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Numerical investigation of non-isothermal phase change phenomena in vertical Bridgman crystal growth

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ABSTRACT

The non-isothermal phase change phenomena during the vertical Bridgman growth process for HgCdTe are numerically investigated using an interface capturing finite element scheme. The influence of the growth parameters such as *Bi*, *Ste*, *U* and the flow parameters including Gr_T and Gr_S on the non-isothermal phase change phenomena are obtained. Some new features about the melt/crystal interface shape, the temperature field near the interface and the flow field are revealed by comparing the non-isothermal phase change with the isothermal phase change. Furthermore, the comparison of the non-isothermal interfacial characteristics between the pure diffusion, the natural convection and the double-diffusive convection is made and the obvious differences are presented.

1. Introduction

II–VI compound semiconductor materials, such as HgCdTe, HgZnTe and CdZnTe, are composed of two, three or four IIB elements such as Zn, Cd and Hg, and VIA elements such as S, Se and Te in the periodic table of the elements. Many applications of these materials are being developed for new kinds of instruments such as lasers, infrared detectors and imaging instruments because of their special electrical, optical and photoelectrical properties. Therefore, significant research has been made focusing on the pure crystal growth process for these materials in the recent years [1–5].

The vertical Bridgman method (VBM) was firstly developed in 1925. Now it is one of the best and popular techniques for the growth of semiconductor crystals. A schematic diagram of a typical vertical Bridgman growth system is shown in Fig. 1(a). This technique has the advantage of steady temperature fields, controllable temperature gradients and purification. The method can yield pure crystals of large sizes by controlling the boundary conditions of heat and mass transfer. Therefore, it is widely used to produce II–VI compound semiconductor crystals. As we all know that the interface shape during the crystal growth process significantly affects the final crystal quality. Plane, stable interfaces are good for high-quality crystal growth; however, in practice, planer interfaces are not easy to obtain. In the phase diagram of HgCdTe [6], the solidus and liquidus lines are far apart, so it is thinkable that there is remarkable concentration segregation of the solvent HgTe from the crystal into the melt at the interface. This segregation makes it difficult to grow large, uniform, high-quality HgCdTe crystals because of multi-elements non-isothermal phase change. Therefore, it is very important to comprehend interfacial characteristics such as interface shape, concentration segregation, melting temperature, and non-isothermal phase change which occur at the melt/crystal interface.

Since II–VI semiconductor materials are not transparent and the practical growth process are implemented at high temperatures and other difficult experimental conditions, visual observation of the phenomena and measurements of temperature, concentration and velocity fields during the growth process are almost impossible. Numerical simulations have special significance in this research field. Capper et al. [7] investigated the shape of the melt/crystal interface by rapidly quenching HgCdTe crystals using VBM. They showed that the interface is a paraboloid with the center lower than the edges. Wang et al. [8] investigated the effects of growth speed on the interface shape by pulling. Their results illustrated that the concave paraboloid becomes more concave with higher growth speeds. Hong and Wei [9] numerically studied steady state Bridgman growth of a CdTe crystal with two ampoule configurations. Their results revealed that although the two ampoule configurations were quite different, they had similar influence on the melt/crystal interface shape with the undesirable concave interface. A numerical simulation was carried by Lan and Ting [10] to study the interface morphology during Bridgman crystal growth illustrated the effects of growth speed, ambient temperature profile and ampoule design. Liu and Chen [11] numerically studied the effects of gravity on the interface in a horizontal Bridgman growth system. The strong convection in the melt can lead to a concave interface in the crystal, but the interface becomes flat when the gravity is small enough. Wei et al. [12] analyzed the influence of the pulling rate and ambient temperature distribution

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Nomenclature

A	aspect ratio $A = H/R$
Bi	Biot number
C	dimensionless solute concentration C^*/C_0^*
C^*	solute concentration (mole fraction)
C_0^*	initial solute concentration (mole fraction)
C_p	specific heat capacity at constant pressure (J/°C g)
D	solute diffusivity (cm^2/s)
Gr_S	solutal Grashof number
Gr_T	thermal Grashof number
g	gravitational acceleration (cm/s^2)
H	height of the enclosure (cm)
k	the thermal conductivity (W/K cm)
K_0	segregation coefficient of CdTe
Pr	Prandtl number
r, z	dimensionless radial and axial coordinates, $r = r^*/R$, $z = z^*/R$
$\hat{r}, \hat{z}$	dimensionless radial and axial coordinates in the transformed coordinate system
R	radius of the enclosure (cm)
Sc	Schmidt number
Ste	Stefan number
T	dimensionless temperature $(T^* - T_L^*)/(T_H^* - T_L^*)$
T^*	temperature (°C)
T_H, T_L	the dimensionless highest and lowest temperature of the ambient temperature
U	dimensionless pulling rate
U^*	the pulling rate (cm/s)
u_r, u_z	dimensionless radial and axial velocity
u_r^*, u_z^*	radial and axial velocity (cm/s)
ΔH	the latent heat of solidification (J/g)

Greek symbols

α	thermal diffusive coefficient (cm^2/s)
β_S	coefficient of solutal expansion ((mole faction CdTe)$^{-1}$)
β_T	coefficient of thermal expansion (°C^{-1})
ν	kinematic viscosity (cm^2/s)
ρ	density (g/m^3)
τ	ratio of melt and crystal thermal conductivities
ψ	dimensionless stream-function
ψ^*	stream-function (cm^3/s)
ω	dimensionless vorticity
ω^*	vorticity (s^{-1})

Superscript

$*$	dimensional quantities

Subscripts

c	crystal
f	furnace
m	melt
S	solutal
T	thermal

on the melt/crystal interface for Bridgman growth of CdTe. The results show that a lower growth rate produces a flatter interface. Increasing the temperature gradients has also proved effective for improving the interface. Wang et al. [13] brought forward the simplified formula of heat transfer coefficient between the furnace and the ampoule under the condition of the three regions temperature design of furnace. Based on this formula and its modified form, Liu et al. [2,3] used the finite difference method (FDM) to analyze the influence of the accelerated crucible rotation technique (ACRT) on the crystal growth by VBM. Bourret et al. [14–16] studied the variation of the phase change interface position with the time, solute concentration redistribution and the melting temperature after the system reached steady state, and so captured one-dimensional non-isothermal phase change phenomena. On the basis of the axial solute concentration redistribution during the HgCdTe growth process, Wang et al. [17] studied the optimal expression of the initial growth rate to lead to decreasing the length of the initial transient region and increasing the length of the steady growth region. Yeckel and Derby [18] performed the theoretical simulations of VB growth of cadmium zinc telluride and got the effects of the ACRT. Kawaguchi et al. [19] simulate the contributions of CCRT, ACRT, thermal and solutal gradients to the convection in the melt during the growth of CdZnTe crystals by the vertical gradient freezing (VGF) method.

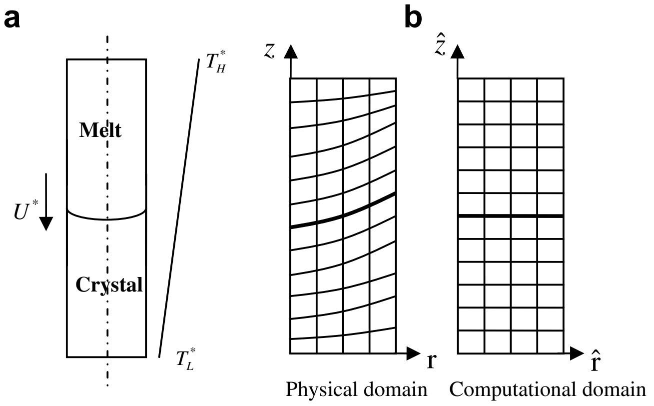

Fig. 1. Schematic diagram of vertical Bridgman growth (a) and sample finite element mesh geometry (b).

As far as authors' knowledge is concerned, crystal growth studies have been mainly done by material's science researchers using experiments or numerical methods. However, the physical processes during crystal growth are very complex; they include multi-elements heat and mass transfer, non-isothermal phase change and double-diffusive convection. The thermophysical analyses of the processes are insufficient, especially non-isothermal phase change phenomena. There have been few studies of factors affecting interfacial characteristics of the non-isothermal phase change phenomena during growth process of II–VI compounds. Shi et al. [20] analyzed the effects of the Bridgman process parameters and the double-diffusive convection in the melt on the melt/crystal interface shape and the solute distribution for the II–VI semiconductor material based on the isothermal phase change. Liu and Lu [21,22] systematically studied one- and two-dimensional non-isothermal phase change phenomena without considering the convection in the melt. The research about two-dimensional non-isothermal phase change phenomena with flow and the influence of double-diffusive convection on the non-isothermal phase change have not been dealt with in the literature. So the current work uses an interface capturing finite element scheme to analyze the two-dimensional non-isothermal phase change phenomena with flow during the II–VI compound crystal growth.

2. Physical model and boundary conditions

The calculating model of vertical Bridgman method in the crystal growth is schematically shown in Fig. 1(a). The material is in a furnace with a linear temperature gradient and a pulling down rate, U^*. So the melt is corresponding to the high-temperature zone