3D ANALYSIS OF MACROSEGREGATION IN TWIN-ROLL CAST AA3003 ALLOY BEFORE AND AFTER HEAT TREATMENT

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Abstract

Twin-roll casting allows production of sheets and strips with required thickness without subsequent homogenization or hot rolling and therefore has a high potential for industrial applications. One of the drawbacks of this method is a formation of inhomogeneous structure with central segregation. Sheet prepared from 3003 aluminum alloy with 1 wt.% Mn, 0.2 wt.% Fe, 0.5 wt.% Si, 0.2 wt.% Cu and small addition of Zr (0.2 wt.%) and Cr (0.1 wt.%) was studied by X-ray computed tomography. Segregations in the material form as manganese, iron and silicon rich channels spread in the rolling direction. The material annealed at 450 °C exhibits diffusion of alloying elements from segregations into their surroundings and formation of voids or a material with lower density in the center of segregation channels due to Kirkendall effect.

Keywords: Al-Mn-Zr alloy, twin-roll casting, central segregation, Kirkendall effect, X-ray computed tomography

1. INTRODUCTION

1.1. Introduction

Because the industry demands a higher production efficiency, price and environment saving, new methods and materials are investigated. Twin-roll casting (TRC) allows production of sheets and strips of a required thickness avoiding subsequent homogenization or cropping known from ingot casting methods. A high cooling rate during TRC (~500 K/s) produces supersaturated solid solution, fine grains and finely dispersed primary particles [1]. On the other hand, inhomogeneities such as central segregations and flattened grains can be found in TRC strips [1, 2].

1.2. Segregations

Central macrosegregations are cylindrical, low melting point regions, oriented in the casting direction and appear as a result of combined solidification and rolling process [2, 3]. Such channels formed in the central plane have almost constant spacing and are at least an order of magnitude larger in size than dendritic grains in their vicinity [2, 4, 5]. Segregated channels reveal predominantly eutectic features. This suggests that a hot rolling component of the TRC process squeezes the mushy zone in between the already solidified skins that become thicker as they move into the roll gap. The liquid, which is enriched in alloying elements, is thus forced to the opposite side of the casting direction [2, 4]. With advancement of the solidification front, the concentration of solutes in liquid phase gradually increases. The liquid is trapped when the two solid skins weld together and well defined solute-rich channels are formed.

The most common alloying elements in 3003 Al are manganese, iron, silicon and copper. These elements are known to have strong influence on the precipitation behavior [2, 6, 7]. Two main phases which are frequently observed in Al-Mn-Fe-Si alloys are orthorhombic $Al_6(Mn,Fe)$ and cubic α-$Al_{12-15}(Mn,Fe)_3Si_{1-2}$ [2, 8, 9]. The main factors influencing the morphology and crystallographic phase are chemical composition and cooling rate [2, 10]. If macrosegregation occurs in the center of a sheet, it cannot be reduced by homogenization. The central eutectic segregates are not affected by a cold-rolling, aluminum matrix deforms around them [11].

1.3. Heat treatment and Kirkendall effect

The Kirkendall effect is a classical phenomenon in metallurgy [12]. It basically refers to a nonreciprocal mutual diffusion process through an interface of two metals so that vacancy diffusion occurs to compensate for the inequality of the material flow and that the initial interface moves. The first experiment was performed by Kirkendall in 1942 [13]. Kirkendall effect was observed on aluminum-silicon interface in semiconductors. During heating silicon diffuses into aluminum layer forming voids instead of eutectic layer [14, 15]. Although Kirkendall effect is generally observed on interface of bulk materials, chemists applied this destructive effect constructively in nanoscale for synthesizing hollow nanostructures e.g. spheres or nanotubes [12, 16], for example Ni-Cr wires in aluminum [17].But as far as we know, Kirkendall effect was not observed on the mostly manganese, iron and silicon central segregations in Al 3003 alloy so far.

1.4. X-ray computed tomography

The conventional methods of a microstructural analysis (such as light optical microscopy or scanning electron microscopy (SEM) are carried out in 2D. Acquirement of 3D information would require analyzing of multiple cuts through the sample, which is hardly feasible, time consuming and of course destructive for the sample. An example of nondestructive method of material characterization is X-ray computed tomography (XCT), which has become very important technique for 3D characterization of materials [18]. It is widely used for detection of cracks and pores, evaluation of parameters of cellular materials and visualization of intermetallic phases in metals [19]. XCT utilizes an X-ray source, a turntable as well as a digital detector. A sample is placed on a rotary table between X-ray source and detector. Projection images are generated at various angular positions, hundreds of projection images are needed to reconstruct a 3D dataset with a mathematical algorithm. The generated volumetric dataset consists of volumetric pixels, so called voxels, which feature a grey value corresponding to the density and the atomic number of the elements within that voxel [19]. The aim of the present study is to evaluate the morphology of central segregation in twin-roll cast AA3003 aluminum.

2. EXPERIMENTAL PART

A modified AA3003 aluminum alloy with 0.17 wt.% of Zr addition with a composition shown in **Table 1** was prepared by TRC method. In this method, molten alloy flows between two water-cooled rotating rolls where it solidifies. TRC material was cold rolled to 85 % thickness and subsequently cut into sample with approximately 11x7 mm^2 cross-section. Afterwards the sample was heat treated with 0.5 °C/min increase up to 450 °C and annealed for 8 hours at 450 °C then quenched into water. The sample was scanned with XCT before and after heat treatment in order to compare central segregation channels before and after heat treatment. Afterwards the sample was cut by a plane containing the most central segregations in order to compare X-ray measurements with SEM images. Scanning electron microscope FEI Quanta FEG 200 SEM was used for observation in back scattered electrons (BSE) at 10 kV. Specimen for SEM was mechanically polished. The sample was further cropped in the perpendicular direction in order to reduce volume for new higher resolution XCT measurement.

Table 1 Nominal composition of studied Al alloy (wt.%)

Al	Mn	Fe	Si	Cu	Zr	Cr
Balance	1.0	0.2	0.5	0.2	0.2	~0.1

XCT measurement with a voxelsize of (7.5 µm)^3 was carried out with 80kV, 200µA and a 0.1 mm Cu prefilter. Using a Nanotom 180NF desktop device from General Electric with a 2304 × 2304 Hamamatsu flat panel detector and a 180 kV nanofocus X-ray tube. The scan parameters for the high resolution measurement with a voxelsize of (0.8 µm)^3 were 80kV and 175 µA without using a prefilter. For this investigation a RX Solutions Easytom 160 device with a 1920 x 1536 Varian flat panel detector and a 160 kV Hamamatsu nanofocus tube was used. For visualization and evaluation the software VGStudio MAX 2.2 was used whereby the surface near region was excluded during evaluation to reduce influence of artefacts. For noise reduction, a 3 × 3 × 3 median filter was applied at the (7.5 µm)^3 dataset and a non-local means filter (value 2 using VGStudio 3.1) at the (0.8 µm)^3 dataset. For segmentation an ISO 50 threshold combined with advanced surface determination was used.

3. RESULTS

In the as-cast material we observed channels of central segregations in accordance with experiments of Šlapáková et al [2] done on the same material. Segregations form as a result of a fast solidification of the strip during twin roll casting. The two solidification fronts progressing from the surface are pushing the melt enriched in alloying elements in front of them. When they finally meet, solute rich segregation of primary particles forms. Such segregations have a form of long channels stretching along the rolling direction [2].The length of each segregation is several mm in the rolling direction. Spacing between each segregation in the transverse direction is approximately 0.5 mm. They are distributed in a band with the width of approximately 1 mm in the central part (**Figure 1**). Some of the segregations contain coarse primary particles with irregular shape and diameter of around 20 µm. EDX analysis of several segregations was performed on several samples of the same material [2].

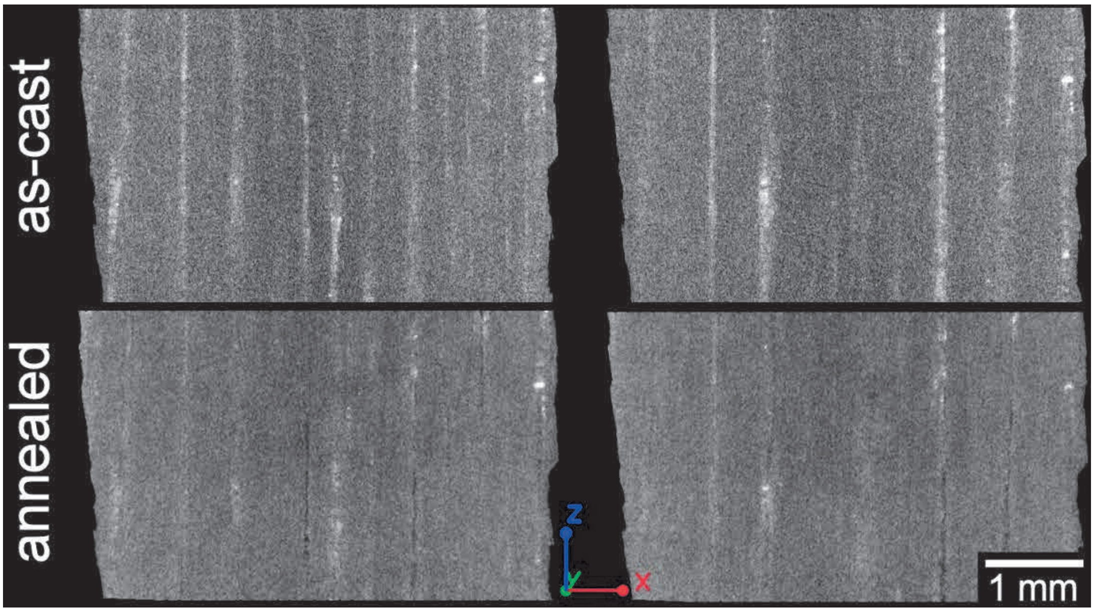

Figure 1 Comparison of central segregation channels before and after annealing of two slices using XCT

Segregation is composed mainly of Mn and Fe located generally in the same particle (**Figure 2**). Silicon is also partially present in the Mn- and Fe-containing particles but mainly appears in their interspace in clusters with size comparable to these particles. The first X-ray measurement of heat treated sample surprisingly revealed the dark areas inside the channels with lower intensity than the matrix material. This could indicate lower density of material or voids inside or in surrounding of channels simmilar to nanoscale Kirkendall effect observed on nanotubes and hollow nanoparticles [12]. SEM investigation confirmed presence of voids (**Figure 2**) and dispelled the possibility of an artifact. The cropped sample was again investigated by XCT with a higher resolution which revealed segmentation of dark areas (**Figure 3**) into multiple voids. Software analysis of XCT enabled to obtain the void size distribution in the volume. The void volume is mostly below 400 μm^3 (**Figure 4**). Voids are dispersed predominantly along central segregation channels. Element analysis of annealed sample revealed higher silicon concentration in the vicinity or at the inner surface of voids.

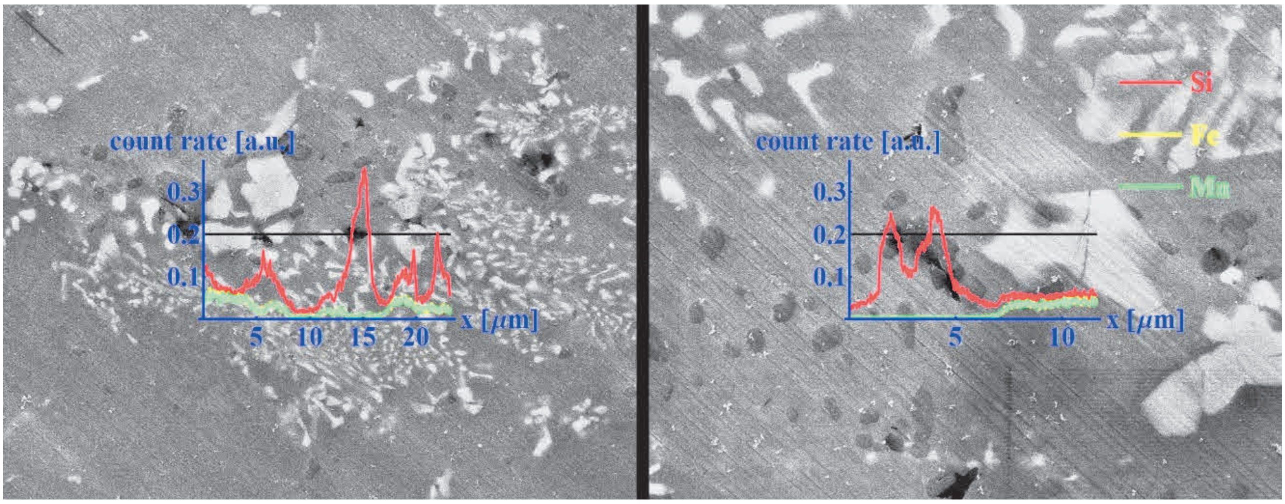

Figure 2 Element line analysis across voids and central segregations observed in SEM using secondary electrons

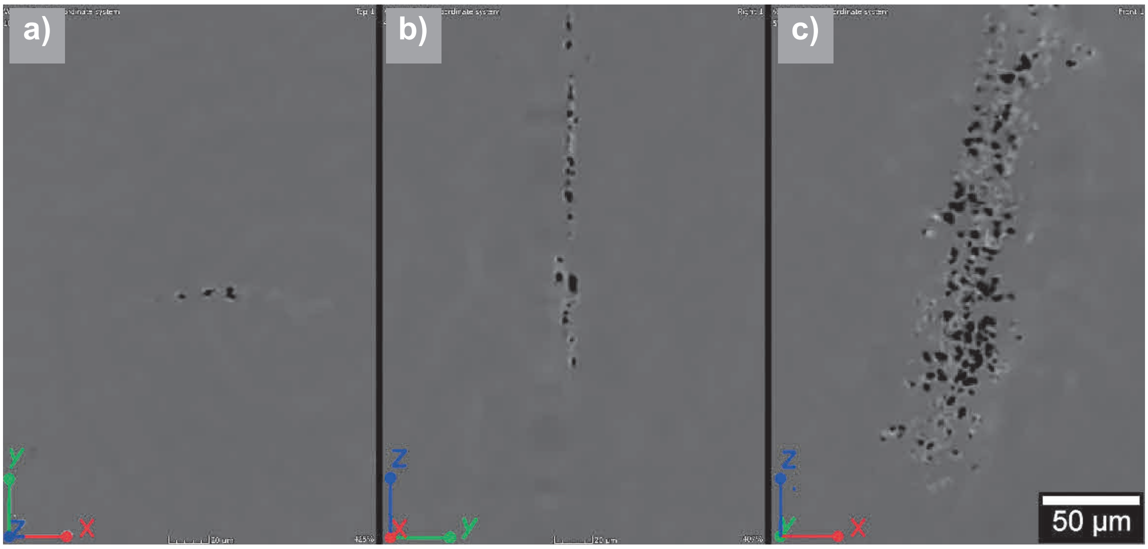

Figure 3 Voids in the annealed sample depicted from 3 different orientations by 3D XCT, a) the rolling direction, b) the transverse direction and c) the normal direction

DISCUSSION

XCT measurement enabled investigation of the identical area of the sample with no mechanical interfering. This method can detect deviations in density thus Mn and Fe rich central segregations in Al matrix can be recognized and their 3D distribution in volume of the sample can be reconstructed. Comparison of identical slices, of this reconstructed 3D volume, before and after annealing revealed decrease of density in areas of central segregations and even formation of low density areas in their places with density lower than Al matrix. With higher resolution we observed separate voids instead of low density areas. These voids are settled in the places where central segregations, now partially or entirely dissolved, were located. This was also observed is SEM (**Figure 2**). Even though SEM displays only thin surface area and sample was mechanically polished some voids were observed. Higher silicon concentration in a proximity of voids seems to be residue of silicon rich particles which surround Mn-Fe rich segregation. Diffusivity of Si is higher then Al and more then 3 orders higher then Fe or Mn [20]. Material apparently diffuses away from interior of Si particles resulting in Kirkendall effect [14, 15]. It is not understood yet if the observed Kirkerdall effect is whole process or part of more complex one and whether Mn-Fe particles also take part in the process. There is relatively little theoretical work on nanoscale Kirkendall diffusion effects caused mostly by the complexity of the reaction system itself compared to bulk planar interfaces [12]. The existing models work almost exclusively with binary systems. Further investigation as comparison of local element composition or phase analysis before and after annealing has to be done as well as computer simulations in order to describe such a complicated system as Mn-Fe-Si central segregation in aluminum alloy where also inhomogeneous shape and surface or other alloying elements must be considered.

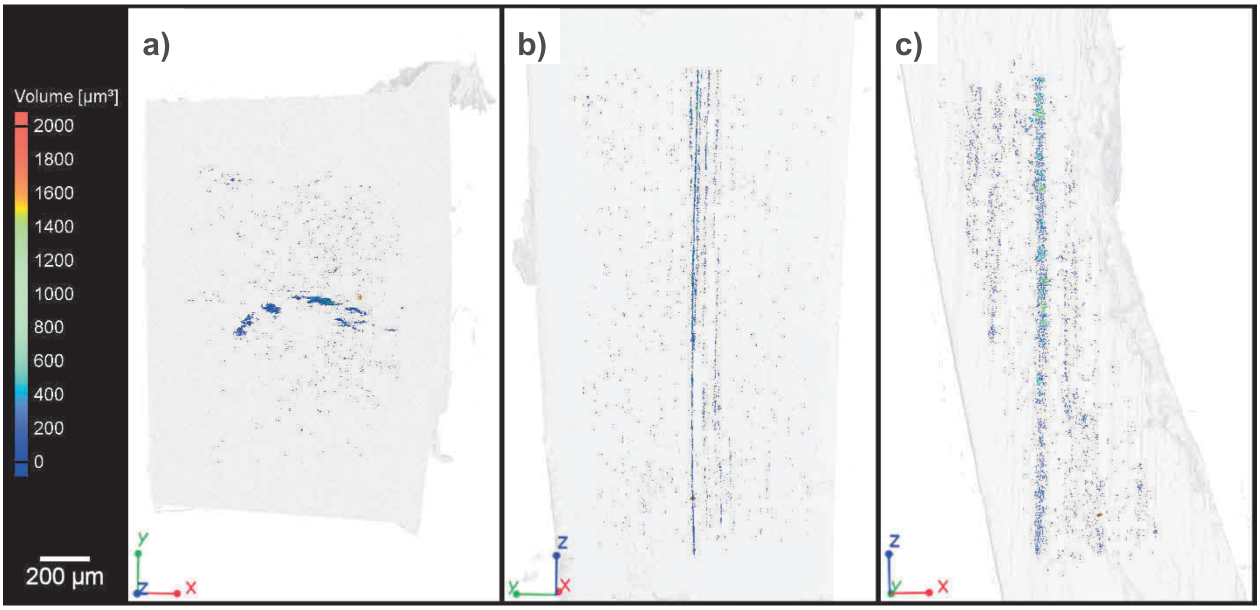

Figure 4 Void segmentation and size analysis depicted from 3 different orientations by 3D XCT, a) the rolling direction, b) the transverse direction and c) the normal direction

CONCLUSION

Twin-roll cast modified-AA3003 aluminum strips contain central segregations. They form channels elongated in the rolling direction with the length of up to several mm and thickness in the order of 100 µm. During heat treatment, material of these central segregations, probably mostly silicon, diffuses into their surroundings. Due to Kirkendall effect, voids are formed as observed in XCT. Their spatial and volume distribution was obtained by computer analysis of XCT data. The volume of each void is mostly below 400 µm³. These results were possible thanks to non-destructive nature of X-ray computed tomography which relatively quickly provides information about the 3D volume. Limitation of this method is that resolution of XCT images decreases with increasing dimensions of the cross-section of the sample.

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