3D characterization by tomography of beta Al$_9$Fe$_2$Si$_2$ phase precipitation in an Al6.5Si1Fe alloy

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Abstract. The microstructure evolution of beta phase during solidification of a synthetic Al6.5Si1Fe (wt.%) alloy has been investigated by in-situ synchrotron micro-tomography and post-mortem tomography. In-situ solidification was observed at a constant cooling rate of 10°C min$^{-1}$, from above the alloy’s liquidus with the melt at 618°C down to 575°C which is just above the (Al)-Si-beta invariant eutectic reaction. Primary (Al) dendrites nucleated at 608°C, followed by the formation of beta-Al$_9$Fe$_2$Si$_2$ phase starting at 593°C. After a rapid growth stage until 587°C as thin plates, beta phase continued to grow at a paced rate. Thickening of the plates was also evaluated and it was observed that the decrease in the lateral growth rate of the plates did not lead to an increase of their thickening rate. It was noted that the interconnectivity between beta precipitates increased as the solidification progressed. While nucleation of beta phase has previously been reported to occur on the alumina scale formed at the outer surface of the material, it is shown from post mortem tomography that bulk nucleation can occur as well.

1. Introduction
Tomography in material science is used nowadays as a non-destructive method to visualize internal structure of objects and acquire information on their three dimensional geometries and properties [1, 2]. Within the past 10 years, the development of in-situ tomography for the study of solidification phenomena has increased significantly. This is due to the development of fast cameras to record the images which facilitate the observation of in situ solidification process [3]. The use of in-situ method with X-ray can be seen in the work by Mathiesen and co-workers [4, 5] who studied dendritic growth and the dynamics of columnar to equiaxed transition. Kim et al. [6] also studied the effect of iron on an Al-Si-Cu alloy with in-situ radiography. Furthermore, the development of in-situ experiments was achieved by allowing 3D imaging by rotating the sample during in situ solidification [7, 8].

The present work considers the solidification of a high purity Al6.5Si1Fe (in mass.%) alloy, and more particularly the precipitation of beta-Al$_9$Fe$_2$Si$_2$ phase that occurs after primary (Al) and before the invariant (Al)-Si-beta eutectic. Early studies on beta phase precipitation and growth morphology using in situ X-ray tomography and in situ synchrotron can be seen in the works by Wang et al. [9], Terzi et al. [10] and Puncreobutr et al. [11] which were all performed on Al-Si-Cu-Fe alloys. Observation by Wang et al. [9] using in situ synchrotron radiography suggested several stages in beta phase formation in a Al17.5Si3.5Cu0.8Fe alloy. They observed a rapid growth in early stage which was...
later constrained by primary aluminum dendrites, followed by attachment and diffusion growth where the plate thickened. Terzi et al. [10] in a Al8Si4Cu0.8Fe alloy showed the beta phase nucleated at or near the surface or the outer skin surface after the primary aluminum nucleation, where the outer surface was most probably \( \gamma - \text{Al}_2\text{O}_3 \). Recent work by Puncreobutr et al. [11] quantified the nucleation of intermetallic as a function of undercooling in Al7.5Si3.5Cu0.6Fe alloy. In this case also, the plates displayed a very rapid lateral growth and slow thickening. They also found that some eutectic silicon precipitates attached to a beta plate or outer oxide when they first appear. Further review regarding the beta phase precipitation can be seen in the work by Ferdian et al. [12]. The objective of this study is to characterize the morphology and the growth of beta phase in high purity Al-Fe-Si ternary system using in-situ as well as post-mortem tomography.

2. Experimental method

Two types of tomography experiments, namely post mortem and in-situ, were conducted for studying the growth of beta phase. The in-situ synchrotron tomography was conducted at European Synchrotron Radiation Facility (ESRF) ID-19, with the holding furnace developed by SIMAP, Grenoble. The Al-6.5Si-1Fe sample was prepared as a cylinder with 1 mm in diameter and 3 mm in height. At the bottom, a 0.55 mm hole was drilled for inserting a thermocouple. A pink beam of 17.6 KeV with 700 projections was used with a scan time of 0.75 sec resulting in a total of 36 tomography data images (1.1 \( \mu \text{m.voxel}^{-1} \)). The experiment was recorded at a constant cooling rate of 10°C min\(^{-1}\) from the fully melt condition at 618°C to 575°C prior to the (Al)-Si-beta eutectic reaction.

Post–mortem tomography has been applied to differential thermal analysis (DTA) samples cooled at various cooling rates, in the range 0.2 to 5°C min\(^{-1}\), DTA samples were 3.9 mm in diameter and 5mm in height. Post mortem tomography was carried out by using GE Phoenix Nanotom equipped with a 180 kV/15W X-ray tube. A monochromatic beam was transmitted and produced 1440 images as the specimen was rotated by 360° (2.5 \( \mu \text{m.voxel}^{-1} \)). An image stack of volume was constructed using Datos X (GE Sensing and Inspection GmbH) and VG Studio Max (Volume Graphic GmbH, Germany).

The tomography data was then reconstructed and post-treated with image analysis by extracting the beta precipitates through series of operation using imageJ (National Institute of Health, USA) and Aphelion (ADCIS, France). Due to the similar contrast of beta phase and the oxide skin, an additional masking operation consisting of Gaussian-blur filtering, thresholding, iteration of erosion and dilation [7], was added to remove the outer oxide layer.

3. Result and discussion

Analysis to the reconstructed volume of in-situ experiment showed the precipitation of primary (Al) dendrites occurred at 608°C, while nucleation of beta phase started at 593°C in several locations as the one marked by a circle in figure 1. The beta phase grew in interdendritic regions along with the continuing growth and coarsening of the primary (Al) dendrites. This precipitation of beta phase in the alloy agrees with DTA experiments and calculations according to scheil model or lever rule [13]. Further analysis on the nucleation of beta phase was performed by defining a region of interest of 1008x1008x500 voxels taken from the middle region. The 3D volume reconstructed rendering shows the nucleation of beta phase with each separate particle coloured differently, as illustrated in figure 2. The first observed nucleation of beta phase appeared in five sites located on the outer oxide skin; however due to oxide skin removal operation and weak phase contrast, some of the initiation sites may have been wiped off during image analysis.

As the solidification progressed, a rapid increase in the number of beta precipitates was detected and this continued until saturation. Observation showed the rapid growth of beta phase and notice was made that no new nucleation occurred below 587°C. Analysis upon the region of interest indicated that approximately 32 nucleation events of beta particles in total were observed during the solidification process.
Figure 1. Image slices extracted horizontally from the middle of the reconstructed volume obtained during in situ solidification of Al6.5Si1Fe.

Figure 2. 3D visualization of beta phase nucleation where each colour represents single beta particle. Note that some particles impinged and connected in a later stage.

The tomography images showed that sometimes dendrite arms of primary (Al) blocked the growth path of the beta precipitates. However, in such a case, it could be seen that the beta phase growth
continued by surrounding the dendrite and some of the precipitates reconnected after passing the obstacle. During beta growth some branching and bending were also observed. Branching was noticed in an intermediate stage growth and the interconnectivity was increased in a later stage as the temperature dropped and the connection between precipitates created clusters of large beta phase.

A series of growth steps of a single beta phase is shown in figure 3. Examination showed a robust growth of beta phase occurred in an early growth stage, where the phase length could be doubled in one tomography step. As well known, the beta phase growth is characterized by a high lateral growth rate that leads to the development of elongated 2D plates. The growth began to slow down as the temperature reached 585°C and this was accompanied by the increase of interconnectivity of beta precipitates.

Figure 3. A series of reconstructed 3D images from a single nucleated beta precipitate taken at several steps during growth. As the solidification continues other precipitates got connected to the selected one (voxel size 1.1 µm).

Lateral length measurements were performed on several beta precipitates at each tomograph step. From these length measurements, five beta phase particles which are labelled 1 to 5 were selected for further analysis of their growth kinetics. In general, we could classify the beta phase growth into three stages based on the length curve shape as: initial stage (I), peak stage (II) and decline or saturation stage (III) as seen in figure 4a. Individual analysis of beta phase particles showed that the lateral growth rate varied among them and also between each step. The highest detected rate in stage I was at 34.5 µm.s⁻¹ and then fluctuated in stage II within the range of 5 - 25 µm.s⁻¹. The variation between steps from a single phase could be caused by the blockage on (Al) dendrites and other obstacles in the interdendritic region that slowed down the growth rate as already mentioned.

Analysis of the thickness of beta precipitates showed it increased during solidification especially in the region close to the surface where the nucleation initiated. Thickness rate was measured from the highest plate thickness from each tomography step. The results showed that the beta precipitates thickened at a rate of 0.76 µm.s⁻¹ during the first stage and this slowed down to 0.21 µm.s⁻¹ during the next stage. The thickening rate in the later stage further decreased to 0.1 µm.s⁻¹.

The thickening mechanism was largely overtaken by the high lateral beta growth which is 20 times faster. As seen from figure 4b which compares both rates, the lateral growth rate was dominant in the early stage, marked with very high values and slowed down as the solidification progressed. However,
the thickening rate did not increase during the later stage, but rather seemed to decrease along with decreasing lateral growth rate.

![Figure 4](image.png)

**Figure 4.** Evolution with time of the maximum length of beta precipitates (a), comparison of lateral and thickening growth rates of beta precipitates (b).

The volume fraction of beta phase taken from in-situ tomography data increases rapidly in the beginning and saturates as the solidification continues as seen in figure 5a. The final value at the end of the experiment, at a temperature just above the ternary (Al)-Si-beta eutectic, agrees with the prediction made following the Scheil’s model using Thermocalc and the TCAL2 database [13]. Comparison of the experimental and predicted evolution shows that the beta phase appeared with an undercooling of 10°C. Such a high undercooling explains the large growth rate experienced by the beta precipitates at the beginning of their growth.

![Figure 5](image.png)

**Figure 5.** Measured volume fraction of beta phase compared with predicted evolution carried out with Thermocalc and the TCAL2 database.

Analysis to the post-mortem tomographs showed the nucleation sites for the beta phase are mostly located again on the outer oxide surface (skin). The growth followed by the beta precipitates expanding in radial direction toward the sample centre, as observed in sample cooled at 0.2°C.min⁻¹ in figure 6. However, observation at low cooling rate also showed small fraction of beta precipitates not associated with the large plates that nucleated on the outer skin. At a higher cooling rate, the initiation of beta precipitates seemed dispersed within the sample, between the (Al) dendrites. Unfortunately, the
image quality generated by tomography is inadequate for detailed analysis, as clearly seen at a higher cooling rate where very thin plates could hardly be detected due to a too low spatial resolution.

Figure 6. Rendering of post-mortem tomograph with various cooling rates (voxel size: 2.5 µm)

4. Conclusion
In situ tomography experiment showed that beta phase growth is dominated by its lateral extension. Initial growth showed a high growth rate which slowed down after saturation level was reached. This evolution could be related with the high undercooling necessary for beta phase nucleation. The slowing down of the lateral growth did not cause the thickening rate to increase. Instead, it appears that the thickening rate also decreased along with lateral growth rate decreasing. Observation on samples solidified at low cooling rates by post-mortem tomography seems showing nucleation of beta precipitates occurs both on the outer oxide skin of the samples and in the bulk of the material.

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