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Simplified numerical approach for estimation of effective segregation coefficient at the melt/crystal interface

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ABSTRACT

A thermo-gravitational convection and impurity transfer in the melt were investigated using a simplified numerical model for Bridgman GaSb(Te) crystal growth in microgravity conditions. Simplifications were as follows: flat melt/crystal interface, fixed melt sizes and only lateral ampoule heating. Calculations were carried out by *Ansys ®Fluent®* code employing a two-dimensional Navier–Stokes–Boussinesq and heat and mass transfer equations in a coordinate system moving with the melt/crystal interface. The parametric dependence of the effective segregation coefficient K_{eff} at the melt/crystal interface was studied for various ampoule sizes and for microgravity conditions. For the uprising one-vortex flow, the resulting dependences were presented as K_{eff} vs. V_{max}—the maximum velocity value. These dependences were compared with the formulas by Burton–Prim–Slichter's, Ostrogorsky–Muller's, as well as with the semi-analytical solutions.

1. Introduction

The effective segregation coefficient K_{eff} is the important parameter for analysis of impurity segregation in melt crystals growth. Its value depends on a structure and intensity of the melt flow that substantially affects the impurity flux from the melt into the crystal. Estimations of K_{eff} can be found in many publications, basically using two approaches. The first one, quite simple, consists of an application of approximate analytical hydrodynamic formulas [1,2] for the value of the flow velocity near the melt/crystal interface (MCI). This approach is widely used now in technological practice [3]. The second approach is based on the complete numerical simulation of the crystallization process, and requires specialized program codes, a significant computational cost, and high user qualifications [4].

In this paper, a thermo-gravitational convection and impurity transfer in the melt for the Bridgman GaSb(Te) crystal growth in microgravity are calculated using a numerical model with the following simplifications: flat MCI, fixed melt volumes, and only lateral ampoule heating. Parametric calculations were carried out employing a program code *Ansys®Fluent®*, which was supplemented by the author's subroutines in C++, taking into account a crystallization model [5,6]. Numerical solutions are compared with the data calculated by a semi-analytical model [5], and using analytical formulas of Burton–Prim–Slichter (BPS) [1] and Ostrogorski–Muller (OM) [2].

2. Simplified numerical model

A crystallization process is considered for the constant MCI velocity $\boldsymbol{V_S}=(V_S,0)$ in a flat melt layer (X,Y) of thickness H and length L at a gravitational field $\mathbf{g}=(0,-g)$ for the following thermal conditions: $T_S=985$ K (the melting point) and different values of $T_W=996\div1058$ K (Fig. 1), which determine the variation of the longitudinal temperature gradient. The equations of Navier–Stokes–Boussinesq and heat and mass transfer in a melt may be written in the coordinate system associated with moving MCI in the vector form [5]:

$$\begin{aligned} &\partial V/\partial t+[(V-V_S)\nabla]V=-1/\rho\nabla P+\nu\Delta V+g\beta_T T,\\ &\mathrm{div}V=0,\\ &\rho C_p\{\partial T/\partial t+[(V-V_S)\nabla]T\}=\lambda\Delta T,\\ &\partial C/\partial t+[(V-V_S)\nabla]C=D\Delta C. \end{aligned} \tag{1}$$

Solving this system, the velocity vector $\mathbf{V}=(V_X,V_Y)$, the pressure P, the temperature T and the impurity concentration C dependence on the spatial coordinates and time are determined.

For the melt boundaries: $\mathbf{V}=0$; T_S, T_W are defined as shown in Fig. 1, and the top and the bottom walls are thermo-isolated.

The initial impurity concentration in the melt: $C=C_0$; the boundary condition for the boundary at the MCI is $D\nabla C=(1-K_0)V_S C$, and

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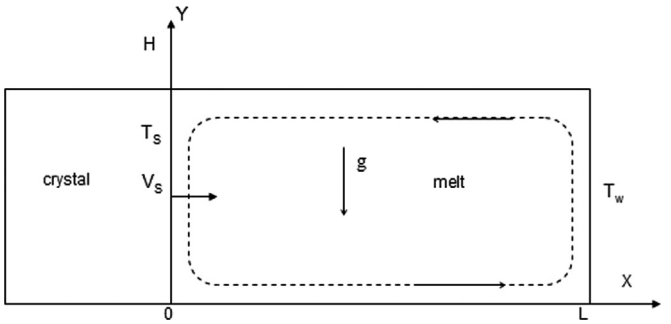

Fig. 1. Scheme of the simplified model: thermal boundary conditions, V_S—velocity of MCI, and the direction of melt flow (dashed line).

$\nabla C=0$—for all other boundaries in the case of a continuous crystallization for the whole ingot length. For the GaSb(Te) melt, the physical parameters are as follows: density $\rho=6.06$ g/cm^3, kinematic viscosity $\nu=0.0032$ cm^2/s, thermal conductivity $\lambda=1.02\times10^6$ erg/cm × K × s, heat capacity $C_P=3.3\times10^6$ erg/g × K, thermal expansion coefficient $\beta_T=9.6\times10^{-5}$ K^{-1}, tellurium diffusion coefficient $D=5\times10^{-5}$ cm^2/s [7], and the equilibrium segregation coefficient $K_0=0.37$. The crystallization rate was constant: $V_S=3\times10^{-4}$ cm/s, and the microgravity level varied as: $g/g_0=1.6\times10^{-5}\div2.2\times10^{-3}$; $g_0=980$ cm/s^2.

For numerical analysis, it is useful to consider the dimensionless similarity criteria that characterize the mode of thermal gravitational convection and impurity transport. They are $Gr=g\beta_T[(T_W-T_S)/L]H^4/\nu^2$—Grashof, $Pr=\nu\rho C_P/\lambda$—Prandtl, $Ra=GrPr$—Rayleigh and $Sc=\nu/D$—Schmidt numbers. For GaSb(Te) melt: $Pr=7.45\times10^{-2}$ and $Sc=64$. The value of Gr was a variable parameter, a function of the microgravity g/g_o level and the melt size $H\times L$.

In this work, a simplified variant of the model was used, that corresponds to the calculation of a discrete stage of crystallization ($H\times L$: 1.5×4, 0.6×1.5 and 3×9 [cm × cm]), and the condition $C=C_0$ at $X=L$ (Fig. 1).

Analysis of the results for space experiment by means of the hydrodynamic similarity criteria and known analytical models [3] has showed an importance of such input parameter, as the velocity V_∞ on the end of the boundary layer. However, the determination of V_∞ and the boundary layer thickness requires a detailed analysis of the velocity and concentration fields calculated on the basis of full Navier–Stokes and heat–mass transfer equations. Therefore, in the present work this task has been simplified. The maximum V_{max} of velocity magnitude $\mathbf{V}_{magn}=(V_X^2+V_Y^2)^{1/2}$ was used for the analytical models instead of V_∞. This simplification was interesting for technological practice. Therefore, the comparisons with known analytical models are present not only vs. the dimensionless criteria (Gr and Re), but also vs. the values of V_{max}.

3. Semi-analytical approach and analytical models

The calculations, using a particular simplified numerical model, were compared with the data calculated by semi-analytical [5] and analytical formulas [1,2].

In the semi-analytical model [5], the equation for the impurity concentration is solved numerically for the following analytical velocity field in the melt:

$$V_X(X,Y)=Gr\left[0.25-(Y-0.5)^2\right](Y-0.5)\{1-e^{-\alpha X}[\cos(\beta X)+(\alpha/\beta)\sin(\beta X)]\}$$
$$V_Y(X,Y)=Gr\left[0.25-(Y-0.5)^2\right]e^{-\alpha X}(\alpha^2/\beta+\beta)\sin(\beta X)/24. \quad (2)$$

Here, $\alpha=4.15$, and $\beta=2.286$.

We investigated a few formulas for K_{eff}, corresponding to the existing analytical models.

For BPS model [1]:

$$K_{eff}=K_0/\left[K_0+(1-K_0)e^{-\Delta}\right], \quad (3)$$

where $\Delta=V_S\delta/D$. Here the name "BPS model" indicates the solution for the problem of convective diffusion in the "stagnant film" approximation obtained in [1]. In this work, the convective flow intensity is accounted via parameter δ associated with the diffusion layer thickness. There are no restrictions on this parameter δ. Although in [1] the found solution was applied for Czochralski crystal growth (there δ is defined in the model of a rotating disk), nothing prevent the use of BPS solution for the Bridgman method, too, while an appropriate formula for δ will be chosen.

The estimation of δ in the Bridgman crystal growth was proposed by means of the formula based on well-known Blasius solution for the flow along a plate [8]:

$$\delta=5(\nu H/V_{max})^{1/2}(D/\nu)^n, \quad (4)$$

where the exponent n is dependent on $Sc=\nu/D$ with $1\geq n\geq1/3$ for $0<Sc<\infty$ [9]. For most cases $n\approx1/2$ [10]; in particular, this value is recommended for semiconductor melts with $5<Sc<50$ [8]. For our case $Sc=64>50$ and formally we should use $n=1/3$. However, these criteria are very approximate, so we have carried out the estimations for both cases $n=1/2$ and $n=1/3$ of the BPS model. The question of the applicability of the formula (4) in the BPS model is considered for the first time.

For OM model [2], a weak convection case was assumed:

$$K_{eff}=(1+\eta)/(1+\eta/K_0), \quad (5)$$

where $\eta=(D/\nu)^{1/2}V_{max}D/V_S^2H$ and V_{max} is a convective velocity beyond the hydrodynamic boundary layer. The coefficient η corresponds to the formulas (23) and (38) in [8] with $V_D=V_\infty(D/\nu)^{1/2}$.

In formulas (3) and (5), the velocity V_{max} is an input parameter. Therefore, for K_{eff} comparison using values calculated by the semi-analytical and numerical models, V_{max} was taken as the maximum V_{magn} in the melt. This value was calculated by the formula (2) and by *Ansys®Fluent®*. The required value of K_{eff} is calculated as follows: $K_{eff}=K_0<C>/C_0$, where $<C>$ is an average impurity concentration at MCI.

The impurity redistribution along MSI (including the diffusion mass transfer) is out of a consideration of the one-dimensional models. Therefore, it is necessary to find a way for verification of these 1D models with the 2D modeling. For this purpose the values of K_{eff} were calculated for the central and both extreme points of MSI, but a good correlation between 1D and 2D models was absent. However the good correlation has been obtained with using the averaged value of K_{eff} along MSI. In this case the value of $\delta(y)$ was calculated with $y=H$ corresponding to "outlet edge" of MSI. In the following sections we present the results of such verification.

4. Microgravity influence

A thermo-gravitational convection was investigated in the ampoule with only lateral heating for a number of gravity levels. For microgravity values: $g/g_0=1.6\times10^{-4}$, 1.6×10^{-3} and 1.4×10^{-2}, the maximum values of a velocity magnitude V_{max} correspond to 7.5×10^{-5}, 8.1×10^{-4} and 4.4×10^{-3} cm/s, respectively. This comparison shows that with increasing g/g_0 the one-vortex flow persists, and its structure is changing only slightly. In Fig. 2a there are shown contours of the V_{magn} in the ampoule for $g/g_0=1.6\times10^{-3}$. A noticeable feature of this weak flows is symmetry of velocity field relatively to the longitudinal ($Y=H/2$) and transverse ($X=L/2$) cross-sections (Fig. 2b).