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A mathematical model coupled to CALPHAD to predict precipitation kinetics for multicomponent aluminum alloys

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Abstract

A mathematical model was developed to simulate the precipitation kinetics during heat treatment of multicomponent aluminum alloys. The model is based on the general numerical framework proposed by Kampmann and Wagner, and features a full coupling with CALPHAD software for the evaluation of the Gibbs–Thomson effect. It also does not rely on the assumption that precipitate phase composition is stoichiometric or uniform, and is therefore applicable for predicting complex precipitation kinetics encountered in industrial practices. Applications of the model to various aging treatments of binary Al–Sc alloys and a ternary Al–Sc–Zr alloy were conducted. It was found that the model predictions for extended time coarsening kinetics are in good agreement with the analytical Lifshitz–Slyozov–Wagner coarsening theory. Its ability to reproduce the complex precipitation pathways in multicomponent alloys was demonstrated by simulation of the precipitation kinetics for an Al–0.09 at.% Sc–0.03 at.% Zr alloy. Comparison of the simulation results with experimental measurement has also highlighted research directions that require further effort.

Keywords: Precipitation modeling; CALPHAD; Multicomponent; Aluminum alloys

1. Introduction and background

The precipitation of a second phase is an important strengthening technique for enhancing the mechanical properties of metallic materials. It has been widely used in commercial aluminum alloys, nickel alloys and steels. The microstructural features of precipitates with a strong influence on the final mechanical properties include their composition, fraction, number density, size distribution, morphologies and the solute level of alloying components in the matrix phase. The evolution of these microstructural features during heat treatment results from three concomitant processes: nucleation, growth and coarsening. There have been significant research efforts to develop numerical models based on classical nucleation, diffusion-controlled growth and coarsening theories to predict microstructure evolution during the entire precipitation process. Such a predictive numerical model was developed by Wagner and Kampmann [1], which is referred to as the KWN model. The essence of this model as well as its later variants is that the continuous particle size distribution (PSD) curve is subdivided into size classes, each of which is associated with a precipitate number. The temporal evolution of size distribution is then tracked by following the size evolution of each discrete size class. More recently, Maugis and Goune [2] extended the original KWN framework to multicomponent alloy systems. Myhr and Grong [3] took the same discretization idea in the original KWN model, but adopted a different strategy in tracking its temporal evolution. Instead of the size evolution of each class, Myhr and Grong tracked the evolution of the precipitate number associated with each class. This extended model has been used in various alloys systems with some variations related to the treatment of the Gibbs–Thompson effect and the

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numerical procedures used to solve the continuity equations [4–7].

In spite of the reported success, there are still challenges hindering the application of these models to multicomponent alloys. The first challenge identified is related to the evaluation of the Gibbs–Thomson effect for multicomponent alloys. The second results from the widely adopted assumption that the composition of the precipitates is purely stoichiometric. These two challenges are described in more detail below, and a KWN model is developed to handle these challenges.

The first challenge is reflected in the fact that research reported in the literature currently relies on one of the following approximations in accounting for the Gibbs–Thomson effect for multicomponent alloys: (i) pseudo-binary approximation [3]; (ii) extension of an analytical expression that is only applicable to binary alloys [8]; (iii) the solubility product method, which is only strictly valid for an ideal dilute solution model [2,5,6].

These approximations bring uncertainties and errors to the model predictions. These uncertainties escalate as the number of alloying components in the alloy increases. A solution to this problem is to use the achievements in the CALculation PHAse Diagram (CALPHAD) community. CALPHAD software such as Thermo-Calc can be adopted to evaluate the Gibbs–Thomson effect for multicomponent alloys dynamically during the simulation, and the only adjustment one has to make to the general phase diagram calculation procedure is increasing the molar Gibbs energy of the spherical-shaped precipitate phase by $2\gamma\kappa V_m$ due to the presence of the matrix–precipitate interface. This treatment was employed by Chen and co-workers [9]. However, Chen et al. applied their treatment only to simulating the precipitation of a single precipitate, and unacceptable computational cost might have prohibited the application of their treatment within the KWN framework. It is the first aim of this work to propose an alternative solution to the coupling of CALPHAD software with the KWN model. The combination of CALPHAD software with a precipitation kinetic model is mutually beneficial. It would allow precipitation models to access those validated thermodynamic models so that they can be applied to various alloy systems without having to develop new thermodynamic models or make over-simplified approximations. It also exposes the thermodynamic databases developed in the CALPHAD research community to the experimental measurement of precipitation states. Therefore, the possibility exists that the equilibrium thermodynamic database can be improved by the experimentally validated kinetic simulations [10].

The second challenge with the reported KWM models emerges when one tries to simulate the heat treatment involving the evolution of the precipitate composition. One example is the precipitation of the ALPHA phase (an Al–Fe–Mn–Si quaternary phase) from AA3003 alloy during a homogenization heat treatment. Both experimental measurement with energy dispersive spectroscopy [11] and the commercial databases FTlite[1] and TTAL6[2] indicate that this precipitate phase is not a pure stoichiometric phase. Another example is the precipitation of carbides in in-service power plant steels. The composition of the carbide has been used as "a built-in recorder" for the effective service temperature [12]. With the stoichiometric approximation in place, it is not possible to model these commonly observed precipitation phenomena where precipitate composition evolves with time. This approximation also excludes the possibility of constructing a model that is fully coupled to a thermodynamic database, owing to their inconsistency on precipitate composition. Removing this assumption for multicomponent alloys increases the mathematical complexity dramatically, as indicated by Kamp et al. [7] and evidenced by the various approximate treatments reported in Refs. [2,5,6]. In particular, one numerical complexity that has received little attention is that the average matrix composition cannot be calculated by solving the global mass balance equation alone. This is because the precipitate composition is not known a priori, and it has to be solved together with all the other equations governing interfacial matrix compositions and average matrix compositions.

The aims of the present paper are to address these two challenges outlined above and develop a fully CALPHAD-coupled multicomponent precipitation kinetics model. To validate the model and demonstrate its abilities, the simulation results on the aging treatment of various binary Al–Sc alloys and ternary Al–Sc–Zr alloy are compared with analytical theory and some reported experimental results. Comparisons with the experimental measurements demonstrate the model's ability to predict complex precipitation pathways in multicomponent alloys. The paper also highlights areas that require further research effort.

2. Mathematical model

Following the original KWN model [1], a fully CALPHAD-coupled model is developed to deal with the precipitation kinetics of non-stoichiometric precipitates in multicomponent alloys. Mathematically, it consists of three sets of equations, which are derived in Section 2.1 without invoking the stoichiometric assumption. They are also written in the form that is suitable for coupling to the CALPHAD software. The nucleation model and the numerical solution are described in Sections 2.2 and 2.3, respectively.

Before proceeding to the specific model descriptions, it is useful to summarize the assumptions adopted in the model: (i) precipitates are of spherical shape and their growth/dissolution is solely controlled by diffusion in the matrix;

[1] The FTlite–FACT light metal database (formerly FSlite) is developed by C.R.C.T., Ecole Polytechnique de Montreal, Canada.

[2] TTAL6 is the Al-based alloys database developed by Thermotech, Surrey, UK.