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Analysis of particle engulfment during the growth of crystalline silicon

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ABSTRACT

To better understand the physical mechanisms responsible for foreign inclusions during the growth of crystalline silicon, steady-state and dynamic models are developed to simulate the engulfment of solid particles by solidification fronts. A Galerkin finite element method is developed to accurately represent forces and interfacial phenomena previously inaccessible by approaches using analytical approximations. The steady-state model is able to evaluate critical engulfment velocities, which are further validated using the dynamic model. When compared with experimental results for the SiC-Si system, our model predicts a more realistic scaling of critical velocity with particle size than that predicted by prior theories. Discrepancies between model predictions and experimental results for larger particles are posited to arise from dynamic effects, a topic worthy of future attention.

1. Introduction

When casting multicrystalline silicon (mc-Si) ingots for solar cells, the silicon melt usually contains a certain concentration of carbon and nitrogen, which both possess relatively low segregation coefficients in silicon [1]. During directional solidification, these two elements will accumulate to supersaturation levels in the liquid silicon and precipitate to form solid particles of silicon carbide (SiC) and silicon nitride (Si_3N_4). Depending upon solidification conditions, these foreign particles in the melt can either be pushed or be engulfed by the advancing solidification front. The engulfed particles, which eventually become inclusions in mc-Si ingots, can lead to lower cell efficiency, wafer breakage and sawing defects. Even worse, when slicing the ingots into wafers, the sawing wire can be broken by these extremely hard inclusions [2], which significantly increases costs.

There is a long history and substantial research on the analysis of particle engulfment during solidification, because this topic is also important in other physical processes, such as fabrication of metal-matrix composites, separation processes, cryogenic preservation of biological materials, and frost heaving. Pioneering studies on modeling the particle engulfment processes have been performed by Uhlmann et al. [3] and Chernov et al. [4,5], and particularly relevant research has been more recently put forth by Stefanescu et al. [6], Rempel and Worster [7,8], Park et al. [9], and Garvin et al. [10,11].

Only a few researchers have specifically focused on the solidification of silicon, due to the difficulties of high temperatures (greater than 1687 K) and small particle sizes (typically ranging between 1 and 400 μm). Among those limited studies on the silicon system, Søiland et al. [12], Trempa et al. [13], and Reimann et al. [14] investigated the occurrence of SiC and Si_3N_4 inclusions by analyzing ingot samples from directional solidification. Recently, Jauss et al. [15] and Azizi et al. [16] have aimed to quantitatively determine engulfment conditions via experiments using a silicon floating zone system pre-seeded with a distribution of SiC particles and grown under microgravity and terrestrial conditions. These studies of inclusions during silicon crystal growth have pointed out that classical theoretical models of engulfment do a poor job of explaining experimental observations. Therefore, we desire to develop new approaches to model particle engulfment and to further our understanding of this complicated phenomenon in this system.

2. Model description

Page limitations prevent an extensive discussion of the engulfment model; rather, we highlight its most important features in the following section. An extended discussion of model development and validation is presented in [17] for interested readers.

A solid particle interacting with a solidification front is depicted in Fig. 1, where a typical example of system geometry and melt flows is depicted on the right, along with quantities defined in the model. The crystal solidifies towards the particle with a growth

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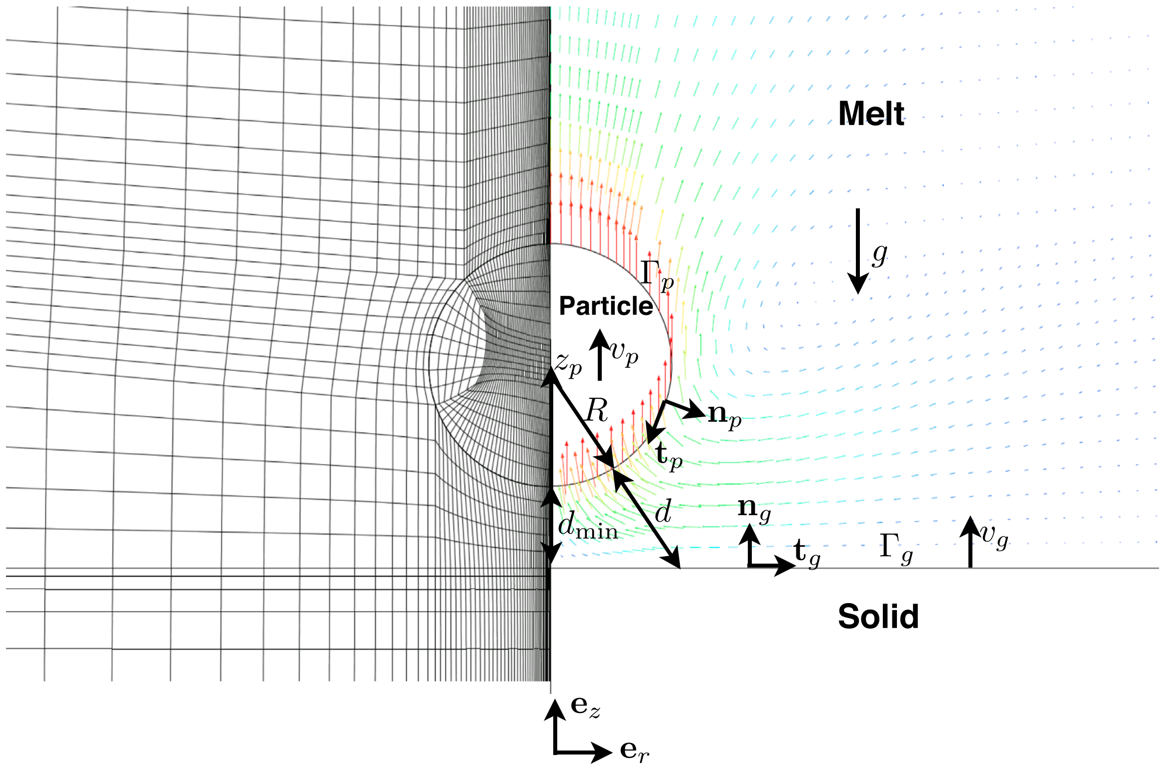

Fig. 1. Schematic of our particle engulfment model. Left side: a representative portion of the computational mesh of biquadratic elements. *Right side*: a typical flow field and some quantities used in the model.

rate, v_g, and when the gap between particle and melt-solid interface decreases (to one micron or less for typical systems of interest), repulsive van der Waals forces [19] push the particle away from the interface, imparting a particle velocity, v_p. Due to the upward particle motion, liquid must flow into the gap to fill the volume previously occupied by the particle, which results in drag forces on the particle. If the drag forces—which hinder the particle from moving together with the advancing solid-liquid interface—dominate, the particle will be engulfed into crystal. Thus, a force balance analysis is the pivotal step in modeling particle engulfment.

In the model, we consider only motion in the *z*-direction and apply Newton's second law, $ma = F$, to describe the time-dependent motion of the particle, written in a detailed form:

$$\rho_p V_p \frac{d^2 z_p}{dt^2} = -\rho_p V_p g - \frac{2AR^3}{3d_{\min}^2 (2R + d_{\min})^2} + \int_{\Gamma_p} \mathbf{e}_z \cdot (\mathbf{n}_p \cdot \mathsf{T})\, dS. \qquad (1)$$

On the left-hand side, the particle density, ρ_p, and volume, V_p, constitute the mass, m, and $d^2 z_p/dt^2$ is the particle acceleration, a. The net force in Newton's law, F, is explicitly computed via the terms on the right-hand-side of the above equation, where g denotes the gravitational constant, A represents Hamaker's constant, T represents the total stress tensor for flow in the melt, and other variables are defined in Fig. 1. These three terms describe the gravitational force, the repulsive van der Waals force, and hydrodynamic forces acting on the particle, respectively. We note that the representation of van der Waals forces is particularly important; we provide an extensive discussion in [17].

To compute the hydrodynamic forces in Eq. (1), the incompressible Navier–Stokes equations and continuity equation are applied in the melt domain to describe the velocity and pressure fields. Theoretical considerations suggest that continuum mechanics accurately describe liquid flows though channels of 1–2 nm in lateral dimension [18], so we expect our approach to be valid for this system, where typically $d_{\min} \geq \mathcal{O}(10)$ nm.

The temperature field is needed to understand the effects of heat transfer on the solidification front, and a energy conservation equation is written over all domains. The location of the solidification front is determined via its temperature, T_i, which is given by [8]:

$$T_i = T_{mp}\left[1 - \left(\frac{\lambda}{d}\right)^3 - \frac{\sigma_{s\ell}\kappa}{\rho_s \Delta H_f}\right], \qquad (2)$$

where T_{mp} denotes the equilibrium melting point for a planar interface. The second and third terms on the right represent the melting point changes due to the premelting [7] and Gibbs–Thomson effects, respectively. Here λ is a length scale proportional to the van der Waals interaction strength [7], d is the gap thickness in Fig. 1, $\sigma_{s\ell}$ denotes the solid-liquid interfacial free energy, κ represents the mean interfacial curvature, and ΔH_f is the latent heat.

The above equations represent a dynamical system, and one approach for analysis is to directly integrate the time-dependent equations. However, another framework for analysis, indeed the approach taken by nearly all prior researchers, is to solve for steady states in the moving reference frame of the solidification front. For the steady-state model, all time derivatives of the above equations are formally set to zero. When the particle acceleration is set to zero, Eq. (1) becomes an implicit function of minimum gap thickness and growth rate, which we will denote as $f(d_{\min}, v_g) = 0$.

The above model consists of a coupled set of nonlinear equations that we choose to solve numerically via the Galerkin finite element method. We emphasize that our numerical solution of the underlying mathematical equations of this system allows for accurate representation of forces and interfacial phenomena previously inaccessible by classical approaches using analytical approximations [3–7,10,9]. A representative illustration of a mesh of biquadratic elements at the early stages of a transient computation is shown on the left of Fig. 1. Validations of computational

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