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Acta Materialia 61 (2013) 3370–3377

Acta MATERIALIA

www.elsevier.com/locate/actamat

Modeling and predicting microstructure evolution in lead/tin alloy via correlation functions and stochastic material reconstruction

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Received 26 December 2012; received in revised form 17 February 2013; accepted 17 February 2013
Available online 7 March 2013

Abstract

The binary lead/tin (Pb/Sn) alloy is widely used as an interconnect in microelectronics. The physical properties of this heterogeneous material critically depend on its complex bulk microstructure containing Pb-rich and Sn-rich phases, which can be both laminar and globular. In this paper, we devise a procedure to model and predict the microstructure evolution (i.e. coarsening) in a Pb–Sn alloy aged at elevated temperatures below its melting point using statistical morphological descriptors, i.e. the two-point correlation functions S_2 associated with the phases. We verify via phase-field simulations that the growing length scale characterizing microstructure coarsening can be well captured by the corresponding correlation functions, which enables us to predict the S_2 of intermediate microstructures given the initial and final microstructures. Stochastic material reconstruction techniques are employed to generate virtual three-dimensional microstructures that are consistent with the predicted correlation functions, which are quantitatively compared with the actual alloy microstructures when available.

Keywords: Microstructure evolution; Heterogeneous materials; Correlation functions; Stochastic microstructure reconstructions

1. Introduction

The binary lead/tin alloy, a two-phase heterogeneous material, has been studied for decades due to its importance in electronic packaging applications. The physical properties of such a heterogeneous material are determined by the associated properties of individual constituent phases as well as the complex morphologies of these phases – the material microstructure – which can be quantitatively characterized by certain statistical morphological descriptors, i.e. the spatial correlation functions of the alloy phases [1–3].

Binary lead/tin alloys are widely used as solders [4]. In particular, a eutectic alloy of 63% tin (Sn), 37% lead (Pb) has been used as an interconnect due to its unique low melting point (183 °C), good wettability and excellent mechanical properties. The eutectic microstructure contains a Pb-rich phase and a Sn-rich phase, which can possess both laminar and globular morphologies. The salient microstructural features such as the width and extent of the laminar phases as well as the size and spatial distribution of the globular phases can significantly affect the overall mechanical properties of the alloy [5–7].

At temperatures below the eutectic melting point, the enhanced diffusion of Pb and Sn atoms can lead to significant coarsening in the alloy, which lowers the total interfacial energy [8]. This coarsening process can be accurately modeled by the Cahn–Hilliard equation [9,10]. A heat treatment (e.g. annealing) can then be employed to "tune" the eutectic microstructure to achieve desirable material performance. On the other hand, coarsening could also produce an

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1359-6454/$36.00
http://dx.doi.org/10.1016/j.actamat.2013.02.026

undesirable degradation of material properties such as loss of strength or the disappearance of grain-boundary pinning effects. Moreover, the rate of coarsening increases with temperature and it is of particular importance to quantitatively model and predict the degree of coarsening in the design of materials for high temperature applications.

Phase-field modeling techniques, which simulate the microstructure evolution according to the Cahn–Hilliard equation (see Section 3.1), are widely used to study coarsening and phase separation in binary systems [11–15]. Specifically, an appropriate length scale λ associated with the evolving microstructure can be defined, which satisfies the following scaling relation for the coarsening processes that are entirely diffusion controlled (i.e. diffusion is the slowest process) [8]:

$$\lambda(t)^3 - \lambda_0^3 \sim t \tag{1}$$

where λ_0 is the length scale associated with the initial microstructure and t is the time duration of the coarsening process. It was first established for the distribution of grain size in a supercooled system that had undergone Ostwald ripening [16]. For systems that are not diffusion controlled (e.g. the association and disassociation of atoms are slower than their diffusion), the exponent associated with the characteristic length scale would be different. Using phase-field simulations, it has been verified that for systems evolving according to the Cahn–Hilliard equation, the associated λ satisfies Eq. (1). It is noteworthy that there is no unique choice of λ. In other words, a variety of different characteristic length scales satisfying Eq. (1) can be chosen for the binary system of interest, which are mutually consistent with one another. For example, for alloys containing particle-like phases, the effective particle radius can be used as λ; while for alloys with laminar phases, the volume-to-surface ratio of the phases can be a natural choice for λ.

Although phase-field modeling has proven to be very powerful and successful in many aspects, it is difficult to directly use simulated microstructures to quantitatively model the ones obtained experimentally. This is because the spatial correlations of the initial density fluctuations leading to phase separation and coarsening in the system are generally not known a priori. Gaussian random fields are often employed to approximate the actual density profiles [13], which might not be sufficient to capture certain subtle initial correlations in the system that persist and are manifested in the coarsened microstructures.

In this paper, we devise a methodology to model and predict microstructure evolution in Pb–Sn alloy aged at elevated temperatures below its melting point, using the two-point correlation function S_2 (defined below) associated with either Pb-rich or Sn-rich phases and stochastic material reconstruction techniques [17]. In particular, using phase-field simulations, we show that a growing length scale $\lambda(t)$ is well defined via the S_2 associated with the microstructure of interest. This in turn enables us to predict the two-point correlation function of intermediate microstructures given the initial and final microstructures. We extract such length scales by analyzing two-dimensional (2-D) images of Pb37Sn63 alloy samples that were isothermally aged at 175 °C up to 216 h and construct a general functional form of S_2 that, when appropriately parameterized with $\lambda(t)$, can accurately approximate and predict the corresponding S_2 of microstructures aged for different time periods. Stochastic material reconstruction techniques are then employed to generate virtual three-dimensional (3-D) microstructures consistent with the predicted S_2, which are quantitatively compared with the experimentally obtained material morphology to verify the accuracy of our approach. Our procedure is complementary to the phase-field modeling approaches. The rest of the paper is organized as follows. In Section 2, we present mathematical definitions of the statistical microstructural descriptors employed in the paper, including the two-point correlation functions S_2 and the lineal-path function L. In Section 3, we describe in detail our procedure for modeling and predicting microstructure evolution in Pb–Sn alloy using S_2 and the stochastic reconstruction techniques, which are used to verify the accuracy of our models and predictions. In Section 4, we offer concluding remarks.

2. Statistical morphological descriptors

In general, the microstructure of a two-phase heterogeneous material (e.g. a binary alloy) can be uniquely determined by specifying the indicator functions associated with each individual phase [1], i.e.

$$I^{(i)}(\mathbf{x}) = \begin{cases} 1 & \mathbf{x} \text{ in phase } i \\ 0 & \text{otherwise} \end{cases} \tag{2}$$

where $i = 1$ and 2. The volume fraction of phase i is then given by

$$\varphi_i = \langle I^{(i)}(\mathbf{x}) \rangle \tag{3}$$

where $\langle \cdot \rangle$ denotes the ensemble average over many independent material samples or volume average over a single large sample if it is spatially "ergodic" [1,2]. The two-point correlation function $S_2^{(ij)}(\mathbf{x}_1, \mathbf{x}_2)$ associated with phases i and j is defined as

$$S_2^{(ij)}(\mathbf{x}_1, \mathbf{x}_2) = \langle I^{(i)}(\mathbf{x}_1) I^{(j)}(\mathbf{x}_2) \rangle \tag{4}$$

which is also the probability that two randomly selected points $\mathbf{x}_1$ and $\mathbf{x}_2$ fall into phase i and j respectively. For a binary heterogeneous material, there are in total three different S_2, including two autocorrelation functions ($i = j$) and one cross-correlation function ($i \neq j$). However, it has been shown that only one of them is independent [1,18] and the remaining two functions can be explicitly expressed in terms of the independent one. Thus, it is sufficient for us to consider the S_2 of either the Pb-rich phase or the Sn-rich phase. In the subsequent discussions, we will only employ S_2 associated with the Pb-rich phase