Available online at www.sciencedirect.com

ScienceDirect

Computational Materials Science 39 (2007) 871–879

COMPUTATIONAL MATERIALS SCIENCE

www.elsevier.com/locate/commatsci

Modeling the microstructural evolution of Ni-base superalloys by phase field method combined with CALPHAD and CVM

J.C. Wang [a,b,*], M. Osawa [b], T. Yokokawa [b], H. Harada [b], M. Enomoto [c]

[a] *State Key Laboratory of Solidification Processing, Northwestern Polytechnical University, Xi'an 710072, Shaanxi Province, PR China*
[b] *High Temperature Materials 21 Project, National Institute for Materials Science, 1-2-1 Sengen, Tsukuba Science City, Ibaraki 305-0047, Japan*
[c] *Department of Materials Science, Ibaraki University, 4-12-1, Nakanarusawa, Hitachi 316-8511, Japan*

Received 16 June 2006; received in revised form 8 October 2006; accepted 11 October 2006

Abstract

A quantitative simulation with phase field method (PFM) in a real alloy system was performed with a new strategy of modeling the microstructure evolution of Ni-base superalloys, in which, the calculation of phase diagrams (CALPHAD) method and cluster variation method (CVM) are combined with the PFM. In this strategy, the four sub-lattice model is used to evaluate the chemical free energy density while CVM to calculate some parameters, such as the lattice misfit and gradient coefficients in phase field equations. With this strategy, the microstructure evolution of a Ni–Al binary alloy at the temperature of 1000 K has been simulated; the elastic energy due to the lattice misfit and elastic inhomogeneity between γ and γ' phases has also been taken into account. Moreover, the directional coarsening phenomenon of Ni–Al alloy has been reoccurred when the various external stress conditions are applied, which shows good agreement with Pineau's directional coarsening map.

Keywords: Ni-base alloys; Phase field modeling; CALPHAD; CVM

---

1. Introduction

As one of the most important classes of high-temperature structural materials, Ni-base superalloy exhibits a truly unique combination of properties at elevated temperatures. The microstructures in a Ni-base superalloy typically are a two-phase mixture of γ and γ' phases, i.e., the disordered fcc phase (Ni solid solution) and the ordered $L1_2$ phase (Ni_3Al intermetallic compound), respectively. Precipitation hardening by the ordered phase is the major strengthening mechanism for Ni-base superalloys. Therefore, the microstructure control of γ phase and γ' phase is the key technique in the development of advanced Ni-base superalloys. However, since the factors that influence the microstructure formation are extensive, which increases the difficulty and complexity of controlling the microstructure, many trial-and-error experiments are often necessary for searching the best combination of desired microstructures and material properties. One obvious choice for overcoming this shortcoming is to use simulation for modeling the microstructural evolution.

Phase field method (PFM) is a newly developed and extremely powerful tool for the simulation and prediction of complex microstructure evolution, which has been demonstrated in quite a lot of areas, such as dendrite growth, spinodal decomposition, grain growth and so on [1]. As to the order–disorder phase transformation, especially for the γ–γ' in the Ni-base superalloys, the PFM has numerous advantages, as it ensures that many factors are taken into account, such as the coherency strain, elastic anisotropy, external stress, and $L1_2$ ordering of the precipitate phase. These advantages make it very convenient to study the microstructural evolution and the coarsening behavior of

---

[*] Corresponding author. Address: State Key Laboratory of Solidification Processing, Northwestern Polytechnical University, Xi'an 710072, Shaanxi Province, PR China.

E-mail address: jchwang@nwpu.edu.cn (J.C. Wang).

0927-0256/$ - see front matter © 2006 Elsevier B.V. All rights reserved.
doi:10.1016/j.commatsci.2006.10.014

the precipitates in a Ni-base superalloy. Therefore, many studies have been done in predicting the microstructure evolution of Ni-base superalloys with phase field method ever since the first phase field model for precipitation coarsening was proposed [2–9].

However, although tremendous success in this area has been achieved, additional progress in applying this method in a real alloy system is further required. The simulation with real parameters is highly anticipated by materials scientists. Quantitative simulation by coupling real thermodynamic databases and kinetic databases with PFM has recently been receiving more and more attention [7–9]. There are two typical ways to couple a thermodynamic or kinetic database with the phase field model. One is to use a thermodynamic model to directly calculate the free chemical energy, as was done by Zhu et al. [7] who coupled the sub-lattice model with the PFM to study the precipitate shape evolution in a Ni-base superalloy. In PFM, however, the free energy form for different phases should be a uniform style. As a result, only a few thermodynamic models (such as the sub-lattice model) can be directly used in this way. Another way is to combine PFM with some thermodynamic software (such as THERMO-CALC) so that the free energy and some kinetic coefficient can be determined. Grafe et al. [8] have used such a method to study the Ostwald-ripening behavior of a Ni-base by accessing THERMO-CALC through its user–programming interface. In addition, Chen et al. [9] performed a quantitative study modeling precipitate growth and dissolution with inputs from a CALPHAD thermodynamic database and a DICTRA kinetic database. Success has been achieved with both methods. However, there does still exist some room for improvement with quantitative modeling. For an instance, some parameters are difficult to obtain regardless of whether thermodynamic or kinetic databases being used in the experiments. The parameters included the lattice misfit and the gradient coefficients of interface energy, which are very important for the simulation but not easily measured in experiments. It is difficult to obtain highly accurate results, especially, the gradient coefficients of the energy. Fortunately, the cluster variation method (CVM) may provide a solution. CVM, one of the most reliable tools used to provide detailed information of the atomic arrangements of a given alloy, can be used to calculate not only the free energy variations with long-range or short-range order parameters, the equilibrium profile of composition and order parameters, but also some of the parameters that are needed in the PFM simulations with high accuracy, such as the lattice misfit and the interface energy. Kikuchi et al. [10] have used the CVM to calculate the inter-phase boundary (IPB) and the anti-phase boundary (APB) energies with multi-atom interactions, in which a super cell with many layers was used. Recently, Asta et al. [11–13] used the CVM to predict the interface properties of Al–Li and Al–Ag alloys, which also demonstrated that the CVM could be used to calculate the interface energy. Therefore, in the present paper, a quantitative simulation will be performed with a new strategy of quantitative simulation, which combines PFM, CALPHAD, and CVM to study the dynamics and morphological evolution of γ and γ' phases in a real Ni–Al binary alloy system.

2. Models

2.1. Phase field model

In the present study, the phase field model proposed by Wang et al. [1] is adopted. In the model, the composition field and three long-range order (LRO) parameter fields have been used to describe the concentration profile and the $L1_2$ order phase structure. The spatial–temporal evolution of the LRO parameters can be obtained by solving the time-dependent Ginzburg–Landau equations, while the temporal evolution of composition field can be described by the non-linear Cahn–Hilliard diffusion equation, which are:

$$\frac{\partial \eta_i}{\partial t} = -L\frac{\delta F}{\delta \eta_i}, \quad i = 1,2,3, \tag{1}$$

$$\frac{\partial c}{\partial t} = M\nabla^2 \frac{\delta F}{\delta c}, \tag{2}$$

where L and M are the structural relaxation and diffusion mobility respectively, c, the composition filed, η_i, the order parameter field, which can describe the four different domains in the $L1_2$ phase structure, and F, the total free energy of the system, which includes the chemical free energy, interface energy, and elastic energy, i.e., $F = F_{\text{chemical}} + F_{\text{interface}} + F_{\text{elastic}}$. The interface energy can be introduced through the gradients of the field variables in the diffuse interface phase field model. According to the Cahn–Hilliard diffuse interface energy model [14], the interface energy i can be written as:

$$F_{\text{interface}} = \int_V \left(\Delta f(c,\eta_1,\eta_2,\eta_3) + \frac{\alpha}{2}(\nabla c)^2 + \sum_{i=1}^{3} \frac{\beta_i}{2}(\nabla \eta_i)^2 \right) \mathrm{d}V, \tag{3}$$

where $\Delta f(c,\eta_1,\eta_2,\eta_3)$ is the free energy of the homogeneous alloy with respect to the free energy of the equilibrium two-phase mixture. And α and β_i are gradient energy coefficients, which can be determined from the total interface energy. The composition-dependent elastic strain energy F_{elastic} due to elastic inhomogeneity can be solved with the method of high-order approximation proposed by Hu et al. [15].

2.2. Chemical free energy calculated with the sub-lattice model (CALPHAD)

Obtaining reasonable simulation results with PFM requires the free energy calculation to be highly emphasized because the free energy of the system, as a function of some order parameters, will influence the driving force and path