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Coupled diffusion-deformation multiphase field model for elastoplastic materials applied to the growth of Cu_6Sn_5

Johan Hektor [a, *], Matti Ristinmaa [a], Håkan Hallberg [a], Stephen A. Hall [a, c], Srinivasan Iyengar [b]

[a] Division of Solid Mechanics, Lund University, Box 118, 221 00 Lund, Sweden
[b] Division of Materials Engineering, Lund University, Box 118, 221 00 Lund, Sweden
[c] European Spallation Source, Box 176, 221 00 Lund, Sweden

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ABSTRACT

A coupled diffusion-deformation, multiphase field model for elastoplastic materials is presented. The equations governing the evolution of the phase fields and the molar concentration field are derived in a thermodynamically consistent way using microforce balance laws. As an example of its capabilities, the model is used to study the growth of the intermetallic compound (IMC) Cu_6Sn_5 during room-temperature aging. This IMC is of great importance in, e.g., soldering of electronic components. The model accounts for grain boundary diffusion between IMC grains and plastic deformation of the microstructure. A plasticity model with hardening, based on an evolving dislocation density, is used for the Cu and Sn phases. Results from the numerical simulations suggest that the thickness of the IMC layer increases linearly with time and that the morphology of the IMC gradually changes from scallop-like to planar, consistent with previous experimental findings. The model predicts that plastic deformation occurs in both the Cu and the Sn layers. Furthermore, the mean value of the biaxial stress in the Sn layer is found to saturate at a level of −8 MPa to −10 MPa during aging. This is in good agreement with experimental data.

1. Introduction

Over the past years phase field modeling has evolved into a powerful tool for computational materials science. One advantage of phase field models is that there is no need to explicitly track the position of interfaces during microstructural evolution. Instead, the position of the interfaces is implicitly given by the evolution of the phase field variables used to describe the microstructure. This makes it possible to simulate complex polycrystalline microstructures without making assumptions on, e.g., the shape of the grains [1]. Phase field models have been used to study, for example, solidification processes and dendritic growth [2,3], recrystallization [4], and martensitic phase-transformation [5]. Another phenomena to which phase field modeling is well adapted is growth of intermetallic compounds (IMC). The focus of the present work is the IMC in the Cu–Sn system. These IMC are of great importance due to their role in soldering of electronic components. The recent transition to lead-free solders, driven by environmental concerns and legislation, have further increased the interest for the Cu–Sn system, both from experimental [6–13] and modeling [14–18] perspectives.

During a soldering process, a layer of the intermetallic compound Cu_6Sn_5 (η-phase) will form at the interface between the Cu substrate and the liquid solder [9]. A small amount of IMC is necessary to achieve sufficient bonding between the substrate and the solder. However, in the presence of too much IMC, the mechanical properties of the solder joint will degrade due to the brittleness of the intermetallic phase [19]. The thickness of the IMC will increase during aging and the growth of the Cu_6Sn_5 phase will introduce stresses in the surrounding microstructure [8]. These stresses are believed to be responsible for another reliability concern in the electronics industry, namely the growth of tin whiskers [20].

The formation and growth of intermetallic compounds in the Cu–Sn system has been previously studied using the phase field method. A one-dimensional model of the growth of Cu_6Sn_5 was

* Corresponding author.
E-mail address: johan.hektor@solid.lth.se (J. Hektor).

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developed by Umantsev in Ref. [17]. In this model the phase fields represent ordering and crystallization rather than individual grains. Huh et al. [14] developed a model for simulating growth of Cu_6Sn_5 between a Cu substrate and molten solder. Based on the model by Huh and coworkers, several additional studies have been presented, e.g. Refs. [15,16]. None of these models, however, take the mechanical behavior of the material into account. In Ref. [18], the stress build-up caused by the growth of IMC is simulated using the finite element method. The model in Ref. [18] is uncoupled and only deformation is taken into account while the growth of IMC is added based on curve fitting of experimental data.

In this paper, we present a coupled diffusion-deformation, multiphase field model capable of simulating both the growth of the Cu_6Sn_5 phase and the associated build-up of stresses. The equations governing the evolution of the simulated microstructure are derived in a thermodynamically consistent way using the concept of microforces [21,22] and the constitutive framework developed by Ammar et al. [23]. The model is used to study the growth behavior of Cu_6Sn_5 during isothermal aging at room temperature.

The paper is structured in the following way: In Section 2 the multiphase field model is derived, starting from a dissipation inequality and a postulated free energy. In Section 3 and Section 4 numerical aspects of the model and the choice of model parameters are discussed. Results from the simulations are presented in Section 5. The paper is closed with some concluding remarks in Section 6.

2. Multiphase field model

In multiphase field models a polycrystal microstructure is represented by a set of non-conserved phase fields $\boldsymbol{\phi} = (\phi_a(t,\mathbf{X}),\phi_b(t,\mathbf{X}),\ldots,\phi_n(t,\mathbf{X}))$, where each phase field represents one grain in the microstructure. The phase fields are functions of time, t, and spatial coordinates, $\mathbf{X}$. Grain boundaries are taken as the regions where two or more phase field variables vary smoothly between 0 and 1. The smooth variation occurs over some distance, creating a diffuse interface region. Based on the properties of each phase present at the interface, the material properties of the interface region can be estimated using the interpolation function introduced in Ref. [15]:

$$h_i(\boldsymbol{\phi}) = \frac{\phi_i^2}{\sum_j \phi_j^2}, \tag{1}$$

where the sum in the denominator is taken over all phase fields, i.e. $j = 1 \ldots n$.

To simulate diffusion, conserved field variables $\boldsymbol{x} = (x_a(t,\boldsymbol{X}),x_b(t,\boldsymbol{X}), \ldots ,x_n(t,\boldsymbol{X}))$, representing the molar fraction in each grain is used. The molar fraction fields are related to the global molar concentration field c through

$$c = \sum_n h_n \frac{x_n}{V_m}, \tag{2}$$

where the interpolation function h_i in (1) is used to interpolate between the molar fraction field corresponding to each phase field and where V_m is a constant molar volume.

The derivation of the equations governing the evolution of the phase fields and the concentration field is presented below. The derivation is based on the framework developed by Ammar et al. [23]. In the present work, this framework is extended to a multiphase setting by making use of the interpolation function (1) and by formulating the free energy and other quantities as summations over all phase fields. Following [21] and [22], a system of microforces is associated with each phase field. These forces represent configurational forces acting on the crystal lattice. The microforce system belonging to phase field a comprises an internal microstress vector ξ_a and a scalar microforce π_a, as well as an external microforce γ_a. The microforces in the other phases are defined analogously. In the same way as the Cauchy stress $\boldsymbol{\sigma}$ is energy-conjugated with the gradient of the displacement $\boldsymbol{\nabla u}$, π_a and ξ_a are energy-conjugated with ϕ_a and $\boldsymbol{\nabla}\phi_a$, respectively. Each microforce system is presumed to follow a balance law, cf. [21], of the form

$$\boldsymbol{\nabla}\cdot\xi_i + \pi_i + \gamma_i = 0. \tag{3}$$

The term microforce system is motivated by (3) having the same form as the equilibrium equation for the Cauchy stress,

$$\boldsymbol{\nabla}\cdot\boldsymbol{\sigma} + \boldsymbol{b} = \boldsymbol{0}, \tag{4}$$

where $\boldsymbol{b}$ denotes the body force vector.

2.1. Dissipation inequality

Following the procedure in Ref. [23], with the enhancement that the power densities are formulated as sums over all phase fields, the first and second laws of thermodynamics result in a Clausius-Duhem inequality,

$$-\sum_i \pi_i\dot{\phi}_i + \sum_i \xi_i^T\boldsymbol{\nabla}\dot{\phi}_i + \boldsymbol{\sigma}:\dot{\boldsymbol{\varepsilon}} - \dot{f} + \mu\dot{c} - \boldsymbol{J}^T\boldsymbol{\nabla}\mu \geq 0, \tag{5}$$

where $\boldsymbol{\varepsilon} = \boldsymbol{\varepsilon}^e + \boldsymbol{\varepsilon}^* + \boldsymbol{\varepsilon}^p$ is the total strain, consisting of elastic strains $\boldsymbol{\varepsilon}^e$, transformation strains $\boldsymbol{\varepsilon}^*$, and plastic strains $\boldsymbol{\varepsilon}^p$. In (5), a tensorial contraction over two indices is denoted by $(\cdot):(\cdot)$. The chemical potential and the diffusion flux are denoted μ and $\boldsymbol{J}$ respectively and a superposed dot denotes differentiation with respect to time. The free energy density $f = f(\boldsymbol{\phi},\nabla\boldsymbol{\phi},c,\boldsymbol{\varepsilon}^e,\boldsymbol{\kappa})$ is taken as a function of the set of phase fields $\boldsymbol{\phi}$, their gradients $\nabla\boldsymbol{\phi}$, the global concentration field c, the elastic strain $\boldsymbol{\varepsilon}^e$ and a set of internal variables $\boldsymbol{\kappa}$ related to plasticity in terms of the evolving dislocation density, to be specified later on. Using the chain rule to calculate the time derivative of f and inserting it into (5) gives

$$\begin{aligned} &-\sum_i\left(\pi_i + \frac{\partial f}{\partial\phi_i}\right)\dot{\phi}_i + \sum_i\left(\xi_i - \frac{\partial f}{\partial\boldsymbol{\nabla}\phi_i}\right)^T\boldsymbol{\nabla}\dot{\phi}_i + \left(\mu - \frac{\partial f}{\partial c}\right)\dot{c} \\ &+\left(\boldsymbol{\sigma} - \frac{\partial f}{\partial\boldsymbol{\varepsilon}^e}\right):\dot{\boldsymbol{\varepsilon}}^e + \boldsymbol{\sigma}:\dot{\boldsymbol{\varepsilon}}^p - \boldsymbol{J}^T\boldsymbol{\nabla}\mu - \sum_i\frac{\partial f}{\partial\kappa_i}\dot{\kappa}_i \geq 0. \end{aligned} \tag{6}$$

To ensure that the second law of thermodynamics is fulfilled, (6) should hold for any combination of $\boldsymbol{\phi},\boldsymbol{\nabla\phi},c,\boldsymbol{\varepsilon}^e$, and $\boldsymbol{\kappa}$. We can therefore extract state laws for the internal microstress, the chemical potential and the Cauchy stress:

$$\xi_i = \frac{\partial f}{\partial\boldsymbol{\nabla}\phi_i}, \quad \mu = \frac{\partial f}{\partial c}, \quad \boldsymbol{\sigma} = \frac{\partial f}{\partial\boldsymbol{\varepsilon}^e}. \tag{7}$$

Inserting the state laws into (6), results in the dissipation inequality

$$D = -\sum_i \pi_i^{dis}\dot{\phi}_i - \boldsymbol{J}^T\boldsymbol{\nabla}\mu + \boldsymbol{\sigma}:\dot{\boldsymbol{\varepsilon}}^p - \sum_i K_i\dot{\kappa_i} \geq 0, \tag{8}$$

where $\pi_i^{dis} = \pi_i + \frac{\partial f}{\partial\phi_i}$ and $K_i = \partial f/\partial\kappa_i$. From the dissipation inequality it is possible to identify three dissipative processes. The first term represents the phase field dissipation, which is related to the rearrangement of atoms during the evolution of the phase fields [23]. The second term is the mass transport caused by diffusion and