Acta Materialia 129 (2017) 229–238

Contents lists available at ScienceDirect

Acta Materialia

journal homepage: www.elsevier.com/locate/actamat

Full length article

Determining material parameters using phase-field simulations and experiments

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ARTICLE INFO

Article history:
Received 7 November 2016
Received in revised form
23 January 2017
Accepted 20 February 2017
Available online 22 February 2017

Keywords:
Phase-field method
X-ray tomography
Coarsening
Al alloys
Temporal evolution

ABSTRACT

A method to determine material parameters by comparing the evolution of experimentally determined 3D microstructures to simulated 3D microstructures is proposed. The temporal evolution of a dendritic solid-liquid mixture is acquired in situ using x-ray tomography. Using a time step from these data as an initial condition in a phase-field simulation, the computed structure is compared to that measured experimentally at a later time. An optimization technique is used to find the material parameters that yield the best match of the simulated microstructure to the measured microstructure in a global manner. The proposed method is used to determine the liquid diffusion coefficient in an isothermal Al-Cu alloy. However, the method developed is broadly applicable to other experiments in which the evolution of the three-dimensional microstructure is determined in situ. We also discuss methods to describe the local variation of the best-fit parameters and the fidelity of the fitting. We find a liquid diffusion coefficient that is different from that measured using directional solidification.

1. Introduction

Computational methods play an important role in accelerating the discovery and development of advanced materials [1]. One of the most promising areas in which computational methods are employed is in Integrated Computational Materials Engineering (ICME), which is receiving increased attention from both academia and industry [2,3]. The establishment of reliable and comprehensive materials databases - the main component of the Materials Genome Initiative (MGI) [3] - is a key to the success of ICME [2,4]. Traditionally, material parameters are measured one at a time by designing dedicated experiments using idealized specimens and specimen geometries (e.g. a planar interface in a diffusion couple experiment for measuring the diffusion coefficient). However, such procedures are often tedious, and typically parameters are measured only in a fraction of the relevant phase space, which may involve materials composition, temperature, pressure, etc. In addition, the idealized geometry may not be representative: industrially relevant microstructures are heterogeneous and artificial surfaces may introduce unwanted boundary effects. Furthermore, for hierarchically ordered materials, effects on different length scales compete and interact. Recently, researchers have begun to calculate material parameters from first-principles, such as the free energy [5] and the diffusion coefficients in the solid phase [6–8] and the liquid phase [9]. However, experimental verification of the calculated material parameters under realistic conditions is needed.

In this work, we propose to determine material parameters directly from structural studies of bulk samples acquired during synthesis or processing. To image material microstructure evolution, various techniques have been used, e.g. Computed Tomography (CT) [10,11], 3D X-Ray Diffraction (3DXRD) [12] and Diffraction Contrast Tomography (DCT) [13]. Using x-rays emitted from a synchrotron source, time-resolved high spatial resolution 3D images can be acquired using tomographic methods, for a review see Ref. [14]. In favorable cases, the temporal resolution may be on the sub-second scale [15]. Some of these techniques are increasingly becoming available in laboratory sources, such as the laboratory-based DCT (labDCT) [16]. At the same time, the rapid increase in computing power and the development of advanced modeling

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http://dx.doi.org/10.1016/j.actamat.2017.02.056

techniques such as quantitative phase-field models [17–20], accurate simulations of microstructure evolution in 3D have become feasible. Therefore, we propose to determine material parameters by direct comparison between the 3D temporal evolution of microstructures determined through experiment and phase-field simulation. We claim that the parameter values that provide the best match between the experimental and the simulated microstructure in a global manner (both in 3D space and in time) correspond to the physically correct ones. The proposed method can be used to verify the calculated material parameters by first-principles and multiscale modeling simulations. Another advantage of this approach is that it permits the measurement of multiple - in some cases potentially all relevant - material parameters from one experiment in a realistic environment. Notice that though this paper focuses on the phase-field method, other modeling techniques relevant to the problem studied can also be used, such as Monte Carlo Potts model [21] and the vertex model [22] for grain growth and the level-set method for solidification [23].

In recent years, several direct comparisons between experiment and phase-field simulations have been performed [24–27], but the comparisons have mainly been qualitative or based on average quantities, such as the average particle size and the interface area per unit volume. Rigorous comparisons of the morphologies are rare. McKenna et al. [25] used a one-to-one comparison to test a grain growth phase-field model, but they did not use it for extracting material parameters. Demirel et al. [28] used a similar approach for grain growth in thin films. Aagesen et al. [24] estimated a value of the liquid diffusion coefficient using a comparison between phase-field simulations and tomography in a heuristic manner. We here introduce a general optimization formalism and discuss key aspects of this fitting approach, such as the cost functions to quantify the similarity between experiment and simulation, the accuracy, the initial and boundary conditions and the computational speed. To the best of our knowledge, this is the first systematic study where phase-field simulations and 3D tomography are combined to extract material properties. Though in general the optimization relies on performing phase-field simulations many times, we predict that one may only need to consider a small fraction of space-time in a given step of the optimization for many relevant problems.

We demonstrate the approach by fitting the liquid diffusion coefficient D^{L} and the capillary length l^{L} in the context of the isothermal coarsening of dendrites in a liquid of composition nearly equal to that of the eutectic composition in the Al-Cu system. It is a well-studied system, and relevant material parameters have been extensively measured by traditional means, e.g. the free energy [29,30], the solid/liquid interfacial energy [31,32] and the liquid diffusion coefficient [33–35]. However, the values determined from the liquid diffusion coefficient measurements display a large scatter in value, argued to be mainly due to convection [33]. Moreover, an existing temperature gradient during directional solidification may alter the measured liquid diffusion coefficient. In section 2, the fitting methodology is presented in detail. In section 3, the results of the demonstration on the Al-Cu system are provided. We discuss limitations and potential applications in section 4 and conclude the paper in section 5.

2. Optimization approach

Initially, we present the mathematical model and the associated terminology and notations. Then two types of cost functions and several ways to define the fitting domain are proposed and compared. Finally, the statistics of the fitting method is discussed. Throughout, for reasons of simplicity, we shall assume a two-phase problem, where the microstructure is characterized by a moving boundary between the two phases.

2.1. The mathematical model

The fitting approach is shown schematically in Fig. 1. Here the symbol $\mathscr{G}$ represents the geometry of the material microstructure. The x-ray experiment provides a series of 3D material microstructures $\mathscr{G}^{\mathrm{exp}}(t)$ evolving with time (shown in the upper solid box in Fig. 1). With one frame of the experimental microstructure (time t_0) as input ($\mathscr{G}^{\mathrm{sim}}(t_0) = \mathscr{G}^{\mathrm{exp}}(t_0)$) and a guess of material parameters $\mathbf{p}$, the simulation method [19] can produce a series of evolving microstructures $\mathscr{G}^{\mathrm{sim}}(t, \mathbf{p})$ (shown in the lower dashed box in Fig. 1). For time $t > t_0$, a cost function f_{cost} is used to measure the dissimilarity between the two microstructures. We claim the real material parameters $\mathbf{p}^{\mathrm{real}}$ should give the least dissimilarity between the experimental and simulated microstructures, i.e. f_{cost} reaches a minimum as shown in Fig. 1 (right).

This fitting process can be described by the following optimization problem:

$$\begin{aligned} &\text{find} && \mathbf{p} \\ &\text{minimize} && f_{\mathrm{cost}}(t, \mathbf{p}) = f_{\mathrm{cost}}\Big(\mathscr{G}^{\mathrm{exp}}(t), \mathscr{G}^{\mathrm{sim}}(t, \mathbf{p})\Big) \\ &\text{such that} && \mathscr{G}^{\mathrm{sim}}(t, \mathbf{p}) \text{ fulfills phase-field equation} \\ & && \mathscr{G}^{\mathrm{sim}}(t_0, \mathbf{p}) = \mathscr{G}^{\mathrm{exp}}(t_0) \\ & && \mathscr{G}^{\mathrm{sim}}(t, \mathbf{p}) \text{ fulfills boundary condition} \end{aligned} \tag{1}$$

The optimization problem can be solved by any appropriate optimization algorithm. Notice here the optimization approach is independent of the geometric representation $\mathscr{G}$, which may thus be discretized like a binary image or be continuous like NURBS (explicit) [36] and level-set methods (implicit) [37]. The flowchart of the fitting algorithm is shown in Fig. 2.

2.2. The cost function

Two types of cost functions are proposed based on the representation of the microstructure geometry. If these microstructures are represented by binary images ($\mathscr{G}^{\mathrm{exp}} = \mathrm{Img}^{\mathrm{exp}}$, $\mathscr{G}^{\mathrm{sim}} = \mathrm{Img}^{\mathrm{sim}}$), the correlation function can be used to construct the cost function (the corr-cost function)

$$f_{\mathrm{cost}}(t, \mathbf{p}) = 1 - \underset{\Omega_{\mathrm{fit}}}{\mathrm{corr}}\Big(\mathrm{Img}^{\mathrm{exp}}(t), \mathrm{Img}^{\mathrm{sim}}(t, \mathbf{p})\Big) \tag{2}$$

where Ω_{fit} is the fitting domain. If a continuous geometry representation like the signed distance function as known from the level-set method is used ($\mathscr{G}^{\mathrm{exp}} = \phi^{\mathrm{exp}}$, $\mathscr{G}^{\mathrm{sim}} = \phi^{\mathrm{sim}}$), the squared 2-norm function can be used:

$$f_{\mathrm{cost}}(t, \mathbf{p}) = \frac{\left\|\phi^{\mathrm{sim}}(t, \mathbf{p}) - \phi^{\mathrm{exp}}(t)\right\|^2_{2,\Omega_{\mathrm{fit}}}}{\left\|\phi^{\mathrm{exp}}(t) - \phi^{\mathrm{exp}}(t_0)\right\|^2_{2,\Omega_{\mathrm{fit}}}} \tag{3}$$

Here, the normalization is used to make the cost function independent of the fitting domain size. By this definition, $\sqrt{f_{\mathrm{cost}}}$ has a physical meaning, namely representing the root mean square migration distance of the simulated interfaces relative to the experimentally determined interfaces if no topological change occurs. As the segmentation applied to the tomographic data in the example case given in the current work is based on the signed distance function [38], ϕ^{exp} is available. However, ϕ^{sim} is not directly available from the phase-field simulation. In this work, the equilibrium profile of a planar interface is used to provide an approximation of the signed distance function from the interpolation function in the phase-field model, and then a reinitialization