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Research paper

Effect of particle shape and size distribution on the dissolution behavior of Al_2Cu particles during homogenization in aluminum casting alloy Al-Si-Cu-Mg

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ABSTRACT

Finite element and finite difference modeling approaches were used in this study. Planar, cylindrical, spherical and elliptical particle shapes were considered. A log-normal distribution function was utilized to evaluate the effect of the particle size distribution on the dissolution behavior of the Al_2Cu particles. The models were validated by conducting homogenization experiments, and using microscopy and image analysis. The dissolution kinetic is not affected by the particle-matrix interface. The assumption of spherical shape results in the best agreement between the modeling predictions and the experimental data provided that the effect of the particle size distribution instead of an average particle size is taken into account.

1. Introduction

The demand for improved fuel efficiency in transportation vehicles without impairing performance has led aluminum alloys to become the most heavily consumed non-ferrous metal (Starke and Rashed, 2017). Homogenization is an indispensable step in processing Al alloys to improve their mechanical properties since there is always microstructural segregation associated with aluminum casting (Shi et al., 2015). The homogenization process also serves as the solution treatment required for subsequent age hardening process (Apelian, 2008). The homogenization temperature should be within a range that does not cause incipient melting of the secondary phases (Mohamed et al., 2012). During the homogenization process, the secondary phases undergo dissolution in two stages that together control the dissolution kinetics: stage I) atom transfer across the particle-matrix interface (interface reaction), and stage II) diffusion of the solutes away from the interface (long-range diffusion) (Brown, 1976). Other important considerations in designing the homogenization processes are the secondary dendrite arm spacing (SDAS) and their aspect ratios, which are controlled by the cooling rate during solidification (Manente and Timelli, 2011). An outcome of SDAS is the distance between different phase particles that affects the homogenization time (Sjolander and Seifeddine, 2014). Another factor that affects the dissolution kinetic is that how far the system is from the equilibrium state (homogenized state), which provides the system with the driving force towards the equilibrium state. This factor is dictated by the casting process (Ashiri et al., 2014a). Casting processes also affect the SDAS, which consequently affect the particle dissolution rate (Ashiri et al., 2014b).

Modeling the homogenization process deepen our understanding of the alloy behavior during heat treatment, which in turn enables process optimisation, and results in lower variability in the final properties of the material. This is of paramount importance when casting large irregular shaped parts that results in different solidification rates and consequently varied microstructural properties at different sections of the parts. Modeling the dissolution kinetics of the secondary phase particles that are in equilibrium with the matrix was first initiated by Thomas and Whelan (1961). They considered that the dissolution kinetic is the reverse of the growth rate. Later, Aaron (1968) suggested that theoretically the dissolution kinetics is not simply the reverse of the growth rate. He modeled the dissolution of a binary phase in one dimension. However, his model only considers the transient part of the dissolution field. Afterward, Whelan (1969) expanded the model developed by Aaron (1968) considering both the transient and steady-stated part of the diffusion field. Nevertheless, the model assumes that dissolution is limited by long-range diffusion (i.e. the particle is in equilibrium with the matrix) and it does not consider the effect of particle-matrix interfacial reactions on the dissolution kinetics. Nolfi et al. (1969) then proposed that the dissolution process is controlled by interfacial reactions at the beginning of the process and it is diffusion controlled at longer times of dissolution. The model was limited to the spherical particle case. After Nolfi et al. (1969), Brown (1976) considered the effect of particle shape on dissolution kinetic. They

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considered spherical, planar and cylindrical geometries. Nonetheless, the model was still capable to predict the dissolution of only binary phases. Subsequently, Rometsch et al. (1999) modeled the dissolution of Mg_2Si binary phase in Al-Si-Mg alloy. They used Fick's first law to model the final dissolution time of the secondary phase and they assumed that the particle-matrix interface is stationary as the particles dissolve. Their model does not consider the segregation of solutes in the matrix that is associated with cast alloys as well as the movement of the particle-matrix interface during dissolution. Following this model, Vermolen and Vuik (2000) modeled dissolution of a multicomponent phase in multicomponent alloys. They assumed that the particle-matrix interface is moving instead of being stationary. They used Fick's second law to model the dissolution process. Recently, Foroozmehr et al. (2012) developed a coupled dissolution-diffusion approach and a finite-element analysis to model the solutionizing process and solute distribution in a macroscale through the interface of the two layers. Most recently, Colley et al. (2014) modeled the dissolution of secondary phase in Al-Mg-Si alloy which is a simpler cast alloy system compared to Al-Si-Cu-Mg alloy studied in this research.

The alloy used in this study is a commercially important aluminum alloy 319 (Al-Si-Cu-Mg), which is a heat treatable casting alloy. This alloy is extensively used for automotive engine block and cylindrical head applications due to the demand for high thermal stability (Ceschini et al., 2016). Different phases that are present in the microstructure are the aluminum dendrites, Si eutectic phase, Cu rich precipitates (θ-Al_2Cu and Q-$Al_5Mg_8Cu_2Si_6$), iron-intermetallic phases such as Al_5FeSi and $Al_{15}(Mn,Fe)_3Si_2$ (Ibrahim et al., 2011). Q-$Al_5Mg_8Cu_2Si_6$ is a low melting point phase, and melts at temperatures above 505 °C (Samuel, 1998). It also increases the possibility of melting of Al_2Cu phase (Han et al., 2008). Morphology and size distribution of different phase particles are affected by the SDAS (Ceschini et al., 2017). Although the models referenced above have been developed for the dissolution of the secondary phases in binary or multicomponent alloys, none of them have been applied to examine the effect of the segregation, particle shape and particle size distribution on dissolution of the particles in Al-Si-Cu-Mg aluminum alloys during homogenization. Recently, the homogenization behavior of this type alloy was modeled (Sadeghi et al., 2017). The novelty of this research is to model the effect of the particle shape particularly elliptical shape, and particle size distribution on the dissolution kinetics. The model predictions were compared to experimental measurements.

2. Experimental methodology

The alloy of this study was a 319-type alloy, which was melted in an open air furnace and gravity cast using a wedge shaped copper mould which allowed for different SDAS at different wedge heights. More details about the casting process can be found elsewhere (Mackay and Sokolowski, 2010a). The chemical composition of the alloy is shown in Table 1.

The wedge cast ingot was sliced, and 1 cm (l) × 1c cm (w) × 1 cm (h) specimens for the homogenization heat treatment were cut horizontally at distances of 25 mm, 60 mm and 115 mm from the end chill perpendicular to the solidification direction. A fluidized sand bath (FB) was used for the solutionizing, which offers high heating rate; therefore, it minimizes the effect of non-isothermal heating to reach the set point temperature (Ragab et al., 2011). The solution heat treatment temperature was 490 °C or 500 °C. Specimens were solution heat treated for different times up to 6 h depending on the solutionizing temperature and SDAS. At solutionizing temperature of 500 °C, samples with SDAS of 14, 22 and 39 μm were soaked for 45 min, 1 h and 4 h. Samples with SDAS of 39 μm were soaked at 490 °C for 6 h. Solution heat treated specimens were then quenched into water at room temperature to preserve their microstructure.

Table 1
Chemical composition of the 319 alloy.

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

Optical microscopy of polished samples was done on sections perpendicular to solidification direction, which was followed by image analysis for quantitative analysis of the SDAS and area fraction of the secondary phase particle. Volume fraction was then assumed to be equal to area fraction, i.e., $\boldsymbol{V_f = A_f}$ (Vander Voort, 2000). Different phases were distinguished due to having different color scales. The particles size was then calculated based on their area measurements. As-cast and heat treated samples were examined using scanning electron microscopy (SEM) in secondary electron (SE) mode and electron dispersive spectroscopy (EDS) to identify different phases present in the alloy.

Phase diagram of the alloy as well as the weight fraction of different phases at different temperatures were obtained using ThermoCalc software. The predicted phase diagram is shown in Fig. 1. The predicted phase diagram by ThermoCalc, showed that the temperature range for the dissolution process is 475 °C to 510 °C implying that solutionizing at higher than 510 °C results in melting of the Al_2Cu phase rather than its dissolution. Solutionizing within this temperature range, results in complete dissolution of the Al_2Cu phase. The equilibrium phases and their weight fraction at different temperatures in which they are present are depicted in Fig. 2. The volume fraction of the Q phase in the studied alloy was negligible compared to the Al_2Cu phase, therefore, the model only considers the dissolution of the Al_2Cu phase.

3. Microstructure investigation

Fig. 3a show typical optical micrographs of the as-cast microstructure. It can be seen that the microstructure is dendritic with intermettalics phases formed at interdendritic regions. The effect of homogenization on the microstructure is shown in Fig. 3b. It can be seen that the dendritic microstructure is eliminated after homogenization and Si phase particles spherodized somewhat. The as-cast microstructure consists of α–Al dendrites, Si eutectic and intermetallic phases including Al_2Cu and AlFeMnSi, as identified using EDS. The morphology of these phases are shown in Fig. 4. The results of the EDS analysis of the different phases present in the alloy are shown in Fig. 5a to d. The average SDASs measured at 25, 60 and 115 mm from the chilled end were 14, 22 and 39 μm, respectively. The as-cast Al_2Cu particle size ranges from 10 μm^2 to 100 μm^2. The as-cast volume fraction of the Al_2Cu particles for SDASs of 14, 22 and 39 μm were 1.8, 1.8 and 2.6%, respectively. The average distance between the Al_2Cu particles were 19.5, 25.2 μm and 28.7 for the SDAS of 14, 22 and 39 μm, respectively.

4. Mathematical model

Solution heat treatment at high enough temperatures can dissolve Al_2Cu phase particles that are present in the as-cast material. The effect of the shape of the particles on the kinetics of the dissolution process was modeled in two and three dimensions using the commercial package COMSOL. Transport of dilute species and deformed geometry modules of the COMSOL were used. For two-dimensional (2D) modeling, finer and fine meshes and for the three-dimensional (3D) modeling, normal and coarse mesh were used. The total number of elements within the diffusion field for finer, fine, normal and coarse meshes are 2072, 1272, 910 and 436, respectively. Four different particle morphologies, i.e. spherical, planar, cylindrical and elliptical geometries were considered. The diffusion equation (Eq. (1)) was solved using the finite element method to find the concentration profile, c_{Cu}, in the diffusion field after each time step.