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3D characterisation of the Fe-rich intermetallic phases in recycled Al alloys by synchrotron X-ray microtomography and skeletonisation

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ABSTRACT

Synchrotron X-ray microtomography and skeletonisation method were used to study the true 3D network structures and morphologies of the Fe-rich intermetallic phases in recycled Al-5.0%Cu-0.6%Mn alloys with 0.5% and 1.0% Fe. It was found that, the Fe-phases in the 1.0%Fe alloy have node lengths of 5–25 μm; while those in the 0.5%Fe alloy are of 3–17 μm. The Fe-phases in the 1.0%Fe alloy also developed sharper mean curvature with wider distribution than those in the 0.5%Fe alloy. Combining SEM studies of the deeply-etched samples, the true 3D structures of 4 different type Fe phases in both alloys are also revealed and demonstrated.

Aluminium (Al) alloys are widely used in the transportation, building and packaging industry because of their lightweight, high specific strength, high corrosion resistance, and excellent recyclability [1]. In modern vehicles, Al alloys are playing increasingly important roles in reducing the weight of vehicles and, hence, fuel consumption and CO_2 emissions in transportation [2]. Approximately 90% of the Al alloys used in land vehicles are from recycled sources for cost reduction and sustainability [3]. In Al alloys, especially recycled Al alloys where Fe concentration is often higher than 0.5% (weight percentage), Fe is the most common impurity element, and it can be easily picked up in sorting and remelting processes during Al recycling [4]. Normally, when the Fe in an Al alloy is >0.05% [4], brittle Fe-rich intermetallic phases (named Fe-phases hereafter) form and their size, morphology and distribution have profound effects on the castability and mechanical properties of the final parts. In most cases, these Fe-phases, especially when the needle-like or plate-like phases, such as β-Al_7Cu_2Fe phase, are detrimental to the alloys [5]. In some alloy systems, neutralisation elements, *e.g.*, Mn and Si, can be used to alter the morphology of the Fe-phases to a less harmful type [6–8].

Quantitatively understanding of the size, morphology and distribution of the Fe-phases are of paramount importance in the physical metallurgy of recycling Al alloys, and in the manufacturing of high-quality components for the transportation industry. In the past, majority of the research on Fe-phases was conducted using 2-dimensional (2D) imaging methods [6–8], *i.e.* optical and/or electron microscopy, which gives very limited information about 3-dimensional (3D) structures/morphologies, and the spatial interconnections and correlations between the different phases. Recently, a number of investigations [9,10] have been made by using scanning electron microscopy (SEM) and focused ion-beam (FIB) tomography to reveal the well-connected and branched 3D network structure in Chinese script type α-Fe-phases in Al-Si alloys (the typical composition is $Al_{14}Fe_{2.8}Si_2$). 3D morphology of Fe-phases has also been characterised using serial sectioning plus optical [11] or electron microscopy [12]. However, FIB is normally used for sectioning sub-micrometre features [12], not for those of length scale in many hundreds, even thousands of micrometres, such as the Fe-phases in present study, and serial sectioning is often very time-consuming. Recently, synchrotron X-ray tomography has been used to study the 3D microstructures of a wide range of multiphase alloys [13–16]. For example, the nucleation and growth of the Fe-phases in 3D in Al-Si alloys were reported in [4,17–20]; and the snapshots of the 3D Fe-phases in Al-Cu alloys were given by Gutiérrez et al. [21]. However, Gutiérrez, et al. did not segment the individual Fe-phases [21]. Hence, the detailed 3D structures of the Fe-phases, and their spatial interconnection with other phases such as Al_2Cu have not been revealed.

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Table 1
The parameters used for the tomography acquisition at TOMCAT, Swiss Light Source.

X-ray beam	Polychromatic radiation
Scintillator	LuAG: Ce 100 μm
Detector	GigaFRoST [29]
Effective pixel size	1.62 μm
Detector area	1392 × 1392 pixels
Exposure time	7.0 ms
Magnification	6.8×
No. of projections	2000
Sample-to-scintillator distance	180 mm

Normally, 4 different types Fe-phases exist in the Al-Cu alloys with Fe concentration of 0.5–1.0% [22,23]. They are plate-shaped phases β-Al_7Cu_2Fe and $Al_3(FeMn)$; and Chinese script-type phases, α-$Al_{15}(FeMn)_3Cu_2$, and $Al_6(FeMn)$. These Fe-phases are very different to the plate-shaped Fe-phases found in the Al-Si alloys [4,17–20]. So far, no reports have been found that describe the true 3D structures of the 4 typical Fe-phases present in the Al-Cu alloys [22,23] and their spatial interconnections and correlations.

In this paper, we used synchrotron X-ray microtomography and skeletonisation method to study the 3D network structures and morphologies of the Fe-phases and the associated Al_2Cu phases in two alloys: Al-5%Cu-0.6%Mn with 0.5% and 1.0% Fe (named 0.5Fe alloy and 1.0Fe alloy, respectively, hereafter). Higher Fe content was deliberately added into the two alloys to mimic those often found in the recycled Al alloys. The complex 3D network structures of the Fe-phases and the Al_2Cu phases, their mean curvature distributions and the inter-dependence between the Fe-phases and the Al_2Cu phases were reported for the first time. Furthermore, the true 3D morphologies of the 4 different types Fe-phases in Al-5%Cu alloys are also revealed, providing more quantitative 3D information for understanding the structures of the Fe-phases.

Both alloys were made by using pure Al ingot (99.9%), Al-20% Cu, Al-10%Mn and Al-10%Fe master alloy with the correct charge weight. The feedstock materials were held inside a clay-graphite crucible and melted at 780 °C in an electric furnace. After temperature homogenisation, the melt was then cooled to 710 °C, and poured into a steel permanent mould (Ø 65 mm × 70 mm) preheated to 200 °C to form an ingot. Cylindrical samples (~Ø10 mm × 20 mm) were cut from the edge of the ingot and then machined into Ø 2 mm × 5 mm for tomography scans. The solidification time at the location where the samples were taken was ~42.5 s with an average cooling rate of ~2.5 K/s [24]. Routine 2D microstructure characterisation was made using a FEI Quanta 200 Field Emission Gun scanning electron microscope (SEM) equipped with an energy-dispersive X-ray analyzer. For the SEM samples, 10% NaOH aqueous solution was used to dissolve the Al matrix (20 min) in order to expose more of the Fe-phases embedded inside the Al matrix. Synchrotron X-ray tomography experiments were performed at the TOMCAT beamline X02DA of the Swiss Light Source (SLS), Paul Scherrer Institute, Switzerland. The experimental parameters used are given in Table 1 [25]. A white beam from a superconducting bending magnet source was used with a 400 μm Al filter to remove the low energy tail of the incident beam and to reduce heat load on the sample and detector. The imaging system consists of a 100 μm LuAG: Ce scintillator (Crytur) coupled to a white-beam compatible microscope with a 6.8× magnification (Optique Peter). For each scan, 2000 projections were acquired over 180° of sample rotation. Tomographic reconstructions were performed on the TOMCAT cluster [26] using the GridRec algorithm [27] coupled with the Parzen filter [28].

Fig. 1a shows the typical cross-sectional slice obtained from the synchrotron X-ray tomoscan, and an area of interest was extracted and showed in Fig. 1b. The corresponding SEM images and the deeply-etched Fe-phases are shown in Fig. 1c and d respectively. Both X-ray images and SEM images show that the pore is in dark, the Al matrix is in dark grey, the Fe-phases are in light grey, and the Al_2Cu phases are in white colour. Open source image processing software, Image J [30] was used to adjust the contrast between the different phases. Then, the 3D bilateral filter was applied to the tomography datasets to increase the contrast and reduce noise. Finally, the pores, Al dendrites, Fe-phases, Al_2Cu phases were manually segmented by using different threshold values (Pore: 0~10688, α-Al: 10689~26438, Fe-phases: 26439~38814; Al_2Cu: 38815~65535), and the boundaries between different phases were manually labelled. 3D segmentation and feature rendering were performed using Avizo Lite v9.0.1 (VSG, France) and the dedicated GUP visualisation node in Viper, the University of Hull's High Performance Computer (HPC) cluster. Normally, a sub-volume of 500^3 voxels with a voxel size of $(1.62\ \mu m)^3$ was chosen for further analyses. However, from the X-ray absorption contrast only, it is not possible to distinguish and segment the 4 different types of Fe-phases in the two alloys. Hence, the segmented Fe-phases from the X-ray tomoscans contain all 4-type Fe-phases. By comparing the SEM images of the Fe-phases revealed by the deeply-etched samples with those showed in the X-ray tomography, we are able to identify the 4 different type Fe-phases as discussed later in the paper.

Fig. 2 shows the 3D colour rendering of the Fe-phases, Al_2Cu phases and α-Al matrix and their mean curvature distributions for the 0.5Fe and 1.0Fe alloys, respectively. The mean curvature H [31] is defined as:

$$H = 0.5 * \left(\frac{1}{R_1} + \frac{1}{R_2}\right) \quad (1)$$

where R_1 and R_2 are the two principal radii of curves respectively. Local curvature is an important geometrical parameter for the interface between two phases (dendrites or intermetallics) formed during the solidification processes, influencing the diffusion of solutes and therefore the final morphology of the phases.

Fig. 2a and b show the complex network and intricate morphology of the interconnected Fe-phases and Al_2Cu phases. Red shows the Fe-phases, green for the Al_2Cu phases, blue for the Al matrix. These phases conglomerate together in the α-Al inter-dendritic region in the chosen volume of 162 μm × 162 μm × 162 μm. Fig. 2c and d show the 3D network of the Fe-phases with their mean curvatures for the 0.5Fe and 1.0Fe alloy, respectively. The Fe-phases form a spatially interconnected

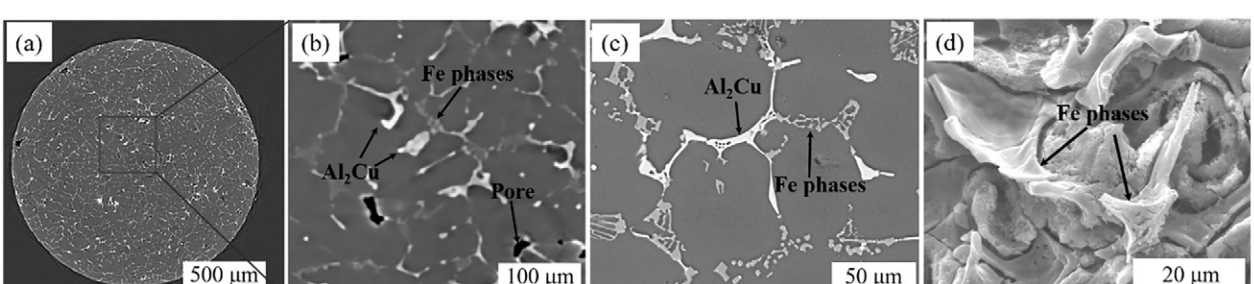

Fig. 1. (a) a typical 2D slice from the tomography scan of the 0.5Fe alloy; (b) the enlarged image of the framed area in (a) and processed using a 3D bilateral filter; (c) a typical SEM image of the 0.5Fe alloy; (d) a SEM image, showing the deeply-etched Fe phases.