Correlative Tomography – Combining X-ray Nanotomography and FIB/SEM Serial Sectioning to analyze Al-Si cast alloys

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Abstract

Aluminium alloys with silicon as their major alloying element are widely used in die or sand casting of machine or vehicle parts. The microstructure of hypereutectic Al-Si alloys (>12% Si) consist of primary silicon particles and the Al-Si eutectic. Elements such as Cu, Mg and Ni are added in order to form a great variety of different intermetallic phases which improve mechanical properties at elevated temperatures. Size, shape and connectivity of these complexly formed intermetallic phases play a decisive role for the properties and are for that reason of great interest. As the complex shapes and connectivity are not accessible by means of classical 2D techniques, 3D analysis of the microstructure is mandatory. Due to the size of the particles in the µm-range, only high resolution 3D imaging techniques are suitable. In this work we used 3D X-ray Microscopy as well as FIB/SEM Serial Sectioning to image the microstructure of an AlSi12Cu4Ni2Mg cast alloy. 3D images were taken at the very same sample volume in order to compare information gained from the different techniques.

Keywords: correlative tomography, X-ray Microscopy, X-ray Nanotomography, FIB/SEM Serial Sectioning, AlSi cast alloys

1 Sample preparation for X-ray Nanotomography

An Aluminium-Silicon die-cast alloy was used for the correlative tomography study. The chemical composition is summarized in Table 1. For high resolution 3D X-ray Microscopy, a cylindrical shaped sample with a diameter of less than 100µm is preferred. From the bulk sample, a matchstick shaped piece with a circular cross section of approx. 1.5 mm and a length of 2 cm was cut by means of wire EDM (electrical discharge machining). The top of the cylindrical piece was turned on a lathe machine to a conical shape. The sample was then mounted in a sample holder for the X-ray Microscope. As the needle tip was not yet cylindrical, the sample was further shaped using a dual beam FIB/SEM (FEI Helios 600 Nanolab). An annular milling pattern with an inner diameter of 60µm and the highest possible milling current of 21 nA was used in an unattended overnight-session. The final shape was then cut with an annular milling pattern with an inner diameter of 50 µm. In a second step, the top of the cylindrical part of the sample was flattened with a cleaning cross section milling pattern from the side. Figure 1 shows images of the different steps of FIB sample preparation and the final sample shape.

Table 1: chemical composition of the alloy AlSi12Cu4Ni2Mg

<table>
<thead>
<tr>
<th>Element</th>
<th>Si</th>
<th>Cu</th>
<th>Mg</th>
<th>Ni</th>
<th>Fe</th>
<th>Mn</th>
<th>Al</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wt.%</td>
<td>11.0–13.0</td>
<td>3.0–4.0</td>
<td>0.5–1.2</td>
<td>1.0–3.0</td>
<td>&lt;0.7</td>
<td>&lt;0.3</td>
<td>balance</td>
</tr>
</tbody>
</table>

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X-ray Microscopy and Nanotomography

X-ray Microscopy as a new emerging field in high resolution computed tomography has the potential to solve numerous problems in materials science. In most cases, the high magnification in X-ray Microscopy is reached by magnifying X-ray optics such as Fresnel zone plates [1-3]. We follow a different approach using only the geometric magnification due to a small source-sample distance and a very small X-ray source [4,5]. For that a 30 kV electron beam in a scanning electron microscope (SEM) is focused on a small tungsten needle target with a tip diameter of less than 100 nm and creates a geometrically limited X-ray spot, achieving a spatial resolution below 100 nm. The sample is positioned in front of this X-ray source and can thus be rotated by 360° for CT scanning. The attenuated X-ray beam is detected by a photon counting, direct converting pixel detector via a thin beryllium window outside the vacuum chamber. Due to the detector pixel size of 55 µm, the fixed focus-detector distance and the actual focus-object distance in the range of 1 mm, a variable voxel-size between 50 nm and 500 nm is accessible. With an integrated energy dispersive X-ray spectroscopy (EDS) detector, chemical information from the sample surface can be recorded and correlated with the reconstruction from X-ray Nanotomography (see Figure 2).

To investigate the prepared specimen the XRM-II nanoCT from ProCon X-Ray was used. For nanotomography mode, the virtual rotation axis needs to be set in the center of the cylindrical specimen to allow eucentric rotation. The 30 kV electron beam was set to an electron current of 300 nA and focused on the target tip. Due to the size of the flat panel detector of 1280 x 514 pixels, a magnification of x365 was chosen, resulting in a voxel-size of 150 nm to record a similar volume like in FIB/SEM Serial Sectioning. Like shown in the X-ray projection in Figure 3, a sample height of about 70 µm was imaged. The size of the detector would even allow to measure a three times wider sample for this specific magnification in the same time. The number of projections was set to 400, which leads to high image quality in 3D volume reconstruction, as shown in previous studies [5]. For image acquisition, a total exposure time of two minutes per projection was chosen. The overall CT process including image
acquisition, specimen rotation and refocusing of the e-beam on the X-ray target, due to e-beam drift, is fully automated and took about 15.5 h.

The XRM-II nanoCT system uses a custom-designed software (see also [5]) to generate a 3D volume based on the measured projections. This software takes the geometry and measurement setup into account, corrects mechanical instabilities, which can occur during data acquisition, and applies a 3D phase-contrast filter. The software uses a total-variation (TV) [6] regularized Simultaneous Algebraic Reconstruction Technique (SART) [7] as reconstruction algorithm. This algorithm is necessary due to the critical signal-to-noise ratio (SNR) caused by a small focal spot and low photon flux. During the reconstruction process, geometric deviations by the mechanical instabilities and thermal drift are corrected. This correction is based on image-correlation and compares the forward-projected volume image with the measured data (see also [8,9]). After the reconstruction process, the image contrast can be improved by applying a Paganin-based phase contrast [10] and deconvolution filter.

3  FIB/SEM Serial Sectioning

Scanning Electron Microscopy (SEM) is a widely used microscopy technique with applications in fields such as material science, life science, and semiconductor research. The specimen is scanned by a focused electron beam and back scattered electrons or secondary electrons are detected with the respective detectors. SEM techniques can be extended to three-dimensional (3D) imaging using a serial sectioning technique. In focused ion beam scanning electron microscopy (FIB/SEM), a specimen is imaged by sequentially removing thin layers of material using a focused ion beam. Each resulting surface is then scanned using a focused electron beam such that 3D information is acquired layer-by-layer (see Figure 4).

The versatile contrast mechanisms in electron microscopy lead to the enormous capability of this method. Standard tomography is done with the help of secondary or backscattered electron imaging and the resolution ranges down to a few nanometers whereas volumes up to 50 µm edge length are still feasible. The integration of Electron Backscatter Diffraction (EBSD) into the serial sectioning process [11] even allows for the detection of different phases and their crystallographic orientation and using X-ray spectroscopy (EDS-signal) allows for a chemical analysis of the sample, as demonstrated for Al-Si alloys [12,13]. Recent developments widened the range of analyzed volumes by using Xe-Plasma FIB [14] or in situ Femto-Laser ablation [15] with higher ablation rates and thus bigger analyzed volumes.
3.1 Sample Preparation

For the FIB Serial Sectioning, the already in X-ray Nanotomography measured sample had to be further processed. For the correct position of the milling pattern for each slice and to compensate for sample drift, a fiducial mark has to be positioned outside the analysed volume. The used software for automated serial sectioning (FEI auto slice&view G2) places this mark either on the left- or righthand side behind the analysed volume. As the sample is freestanding and the whole volume of the cylindrical part should be analysed, there is no sample, where the fiducial mark has to be positioned. To compensate for that, we followed an experimental route compared to FIB-based in-situ liftout TEM-sample preparation. A thin slab of a silicon wafer was cut out and transferred with an Omniprobe micromanipulator to the sample. Then, the sample was positioned on a pre-tilt holder to orient the sample axis perpendicular to the ion beam. The side of the cylindrical part of the sample facing the ion beam was covered with a 1µm platinum layer in order to protect the surface and avoid curtaining in the following serial sectioning process. Figure 5 shows the sample prepared for FIB/SEM Serial Sectioning.

![Figure 5: prepared sample for serial sectioning. Left: Overview (electron-image), Middle: example for an ion image in the serial sectioning process. The ROI is covered with a thin platinum-layer and a fiducial mark was placed at the upper left corner on the transferred silicon slab. Right: example of an electron image in the serial sectioning process.](image)

For serial sectioning, an ion-current of 21 nA and a slicing distance of 62.5 nm were chosen. 1000 serial images with a pixel size of 62.5 nm x 62.5 nm were recorded. The tilt of 38° between electron beam and normal of the cross section (results from the angle of 52° between electron- and ion-column) was compensated during imaging with a narrower line scanning and thus the final voxel-size is isotropic with 62.5 nm in all directions. For imaging, an electron voltage of 5 kV and an electron current of 1.4 nA were chosen. Image resolution is 1024 x 884 pixel and dwell time (time per pixel) is 10 µs. Time per slice (cutting and imaging) was approx. 60 s and the whole process took 17.5 h in an unattended overnight session.

Reconstruction of the sample was done using Amira 5.3 software. The images were aligned using the least-square method. For segmentation the inner volume of the sample with a diameter of approx. 45 µm was extracted and the image was filtered using a mean filter. Segmentation was done using thresholding for the different phases with some manual corrections. The segmented image was smoothed in xy, yz and xz planes.

4 Comparison of results

4.1 Artefacts

Due to the different mode of data acquisition, both datasets suffer from different artefacts in the reconstruction. The sample diameter of 50 µm is quite big for a serial sectioning tomography using a Ga-FIB, so the ion currents have to be relatively high (21 nA in our case). At higher currents the so called curtaining effects [18] occurs, where irregularities at the surface or at the cross section continue downwards and produce grooves. These grooves interfere with segmentation and may lead to errors and the need for manual corrections. Curtaining may be further reduced by cutting under different angles (rocking polish), which was not implemented in the software for automated serial sectioning. Reduction of curtaining effects by image processing is also possible [19] but sometimes also leads to changes in the real structures in the image. As curtaining was not severe in the experiment, the negative effects during segmentation were corrected manually. In the X-ray-CT image streak artifacts and beam hardening artefacts are visible, but not severe. Due to imperfections in the rotational axis of the stage, the sample shifts during the measurement, which leads to slightly different magnifications during rotation and blurring in the 3D volume. Using a high number of projections for reconstruction blurring gets more pronounced but signal-to-noise ratio improves.
4.2 Phases

In the reconstruction it is differentiated between the Al-matrix, silicon, intermetallic phases and cracks/pores. In the AlSi12Cu4Ni2Mg alloy, many intermetallic phases such as Al7Cu4Ni, Al2Cu, Al15Cu4Mg8Si7 and Al15Si2(FeMn)3 [20] may form and it is impossible to unambiguously differentiate between them neither in FIB/SEM Serial Sectioning (without EBSD or EDS mapping) nor in X-ray Microscopy with absorption contrast. Figure 6 shows a reconstructed slice from X-ray Nanotomography (denoted as “XRM image” in the following) and one imaged slice from FIB/SEM Serial Sectioning (denoted as “SEM image” in the following) from the same region of the sample. In the SEM image we can differentiate between the Al-matrix, silicon (dark) and different intermetallic phases. As the exact chemical composition of the phases cannot be determined from the SEM image or the XRM image, the different phases are named IP1 – IP5 (from lightest to darkest in SEM image). The grey-level of different phases in the XRM image is not uniform and the boundaries are not clearly visible. One has to keep in mind, that the information depth in the SEM image is in the range of a few nanometers and the XRM image shows an integration over the whole slice thickness of 150nm and thus a sharp interface will be more blurred in the XRM image than in the SEM image. All intermetallic phases seen in the SEM image can be differentiated in the XRM image as well. IP5, which has in the SEM image a very similar grey-level than the aluminium matrix is markedly darker than the matrix in the XRM image. The IP5 particle is surrounded by another intermetallic phase and thus visible in the SEM image. The silicon particles are not visible at all in the XRM image due to the very similar attenuation coefficient to the matrix.

Figure 6: Comparison of an imaged slice from FIB/SEM Serial Sectioning (left) and a reconstructed slice of X-ray Nanotomography (right). Intermetallic phases can be differentiated in both datasets whereas silicon is only visible in the SEM image.

Figure 7: Comparison of an imaged slice from FIB/SEM Serial Sectioning (left) and a reconstructed slice of X-ray Nanotomography (right). Whereas IP6 shows good contrast in the SEM image and poor contrast in the XRM image, IP7 shows good contrast in the XRM image but poor contrast in the SEM image.
In Figure 7 it is obvious, that the contrast situation in the SEM image and the XRM image is not always comparable. The complexely formed intermatllic phase IP6 in the center of the image (probably the “script phase” Al$_{15}$Si$_2$(FeMn)$_3$ [21]) has good contrast in the SEM image but is only indistinctly visible in the XRM image. On the other hand, a part of the intermetallic particle in the lower part of the image IP7 shows only indistinct contrast in the SEM image but good contrast in the XRM image.

4.3 Cracks

Inside the sample some cracks within the silicon phase were found (Figure 8). As the secondary electron signal used in SEM imaging is always high at sample edges, these cracks have a pronounced white border and a dark inner part. Thus cracks and pores are difficult to binarize using simple thresholding and a manual segmentation has to be performed. In the XRM image cracks show good contrast due to their high differences in attenuation coefficient. However the surrounding silicon phase is not visable and the cracks appear to be located in the aluminium matrix.

4.4 3D Reconstruction

Due to better resolution and contrast, only the volume measured with FIB/SEM Serial Sectioning was reconstructed as described in chapter 2. The silicon phase is shown in green. Different interpetallic phases are shown in different colours but as the chemical information is missing, a clear allocation is not possible. The aluminium matrix is not shown for reasons of better visibility. A volume rendering view of the XRM dataset is shown for comparison (Figure 9). Volume rendering of the FIB/SEM dataset does not provide a good overview due to the light background in the images.
5 Summary and Outlook

Correlative tomography measurements from the very same sample were done using FIB/SEM Serial Sectioning and X-ray Nanotomography. Whereas the silicon phase shows good contrast in the SEM images, the absorption contrast in the XRM dataset is not sufficient to differentiate between aluminum matrix \((Z=13)\) and silicon \((Z=14)\). Contrast situation in the SEM image and the XRM image is not always comparable, as some intermetallic phases show different contrasts in both measurements. Heavy intermetallic phases and cracks can be distinguished in both datasets. SEM images show more details of the microstructure and are easier to segmentate. Nevertheless serial sectioning is a destructive technique and thus X-ray Nanotomography is essential for in-situ observations. As both experimental setups allow for additional EDS measurements, further studies combining serial sectioning with EDS mappings and a correlation of EDS Mapping with X-ray Nanotomography should help to separate different intermetallic phases in the reconstructed volume.

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