



Physical vapor deposition of multiphase materials with phase nucleation via a coupled phase-field approach



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ABSTRACT

A coupled phase-field model is presented for simulating physical vapor deposition (PVD) of multi-phase materials, including the effects of phase nucleation. This model is utilized to study the role of initial substrate phase and temperature distributions, which are important experimental deposition parameters, on PVD of a generic allotropic metal with two stable phases. PVD simulations are performed for the deposition of a high temperature phase below its phase transition temperature, and for bicrystal and amorphous substrates with a Gaussian temperature distribution. Several general observations are made from these simulations. During the initial stages of PVD, the substrate phase distribution acts as a template for the growing phases. As the thin film continues to grow, the release of latent heat due to deposition creates a temperature gradient within the film, i.e., regions near the film surface become hotter than near the substrate. Additionally, a substrate with a defined temperature distribution that encompasses temperatures above and below the phase transition temperature, allows for distinguishable regions within the thin film where different phases are stable and continue to grow. Three-dimensional simulations provide additional insight into the role of substrate temperature and phase distribution on the resulting microstructure with different flux rates.

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1. Introduction

Considerable research has been devoted to thin films due to their application in optoelectronic and microelectronic devices, nanoelectromechanical systems, and protective coatings, among others. The microstructure of a thin film may be amorphous or consist of nanometer to micron-sized regions that differ in phase (i.e., crystal structure), orientation, and chemical composition. The exact details of the surface morphology and subsurface structure generally influence its mechanical, electrical, and chemical properties.

Thin films are typically grown using vapor deposition techniques where the surface morphology and underlying microstructure (e.g., phase distribution, grain size, etc.) are a function of the deposition conditions and materials used [1–9]. During vapor deposition in some materials, multiple stable or metastable phases may nucleate and coarsen that will enhance or diminish the desired thin film properties (cf. [10–14]). As such, to elucidate links between vapor deposition processing conditions and thin film

phase distribution or evolution, a predictive model must specifically consider solid-state phase transformations and phase nucleation in addition to critical aspects of thin film growth for the chosen deposition technique [9,15–17].

The objective of this work is to present a “first treatment” phase-field model to simulate simultaneous thin film growth and subsurface phase-evolution in a multiphase thin film with isotropic growth kinetics. Physical vapor deposition (PVD) is the growth process considered in this work because the composition of the depositing target material is conserved during transport to the substrate, and physical processes dominate over chemical reactions during vapor transport and thin film growth [2,3]. Therefore, chemical reactions and composition changes can be neglected in a phase-field model for multiphase PVD (provided the thin film is composed of a single element or a compound where all of the phases have the same stoichiometry, e.g., Al_2O_3). Within the PVD process, the critical aspects of thin film growth to be considered are surface tension, diffusion, non-local shadowing effects, and re-emission [1–3,18]. To the authors’ knowledge, there has been no attempt in the literature to address concurrently thin film growth, subsurface phase evolution, and phase nucleation during PVD of multiphase materials.

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Recently, Stewart and Spearot [19,20] developed two phase-field models for simulating PVD of a single-phase polycrystalline material. In that work, a traditional single free energy functional approach for describing the energetics of a PVD thin film system was compared to a coupled approach that treated the PVD growth process as being separate from the subsurface grain evolution process (i.e., different free energy expressions for each process). It was shown that the coupled phase-field approach is sufficient for determining qualitative descriptions of microstructure evolution during PVD (e.g., the influence of surface features on GB migration) which can act as a guide for developing more complex models, while the single free energy approach is necessary for a physically consistent description of these inherently connected PVD process mechanisms. In addition to prior work by the current authors, this work leverages previous modeling efforts on PVD growth dynamics [18], evolution in multiphase materials [21], and phase nucleation [15,16], to incorporate the relevant aspects of the PVD growth process and subsurface phase evolution into a coupled phase-field model.

The proposed phase-field model is applied to simulate PVD of a generic allotropic metal with two stable phases and isotropic growth kinetics. These simulations allow for a study of initial substrate phase distribution and substrate temperature on the resulting phase distribution within the thin film. The deposition conditions simulated in this work are chosen to highlight the utility of the coupled PVD phase-field model in capturing general features of thin film growth with subsurface nucleation and evolution of multiple phases.

2. Simulation methodology

In this section, the major components for modeling PVD, multiphase evolution, and classical phase nucleation within a coupled phase-field simulation framework are presented; full analyses of the motivating literature can be found in [15,16,18,21]. The formalism developed in this work to sequentially couple PVD thin film growth and subsurface phase evolution for modeling both processes is also presented.

2.1. Physical vapor deposition

To capture arbitrary surface morphology formation, surface tension and diffusion, and nonlocal shadowing effects during PVD, this work utilizes the interfacial growth model of Koblinski et al. [18], which was also used in [19,20] for capturing PVD processes of a single-phase material. To model vapor deposition, two field variables are introduced: $f(\mathbf{r}, t)$ and $g(\mathbf{r}, t)$. The first variable, $f(\mathbf{r}, t)$, describes the growing thin film where $f(\mathbf{r}, t) \approx 1$ defines a solid region, $f(\mathbf{r}, t) \approx -1$ defines a vapor region, and $f(\mathbf{r}, t) \approx 0$ defines interfaces. The second variable, $g(\mathbf{r}, t)$, describes the incident vapor density where $g(\mathbf{r}, t) \approx 0$ defines a region of no incident flux and $g(\mathbf{r}, t) > 0$ defines the local density of incident vapor being transported to the thin film surface. The time evolution of these field variables is given in the following coupled non-dimensional equations of motion,

$$\frac{\partial f}{\partial t} = \nabla^2 \left(-f + f^3 - 2a(\nabla^2 f) \right) + B(\nabla f)^2 g + C\sqrt{(\nabla f)^2} \eta \quad (1)$$

$$\frac{\partial g}{\partial t} = \nabla[D\nabla g - \mathbf{A}g] - B(\nabla f)^2 g \quad (2)$$

Eq. (1) describes the growth and evolution of the solid, $f(\mathbf{r}, t)$, at the expense of the vapor, $g(\mathbf{r}, t)$, while Eq. (2) describes the evolution of the incident vapor, $g(\mathbf{r}, t)$. The first term in Eq. (1) is formally the Cahn-Hilliard dynamics where the variational derivative of the double well free energy functional for this model

has been calculated, providing an energy barrier between the vacuum and solid phases. This term allows the formation of arbitrary surface morphologies and accounts for both surface and bulk diffusion during thin film growth, where a is the surface diffusion coefficient. The second term, coupling Eqs. (1) and (2), serves as the source term that leads to growth of the thin film at the expense of incident vapor, where B controls the rate of vapor-to-solid aggregation. The last term provides thermal fluctuations following an uncorrelated Gaussian distribution, $\eta(\mathbf{r}, t)$, where the parameter, C , controls the overall strength of the noise. In Eq. (2), the first term is the diffusion equation modified for the presence of an external force, \mathbf{A} , where D is the diffusion coefficient and \mathbf{A} is the strength and direction to the incident vapor flux. The second term, opposite the second term in Eq. (1), is the sink that removes vapor in regions that have been converted to solid. Finally, an additional parameter, b , not explicitly included in Eqs. (1) and (2) is used to prevent solid growth in regions away from the interface in the vacuum region (i.e., $f(\mathbf{r}, t) < b$) and is constrained to the interval $-1 < b < 0$.

2.2. Multiphase evolution

The description of multiple phases within the thin film is motivated by the phase-field model for multiphase systems developed by Steinbach et al. [21], which can be used to quantitatively model solid-solid and solid-liquid transformations. In later work, Steinbach et al. [22] extended this model to account more accurately for the conservation of interfacial stress. In this multiphase model, each of the N phases are assigned a unique field variable, $p_i(\mathbf{r}, t)$, that varies between 0 and 1. These variables correspond to the volume fraction of each phase at a given location, thus allowing for different phases to be modeled and distinguished. Therefore, at any given location in a N phase system, the following constraint must be satisfied,

$$\sum_{i=1}^N p_i(\mathbf{r}, t) = 1 \quad (3)$$

A free energy functional for an N phase system is constructed by considering all pairwise interactions, thus providing kinetic and potential energy terms that are dependent on the local $p_i(\mathbf{r}, t)$ values, their gradients, and temperature. These kinetic and potential energy terms capture bulk and interfacial energies and their differences for phases present in the local volume; defining the energy barriers for phase transformations. Since phase transformations are non-conserved phenomena, the equations of motion utilizing a free energy functional of this type are determined by Allen-Cahn dynamics, giving the following set of equations,

$$\frac{\partial p_i}{\partial t} = \sum_{k \neq i}^N \frac{1}{\tau_{ik}} \left[\varepsilon_{ik}^2 (p_k \nabla^2 p_i - p_i \nabla^2 p_k) - \frac{p_i p_k}{2a_{ik}} (p_k - p_i - 2m_{ik}) \right] \quad (4)$$

The parameters in Eq. (4) are as follows: τ_{ik} is a kinetic coefficient, ε_{ik}^2 defines the numerical interface thickness, a_{ik} is a positive constant, and m_{ik} is the coefficient for deviation from thermodynamic equilibrium that provides the local driving force as a function of temperature. These numerical parameters are related to the following physical thermodynamic quantities,

$$\tau_{ik} = \frac{L_{ik} \lambda_{ik}}{T_{ik} \mu_{ik}} \quad \varepsilon_{ik}^2 = \lambda_{ik} \sigma_{ik} \quad a_{ik} = \frac{\lambda_{ik}}{72 \sigma_{ik}} \quad m_{ik} = \frac{6a_{ik} L_{ik} (T_{ik} - T)}{T_{ik}} \quad (5)$$

where L_{ik} is the latent heat released or consumed during the i - k phase transformation, λ_{ik} is the i - k interface thickness, T_{ik} is the temperature at which the i - k phase transformation takes place, μ_{ik} is the i - k interface mobility, and σ_{ik} is the i - k interfacial energy. All of these parameters depend on the phases that comprise the

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