A Model of Binary-Alloy Solidification in the Gravitational Field

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ABSTRACT

The paper presents a theoretical model of binary-alloy solidification where gravity acts as the driving force of the convection in the melt and affects chemical diffusion as well. The significance of the model for studying solidification processes under different gravity conditions is discussed. A simplified version of the model has been used to simulate the solidification of a two-dimesional sample of the pseudo-binary mercury-cadmium-telluride alloy. The influence of gravity on the chemical diffusion was studied and a series of numerical experiments was performed which show the effects of the accuracy with which the material parameters have been measured.

INTRODUCTION

The solidification processes are driven by heat and mass transport. The transport itself can be due to the conduction of heat and the diffusion of solute and, in addition, convection and radiation may be involved. The phenomenological models of processes of this type should contain the balance equations

for mass, momentum, and energy, the constitutive relations for generalized fluxes, and the equations of state. We restrict ourselves to the constitutive relations which follow from the axioms of non-equilibrium thermodynamics (Vodák, 1978). Using the hypothesis of local thermodynamic equilibrium we express equations of state from the equilibrium phase diagram of the binary alloy under consideration.

We distinguish three phases in the system undergoing solidification: solid, liquid, and inter-phase (called mushy-zone by some authors), the last of them being understood as a mixture of solid and liquid. There are two main phenomenological approaches to modelling such a system. First, there is a generalization of the classical Stefan problem where the inter-phase is modelled as a discontinuity surface (Vodák, 1982). The other approach formulates the balances globally over the whole region occupied by the alloy and treats them in a distributional (weak) sense. A useful example of the model of this type has been presented by Alexiades, Wilson and Solomon (1985). Our theoretical model uses similar approach and, in addition, takes the convective phenomena and the influence of gravitational field into account.

A MODEL OF BINARY-ALLOY SOLIDIFICATION

Basic assumptions and the balance equations

We consider a binary alloy consisting of components A and B supposing it to be capable of forming solid solutions in all proportions. The alloy is assumed to be situated in an exterior gravitational field. The mass transfer due to diffusion and heat transfer due to conduction and radiation are taken into account in the whole system under consideration. In addition, convective heat and mass transfer is supposed to occur in the liquid and the inter-phase. We assume that these two phases are quasiincompressible Newtonian fluids. Hence, the Boussinesq approximation and the Navier-Stokes law for the stress tensor may be applied there. However, since we suppose no convection in the solid phase, the Navier-Stokes law may be applied even there formally.

We further assume that there are no chemical reactions and that the interactive forces between the components of the alloy are negligible. Finally, we model the alloy as an isotropic material.

Theoretical studies of alloys employ the model of a mixture usually (Atkin and Craine, 1976a, b) and this is our approach as well. The binary alloy in a solid phase (S), liquid phase (L), and an inter-phase (I) is then described by the respective continua $E^k, k = S, L, I$, each of which may be viewed as a superposition of two one-component continua $E_\alpha^k, \alpha = A, B$. Due to space limitations it is impossible to present the derivation of the model here, the

interested reader being referred to the paper by Vodák, Černý and Přikryl (1992). We only list some assumptions on which the model is based and indicate the main lines of its development.

We start with formulating the basic conservation laws for the continua in question taking no care about the phase change process temporarily. The mass balance is written in the form

$$\text{div } \mathbf{v}^k = 0, \tag{1}$$

$$\rho^k = \bar{\rho}^k - \bar{\rho}^k \alpha^k (T^k - \bar{T}^k) - \bar{\rho}^k \gamma^k (C_B^k - \bar{C}_B^k), \quad k = L, S, I, \tag{2}$$

where $\mathbf{v}$ is velocity, ρ density, α and γ are expansion coefficients, T is temperature, C_B concentration of the component B, and the bar denotes appropriate reference values. Since the incompressibility of E^k does not imply that of E_α^k we write the mass balance equations for the individual components of E^k as well. We get

$$\rho^k \frac{dC_\alpha^k}{dt} + \text{div } \mathbf{j}_\alpha^k = 0, \quad k = L, S, I, \tag{3}$$

where C_α denotes the concentrations, t is time, $\mathbf{j}_\alpha$ the diffusion fluxes and d /d t denotes the material derivative.

To obtain the balance of momentum in the liquid phase we use the "weak diffusion approximation", suppose that the pressure gradient is due to the weight of the fluid only, apply the Navier-Stokes law and linearize. Assuming formally that the coefficient of dynamical viscosity of the solid phase is infinitely large and supposing that the inter-phase is a fluid whose properties are determined by the lever rule from the corresponding properties of the solid and liquid phases, we may write the balance of momentum in the form

$$\rho^k \frac{d\mathbf{v}^k}{dt} = \text{div } (\eta_v^k \text{grad } \mathbf{v}^k) - \bar{\rho}^k \mathbf{g}[\alpha^k (T^k - \bar{T}^k) + \gamma^k (C_B^k - \bar{C}_B^k)], \quad k = L, S, I. \tag{4}$$

Here, η_v denotes the coefficient of dynamical viscosity and $\mathbf{g}$ is the acceleration of gravity. The balances of momentum for the individual components E_α^k, $\alpha = A, B$, are not taken into account since they would be combinations of the balance written for the mixture and a constitutive relation for the diffusion flux which we introduce later.

Writing the balance of internal energy we employ the results of Atkin and Craine (1976a) in the liquid and inter-phase. We neglect the viscous dissipation of the energy as most authors do and assuming $\mathbf{v}^S = 0$, $\mathbf{v}_\alpha^S \approx 0$ we write the same balance for the solid as well. In addition, we suppose the alloy to be diluted so that $C_B^k << 1$ and denoting the internal energy density by u we finally arrive at

$$\rho^k \frac{du^k}{dt} + \text{div } (\mathbf{q}^k + \eta^k \mathbf{j}_B^k) + (C_A^k)^{-1}(\mathbf{j}_B^k \cdot \mathbf{g}) = \Phi^k, \quad k = L, S, I, \tag{5}$$

where **q** is the heat flux, $\eta = h_B - h_A$ is the difference of the specific enthalpies and Φ stands for radiation heat sources. Note that owing to our assumptions we have $\mathbf{j}_B^S \approx 0$ in the solid.

The phenomenological laws of linear (irreversible) thermodynamics yield the constitutive relations

$$\mathbf{j}_B^k = -\rho^k D^k \text{grad } C_B^k + M^k \mathbf{g} + \rho^k \delta^k \text{grad } T^k, \tag{6}$$

$$\mathbf{q}^k = -k^k \text{grad } T^k + \beta^k \text{grad } C_B^k - \frac{\beta^k M^k}{\rho^k D^k} \mathbf{g}, \quad k = L, S, I, \tag{7}$$

where D is the diffusivity of component B, M is the mobility, δ the Soret, β the Dufour coefficient, and k is the thermal conductivity. We note that the mobilities are introduced as

$$M^k = \frac{\rho^k D^k}{C_A^k} \left(\frac{\partial \mu^k}{\partial C_B^k} \right)^{-1}, \tag{8}$$

where μ denotes the chemical potential. To provide consistency with our previous assumptions we suppose that $D^S, M^S, \delta^S \approx 0$ and, moreover, that $\beta^S M^S / D^S \approx 0$.

After substitution for $\mathbf{j}_B^k$ and $\mathbf{q}^k$ from (6–7) we obtain the final set of balance equations valid in each of the phases ($k = L, S, I$). These balance equations consist of (1–2), (4), and

$$\rho^k \frac{dC_B^k}{dt} = \text{div } (\rho^k D^k \text{grad } C_B^k) - \text{div } (M^k \mathbf{g}) - \text{div } (\rho^k \delta^k \text{grad } T^k), \tag{9}$$

$$\rho^k \frac{du^k}{dt} = \text{div } \left[(k^k - \eta^k \rho^k \delta^k) \text{grad } T^k \right] + \text{div } \left[(\eta^k \rho^k D^k - \beta^k) \text{grad } C_B^k \right] +$$

$$\text{div } \left[M^k \left(\frac{\beta^k}{\rho^k D^k} - \eta^k \right) \mathbf{g} \right] - (C_A^k)^{-1} \left[(-\rho^k D^k \text{grad } C_B^k + \rho^k \delta^k \text{grad } T^k) \cdot \mathbf{g} \right] -$$

$$(C_A^k)^{-1} M^k \mathbf{g} \cdot \mathbf{g} + \Phi^k. \tag{10}$$

To complete the model it is necessary to add the Gibbs relation and to specify the equations of state.

The Gibbs relation and equations of state

Developing the conservation laws and constitutive relations we have not paid attention to the phase change process up to now. The particular phases can only be distinguished by introducing the equation of state which would be subject to the phase diagram and the Gibbs relation. Modelling the process

at a macroscopic level, we employ the assumption of local thermodynamic equilibrium and suppose the local validity of equilibrium phase diagram.

Consider an elementary cell P which is small enough to possess nearly uniform properties so that it belongs to one of the three continua, E^L, E^S, E^I. Assume for simplicity that the liquidus and solidus curves can be written as $C_B = f^k(T)$, $k = L, S$, where f^k are continuous functions. For definitness we suppose that $P \in E^L \Leftrightarrow C_B \leq f^L(T)$, $P \in E^S \Leftrightarrow C_B \geq f^S(T)$, and $P \in E^I$ otherwise.

Being not a pure phase, E^I is viewed as a mixture of liquid and solid and it is treated as a superposition of E^L and E^S. We apply the "lever rule" and introduce the liquid fraction λ as

$$\lambda = \begin{cases} 1 & \text{for } P \in E^L, \\[2mm] \frac{f^S(T) - C_B}{f^S(T) - f^L(T)} & \text{for } P \in E^I, \\[2mm] 0 & \text{for } P \in E^S \end{cases} \qquad (11)$$

in accordance with Alexiades, Wilson and Solomon (1985). Then, any specific quantity X^I defined in the inter-phase can be written as $X^I = \lambda X^L + (1 - \lambda) X^S$. Owing to the definition of λ this equation holds in E^L, E^S as well.

In the liquid or solid the Gibbs relation reads

$$du = T \, ds - \sum_i A_i \, da_i, \qquad (12)$$

where A_i are generalized forces (the pressure p and chemical potential $-\mu$ in our case), a_i generalized coordinates (the volume V and concentration C_B), and s is the specific entropy. However, we want to treat the specific internal energy as a function of T, V, C_B instead of s, V, C_B and we thus transform (12) into

$$du = c_V \, dT - \left[p - T \left(\frac{\partial p}{\partial T} \right)_{V, C_B} \right] dV + \left[\mu - T \left(\frac{\partial \mu}{\partial T} \right)_{V, C_B} \right] dC_B, \qquad (13)$$

where c_V denotes the specific heat under constant volume, using some standard thermodynamic relations.

The next step is to eliminate the derivative $(\partial \mu / \partial T)_{V, C_B}$ from (13). To achieve this we express the Gibbs specific energies as functions of ρ, T, C_B and apply the second Gibbs-Helmholtz equation which expresses the specific enthalpies in terms of these Gibbs potentials. We thus arrive at

$$du = c_V \, dT - \left[p - T \left(\frac{\partial p}{\partial T} \right)_{V, C_B} \right] dV + \left[\eta - T(V_B - V_A) \left(\frac{\partial p}{\partial T} \right)_{V, C_B} \right] dC_B, \qquad (14)$$

where η is the above introduced difference of specific enthalpies. Since we have assumed local thermodynamic equilibrium and our equation of state is being

established from the equilibrium phase diagram we may put $dp = 0$ locally. Using now a generalized form of the Gibbs-Duhem equation we obtain, after some manipulations, the Gibbs relation for the liquid and solid phase in its final form

$$du^k = c_p^k \, dT - p^k \, dV^k + \eta^k \, dC_B, \quad k = L, S, \tag{15}$$

where c_p denotes the specific heat under constant pressure.

In the inter-phase, we apply the lever rule, differentiate, and express du^I as combination of $du^L, du^S, d\lambda$. Substituting for du^L, du^S, expressing the specific properties by the lever rule and taking the assumption $dp = 0$ locally into account we obtain $(k = I)$

$$du^k = c_p^k \, dT - p^k \, dV^k + \eta^k \, dC_B + L_M \, d\lambda^k, \tag{16}$$

where L_M is the latent heat of melting. Since $d\lambda = 0$ in the liquid or solid the relation (16) is valid in each of the phases, $k = L, S, I$, in fact. Recalling the process is isobaric locally we note that $V = V(T, C_B)$, $\lambda = \lambda(T, C_B)$ so that (16) can be given the form

$$du^k = \left(c_p^k + L_M \frac{\partial \lambda^k}{\partial T} - p^k \alpha^k V^k\right) dT + \left(\eta^k + L_M \frac{\partial \lambda^k}{\partial C_B} - p^k \gamma^k V^k\right) dC_B. \tag{17}$$

The equation of state in the form $u = u(T, C_B)$ could now be obtained by the same integration procedure as used by Alexiades, Wilson and Solomon (1985).

The model of solidification

Combining the balance equation and the Gibbs relation obtained we arrive at the following system of equation that the unknown quantities $\rho, \mathbf{v}, T$ and C_B should satisfy:

$$\rho = \bar{\rho}[1 - \alpha(T - \bar{T}) - \gamma(C_B - \bar{C}_B)], \tag{18}$$

$$\text{div } \mathbf{v} = 0, \tag{19}$$

$$\rho \frac{d\mathbf{v}}{dt} = \text{div } (\eta_v \text{grad } \mathbf{v}) - \bar{\rho}\mathbf{g}[\alpha(T - \bar{T}) + \gamma(C_B - \bar{C}_B], \tag{20}$$

$$\rho \frac{dC_B}{dt} = \text{div } (\rho D \text{grad } C_B) - \text{div } (M\mathbf{g}) - \text{div } (\rho \delta \text{grad } T), \tag{21}$$

$$\rho \left(c_p + L_M \frac{\partial \lambda}{\partial T} - p\alpha V\right) \frac{dT}{dt} = \text{div } [(k - \eta\rho\delta) \text{grad } T + (\eta\rho D - \beta) \text{grad } C_B] +$$

$$\left(\eta + L_M \frac{\partial \lambda}{\partial C_B} - p\gamma V\right) [\text{div } (\rho \delta \text{grad } T) + \text{div } (M\mathbf{g}) - \text{div } (\rho D \text{grad } C_B)] +$$

$$\operatorname{div}\left[M\left(\frac{\beta}{\rho D}-\eta\right)\mathbf{g}\right]-(C_A)^{-1}(-\rho D\operatorname{grad} C_B+M\mathbf{g}+\rho\delta\operatorname{grad} T)\cdot\mathbf{g}+\Phi. \quad (22)$$

This system of equations is understood to hold in distribution sense in the entire domain occupied by the alloy. The material properties $\bar{\rho}, \alpha, \gamma, \eta_v, D, k, \delta$, β, M, L_M and the heat sources or sinks are to be determined experimentally. Furthermore, the equilibrium phase diagram is supposed to be known so that it is possible to express $\lambda = \lambda(T, C_B)$. The initial and boundary conditions for equations (18–22) follow from the experimental situation modelled. After solving the corresponding initial-boundary value problem for $\rho, \mathbf{v}, C_B$, and T, the location of the inter-phase can be determined as the subdomain where $0 < \lambda < 1$.

COMPUTER SIMULATION WITH SIMPLIFIED MODEL

A two-dimensional model without convection

The model of solidification based on the system of equations (18–22) is quite complicated and the computing resources we could use had forced us to restrict our study to a considerably simplified version of it. This version is based on three additional assumptions: (i) the density of each of the two basic phases (S, L) is constant, (ii) the convective velocity of the mixture is zero everywhere, (iii) there are no heat sources or sinks. Applying these assumptions to the full version of the model we obtain the following system of partial differential equations

$$\rho\frac{\partial C_B}{\partial t} = \operatorname{div}(\rho D\operatorname{grad} C_B) - \operatorname{div}(M\mathbf{g}) - \operatorname{div}(\rho\delta\operatorname{grad} T), \quad (23)$$

$$\rho\left(c_p + L_M\frac{\partial\lambda}{\partial T}\right)\frac{\partial T}{\partial t} = \operatorname{div}[(k - \eta\rho\delta)\operatorname{grad} T + (\eta\rho D - \beta)\operatorname{grad} C_B] +$$

$$\left(\eta + L_M\frac{\partial\lambda}{\partial C_B}\right)[\operatorname{div}(\rho\delta\operatorname{grad} T) + \operatorname{div}(M\mathbf{g}) - \operatorname{div}(\rho D\operatorname{grad} C_B)] +$$

$$\operatorname{div}\left[M\left(\frac{\beta}{\rho D}-\eta\right)\mathbf{g}\right] - (C_A)^{-1}(-\rho D\operatorname{grad} C_B + M\mathbf{g} + \rho\delta\operatorname{grad} T)\cdot\mathbf{g}. \quad (24)$$

We simulated the solidification process of a two-dimensional sample of the pseudo-binary mercury-cadmium-telluride alloy $(HgTe)_{1-x}(CdTe)_x$, for which the phase diagram (Fig. 1) is known with confidence and many of its thermophysical properties have been measured. We used the temperature and concentration dependent data presented by Alexiades, Geist and Solomon (1985). The numerical method employed was standard semi-Galerkin finite-element procedure with linear isoparametric quadrilateral elements in space.

The resulting system of ordinary differential equations was then discretized by an explicit method using linear finite elements in time (Zienkiewicz, 1971).
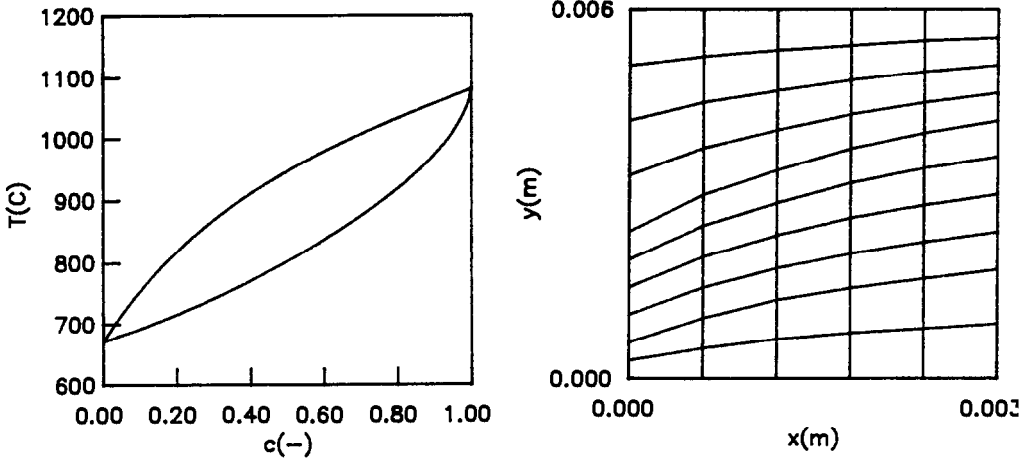

Fig. 1. Equilibrium phase diagram of the alloy.

Fig. 2. The finite-element mesh.

Our numerical experiments were performed on a rectangular domain $R = [0, 0.003] \times [0, 0.006]$ m. The finite element mesh used in the calculations is shown in Figure 2. A Dirichlet boundary condition was prescribed for the temperature at $y = 0$, whereas the remaining sides of the sample were supposed to be heat insulated. Zero mass flux condition was the condition for concentration on the whole boundary of the rectangle. Our initial and boundary conditions implied that the whole sample consisted of the liquid phase only at $t = 0$. The gravitational field was supposed to act in the reversed direction of the y-axis. For $t > 0$, the sample starts to solidify from the lower end upwards due to the boundary conditions prescribed for $y = 0$. The solidification proceeds faster at the $x = 0$ end, where the temperature is lowest. The starting finite-element mesh deforms in the course of time, reflecting the changes in the domains occupied by the individual phases.

The purpose of the computational experiments performed was to study the time development of the temperature and concentration fields under various gravity conditions. In addition, we studied the influence of certain material parameters on the solidification process. A more detailed description of the results obtained will be published elsewhere. Here, we present some of the results for illustrative purposes only.

Results of computational experiments

Figure 3 shows the history of the solidification process at the cross section $x = 1.2 \times 10^{-3}$ m under terrestrial conditions. The development of the domains occupied by the particular phases can be seen in the figure. At the beginning of the process (up to $t = 5$ s approximately) the mushy-zone is very narrow (about 1/100 of the sample height). Then, it becomes still wider and wider, and at $t = 450$ s it occupies 2/3 of the sample and the liquid phase vanishes. Further, the solidification proceeds rather slowly so that the entire sample is solid only at $t = 11050$ s.
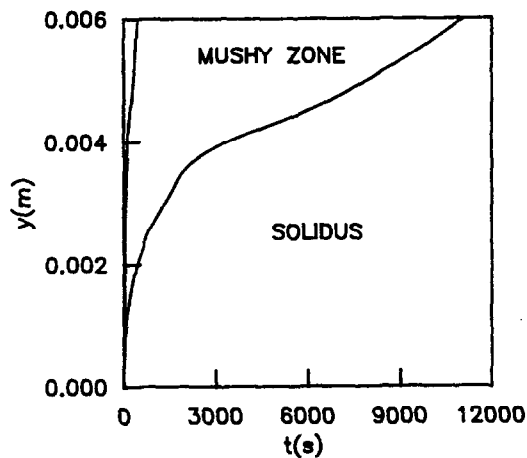

Fig. 3. History of the solidification process.

Figure 4 contains the compositon profile along the above cross section at $t = 5.15$ s. The profile exhibits marked peaks at the values of y corresponding to the phase boundaries. There are almost no changes in concentration in the liquid phase which forms some 90% of the sample at that time. Later, the function $C_B(y)$ becomes smooth and at $t = 1900$ s the composition profile is almost constant. Both the corresponding temperature profiles are in Figure 5. The profiles exhibit distinct jumps in the temperature gradients at the phase boundaries, quite in accordance with the underlying theory.

Studying the influence of a dynamic gravitational fields in the solidification of our sample we considered two cases. First, we used a field which was continuous in space and time, second, we considered discrete local changes of gravity. The results obtained for various values of $g = |\mathbf{g}|$ were compared with those obtained for $g = g_0 = 9.81$ m s^{-2}. The results of our calculations have

shown that it would be necessary to take $g = 10^8 g^0$ to change the reference concentration and temperature fields by 0.1%. This confirms the theoretical studies of the stability of the phase interface under microgravity conditions performed by Přikryl and Vodák (1984), which have indicated that the influence of different gravity conditions on the diffusion of solute itself cannot account for changes in the structure of the solidifying material.

Studying the influence of thermophysical parameters we concentrated on those which are usually determined inaccurately (D^L, c_p^L) or on those parameters that are even not measured at all and their influence is mostly neglected (M^L, η, δ^L). Our numerical experiments revealed that the accuracy in M^L and η is not critical, the effect of these two material properties being of second order apparently. On the other hand, changing D^L or c_p^L by 50% resulted to up 7% changes of the respective concentration or temperature field in a rather short 5s interval of time. In addition, the rate of solidification exhibited marked changes: having increased D^L ten times we observed the phase interfaces to move 4 to 6 times faster.
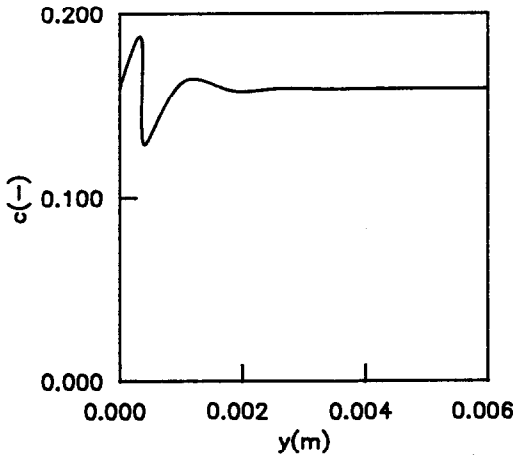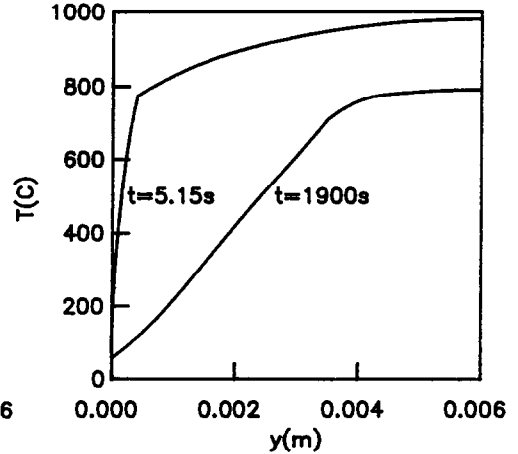

Fig. 4. Composition profile.

Fig. 5. Temperature profiles.

The accurate determination of the Soret coefficient δ^L has proved to be of importance in our experiments. Putting $\delta^L = 5 \times 10^{-12}\,\mathrm{m^2 s^{-1} K^{-1}}$, i.e. $\delta^L = 10^{-3} D^L$, instead of $\delta^L = 0$, we observed up to 30% changes in concentration and 5% changes in temperature at $t = 5\,\mathrm{s}$. In this short time interval the average velocity of the solid/mushy-zone interface was almost twice so large as with zero Soret, whereas the average velocity of the liquid/mushy-zone interface was even 3.5 times so large.

Finally, we studied the effect of the concentration and temperature dependency of the material parameters on the results, comparing the values obtained for the case where the material parameters are concentration and temperature dependent and for the case where they are constant. For the respective constant values we took those corresponding to the solid/mushy-zone interface in the solid phase and those corresponding to the liquid/mushy-zone phase boundary in the liquid. Again, at $t = 5$ s we observed distinct changes in concentration (up to 5%) and in the liquid temperature (about 10%). Moreover, about 20% changes in the rate of solidification were seen.

CONCLUSIONS

The results of the experiments with solidification in space imply that the effect of gravity conditions should be included into the theoretical and computational models of solidification processes. The model presented in this paper enhances the well-known model constructed by Alexiades, Wilson and Solomon (1985) by introducing convective heat and mass transport as well as taking the forced diffusion due to gravitational forces into account.

The results of numerical experiments performed with a simplified version of the model have shown that changes in convective mechanisms play the important role in space experiments, whereas the influence of gravity on the chemical diffusion is a second-order effect. Hence, to be able to simulate space experiments adequately it is necessary to employ the full version of the model despite of its theoretical and practical complexity. This will be the subject of our further research.

However, even the numerical experiments with the simplified version have shown that the inadequate accuracy of material parameters may bias the results of computer simulation to such an extent that our effort to model the influence of the gravitational fields and its possible dynamic changes could not be successful. The results of the computations have shown that the simulation can be seriously biased not only by insufficient accuracy in determining the "primary" material properties (k, c_p, or D) but that also the Soret coefficient δ^L can have marked influence on the results obtained. The usual neglection of thermodiffusion in models of this type is thus questionable.

Another important aspect of our experiments consisted in the use of temperature and concentration dependent material properties. This has shown to be quite important and our computations have revealed that neglecting this dependency may lead to serious errors.

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