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Modeling over-ageing in Al-Mg-Si alloys by a multi-phase CALPHAD-coupled Kampmann-Wagner Numerical model

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ABSTRACT

The formation of the equilibrium precipitation phase during ageing treatment of Al-Mg-Si alloys is preceded by a series of metastable phases. Given longer ageing time, higher ageing temperature or elevated temperature service condition, β″, the main hardening phase, would be replaced by the more stable metastable phases such as β′, B′, U1 and U2. The post-β″ microstructure evolution, called “over-ageing”, leads to a steep drop in the hardness evolution curve. This paper aims to predict directly over-ageing in Al-Mg-Si alloys by extending a CALPHAD-coupled Kampmann-Wagner Numerical (KWN) framework towards handling the coexistence of several different types of stoichiometric particles. We demonstrate how the proposed modeling framework, calibrated with a limited amount of experimental measurement data, can aid in understanding the precipitation kinetics of a mix of different types particles. Simulation results are presented with some earlier reported transmission electron microscopy measurements [1,2] to shed light on how the alloy composition and ageing treatment influence the post-β″ phase selection.

1. Introduction

The precipitation kinetics in heat-treatable aluminum alloys is quite complex. The formation of the equilibrium precipitate phase is preceded by a series of metastable ones due to their ease of nucleation. Examples include the needle β″ precipitate in Al-Mg-Si [3], the plate θ′ precipitate in Al-Cu [4], the platelet η′ precipitate in Al-Zn-Mg [5], and the lath S′ precipitate in Al-Cu-Mg alloys [6]. It is these metastable precipitates rather than their stable counterparts that are contributing to peak hardening. However, given longer ageing time, higher ageing temperature or extended service time at an elevated temperature the precipitates responsible for the peak hardness will be replaced by other more stable precipitates, and eventually the equilibrium phases will form. This phenomenon is termed “over-ageing”. During over-aging the precipitate density will decrease and the precipitate size will increase. This microstructural change would results in the deterioration of the material strength [2].

There is a considerable industrial and academic interest in understanding the details of the transition from metastable to stable phases, especially concerning the industrially important Al-Mg-Si alloys. It has been established from Transmission Electron Microscopy (TEM) and High Resolution (HR) TEM investigations [3,7] that the precipitation sequence from the quenched supersaturated solid solution (SSSS) at room temperature up to the formation of thermodynamically stable Mg_2Si (β) for most common Al-Mg-Si alloys is:

$$\text{SSSS} \rightarrow \text{Clusters} \rightarrow \text{Co-custers, GP}(Mg_4AlSi_6) \rightarrow \beta\text{^}''(Mg_5Si_6) \rightarrow \beta'(Mg_9Si_5),\ B'(Mg_9Al_3Si_7),\ U1(MgAl_2Si_2),\ U2(MgAlSi) \rightarrow \beta\ (Mg_2Si).$$

It should be noted that although the precipitate structures have been identified experimentally, the mixing of minor alloying elements and impurities (such as Cu) can alter their compositions, incite other precipitate structures or cause disorder, in addition to the effects from the major alloying components. For example, β″ as well as β′ precipitates can be disordered, containing regions of each other's structure and the U2 structures in the same needle [8]. In this paper we neglect such complications, and use the published compositions as our focus is on modeling the post-β″

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transformation, i.e., how the β'' particles dissolve and post-β'' particles grow in an over-ageing condition. As discussed in Ref. [9], in Al-Mg-Si alloys the fully coherent monoclinic needle-shaped β'' phase, which dominates at peak hardness, is replaced by four less coherent (but still needle-shaped) phases during over-ageing: these are the hexagonal β', the trigonal U1 (TYPE-A), the orthorhombic U2 (TYPE-B) and the hexagonal B' (TYPE-C). Please note that instead of Type A, B and C as introduced by Matsuda et al. [10], we use U1, U2 and B' as the notation in this paper. The main equilibrium phase is the cubic β, but the Si phase may also form. Whether a particular phase forms depends on the ratio between Mg and Si in the chemical composition of the alloy, the ageing temperature and the thermo-mechanical processing history prior to the ageing treatment as revealed by the reported TEM characterizations [1]. For instance, Si-rich alloys tend to form U1, B' and Si particles during a prolonged over-ageing treatment, while Mg-rich alloys gain larger fractions of β' and U2. As found earlier, an alloy tends to select a post-β'' phase having the Si/Mg ratio closest to its own Si/Mg ratio [1]. Due to the complex interactions among the many factors involved, it is desirable to have a model to predict directly this complex post-β'' microstructure evolution.

The development of such a predictive model requires understanding of the physical mechanisms behind the post-β'' transformations. It has been observed under TEM that the number densities of post-β'' phase particles are much less (about 1/1000 to 1/10) than those of β'' [1,2]. In contrast to the pre-$\beta'' \rightarrow \beta''$ transformation, which is mainly achieved by changing the preceding phase composition, with minimal structural modifications [7], the $\beta'' \rightarrow$ post-β'' transformation must involve the dissolution of most of the β'' particles to provide solute for the growth of the new, and probably separate nucleation of β', B' or U phase particles. In other words, the post-β'' transformation clearly requires diffusional transportation of solute between the dissolving and growing particles. If one put aside the nucleation, it differs from the classical particle coarsening, i.e., Ostwald ripening, only in the origin of the thermodynamic driving force. The driving force for Ostwald ripening is interfacial energy, while the main driving force for the post-β'' transformation is the Gibbs energy difference between the dissolving (metastable) and growing (more-stable or stable) phases. This observation indicates that the competition among particles for solute during the post-β'' transformation could be modelled by extending the approach employed to model Ostwald ripening. The idea of transformation from GPI to GPII zones (metastable phases) via coarsening in Al-Cu alloys has already been suggested some years ago [11,12]. This interesting topic is further explored in this paper to demonstrate how a coarsening model could be extended to simulate the competitive growth and dissolution of multiphase particles during an over-ageing treatment of Al-Mg-Si alloys.

Many approaches, including the phase field method and the Kampmann-Wagner Numerical (KWN) approach, have been proposed in the literature for modeling coarsening. In contrast with the accurate but computationally expensive phase field approach [13], the KWN approach has been gaining popularity recently [14–26]. This is due to its mathematical simplicity and convenient coupling with the CALPHAD database, enabling an efficient treatment of multi-scale, multi-component industrially significant problems [14–25]. It should be noted that the coupling of the KWN model with the thermodynamic databases developed in the CALPHAD research community is a scale-bridging feature as the databases could be established on the base of first principle calculations [27,28]. It has been pointed out by Kozeschnik et al. that the coupling with the CALPHAD could bring the KWN approach's predictive power to a tuning parameter free level [29]. The KWN approach fits into our research purpose; therefore it is chosen to treat the post-β'' microstructure evolution.

The methodology of the KWN approach, initially proposed by Kampmann and Wagner in Ref. [19] for modeling precipitation kinetics, is also found in the seminal work of Maxwell and Hellawell published earlier, i.e. in 1975 on as-cast grain size prediction [26]. The essence of this approach is that the precipitate size distribution curve could be subdivided into size classes, each of which is associated with a number of identical precipitates. The temporal evolution of the size distribution is then tracked by following the size evolution of each discrete size class. This modeling framework and its CALPHAD-coupled multi-component extensions have been seen as a key microstructure chain model in an Integrated Computational Materials Engineering (ICME) modeling framework to optimize alloy chemistry and heat treatment parameters for many industrial metallic materials [14–26]. In the most recent extension of the KWN approach, the assumption of the precipitate particles being spherical has been released enabling a better treatment of needle-shaped particles' precipitation kinetics in aluminum alloys [30,31]. Another extension is reported by Myhr et al. in Ref. [32], where the KWN model is extended to accommodate the dislocation generation, the cluster formation and the competitive nucleation of β'' and β' precipitates during cold deformation, natural ageing and artificial ageing of aluminum alloys, respectively. All of these works reveal the versatile and generic nature of the KWN modeling framework. It should also be mentioned that rapid progress in the CALPHAD community on thermodynamic databases for metastable and stable phases, i.e. the work reported in Refs. [27,28] on Al-Mg-Si alloy system, enables the direct applications of the KWN model to industrial alloys.

In this paper, we will extend the multi-component KWN model reported in Refs. [16,30,31] to treat concurrent nucleation, growth and coarsening of multi-phase precipitate particles. The coupling of the extended model with the reported Al-Mg-Si CALPHAD databases [27,28] will be implemented. Then the new model will be applied to predict the post-β'' microstructural evolution during over-ageing of Al-Mg-Si alloys. The simulations will be discussed together with some reported TEM measurement to shed light on how the alloy composition and ageing temperature influence the post-β'' phase selection.

2. Model description

In this section the assumptions in deriving the multi-phase KWN model is listed and the adaptions to the KWN model toward the multi-phase extension are described.

2.1. The assumptions

Our idea is to consider the competition of the multi-phase particles for solute during the post-β'' transformation as Ostwald ripening driven by the Gibbs energy difference between the dissolving (metastable) and growing (more-stable or stable) phases instead of interfacial energy. The nucleation of the new particles during the over-ageing stage is assumed to follow the classical nucleation law. Using the CALPHAD-coupled multi-component version of the KWN model reported in Refs. [16,30,31] as a starting point, the extension required to treat concurrent nucleation, growth and coarsening of multi-phase particles is adding one more interfacial phase composition relation equation for each extra precipitation phase. Before proceeding to the multi-phase KWN model description, it is useful to summarize the assumptions adopted below. For the calculation of precipitate growth rate, the assumptions include:

- The precipitate shape is assumed to be spherical.