excluded that voids or cavities combine to cracks due to the applied mechanical stress.

Major challenges are expected in cases where the microstructure of interest exhibits higher complexity comprising, e.g., interpenetrated areas of light precipitates and pores. Here, the limitation of nano-CT regarding the discrimination of such features (from image contrast alone) comes into play and requires, in principle, a complete FIB tomography for reliable segmentation. However, in many cases, different types of precipitates and pores appear with different characteristic morphologies regarding size, aspect ratio, faceting, etc. which can be exploited for segmentation of nano-CT data. A promising way to make use of such characteristics is machine learning which has recently been shown to be a powerful tool for segmentation of X-ray tomography data. Applying machine-learning or deep-learning techniques to segment PC data sets requires a better understanding of PC formation. This will be addressed in future research by systematic studies of model structures and comparable data sets. To ensure precise data output of said techniques, it is crucial to supply multiple data sets for a given material system. The correlative nano-CT and FIB tomography approach presented in this work is expected to be highly useful in such procedures by providing reliable data for model training. Machine learning constitutes only one procedure toward an automated segmentation which will be a crucial step in the process when striving for quantitative results and higher throughput. Therefore, various not yet understood transport processes in the field of high-temperature oxidation and corrosion can be elegantly addressed by utilizing the presented combination of techniques.

4. Conclusion

In summary, the present work demonstrates that the utilization of nano-CT for 3D information, SEM for segmentation calibration, and EDXs for phase identification is a promising characterization routine suitable for a wide range of material systems. Depending on the underlying research question, nano-CT enables the usage of PC and/or AC imaging. As demonstrated for the example of an oxide scale in this work, SEM/EDXs-informed image segmentation of nano-CT data yields reliable quantitative 3D data. The most important advantage of nano-CT compared with conventional FIB tomography is its nondestructive nature. This allows interrupted Nano-CT analysis combined with, e.g., ex situ heating, oxidation, or mechanical testing. We anticipate that segmentation calibration by FIB/SEM can often be performed on a sample volume comparable (e.g., adjacent) with the volume used for in situ nano-CT studies. Only performing selected FIB cuts within the pillar after having finished nano-CT analysis or carrying out the segmentation calibration on a comparable sample volume moreover offers the possibility of site-specific extraction of FIB lamellae or pillars for subsequent transmission electron microscopy (TEM), electron tomography (ET), or atom probe tomography (APT) studies. Such techniques provide information at even higher resolution enabling, e.g., phase identification of tiny precipitates and in-depth characterization of interfaces and defects.
5. Experimental Section

The alloy investigated in this study is a multinary single-crystalline Co-base superalloy with a nominal composition of Co32Ni8Al6Cr5Si2W. 5Ti0.5Ta, hereafter referred to as ERBOCo-1. This material was developed within the Collaborative Research Centre SFB/TR-103 “From Atoms to Turbine Blades—A Scientific Approach for Developing the Next Generation of Single Crystal Superalloys” of the German Research Foundation (DFG) and was casted as a single-crystalline plate (8 mm × 53 mm × 71 mm) in a standard Bridgman casting procedure. The single crystal was subjected to a three-step heat and aging treatment (1280 °C/8 h, 1050 °C/5 h, 900 °C/16 h) to tailor the γ/γ′ microstructure. For more details on the investigated alloy, the reader is referred to further literature.[21,14] For the oxidation study presented in this work, a 10 mm × 10 mm × 1 mm sample with [001] surface normal was cut, mechanically ground, and polished down to a surface finish of 1 μm. Subsequently, the sample was oxidized in a Setaram Evolution 1650 thermogravimetric analyzer with a constant gas flow of 20 cm³ min⁻¹ of synthetic air (80 vol% N₂, 20 vol% O₂) at 900 °C for 12 h. Further details on the oxidation experiment can be found in studies by Weiser et al.[57,58] After oxidation, the surface was coated with a protective Ni layer via electrochemical deposition to preserve the oxide scales during metallographic preparation. To obtain cross sections for SEM imaging of the oxide scale, the sample was cut and ion polished using a Hitachi IM4000 X-ray nano-CT was performed using a ZEISS Xradia 810 Ultra equipped with a 5.4 keV rotating anode Cr source and a Zernike phase ring for PC imaging. Two magnification modes can be selected, either large field of view (LFOV) or high-resolution (HR) mode, allowing imaging of 64 × 64 μm and 16 × 16 μm large areas with optical resolutions down to 150 nm (pixel size of 64 nm) and 50 nm (pixel size of 16 nm), respectively. The diameter of the pillar sample (~15 μm) studied in this work was chosen to fit into the FOV of the HR mode. For the data sets, a tilt series with a total number of 901 transmission images was recorded with an acquisition time of 240 s/frame for PC and 120 s/frame for AC imaging. The image series was aligned along the rotational axis and the complete 2D data set was modified to fit the reconstruction geometry. After processing, the reconstructed 3D pillar exactly matched the prepared sample volume. Acquisition and alignment were done in the native ZEISS microscope software (XMController and Scout&Scan). 3D reconstruction was performed employing the Simultaneous Iterative Reconstruction Technique (SIRT).[55] A combination of phases was used for visualization, segmentation, and quantitative 3D analysis. SEM imaging, FIB/SEM tomography, and sample preparation for X-ray microscopy (XRM) were performed using an FEI Helios NanoLab 660 dual-beam FIB/SEM system.

The data processing and analysis were realized by a combination of in-house Java and Python coding together with the commercial software package Arivis Vision4D. The data set was modified (rotation and tilting) by in-house Java scripting to achieve comparable 3D volume data sets. The porosity calculation was performed by setting the threshold to a specific SEM-informed value combined with a size-filtering process (not considering volumes smaller than 0.065 μm³) to suppress noise artifacts in Arivis Vision4D and improve the segmentation of real pores in the 3D volume. A Python-based script calculates the porosity value (area fraction of the segmented pore area in 2D) for each slice of the volume data set with its corresponding position (distance to the unoxidized superalloy) in the data set.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

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