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.

Keywords: A1. Computer simulation, A1. Magnetic fields, A2. Traveling heater method growth, A2.

Microgravity conditions, B2. Semiconducting II-VI materials

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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 $\text{Cd}_{0.9}\text{Zn}_{0.1}\text{Te}$ is used to manufacture room-temperature X-ray and γ -ray detectors [2], and $\text{Cd}_{0.96}\text{Zn}_{0.04}\text{Te}$ is used as 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, 9, 10, 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 $\text{CdTe}_{0.9}\text{Se}_{0.1}$ [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 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 Figure 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 Figure 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].

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 $\text{Cd}_{0.9}\text{Zn}_{0.1}\text{Te}$, 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. Figure 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, Figure 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. Figure 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 melting and growth interfaces do not lie along an isotherm nor along a surface of constant composition.

Figures 2(b1–b4) show the system for a faster growth rate of 2.5 mm/day, again without the application of RMF. The strikingly different zone geometry arises from the interactions among heat and mass transfer and the phase diagram. Let us first consider the changes in the temperature field for the two cases. The maximum temperature regions are located near the ampoule wall, which is heated from outside. The minimum temperature regions are located near the growth interface, as the segregation and accumulation of tellurium leads to a low melting point. In both cases, the maximum temperature is approximately 1,017 K, but the minimum temperature decreases from 970 K to 932 K when the growth rate increases from 1 to 2.5 mm/day.

While there is a minor effect of latent heat (absorbed at the dissolution interface and released at the growth interface), the significant change of temperature distribution is primarily caused by the strong interaction of zone geometry and the tellurium compositional field, which changes significantly due to segregation effects at the different growth rates.

The distributions of tellurium in the melt region for different growth rates are shown in Figure 2(a3) and (b3). Tellurium is rejected into the melt at the growth interface, raising its concentration there, and the dissolution of CZT feedstock dilutes the Te-enriched melt at the melting interface. Therefore, the maximum and minimum concentration regions appear near the growth and melting interfaces, respectively. The maximum tellurium concentration increases at the growth interface when the growth rate increases from 1 to 2.5 mm/day, due to increased tellurium segregation at the faster growth rate. However, the melt flow is very weak and cannot take the tellurium away from the interface in large amounts, thus leading to the tellurium accumulation. As a result, the melting point decreases, forcing the growth interface downward to become more concave. On the other hand, the melting point rises at the dissolution interface due to the dilution effect, tending to move the interface downwards toward the hotter region as the growth rate increases. The segregation and accumulation of tellurium, as well as the lack of convection, lead to a large concentration gradient in the melt region for the growth rate of 2.5 mm/day. This is evidenced by the denser contours in Figure 2(b3) compared with those in Figure 2(a3). Importantly, the high gradient of tellurium in the case of faster growth produces constitutional supercooling. This is indicated by the blue region in Figure 2(b3), where local conditions of temperature and tellurium concentration result in a region that is predicted to be a solid via the thermodynamic phase diagram (or, equivalently, an unstable, supercooled liquid).

Figure 2(a4) and (b4) show the distributions of zinc in the melt region for different growth rates. Radial segregation arises in both cases primarily due to the nonplanar shape of the growth interface; however, compositional inhomogeneity is more severe at the higher growth rate.

Since buoyant melt convection is very weak under microgravity, it is impossible for it to contribute meaningfully to heat or species transport. In particular, species transport is then dominated by diffusive effects, leading to significant composition gradients. As a result, the interface shapes are very sensitive to the growth rate, and high growth rates are plagued by constitutional supercooling. Prior modeling studies have alluded to similar effects [9, 17] and have concluded that additional mixing, such as that provided by the application of a rotating magnetic field may be beneficial. We assess this strategy in the cases presented in the following section.

3.2. CZT growth with RMF

To study the effects of RMF on THM growth of CZT, an appropriate intensity of magnetic field should be chosen to increase melt flows toward moderate mixing. Too weak of a magnetic field cannot mix the melt effectively, and too strong of a magnetic field can lead to highly deformed melting and growth interfaces. Based on prior simulations [19], we choose a field strength of 10 mT for the cases considered here.

Figure 3 shows THM cases for same growth rates considered previously, but with an applied magnetic field of 10 mT rotating at 50 Hz. The primary outcome of the RMF is the driving of a strong azimuthal flow, accompanied by a multi-cellular meridional flow consisting of two vortices flowing outward at the midpoint of the zone and inward along both solid-liquid interfaces, as indicated in Figures 3(a2) and (b2). These flows are over three orders of magnitude faster than the background flows driven by microgravity.

One outcome of these stronger flows is that the strong dependence of melt zone geometry on growth rate, as exhibited by the THM system without RMF, is mitigated. In Figure 3, the melting and growth interface shapes only change slightly when the growth rate increases. This is also reflected in the temperature fields for these cases, shown by Figures 3(a1) and (b1), which have a similar form. Bulk mixing of species is also comparable for each case, with two, relatively well-mixed cores corresponding with the meridional vortices, as indicated in the composition fields for excess tellurium and zinc in the right-most plots. These cases show that the application of RMF under microgravity can mix the melt and make the overall melt composition significantly more uniform.

However, all is not well in this system at the higher growth of 2.5 mm/day, shown in the lower images. Clearly, there remain strong segregation effects near the interfaces, with thin compositional layers forming at both dissolution and growth surfaces. Focusing on the melt region just above the growth interface and near the centerline, the upward circulation of the flow cell transports heat away from the interface, thereby lowering the axial temperature gradient. Simultaneously, the inward flow across the growth interface sweeps rejected tellurium toward the centerline, where it accumulates. While these two effects are visible in the case of 1 mm/day growth in (a1–a4), they do not adversely impact the system. Conversely, these effects are strong enough in the 2.5 mm/day growth system to result in a highly concave growth interface at the centerline, arising from the local accumulation of tellurium and the phase diagram.

Even more significant, constitutional supercooling occurs at the higher growth rate, as shown by the blue region of supercooled liquid in Figure 3(b3), plotted using the same approach as described in the prior section. This region of supercooling arises from the imbalance caused by a decreased axial temperature gradient and a stronger accumulation of rejected tellurium along the growth interface, both driven by the interaction of segregation with the RMF flows. Thus, the mechanism leading to supercooling is different from that in the microgravity growth system without RMF considered in the previous section.

Finally, Figures 3(a4) and (b4) show the distributions of zinc in the melt. The contours are consistent with the flow pattern and are divided into a upper, high-concentration region and a lower, low-concentration region. The radial distribution of zinc is relatively uniform over much of the interface for the case of 1 mm/day growth; however, the deeply convex interface at the centerline for the higher growth rate results in a significant variation of zinc incorporation.

4. Conclusions

We carried out fully-coupled, numerical simulations to study the effects of RMF on the THM growth of CZT under microgravity conditions. The results clearly show that the thermal and flow fields, growth interface, and composition distributions are very sensitive to the growth rate without an applied magnetic field. Under microgravity alone, increasing the growth rate will lead to tellurium accumulation, a concave growth interface, and constitutional supercooling.

RMF does mix the melt via convection, but the outcomes are not always better. Bulk mixing can reduce the sensitivity of growth interface shape to the growth rate and homogenize the composition field—improvements compared to microgravity growth without RMF. However, the specific melt flow pattern driven by the rotating magnetic field can lead to undesirable effects along the growth interface. RMF flows can accentuate local imbalances between the tellurium and temperature distributions near the growth interface, resulting in highly concave growth interfaces near the axis and morphological instability due to constitutional supercooling.

Future optimization of microgravity THM growth will require a comprehensive understanding of the many outcomes of applied rotating magnetic fields. Toward these goals, analysis using computational models, such as that presented here, will be indispensable.

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6. References

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Figure 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.

Figure 2: Distributions of temperature, stream function, tellurium, and zinc in the melt region for different growth rates, without magnetic field: (a1-a4) 1 mm/day; (b1-b4) 2.5 mm/day. Blue overlay indicates supercooled liquid.

Figure 3: Distributions of temperature, stream function, tellurium, and zinc in the melt region for different growth rates, with rotating magnetic field: (a1-a4) 1 mm/day; (b1-b4) 2.5 mm/day. Blue overlay indicates supercooled liquid.