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Modeling precipitate growth in multicomponent alloy systems by a variational principle

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Abstract

A physically based state variable model is proposed to simulate precipitate growth encountered in many industry-relevant precipitation processes. The model is derived from the same variational principle used in deriving the phase field method, and is able to efficiently model diffusion-controlled precipitate growth in multicomponent alloy systems. One of the new features of the model is its treatment of the propagation of the diffusion boundary layer surrounding a growing precipitate. The model was applied to simulate spherical precipitate growth in Al–Mg–Si, Ni–Al–Ti and Fe–C alloy systems, and the simulation results were verified by a direct detailed finite volume-based approach. Compared with the approximate growth models that have been reported, e.g., the invariant field model, the SFFK model and the model by Chen et al., the proposed model's predictions are much closer to the direct detailed approach, especially for growth under high supersaturation conditions. It is concluded that the proposed model is valuable and could be embedded into a multiscale modeling framework to deal with concurrent nucleation, growth, coarsening and macroscopic transportation. This work is also a successful case study demonstrating the potential offered by the variational principle for multiscale microstructural evolution modeling.

Keywords: Precipitate growth model; Multi-component alloys; Thermodynamic extremum principle; Variational principle; Diffusion-controlled phase transformation

1. Introduction

Physically based state variable methods are more suitable for simulating many industry-relevant precipitation processes than direct detailed numerical approaches represented by the popular phase field method [1]. Examples include modeling the up-quenching of AA6060 alloys [2], the formation of a dispersoids-free zone during homogenization heat treatment of AA3XXX alloys [3], the coarsening of gamma prime precipitates during heat treatment of multicomponent Ni-based superalloys [4] and the computation of time–temperature–precipitation diagrams of industrial alloys [5]. A common feature of these problems is concurrent nucleation, growth and coarsening of a large number of precipitates, which may be accompanied by larger-scale macroscopic transportation phenomena (diffusion, heat transfer). The precipitation can be addressed in an adequate manner by the physically based mean field model pioneered by Wagner and Kampmann [6]. The mean field precipitation model's embedded precipitate growth model is the key to its success.

The most widely used embedded precipitate growth model is that obtained from invariant field approximation [2,5,7]. However, the invariant field model (IFM) is in principle only applicable to the growth driven by vanishing supersaturation [8]. Another interesting alternative was recently proposed by Chen et al. [9]. Chen's model has better accuracy than IFM owing to the introduction of an extra parameter called "effective diffusion distance". Although Chen et al. highlighted the necessity of introducing the size parameter to treat precipitate growth, their model derivation is based on the ad hoc extension of an

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existing analytical solution to the isolated binary precipitate growth problem.

A rigorous way of introducing the diffusion distance parameter is to employ the variational principle called the "thermodynamic extremum principle" or the "Onsager maximum entropy production principle". It is necessary to give a brief introduction of this principle before discussing its use in precipitation modeling. This principle has attracted much research interest during the past 15 years for microstructural modeling [10–14]. Carter et al. [11] pointed out that it helps to provide general principles for microstructural evolution. They and other researchers [15] demonstrated that the Allen–Cahn equation and Cahn–Hillard equations could be directly derived from this principle, using the principle as the basis for the popular phase field method. The variational principle could also be used in deriving physically based internal state variable models [1]. Hillert and Ågren [12] envisioned its great value in irreversible thermodynamics by drawing an analogy to the powerful and successful Hamilton's extremum principle in classical mechanics. Svoboda and co-workers [14] pioneered its use and demonstrated its versatility in many interesting applications. It is clear from Svoboda and co-workers' work on diffusion-related studies [16,17] that the variational principle is able to integrate many interacting processes, such as vacancy diffusion, substitutional diffusion, interstitial diffusion, non-ideal generation and annihilation of vacancies, bulk diffusion and grain boundary diffusion, into a single framework, therefore enabling the applications of these fundamental diffusional phase transformation knowledge gained from carefully designed experimental studies into industrial process modeling.

The use of the variational principle in deriving an efficient precipitate growth model is due to Svoboda et al. (SFFK) [18,19]. Kozeschnik [20], one of the SFFK model developers, showed that the SFFK model yields a higher growth rate than an analytical solution by a factor of 2 in the region of low supersaturation and by a factor of 5 for high supersaturation. Since precipitate volume fraction is proportional to the cube of average precipitate radius, SFFK's higher growth rate leads to much larger deviations of precipitate fraction, i.e., by a factor of 8 and 125 in the low and high supersaturation region, respectively.[1] The deviation, as pointed out by Chen et al. [9], was attributed to the fact that the SFFK model employed even fewer independent state parameters than IFM in describing the diffusion profile in front of the migrating interface. Chen et al. proposed a new treatment of the diffusion during precipitate growth for multicomponent alloys by introducing a parameter, called "effective diffusion distance". The parameter characterizes the thickness of the diffusion boundary layer, and plays an important role in determining the precipitate growth rate. Indeed, together with interface friction and cross-interface diffusion, it is an essential physical mechanism involved in general phase transformation [21]. However, as mentioned above, Chen et al. introduced this parameter in an ad hoc way, i.e., by generalizing the analytical expression obtained in isolated growth to interrupted growth. A natural question arises here: what would happen if one introduced the "effective diffusion distance" parameter via the variational principle? This paper shows that it would lead to a more rigorous and physically sound precipitate model.

In addition to the treatment of diffusion, the other important aspect of a multicomponent precipitate growth model is contact conditions at the migrating precipitate–matrix interface. Although it was reported that contact conditions could be derived from the variational principle [22], the practical use of such a treatment is unwarranted, as the value of a key input parameter, the interface mobility, is not known with an acceptable degree of accuracy. In addition, the derivation did not consider other factors (other than the interface friction) that cause deviation from the local equilibrium, such as the trans-interface diffusion and low diffusivity in the parent phase [23]. In view of the success of the direct prescription of the local equilibrium contact condition in modeling industry-relevant diffusion-controlled phase transformation [24], the present authors set out to explore its combination with the variational principle for the treatment of precipitate growth. The goal of the study is to derive the model and demonstrate its advantages over the three reference approaches, i.e., the IFM, the SFFK model and Chen's model [9]. The model is applied to Al–Mg–Si, Ni–Al–Ti and Fe–C alloy systems, and the simulation results are verified by a direct detailed finite volume-based approach, which is the most accurate (but much more computationally expensive) solution. The implications of the proposed model for further understanding of diffusion-controlled phase transformation are discussed.

2. Model descriptions

As well summarized and demonstrated by Svoboda and co-workers [16], for the diffusion concerned in this paper, i.e., the diffusion in single-phase multicomponent crystalline system, there are three different categories: (i) the vacancy mechanism for "slowly" diffusing substitutional components; (ii) the "quick" diffusion of interstitial components; and (iii) the existence of non-ideal sources and sinks for vacancies. Although their different attributes lead to different constraints in their mathematical treatment, all three categories could be treated by the variational principle. For the sake of simplicity and the nature of precipitate growth encountered in the investigated alloy systems, the study is restricted to diffusion in dilute substitutional (Al–Mg–Si and Ni–Al–Ti) and interstitial alloys (Fe–C) with ideal sources and sinks for vacancies. A common feature

[1] The case Kozeschnik selected for his model evaluation is the growth of an isolated precipitate, whose analytical solution had been derived. But it does not represent the typical scenario in which the SFFK model is intended to apply. SFFK model will be evaluated in this paper for interrupted growth.