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Analysis of the effects of a rotating magnetic field on the growth of cadmium zinc telluride by the traveling heater method under microgravity conditions

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ABSTRACT

We present a fully coupled model for the traveling heater method (THM) under microgravity conditions and a rotating magnetic field (RMF) and apply it to analyze the growth of cadmium zinc telluride (CZT). The model provides a self-consistent representation of fluid flow and heat and mass transfer in a liquid zone shaped by dissolution and growth interfaces that are computed to satisfy local transport and thermodynamic equilibrium conditions. The temperature, stream function, tellurium, and zinc profiles in the liquid are analyzed with and without the rotating magnetic field. Results show that the system is very sensitive to the growth rate under microgravity alone, leading to tellurium accumulation, a concave growth interface, and constitutional supercooling at faster growth rates. While RMF-induced convection mixes the zone, creates a more uniform composition, and makes the microgravity system less sensitive to growth rate variations, RMF can also lead to undesirable outcomes. In particular, for stronger RMF fields, flows are driven inward along the growth interface, and the resulting accumulation of tellurium near the centerline results in localized interface concavity and liquid supercooling. The mechanisms behind the above phenomena are clarified, and some advice is provided for applying the RMF appropriately to THM CZT growth under microgravity conditions.

1. Introduction

Cadmium zinc telluride (CdZnTe, CZT) is an important semiconductor crystal with properties that make it suitable for radiation detection applications [1]. For example, single crystal $Cd_{0.9}Zn_{0.1}Te$ is used to manufacture room-temperature X-ray and γ-ray detectors [2], and $Cd_{0.96}Zn_{0.04}Te$ is used as a substrate for epitaxial growth of the infrared detection material HgCdTe [3]. In particular, there has been much recent effort toward improving the growth of large, single crystals of CZT for the fabrication of portable, low-cost, sensitive devices to detect radioactive materials. However, CZT is a particularly difficult material to grow [4], which necessitates improvement in existing methods.

The traveling heater method (THM) is a promising technique to grow large-diameter CZT crystals with good composition uniformity and better yield than competing processes [5]. The THM allows for CZT to be grown at a lower temperature than in other methods, and therefore the problems related to vapor pressure are avoided and contamination from ampoule is decreased. However, the low solid thermal conductivity and low activation energy for stacking faults make it challenging to control the shape of growth interface while growing large grains with few defects [4]. Additionally, the levels of tellurium inclusions in many THM-grown ingots are unacceptable for detector applications [6], though they are often lower compared to other growth methods [7]. To overcome these disadvantages, different approaches have been proposed to control the THM parameters and optimize the growth process [8–11].

Growth of CZT under microgravity conditions has resulted in improved material properties [12,13]. Furthermore, prior experiments indicated that a rotating magnetic field (RMF) better mixed the melt and improved microgravity crystal growth of CdTe and $CdTe_{0.9}Se_{0.1}$ [14,15]. Stelian and Duffar [9] carried out numerical simulations of THM under microgravity conditions (without RMF) and predicted favorable conditions for a stable growth interface in the absence of convection; terrestrial conditions were postulated to be less stable. Recently, Stelian and Duffar [16] extended their

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analyses to consider the effects of a RMF on microgravity and terrestrial THM CZT growth and found that higher RMF levels promoted interface instability.

It is well-known that the RMF produces Lorentz forces and induces a basic azimuthal flow, which generates secondary meridional flows in the form of two vortices that rotate in opposite directions. These secondary flows can modify heat and mass transfer, influencing segregation along and the shape of the melt–solid interface [17]. Since the applied RMF can be instantaneously changed, it provides for a potentially ideal control action, if its complicated effects can be predicted and understood. In this paper, the effects of RMF on CZT crystal growth by the THM under microgravity conditions are numerically analyzed and briefly summarized. The thermal and flow fields, growth interface shapes, and composition distributions for crystal growth with and without RMF are compared. Future papers will present a more detailed explanation of the underlying THM model [18] and our approach to solve for flows driven by a rotating magnetic field [19].

2. Model description

A schematic diagram of the THM system with RMF is shown in Fig. 1. In this configuration, crystalline CZT is grown from a liquid phase that contains excess tellurium. This solvent phase is produced in a molten zone that is simultaneously dissolving a feed solid while growing single-crystal material as it is moved via a traveling heater. The heater profile, also indicated in Fig. 1, is derived from a global heat transfer analysis [18].

Our THM model considers heat transfer through the solid feed, liquid zone, growing crystal, and ampoule domains, along with fluid flow and mass transport in the liquid zone. The dissolution and growth interfaces are solved using equilibrium phase-change conditions as functions of composition via the CdTe phase diagram. In addition, the segregation of species and heat generation via phase change are enforced along both solid–liquid interfaces. We assume that this cylindrical system gives rise to two-dimensional, axisymmetric behavior. To define the steady-state zone size, we set the total amount of excess tellurium contained in the liquid zone. A more detailed discussion of our THM model is given in [18].

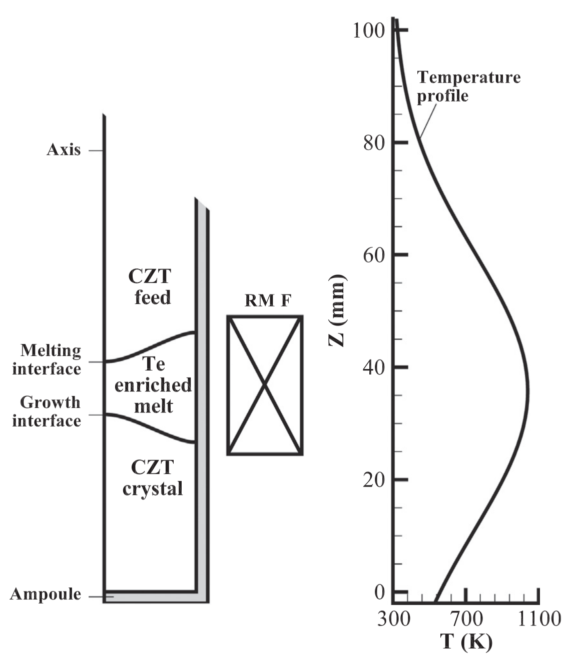

Fig. 1. *Left*: Schematic diagram of the THM system for CZT growth under an applied rotating magnetic field (RMF). *Right*: Heater temperature profile used in the model.

The effects of the RMF are described by solving for the Lorenz forces that arise from the induced magnetic and electric fields in the liquid zone. Contrary to prior considerations of a RMF on the THM [16,17], we directly solve for the time-periodic scalar electric potential through the system using the $\Phi_1 - \Phi_2$ model presented by Barz et al. [20]; this is more fully described in [19]. We compute pseudo-steady states of this mathematical model using a finite-element numerical method [21] that simultaneously solves for melt flow, heat transfer and species transport, and electric scalar potential, along with the shapes of the dissolution and growth interfaces.

In the following sections, we present results for a growth system with an ampoule inner diameter of 32 mm. We assume that the RMF applied externally to the melt can be expressed as a constant magnetic field rotating with a frequency taken to be $\omega = 50$ Hz in this study. Microgravity conditions are considered with an axial acceleration set to be 10^{-6} of the normal gravity and directed toward the growing crystal (downward in all figures). We consider the composition of the feed to be $Cd_{0.9}Zn_{0.1}Te$, with a specified excess amount of tellurium to form the melt zone. Physical properties and other operating parameters needed for the computations are provided in [18,19].

3. Results and discussion

3.1. CZT growth without RMF

CZT growth under microgravity without an applied magnetic field is first considered. Fig. 2 shows the distributions of temperature, stream function, tellurium and zinc in the melt zone for growth rates of 1 mm/day and 2.5 mm/day in plots (a1–a4) and (b1–b4), respectively. In these and all subsequent images, the left boundary represents the system centerline, the right boundary is the inner ampoule wall, and the top and bottom boundaries are the melting and growth interfaces, respectively. All fields are cylindrically symmetric about the centerline axis.

Focusing on the case of 1 mm/day growth, Fig. 2(a1) shows the temperature field through the melt zone, with heat diffusing inward from the heated, outer portion of the melt. The melt velocity, shown in (a2) via streamlines, consists of a very slow, torroidal cell that flows up along the outer boundary with the ampoule and downward along the centerline. Without an applied magnetic field, there is no azimuthal velocity and the meridional flow is driven solely by buoyancy, its specific form determined by our choice of the direction of microgravity acceleration (downward in the figure). This flow is too weak to noticeably affect the temperature field through the molten zone. The streamlines intersecting the melting and growing interfaces indicate an axial flow across the zone, driven by the difference in density from solid to liquid, often referred to as the solidification velocity. Fig. 2(a3) shows excess tellurium rejected at the growth interface and diffusing upward to the dissolution interface, where it can combine with the stoichiometric feed. Similarly, diffusion of zinc occurs in the opposite direction (a4), with the freshly melted feed providing zinc to diffuse downward to the depletion zone adjacent to the lower interface (the depletion zone arises due to zinc segregation during the growth of CZT crystal). While both compositional fields are nearly one-dimensional, there is some radial distortion of the iso-concentration contours due to the effects of weak convection. The geometry of the melt zone arises from the complicated interactions among the temperature and tellurium concentration fields and the liquidus curve of the phase diagram. Note that the