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Review

Fluid dynamics in crystal growth: The good, the bad, and the ugly

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Abstract

Fluid dynamics are important in processes that grow large crystals from a liquid phase. This paper presents a primer on fluid mechanics and convection, followed by a discussion of the physics and scaling of flows in such processes. Specific examples of fluid flows in crystal growth systems are presented and classified according to their impact on outcomes, good or bad. Turbulence in crystal growth is discussed within the limited extent of our understanding, which is incomplete, or ugly.

Keywords: fluid mechanics; crystal growth; convection; segregation; instabilities

1. Introduction

This paper is meant as an introduction toward understanding fluid dynamics and the effects that flow can bring about during the growth of large, single crystals. The title of this article is chosen in a shameless attempt to attract your attention by alluding to an epic film of the 1960s, directed by Sergio Leone and starring Clint Eastwood. However, this title also means to emphasize that flows in large-scale crystal growth systems are always important and that their effects can be beneficial (good) or detrimental (bad). Indeed, one motivation for their study is to understand their effects so that changes in process design or operation may produce better outcomes. What about the "ugly" in the title? Well, please continue reading to find out …

The literature on flows in crystal growth is vast, and no attempt will be made to present a comprehensive summary of it. Some recent reviews on this topic include those of Derby et al. [1], Kakimoto and Gao [2], Tsukada [3], Vizman [4], and Capper and Zharikov [5]. In this paper, we will first focus on some essentials on flows and their effects, followed by a series of examples of interesting and important flows in crystal growth systems. We will focus on the growth of single crystals from liquids, with examples drawn from both solution and melt growth systems. We will not consider the many, interesting complications that may arise in vapor crystal growth processes [6] or during the crystallization of many crystals in purification operations [7].

2. Background

2.1. A primer on fluid mechanics

While fluid mechanics can become horrendously complicated, it is important to remember that fluids follow the same, basic kinematic rules that every scientist or engineer learned long ago, namely, Newton's laws of motion. We write Newton's second law backwards (to make the next equation more clear) as,

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$$\underset{\text{mass}\times\text{acceleration}}{m\mathbf{a}} = \underset{\text{Net force}}{\mathbf{F}_{\text{net}}} \tag{1}$$

where the bold characters indicate vector quantities. This simple form is applicable to a rigid body. However, if we instead imagine a dollop of fluid upon which forces push and pull, we can carefully shrink it to an infinitesimal size and re-express the above expression in terms of the velocity of the fluid,

$$\underset{\substack{\text{mass per unit volume}\\ \times\text{ acceleration}}}{\rho\left(\frac{\partial\mathbf{v}}{\partial t}+\mathbf{v}\cdot\nabla\mathbf{v}\right)} = \underset{\substack{\text{pressure forces}\\ \text{per unit volume}}}{-\nabla p} + \underset{\substack{\text{viscous forces}\\ \text{per unit volume}}}{\mu\nabla^2\mathbf{v}} + \underset{\substack{\text{buoyant force}\\ \text{per unit volume}}}{\rho_0\mathbf{g}\left[1-\beta(T-T_0)\right]} + \underset{\substack{\text{applied body force}\\ \text{per unit volume}}}{\mathbf{F}(\mathbf{v},\mathbf{x},t)}, \tag{2}$$

where the correspondence between eqs. (1) and (2) should be readily apparent. To define our nomenclature, ρ is the density of the fluid, $\mathbf{v}$ is the velocity field, t is time, ∇ is the gradient operator representing derivatives over spatial dimensions $\mathbf{x}$, p is the pressure field, μ is the fluid viscosity, $\mathbf{g}$ is the gravitational vector, β is the thermal expansivity, T is temperature, and the subscript zero denotes the reference state about which the linear dependence of fluid density on temperature is approximated.

The first two terms on the right-hand-side of eq. (2) represent different components of the net force acting among fluid elements. The pressure field, represented by the variable p, transmits forces acting *normal* to an element, while viscosity transmits forces via *shear*, i.e., momentum transferred by fluid sliding over adjacent elements. This expression arises from the work of Newton, who postulated that shear stresses are linearly proportional to velocity gradients. Fluids that obey this constitutive relation are referred to as Newtonian fluids.

The terms on the following line are *body forces* that act over the volume of the fluid. Gravity alone results in a hydrostatic pressure field that varies in elevation, whereas buoyant forces arise from gravity acting over density differences, represented here by the term involving $\beta(T-T_0)$. Buoyancy acts as a lever arm, whereby the net force arises from density differences perpendicular to the direction of gravity. The expression shown in eq. (2) arises from the Boussinesq approximation, which represents changes in density as a linear function of temperature (or composition, which is not shown above). The last term, $\mathbf{F}(\mathbf{v},\mathbf{x},t)$, is a catch-all for additional body forces that may act on the fluid. Some particularly useful outcomes can arise from forces of this type, such as Lorentz effects from the application of a magnetic field to a conducting fluid.

In the process of derivation of the previous equation, we made an additional, important assumption that the fluid itself is incompressible, i.e., that its density does not change appreciably with pressure, an assumption that is extremely good for a liquid and sometimes reasonable for a gas. While the above application of Newton's second law to a fluid manifests itself in the conservation of momentum, we must specify an additional constraint to guarantee continuity, i.e., the conservation of mass. This is written for an incompressible fluid as,

$$\nabla\cdot\mathbf{v}=0. \tag{3}$$

Collectively, eqns. (2) and (3) constitute the celebrated Navier–Stokes equations.

2.2. Convection – the effects of fluid flow

A flowing liquid typically has a significant effect on local temperature and composition via *convection*, or *advection* in some fields. Convection is the transport of heat (thermal energy) and material (species) by flow. This is readily seen in the conservation equations derived for the temperature,

$$\underset{\substack{\text{accumulation of}\\ \text{thermal energy}\\ \text{per unit volume}}}{\rho C_p\frac{\partial T}{\partial t}} + \underset{\substack{\text{convective transport}\\ \text{of thermal energy}\\ \text{through unit volume}}}{\rho C_p\mathbf{v}\cdot\nabla T} = \underset{\substack{\text{conductive transport}\\ \text{of thermal energy}\\ \text{through unit volume}}}{\kappa\nabla^2 T}, \tag{4}$$

where C_p is the heat capacity and κ is the thermal conductivity of the fluid. A more extensive discussion of heat transfer in melt crystal growth is provided in [8]. A similar equation is written for species conservation,

$$\underset{\substack{\text{accumulation of}\\ \text{a dilute species}\\ \text{per unit volume}}}{\frac{\partial c}{\partial t}} + \underset{\substack{\text{convective transport}\\ \text{of a dilute species}\\ \text{through unit volume}}}{\mathbf{v}\cdot\nabla c} = \underset{\substack{\text{diusive transport}\\ \text{of a dilute species}\\ \text{through unit volume}}}{\mathcal{D}\nabla^2 c}, \tag{5}$$

where c denotes species concentration and $\mathcal{D}$ is the diffusion coefficient of the species in the fluid.

Fluid dynamics in crystal growth systems are most often important not because of the momentum carried by the flow but because of the effects of convection. Specifically, fluid flows modify heat and species transport in extremely important ways, and several examples will be discussed later in this paper.

2.3. Understanding through scaling

Finding analytical solutions to the Navier–Stokes equations is extremely challenging, and such solutions are generally available only for very simple systems [9].