Materials & Design 128 (2017) 241–249

Contents lists available at ScienceDirect

Materials & Design

journal homepage: www.elsevier.com/locate/matdes

Modeling homogenization behavior of Al-Si-Cu-Mg aluminum alloy

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ARTICLE INFO

Keywords:
Modeling
Dissolution
Homogenization
Secondary dendrite arm spacing
Al_2Cu phase

ABSTRACT

The effect of the as-cast secondary dendrite arm spacing and homogenization temperature on the secondary phase dissolution and solute redistribution during homogenization of cast Al-Si-Cu-Mg aluminum alloy was investigated. Particularly, the concurrent dissolution of θ-Al_2Cu and Q-$Al_5Cu_2Mg_8Si_6$ phases was studied for the first time. Two modeling approaches were developed using finite-difference analysis to predict the dissolution kinetics; (i) a stationary interface modeling approach in which the interface is considered to be stationary and (ii) a moving boundary modeling approach in which the particle-matrix interface is moving. The dissolving particle shape was considered to be spherical and the dissolution process to be diffusion controlled. The two model predictions were compared against the experimental data obtained from microstructural characterizations. It was shown that although the stationary interface approach provides reasonable results in predicting the dissolution behavior at higher volume fractions of the secondary phase, the results deviate from the experimental measurements at lower volume fractions. On the other hand, the moving boundary approach was capable of predicting the final dissolution time of the secondary phase. It was also shown that the Q-phase does not dissolve completely as opposed to the θ-phase.

1. Introduction

Aluminum is the most heavily used non-ferrous metal in the world due to the demand for improved fuel efficiency in automobiles without impairing performance. Cast aluminum alloys are important classes of Al alloys with a multitude of automotive application, such as engine blocks and cylinder heads due to their light weight, well-established casting, shaping and recycling technologies [1,2]. Due to the significant segregation associated with the casting process [3], the homogenization treatment is an indispensable step in processing these alloys to ameliorate their mechanical properties. It is well established that the homogenization temperatures should be closely controlled to avoid incipient melting of the low temperature Cu-rich phases [4]. A key metallurgical reaction that occurs during the homogenization process of these alloys is the dissolution of the secondary phases. The other reactions that also take place during homogenization process are spherodization of the Si eutectic phases, fragmentation of the iron intermetallic phases, solute redistribution associated with the secondary phase dissolution and the removal of micro-segregation and finally the dendritic microstructure. Another important factors in designing the homogenization processes are the knowledge of the secondary dendrite arm spacing (SDAS) of the as-cast microstructure, which itself is controlled by the cooling rate during solidification [5,6]. SDAS dictates the distance between Cu-rich phase particles as well as the distance between the iron intermetallic phase and Cu rich phase, which affect the dissolution and homogenization times [7].

Modeling the dissolution of the secondary particles during high temperature treatments has been of significant research interest in the past. The work by Aaron [8] was among the very first attempts to theoretically analyze and model the dissolution of a second phase in equilibrium with a solid solution matrix. Later, Whelan [9] modeled the dissolution kinetics of a secondary phase particle. Subsequently, Nolfi et al. [10] expanded the model developed by Whelan by considering the effect of the interfacial reactions as well. Further efforts included the work by Brown [11], who considered the effect of particle shape (spherical, cylindrical and planar shapes) on the dissolution kinetics, and Singh and Flemings [12] whose dissolution model took into account the concentration gradient across the dendrite. The more recent works include Rometsch [13], who developed a numerical finite difference model to predict the dissolution and homogenization time in aluminum A356 and A357, as well as several other studies by Vermolen et al. [14–23] to model the dissolution of multicomponent phases. Then, Foroozmehr et al. [24] used a coupled dissolution-diffusion approach and a finite-element analysis to model the solutionizing process and solute distribution in a co-cast bi-layer Al alloy system.

The alloy of this study is an Al-Si-Cu-Mg aluminum alloy 319, which is a heat treatable casting alloy. Al-Si-Cu-Mag alloys have gained attention due to the demand for high thermal stability imposed by

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http://dx.doi.org/10.1016/j.matdes.2017.05.006
Received 20 January 2017; Received in revised form 20 April 2017; Accepted 4 May 2017
Available online 05 May 2017

stringent emission standards [25]. The complex microstructure of a 319 aluminum casting consists of aluminum dendrites, the Si eutectic phase, Cu rich precipitates (mainly θ-Al_2Cu), iron-intermetallic phases such as Al_5FeSi and $Al_{15}(Mn,Fe)_3Si_2$ as well as Mg-containing phases Mg_2Si and Q [26–29]. The size and morphology of the reported phases are dictated by the cooling rate during solidification rate [30–33]. It should be noted that there is an uncertainty for the stoichiometry of the Q phase and several stoichiometric compositions of the Q phase have been reported in different alloys including $Al_4Cu_2Mg_8Si_7$, $Al_5Cu_2Mg_8Si_6$, $Al_4Cu_1Mg_5Si_4$ and $Al_3Cu_2Mg_9Si_7$ [34,35].

Generally, the cast Al-Si-Cu-Mg alloys have been extensively studied in terms of the optimum heat treatment process from solution heat treatment to aging. However, relatively little work has been done to model their homogenization processes and the effect of microstructural variables such as dendrite arm spacing on the metallurgical reactions that occur. Although, Colley et al. [36,37] modeled the dissolution behavior of Al-Mg-Si alloy which was by comparison much simpler as it only involved the dissolution of Mg_2Si. The aim of this research is to model the (i) dissolution of the secondary phases (θ and Q), and (ii) the removal of segregation that occur during the homogenization process of Al alloy 319. The effects of the SDAS and the homogenization temperature as well as the effect of a two-step heat treatment are also investigated. The modeling results are also evaluated through comparison with experimental data. The validated modeling approach can provide useful information for understanding the cast alloy behavior during homogenization treatments and predict the alloy behavior under relevant processing conditions.

2. Experimental methodology

Experimental work was done on a wedge cast 319-type alloy. The overall alloy composition, measured by an optical emission spectroscopy technique, is reported in Table 1. Metallographic samples for the investigation of the as-cast microstructure were cut at distances of 12 mm, 25 mm, 50 mm, 75 mm and 100 mm from the chilled end perpendicular to the solidification direction. The optical micrographs taken from these samples are shown in Fig. 1. The coarseness of the microstructure is characterized by SDAS, which in turn is dictated by the cooling rate from solidifying temperature.

Samples for heat treatment investigations were cut from horizontal cross-sections of the casting located at 25 mm, 60 mm and 115 mm from the chilled end. Samples dimensions were approximately 15 mm (l) × 15 mm (w) × 10 mm (h). Single-step and two-step solution heat treatments were done using a fluidized sand bath (FB). For the single-step solution heat treatments, samples were soaked at 490 °C or 500 °C for various times up to 6 h to investigate the effect of homogenization on dissolution of particles. For the two-step solution heat treatments, samples were soaked for 1 h at 490 °C or 500 °C and then the temperature was raised to 510 °C. Samples were soaked for various times at 510 °C depending on the first step temperature. It should be noted that samples are exposed to an unavoidable non-isothermal heating process to reach the desired isothermal treatment temperature. However, the use of the FB minimizes this non-isothermal heating (at the centre of the sample). Considering the length of the overall homogenization process, this initial non-isothermal heating (2 min long) is assumed to be negligible. Samples were removed from the furnace and immediately quenched into water at room temperature. All metallographic examinations were carried out on horizontal sections perpendicular to the solidification direction. The samples were polished using a MD-Nap polishing cloth with diamond suspension and MD-Chem cloths with colloidal silica suspensions to finish.

Table 1
The chemical composition of the 319 Al alloy.

Element	Al	Si	Cu	Mg	Fe	Mn
Amount (wt%)	Bal.	8.3	2.8	0.5	0.45	0.34

Thermodynamic analysis was conducted using ThermoCalc software to obtain phase diagram and the stability of different phases as well as the equilibrium concentrations at the particle-matrix interface in the matrix. Fig. 2 illustrates the results of this analysis. According to this analysis, the temperature range for the dissolution is 475 °C–510 °C. As shown in Fig. 2, the equilibrium phases and the temperature ranges in which they are present are the θ phase, up to 465 °C; the Q phase, up to 534 °C; the Si phase, up to 582 °C and the iron phase, up to 616 °C. This implies that by heating at 490 °C or 500 °C for a long holding time, θ will completely dissolve but the iron phase, as well as a fraction of the Q phase will still be present. It should also be noted that the homogenization treatment for these alloys should not be conducted at temperatures over 510 °C as the Cu rich phases will start to melt. The parameters that are used in the model as inputs are shown in Table 2 in which c_i^{sol}, ρ, D_0 and Q are the concentration of the Cu at the particle-matrix interface, the density, the diffusivity factor and the activation energy, respectively.

Phase characterization was done using X-ray diffraction (XRD). The scan was conducted using a Cu-K_α radiation, a voltage of 40 kV and an aperture size of 2 mm. Microstructural investigations were done using optical microscopy (OM) and scanning electron microscopy. Scanning electron microscopy (SEM) analysis was conducted in secondary electron (SE) mode using an accelerating voltage of 20 keV and a working distance of 17 mm. Electron dispersive spectroscopy (EDS) along with were used to characterize different phases present in the as-cast alloy. Secondary dendrite arm spacing and area fraction of particles in the as-cast and heat treated samples after different times were measured using OM and image analysis. Different phases were distinguished based on their color difference under the microscope. To measure the area fraction 50 fields in a straight line were examined and the fraction of the area of the particles to the total area of measurement was calculated through manual image analysis. Volume fraction was then approximated by area fraction, i.e., $\boldsymbol{A_f} = \boldsymbol{V_f}$ as was suggested by Delesse [38].

The volume fraction of the Q phase has been assumed to be 0.006 according to the ThermoCalc analysis. Densities of the θ, Q and Fe phases are 4.34 g/cm^3 [40,41], 2.79 g/cm^3 [40,42] and 3.3–3.6 g/cm^3 [40,41], respectively. The density of the 319 Al alloy is 2.73 g/cm^3 [43].

3. Microstructure investigation

The average measured primary dendrite arm spacing (PDAS), SDAS, particle sizes, inter-particle spacing and initial volume fraction of different phases at sections distanced 25, 60 and 115 mm from the end chill are reported in Table 3.

A typical SEM micrograph of the as-cast sample with clearly observable dendritic microstructure and particles solidified at the inter-dendritic regions is shown in Fig. 3.a. According to the EDS, different phases were characterized. The EDS analysis results are reported in Table 4. Two types of iron intermetallics can be seen in the micrograph; Needle-like (deleterious for mechanical properties [44]) and more rounded skeleton-like $Al_{17}(Fe_{3.2}Mn_{0.8})Si_2$ (Fig. 3.b). θ and Q phases are shown in Fig. 3.c. It is seen that the Q phase forms besides θ particles in the microstructure. Al_2Cu particles have two different types, blocky and eutectic, as observed in Fig. 3.d. Eutectic Al_2Cu itself has two types; coarse and fine (Fig. 3.d). The average Al_2Cu particle size is 43.7, 56.4 and 90.2 μm^2 for the SDAS of 14, 22 and 39 μm, respectively. The Q phase stoichiometry suggested by EDX is $Al_5Cu_2Mg_8Si_6$. The effect of homogenization on the microstructure of the as-cast alloy is further shown in Fig. 4. As Fig. 4.b suggests, the Al_2Cu phase has been dissolved during homogenization, while, the iron-containing intermetallic phases still exist in the matrix. XRD was done as a complementary test to confirm the presence of different inter-metallics. Fig. 5 depicts the result of a XRD test on the initial as-cast alloy. Different phases including θ-Al_2Cu, Q, as well as iron ($Al_{17}(Fe_{3.2}Mn_{0.8})Si_2$) and Si phases were identified. The Q phase has