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Microstructure evolution during homogenization of Al–Mn–Fe–Si alloys: Modeling and experimental results

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Abstract

Microstructure evolution during the homogenization heat treatment of Al–Mn–Fe–Si, or AA3xxx, alloys has been investigated using a combination of modeling and experimental studies. The model is fully coupled to CALculation PHAse Diagram (CALPHAD) software and has explicitly taken into account the two different length scales for diffusion encountered in modeling the homogenization process. The model is able to predict the evolution of all the important microstructural features during homogenization, including the inhomogeneous spatial distribution of dispersoids and alloying elements in solution, the dispersoid number density and the size distribution, and the type and fraction of intergranular constituent particles. Experiments were conducted using four direct chill (DC) cast AA3xxx alloys subjected to various homogenization treatments. The resulting microstructures were then characterized using a range of characterization techniques, including optical and electron microscopy, electron micro probe analysis, field emission gun scanning electron microscopy, and electrical resistivity measurements. The model predictions have been compared with the experimental measurements to validate the model. Further, it has been demonstrated that the validated model is able to predict the effects of alloying elements (e.g. Si and Mn) on microstructure evolution. It is concluded that the model provides a time and cost effective tool in optimizing and designing industrial AA3xxx alloy chemistries and homogenization heat treatments.

Keywords: Homogenization heat treatment; Microstructure modelling; CALPHAD; Aluminium alloys

1. Introduction

Al–Mn–Fe–Si, or AA3xxx, alloys are important commercial aluminum alloys. They have been used in many industrial sectors and in particular in the rapidly growing aluminum heat exchanger market. The homogenization heat treatment is an important step in the manufacturing route for these materials, as the microstructural changes during homogenization influence the deformation, recovery and recrystallization behavior (i.e. grain size, grain morphology, and crystallographic texture) during subsequent thermo-mechanical processes, i.e. high temperature extrusion. Therefore both industrial and academic research efforts have been made towards understanding microstructure evolution during this process [1–7].

The starting microstructure for the homogenization treatment is the as-cast microstructure, which consists of primary aluminum dendrites and a small amount of interdendritic eutectic (typically 1–2%). Segregation is observed to occur at two levels: (i) macrosegregation from the surface to the center of the billet, over the length scale of hundreds of millimeters; (ii) microsegregation over the length scale of the average secondary dendrite arm spacing, typically in micrometers. The interdendritic secondary eutectic phases are a mixture of Al_6(Mn,Fe) and α-Al(Mn,Fe)Si in

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the form of rod- or plate-like particles with a size of ~1–5 μm [1]. These are referred to as constituent particles. The primary aluminum dendrites are typically supersaturated, particularly in manganese, which has a low diffusivity in aluminum. The purpose of the homogenization heat treatment is to reduce microsegregation and produce an optimized microstructure for extrudability. This will result in an improved processing window for extrusion by (i) minimizing low melting point regions in the microstructure where incipient melting can occur during extrusion, and (ii) spherodization of the consitutent eutectic regions.

There are two major microstructural changes that occur during homogenization. The first is the growth and transformation of constituent particles. Their growth has been carefully examined using transmission electron microscopy (TEM) [1,3]. The changes in the constituent particles involve diffusional transport of Fe, Mn, and Si from the primary aluminum dendrites to the interdendritic region. The relevant length scale for diffusion of these alloying components changes is the scale of secondary arm spacing (μm) and in this paper it will be referred to as long-range diffusion. Transformation of the different constituent particles (e.g. Al_6(Mn,Fe) to α-Al(Mn,Fe)Si) is complex [8]. Based on experimental measurements of the constituent particle phase proportions Alexander and Greer [3] concluded that nucleation was the main controlling factor for the reaction, while Li and co-workers considered Si diffusion as essential for this transformation on the basis of their TEM study [9].

The second microstructural change which occurs in the as-cast material during homogenization is precipitation within the primary aluminum dendrites. This occurs via the nucleation, growth, and coarsening of small Fe-, Mn-, and Si-bearing particles (typical diameter 50–200 nm), which are referred to as dispersoids. This precipitation reaction leads to a reduction in the level of super-saturation in the primary aluminum dendrites. For these dispersoids diffusion at the scale of the inter-dispersoid spacing (~100 nm), hereafter referred to as short-range diffusion, controls their growth and coarsening. The composition and crystal structure of the dispersoids has been found to depend on the chemistry [1]. In an experimental study of an AA3003 alloy similar in chemistry to the material in this study (1.15 wt.% Mn, 0.58 wt.% Fe, 0.2 wt.% Si and 0.08 wt.% Cu) the dispersoids were identified as the α-Al(Mn,Fe)Si phase [1]. It was observed that the dispersoids could grow/coarsen to sizes of 100–200 nm and the peak number density can exceed 1000 μm^{-3} [1].

These experimental observations suggest that the starting chemistry and as-cast structure, as well as the temperature/time history experienced by the alloy, can change the nature of the dispersoids and constituent particles. Although some questions remain to be clarified and need further exploration, the essential knowledge is in place to understand the evolution of the microstructure, and thus it is possible to envision developing a chemistry-dependent predictive microstructural model for homogenization. A model would be very useful for the design of industrial processing, but will also provide an insight into the mechanisms involved in this complex microstructural evolution problem. For example, it has been experimentally observed that a dispersoid-free zone (DFZ) forms around the constituent particles during homogenization. One hypothesis is that the formation of DFZ is due to the interaction of long- and short-range diffusion of solute [10]. This hypothesis can be tested by comparing the prediction of a multi-length scale model with the experimental measurement results.

A number of models for homogenization in AA3xxx alloys have been developed. Lok et al. attempted to develop a semi-empirical two length scale model [11]. It is limited to a single diffusing component (Mn) and does not take into account transformation of the constituent particles during homogenization, i.e. from Al_6(Mn,Fe) to α-Al(Fe,Mn)Si for typical AA3003 chemistries. This transformation has been modelled by Li et al. [9] with the assumption that long-range diffusion of Si is the rate controlling factor. Alternatively, Gandin and Jacot [10] proposed a model which considered precipitation in both the intragranular and intergranular regions. The model is able to capture the interactions between the diffusion of all alloying components at the two different length scales. However, one drawback of their model is that they employed a solubility product method to approximate the thermodynamic data. Given the importance of accurate phase diagram knowledge for the selection of heat treatment parameters and alloy chemistry design, the solubility product approximation limits the predictive capability of the model.

The current work extends the approach of Gandin and Jacot by fully coupling the models for (i) the precipitation/dissolution of dispersoids and (ii) the long-range diffusion of solute to the constituent particles using a multicomponent CALPHAD database. To tackle the two length scale diffusion problems encountered in homogenization modeling it employs the mixing reaction methodology adapted from multiscale combustion modeling [12]. The new model will be validated by comparing its predictions with detailed experimental measurements conducted on AA3xxx alloys during homogenization. This work is part of a larger project to develop a through process model for microstructure and mechanical property evolution for extruded aluminum parts which are brazed together to manufacture heat exchangers for automotive applications. The outputs of the current homogenization model (e.g. the volume fraction, size, and spatial distribution of dispersoids/constituent particles, and the solid solution levels in the primary aluminum dendrites) are important inputs for subsequent extrusion and brazing models.

Development of the model and the results from experimental studies are described in Sections 2 and 3, respectively. The model was applied to simulate microstructure evolution during an industrially relevant homogenization