NASA-TM-112925

IN-27-TM
50206

Numerical modeling of HgCdTe solidification: effects of phase diagram, double-diffusion convection and microgravity level.

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1. INTRODUCTION

Melt convection, along with species diffusion and segregation on the solidification interface are the primary factors responsible for species redistribution during HgCdTe crystal growth from the melt. As no direct information about convection velocity is available, numerical modeling is logical approach to estimate convection. Furthermore influence of microgravity level, double-diffusion and material properties should be taken into account. In present study HgCdTe is considered as binary alloy with melting temperature available from phase diagram. The numerical model of convection and solidification of binary alloy is based on the general equations of heat and mass transfer in two-dimensional region. Mathematical modeling of binary alloy solidification is still challenging numerical problem. Rigorous mathematical approach to this problem [1] is available only when convection is not considered at all. Proposed numerical model was developed using finite element code FIDAP [2].

It is well known, that solidification interface shape is controlled primarily by the heat fluxes via interface as well as via ampoule walls. The appropriate temperature boundary conditions on the ampoule walls was obtained from the global model of AADSF furnace [3], that includes radiative, conductive and convective heat transfer. Alternatively experimental temperature measurements were used to generate temperature boundary conditions for the numerical model. Two complimentary solutions where obtained: one for melt region with a fixed boundary and other for melt region with an unknown boundary. In the former case, numerically converged solutions where typically easily obtained with standard FIDAP settings, while in the latter case a special choice of relaxation factors for each degree of freedom was required. The simplified model with fixed interface shape was especially useful for parametric study of convection, as the steady state solution was available for a broad range of parameters.

In the complete numerical model with unknown boundary the following thermophysical properties of HgCdTe was taken into account: the pseudobinary HgTe-CdTe phase diagram [4], the segregation coefficient, an assumed diffusion coefficient for mass transport in HgCdTe, the temperature and composition dependence of the melt density (to calculate buoyancy force) and the melt viscosity and its temperature dependence [5]. The composition dependence of diffusion coefficient was not considered, although it might be important for double-diffusion convection. In the simplified model with the fixed boundary melting temperature was fixed instead of being calculated from phase diagram.

The basic physics that controls shape of the solidification interface is as follows. Due to the large separation between the liquidus and the solidus, there is a wide variation of composition through the boundary layer adjacent to the interface. Cooling from below and the segregation at the solid-melt interface of the heavier HgTe-rich solute each tend to suppress convection. However, the thermal conductivity of the solid HgCdTe is a factor of eight times smaller then the melt and there is a resulting thermal short circuit through the ampoule (fused silica); this results in curved isotherms in this region as the heat cannot travel parallel to the axis within the center of the sample. The interface is therefore curved and, due to the thermal and solutal convection is neither an isotherm nor a line of isoconcentration. Double diffusion also contributes to the mass transport. In the present study, the numerical model is used to consider both these source of mass transport.

2. MATHEMATICAL MODEL AND NUMERICAL PROCEDURE

The main features of the model are double-diffusion convection in the melt and solidification. The set of steady state Navier-Stokes equations in Boussinesq approximation along with energy and solutal balance equations are used [2]. In the following **u** is melt convection velocity, **R** is sample pulling velocity (assumed at the steady state to be equal to crystal growth velocity), **p** - pressure, ρ - density, **g** - microgravity acceleration, β_T and β_C - volumetric expansion coefficients due temperature and concentration change respectively, $\mathbf{T_0}$ and $\mathbf{C_0}$ -reference temperature and concentration, μ - dynamic viscosity, **D** - mass diffusion coefficient, **k** - thermal conductivity, $\mathbf{C_p}$ - specific heat at constant pressure. Furthermore indexes "**s**" and "**m**" are used to refer to solidified and molten parts of the sample.

Momentum conservation equation

$$\rho(u\nabla u) = -\nabla p + \mu\Delta u + \rho g[1 - \beta_T(T - T_0) + \beta_C(C - C_0)] \tag{1}$$

Mass conservation equation (Boussinesq approximation) $\nabla u = 0$ (2)

Energy conservation equation $\rho C_p(u\nabla T) = \nabla(k\nabla T)$ (3)

Species conservation equation $u\nabla C = D\Delta C$ (4)

The boundary conditions incorporate major physical assumptions in order to describe solidification of an unknown interface between molten and solidified HgCdTe. These phase change boundary conditions are as follows.

The melting temperature is determined by

$$T_m = T_s = T_0(C_0) + m(C)C_m \qquad where \qquad m(C) = \frac{dT}{dC} \tag{5}$$

The heat balance on the interface, including latent heat release (**L**) is

$$k_m\nabla T_m \cdot \vec{n} - k_s\nabla T_s \cdot \vec{n} = \rho_s L\left(R - \frac{dS}{dt}\right) \cdot \vec{n} \tag{6}$$

Here **t** stands for pseudo-time, i.e. artificial melting time, used in the model to allow interface adjustment in accordance with temperature field and phase diagram. Notation **n** is unit vector normal to the interface.

The balance of the mass flux across interface is $\rho_m\left(u - \frac{dS}{dt}\right) \cdot \vec{n} = \rho_s\left(R - \frac{dS}{dt}\right) \cdot \vec{n}$ (7)

The no-slip boundary condition at the interface is $(u - R) \times \vec{n} = 0$ (8)

The mass balance for solute transport across the interface is

$$\left(\frac{\rho_m}{\rho_s}\right)D_m\nabla C_m \cdot \vec{n} - D_s\nabla C_s \cdot \vec{n} = C_s\left(R - \frac{dS}{dt}\right) \cdot \vec{n} - C_m\left(u - \frac{dS}{dt}\right) \cdot \vec{n} \tag{9}$$

The chemical equilibrium on the interface is $C_s = \kappa C_m$ (10)

Value of segregation coefficient κ can be obtained from phase diagram as it represents ratio of solidus to liquidus concentration values. In case of HgCdTe value of 4.0 was good estimation. To perform actual calculations initial temperature and solutal field, convection velocities and interface position are required. In order to obtain a steady state solution of the problem (1-4) with boundary conditions (5-11) a three step numerical procedure was used. On the first computational step the position of interface was flat and fixed, and so variable S = 0. Initial zero velocity field and constant initial distribution of temperature and concentration were used to obtain the solution of double-diffusion convection problem in two-dimensional area with fixed boundaries, that satisfies temperature boundary conditions. This solution was used as initial conditions during the second step, when variable **S** was released and the interface shape was obtained with proper adjustment of the computational mesh. A final third step was required to obtain a convergent solution for all variables involved. Two-dimensional 9-node quadrilateral finite elements were implemented. Due to strong nonlinearity of the problem, a relaxation factor value as high as 0.9 was used for surface elevation variable **S**. A first-order upwinding numerical scheme was used [2]. For parametric studies a model was simplified by elimination of independent variable **S** with appropriate boundary conditions on the fixed solid/ melt interface.

3. NUMERICAL TEST PROBLEM: DOUBLE-DIFFUSION CONVECTION

Double-diffusion convection governed by set of equations 1-4 occurs in a two-dimensional square enclosure which is heated and salt rich at the bottom. The vertical boundaries remained isolated. This is a case of opposing or counteracting flow, so that as the heat impact tends to start convection, the salt introduction from below tends to suppress it. It is also known as a "diffusive" regime, as the component with a higher diffusivity (heat) has a destabilizing effect, while the component with a lower diffusivity (salt) has a stabilizing effect on the local density gradient. The complete set of nondimesional parameters for this problem include thermal and solutal Rayleigh numbers, based on enclosure height **H** and top to bottom differences of temperature and salt, Prandtl and Lewis numbers and aspect ratio. Results presented on fig 1, indicate good agreement with data from [6].

4. RESULTS AND DISCUSSION

Two complimentary solutions are presented here.

The first one is based on the complete model described in section 2. In this case typical distribution of temperature field along with pattern of melt flow is presented on fig2a. For comparison experimental photograph featuring "quenched" interface is presented on fig

2b. Good agreement in shape is mainly due proper choice of thermal conductivities of solidified and molten material as well as silica ampoule thermal conductivity and wall thickness. The strict vertical orientation of the sample manifests itself in a symmetrical pattern of the flow. In other simulated cases deviation from the vertical of only 1 degree cases loss of symmetry and formation of one cell pattern of convection. These cases are featured in the video film. The shape of interface closely fits and is almost the same for various levels of gravity. This strongly suggests that convection in the melt virtually has no effect on solidification interface curvature in this particular case. Further efforts are required to investigate this finding in more detail. It should be noted, that simulation of this problem using non-dimensional parameters, like Rayleigh number, can be confusing in this case, as the driving force for convection is not a vertical or a simple radial gradient, but rather a radial temperature gradient created by the curvature of the solidification front. Otherwise the cooling of the melt from below and rejection of the heavier component (HgTe) would suppress convection. The radial variation in concentration is only in fair agreement with experimental observation. It appears, that the calculated temperature radial variation is about 7°C less than expected variation of 20 °C, as was estimated using experimental data.

The second set of solutions is based on simplified model with fixed solid / melt interface. Parametric study of maximum convection velocity depending on microgravity conditions is presented on fig 3. It is clear, that the maximum velocity of convection occurs, when ampoule is inclined at 90^0, while if ampoule is strictly vertical convection is less by factor of 50%. These data strongly suggest, that in order to reduce convection in the melt, vertical orientation of the ampoule is of utmost importance. This is valid till gravity level of order 10^{-6} of gravity on Earth. However at very low gravity, of order 10^{-7} and 10^{-8} of earth gravity the tilt of the ampoule has much less effect in absolute values of convection velocity.

5. CONCLUSIONS

Prediction of solidification interface shape and flow pattern in molten HgCdTe for various gravity conditions are available using a 2-D model of melt convection and solidification. This model utilizes temperature boundary conditions from results of modeling of heat transfer in AADSF furnace, and incorporates all basic modes of heat transfer, such as conduction, convection and radiation. Alternatively, a measured experimental temperature profile can be used.

It appears that crystal/melt interface shape, as well as concentration distribution on the interface depend primarily on heat flow and concentration distribution in close vicinity of interface. The models ability to predict correctly the interface shape and concentration distribution heavily depends on known material properties, such as conductivity and segregation coefficient.

Maximum convection velocity in the ampoule is extremely sensitive to gravity vector orientation and can be reduced at least by factor of 50% if vertical orientation of ampoule is strictly observed. This conclusion is valid to microgravity level of about 10^{-7} compared to earth gravity. If microgravity is about 10^{-8} absolute values of convection in the melt are very low, and convection can be considered as not sensitive to the ampoule orientation.

ACKNOWLEDGEMENTS

The authors thank James B. Hargrave for instant software support and help in video film production. The authors thank Dr. Narayanan Ramachandran for suggesting numerical test problem for double-diffusion convection. The work was supported by the Microgravity Science and Applications Division of the National Aeronautics and Space Administration and by National Research Council Research Associateship Programs.

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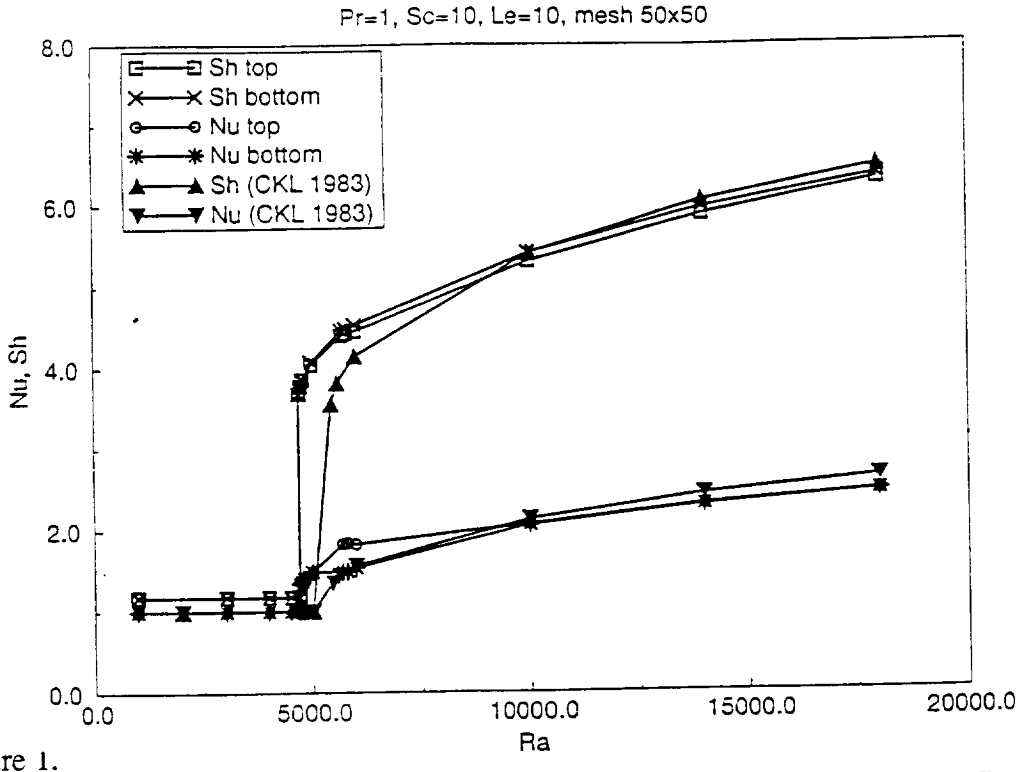

Figure 1.
Nusselt and Sherwood number dependence versus thermal Rayleigh number.Results of test problem appears in agreement with data from [6].

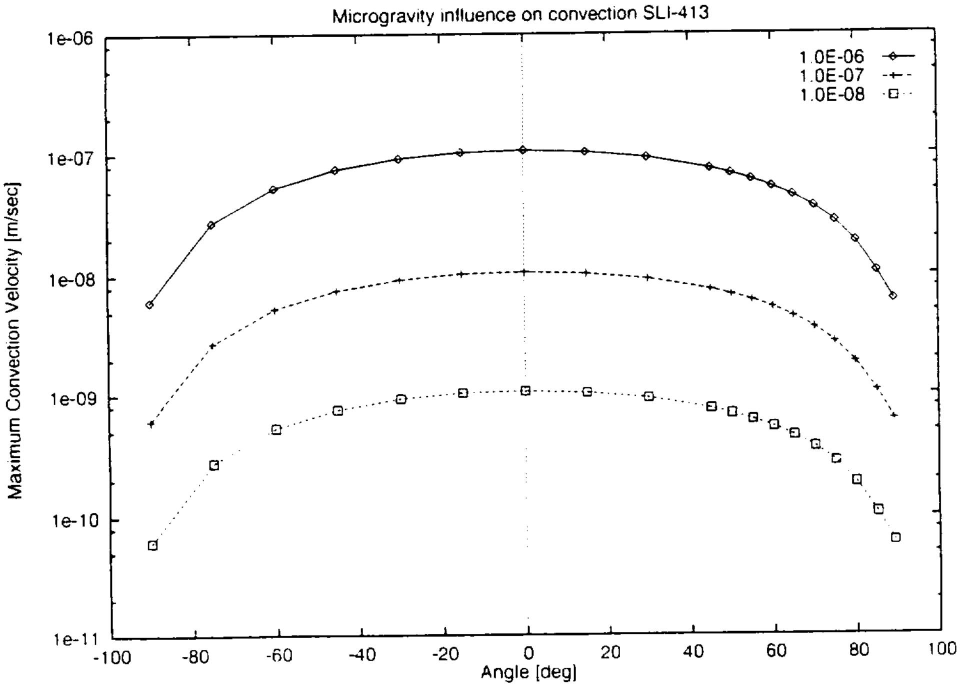

Figure 3.
Maximum convection velocity in the HgCdTe melt as function of gravity vector inclination. Angle of 90 degree corresponds to the strict vertical orientation of the sample.

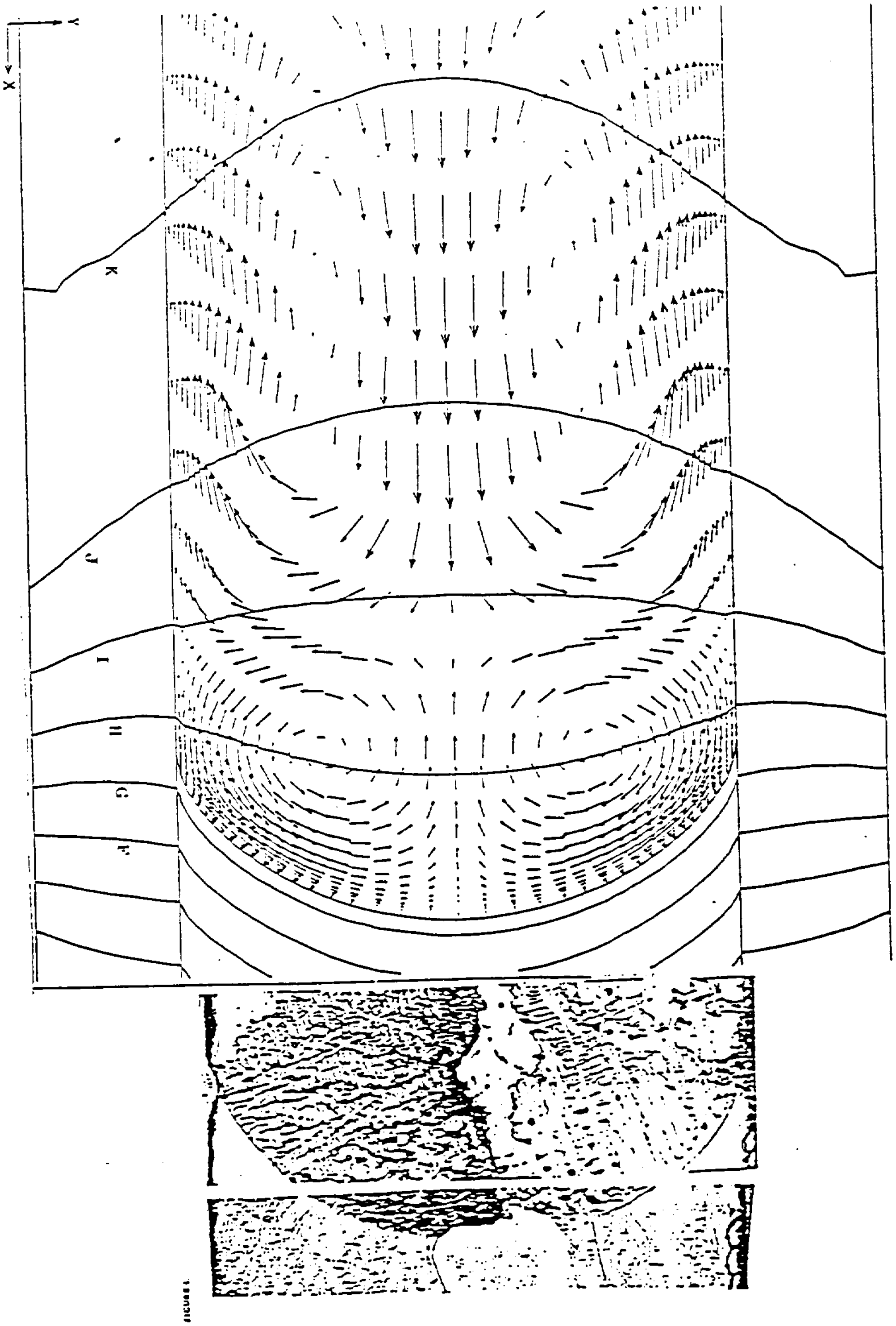

Figure 2.

a) Temperature field and convection patterns in the case of strict vertical orientation of the sample.

b) Experimental photograph of the interface shape. Case MCT-16Q.