Journal of Crystal Growth #vol# (xxxx) xxxx–xxxx

Contents lists available at ScienceDirect

Journal of Crystal Growth

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

Particle engulfment dynamics under oscillating crystal growth conditions

Yutao Tao[a], Tina Sorgenfrei[b], Thomas Jauß[b], Arne Cröll[b], Christian Reimann[c], Jochen Friedrich[c], Jeffrey J. Derby[a,*]

[a] Department of Chemical Engineering and Materials Science, University of Minnesota, Minneapolis, MN 55455, USA
[b] University of Freiburg, Crystallography, Hermann-Herder-Straße 5, 79104 Freiburg, Germany
[c] Fraunhofer IISB, Schottkystraße 10, 91058 Erlangen, Germany

ARTICLE INFO

Communicated by S. Uda

Keywords:
A1. Computer simulation
A1. Fluid flows
A1. Heat transfer
A2. Particle engulfment
B2. Multicrystalline silicon
B3. Solar cells

ABSTRACT

To better understand the physical mechanisms behind particle engulfment dynamics under fluctuating solidification velocities, transient simulations are performed for a SiC particle in a silicon solidification system with oscillating growth rates using a rigorous finite-element model developed previously. Simulations reveal complicated behaviors that require a re-examination of the classical notion of a steady-state, critical growth velocity, v_c, for particle engulfment. Under sinusoidal growth variations at a frequency representative of turbulent fluctuations in a large-scale melt, stable pushing states featuring nonlinear particle-growth front oscillations can arise, even when the maximum growth velocity slightly exceeds v_c. However, higher-amplitude growth oscillations at the same frequency are shown to result in particle engulfment. Significantly, engulfment under such dynamic conditions can occur at average solidification rates far below the steady-state critical velocity, a behavior consistent with many experimental observations.

1. Introduction

During the crystallization of multicrystalline silicon (mc-Si) via directional solidification method, carbon, coming from the original silicon feed or from furnace parts, will inevitably accumulate in the silicon melt due to its relatively low segregation coefficient [1]. Silicon carbide (SiC) particles precipitate and grow as the concentration of this element surpasses supersaturation levels. While smaller particles may be pushed by the solidification front, many larger particles are engulfed, leading to inclusions in mc-Si ingots. The detrimental effects of these hard inclusions include lower cell efficiency, wafer breakage, sawing defects and even sawing wire breakage [2].

Recently, microgravity and terrestrial experiments conducted by the ParSiWal project [3–6] have attempted to quantitatively determine engulfment conditions using a silicon floating zone system pre-seeded with a distribution of SiC particles. The classical, steady-state analysis of Søiland et al. [7] was applied to predict the critical growth velocities at which particles of a certain size were engulfed, but experimental observations deviated considerably from theory. In particular, the model substantially over-predicted velocities needed for engulfment. In the experiments, engulfment was observed to occur at much smaller growth velocities.

Tao and Derby developed a rigorous numerical model for particle engulfment [8] and obtained much better agreement in describing the ParSiWal data [9]. However, this steady-state model also began to deviate from experiment for particle sizes of approximately 40 μm and larger, over-predicting the growth rate needed for their engulfment. While particle sedimentation effects caused by gravity provide a partial explanation for this discrepancy, these prior results motivate the question of what is responsible for engulfment of these larger particles at growth velocities less than predicted? Here, we pursue the idea that a single, critical velocity for the engulfment of a particle of a given size may be too idealized to describe many experimental conditions. Namely, we posit that dynamical effects may override the expected behaviors predicted by steady-state analysis.

We first note that video data from ParSiWal growth experiments indicated that solidification rates often fluctuated over periods of $O(10 \sim 100)$ seconds [4]; see, e.g., Fig. 1. We also note that turbulent flows in large-scale silicon melts also result in unavoidable oscillations in crystal growth rates due to temperature fluctuations with frequencies on the order of $0.1 \sim 1$ Hz [10,11]. We follow below with theoretical analyses that explicitly consider time-dependent growth rates and their effect on engulfment behavior in the SiC–Si system.

2. Model description

There is a vast body of prior work on the modeling of particle engulfment processes. Pioneering analyses were performed by

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[*] Corresponding author.
E-mail address: derby@umn.edu (J.J. Derby).

http://dx.doi.org/10.1016/j.jcrysgro.2016.10.049
Received 31 August 2016; Received in revised form 18 October 2016; Accepted 19 October 2016
Available online xxxx

Please cite this article as: Tao, Y., Journal of Crystal Growth (2016), http://dx.doi.org/10.1016/j.jcrysgro.2016.10.049

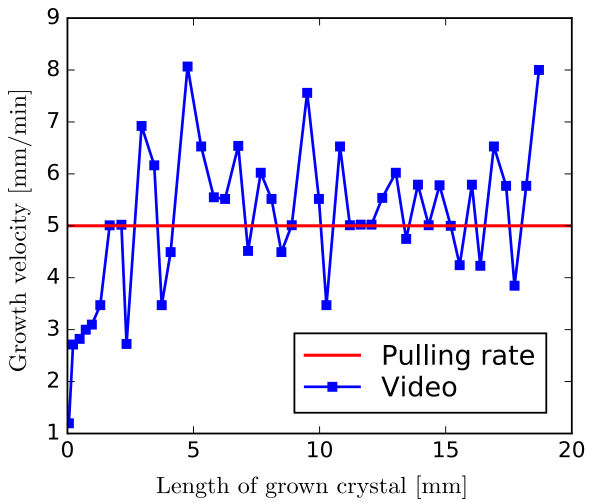

Fig. 1. In the ParSiWal experiments to measure SiC particle engulfment during the solidification of silicon, the growth rate, as measured by video, oscillates in time, even though the pulling rate is set to constant.
Adapted from [4].

Uhlmann, Chalmers, and Jackson [12] and Chernov, Temkin, and Mel'nikova [13,14], with more recent advances by Stefanescu et al. [15], Rempel and Worster [16,17], Park et al. [18], and Garvin et al. [19,20]. Only Garvin et al. [19,20] formulated engulfment as a time-dependent phenomenon; however, they considered just a few examples of dynamic behavior in a model ceramic-metal system. Our model, which is free from most of the simplifying assumptions that have been employed in previous approaches, has been formulated to describe both steady-state and dynamic behavior and is detailed in [8,9,21]. We highlight its most important features in the following discussion.

Fig. 2(a) shows the schematic of a typical particle-solidification front system, along with the quantities defined in our two-dimensional model. The particle is constrained to only move in the z-direction, and its dynamics are described by applying Newton's second law, $ma=F$, which is written in a detailed form:

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

On the left-hand side, the particle density, ρ_p, and volume, V_p, compute particle mass, m, and $\mathrm{d}^2 z_p/\mathrm{d}t^2$ is the axial particle acceleration, $\mathbf{a}_z$. On the right-hand side, the myriad forces acting on the particle are specified. The first term denotes the gravitational force, where g is the gravitational constant. The second term has the exact form of Hamaker force law [22], with A representing Hamaker's constant. This term computes the repulsive van der Waals force, which is the force that pushes the particle away from the interface, imparting the particle with velocity $v_p \equiv \mathrm{d}z_p/\mathrm{d}t$. As the particle is pushed, liquid must flow into the gap to fill the volume previously occupied by the particle, and this gives rise to a drag force, which impedes further motion of the particle. This hydrodynamic force is computed in the third term, where T represents the total stress tensor for flow in the melt. The particle pushing/engulfing transition is determined by the competition among these forces, thus accurate representation is particularly important; a more detailed discussion is presented in Refs. [8,21].

Computing the hydrodynamic forces in Eq. (1) requires solving for the flow in the melt domain. Therefore, the Navier–Stokes and continuity equations are applied in the model to describe the velocity and pressure fields. Simultaneously, the fluid flow is coupled with heat transfer by applying the energy conservation equation over all domains in the system. With the resultant temperature field, the solidification front is represented as a sharp interface, whose location is determined by its temperature, T_i,

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

where T_{mp} denotes the equilibrium melting point for a planar interface, λ is a length scale proportional to the interaction strength arising from van der Waals effects [16], d is the gap thickness in Fig. 2(a), $\sigma_{s\ell}$ denotes the solid-liquid interfacial free energy, κ represents the mean interfacial curvature, and ΔH_f is the latent heat. In Eq. (2), the second and third terms on the right represent the melting point changes due to the premelting [16] and Gibbs-Thomson effects, respectively.

While much of our prior work has considered steady states of this system, we focus here on cases where the solidification growth rate, and the accompanying system, is dependent upon time. We achieve an oscillating growth rate by specifying time-dependent thermal boundary conditions at the top and bottom of the model domains, simulating sinusoidally varying temperatures driven via strong melt flows.

The Galerkin finite element method and trapezoid rule are employed to solve the model equations numerically. Please refer to [8,21] for mesh refinement studies and validations of computational accuracy.

3. Results and discussion

For all of the computations presented here, we consider silicon solidifying upward in an applied vertical thermal gradient of 30 K/cm, with a 40 μm-diameter SiC particle in the melt. We consider gravity to be acting in the system, so there is a sedimentation force acting on the particle. All other physical parameters of the simulations can be found in [21]. For interpretation of ensuing figures, we define the particle velocity, v_p, and two velocities for the growth interface, one measured directly underneath the particle at the system centerline, $v_{g,0}$, and the other measured far away from the particle, denoted as $v_{g,\infty}$; see Fig. 2(a).

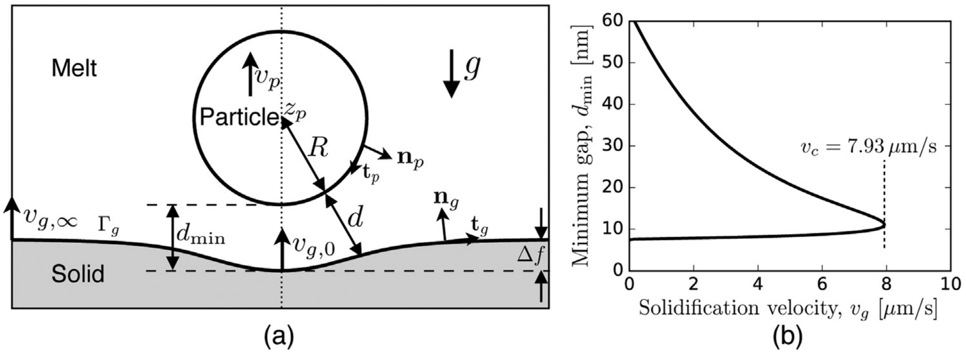

Fig. 2. (a) A depiction of particle engulfment by a solidification interface, with symbols used in paper. (b) Steady states of the SiC-Si system are plotted as a curve of minimum gap thickness, $d_{\min}$, versus applied solidification velocities, v_g. The upper branch of the curve represents stable steady states in which the particle is pushed ahead of the interface, while the bottom branch represents unstable steady states. The maximum extent of the curve establishes the critical velocity, v_c, for this system.