A generalized algorithm for modelling phase change problems in materials processing

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Phase change problems form an integral part of several manufacturing and materials processing applications. Such problems are computationally challenging because of the presence of multiple length scales and morphologically complex interfaces. In this work, we address some of those challenges through a generalized algorithm for computational modelling of solidification processes of binary alloys along with the associated transport phenomena. Different approaches for modelling phase change problems are discussed, and a fixed grid enthalpy-based model is presented. The phase change model is integrated with a pressure-based finite volume algorithm to obtain a complete solution of alloy solidification along with the associated flow, heat and mass transfer. Special algorithms related to non-equilibrium solidification issues are also discussed.

1. Introduction

WITH the advancement of technology, there is a growing need for developing new materials and manufacturing techniques. Quite often, thermal issues are dominant factors in the development and improvement of a broad variety of manufacturing processes. The examples are numerous, including new fabrication and bonding techniques applied in the microelectronic manufacturing industry, materials joining processes for traditional as well as new advanced materials, micro-joining processes for the manufacture and assembly of electronic devices, high energy beam (such as laser) manufacturing processes, and also in the growth of semiconductor crystals. Frequently, one is faced with the challenging task of modelling and analysis of various transport phenomena (fluid flow, heat, and mass transfer) involved in manufacturing and materials processing. Such problems are computationally challenging and require special algorithms to address the presence of multiple length scales and morphologically complex interfaces during phase change. The problems are often CPU intensive, too, because of their inherent three dimensionality and high degree of nonlinearity.

In each of the above applications, and in several others, the process primarily involves one or more physical and/or chemical changes through heat addition or removal to the feed material(s) and to produce a product of desired composition, properties, and physical state. In order to achieve a desired quality of the final product, it is essential to predict the microstructure and physical properties based on input parameters of the process. This necessitates a complete thermo-fluid analysis of the process. For most of the processes mentioned above, there are several features that are common such as melting and solidification, heat and fluid flow, species transport, microstructure development, and so on. Hence, a unified approach is desirable for thermal analysis and modelling of many of the above processes in order to establish a correlation between the thermal behaviour, microstructure development and the final properties of the resulting product.

Among the various thermo-physical processes in materials processing, phase change (especially solidification) is, perhaps, the most complex one in terms of physical understanding and computational modelling. The modelling of it is also the most important since it directly affects the prediction of microstructure development and physical properties of the final product[1]. However, there are several computational challenges involved in phase change modelling, such as the presence of distinct length scales (macroscopic and microscopic). Tracking a complex interface shape and its dynamics is also a difficult task, especially for binary and higher order mixtures. The aim of the present work is to present a generalized algorithm for computational modelling of solidification processes of binary alloys along with the associated transport phenomena. This would allow us to model a large class of solidification problems under the same framework. Special numerical algorithms have been formulated to capture the physics of the models of solidification which contain important features of non-equilibrium effects, convection-modified partitioning, rationalization of latent heat function, and so on.

2. Physical and computational issues

To appreciate the kind of physical issues involved, we consider the case of crystal growth. During crystallization, the interfaces separating the phases present can undergo morphological changes having serious technological implications. Such changes in morphology can contribute to the inhomogeneous distribution of the solute that is re-

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jected from the solid and can be greatly enhanced by non-equilibrium phase change effects. The rejected solute accumulates in the regions around the irregularly shaped phase front, which may result in both microscopic and macroscopic irregularities in the solute distribution. This phenomenon of solute rejection and the associated transport is called macro- or micro-segregation, depending on the scale at which such processes occur[2]. Thus, in such moving boundary problems, not only are the transport of momentum, energy and species coupled, but also the formation, evolution, and dynamics of the interface play major roles in defining the behaviour of the system. Apart from the inherent nonlinearity of these diverse phenomena, the interfacial deformation in itself can be a highly complex and intractable feature. Coupled with these, the physical phenomenon of phase change brings in certain instabilities of flow. It is quite obvious that such an involved physical process occurring in a wide range of length scales (both macroscopic and microscopic) can pose serious challenges on the mathematical modelling and numerical techniques to solve the problem.

The difficulties associated with the computer simulation of such problems are of varied nature. One of the major difficulties is that there are two distinct length scales involved (namely macroscopic and microscopic) for the entire physical problem. To illustrate the relevant issues clearly, let us consider the solidification of an ammonium chloride–water solution in a square cavity as shown in Figure 1. This binary mixture in the configuration shown is commonly used for such basic studies on solidification. The solidification of such a substance takes place over a range of temperature as determined by its phase diagram (Figure 2). Hence, instead of a sharp solid–liquid interface we have an interfacial region, the state of which varies from a pure liquid at the liquidus temperature to a pure solid at the solidus temperature. The morphology of this region (popularly known as the 'mushy' region) depends on several factors such as the composition, cooling rate and temperature gradient. The morphological structure, growth and evolution of the mushy region is at a microscopic scale, while the overall shape of the interface and transport within the bulk fluid region is at a macroscopic scale. Hence the computational technique, in addition to solving for fluid flow, heat and mass transfer, must be capable of performing the following two important tasks: (1) determining the complex and dynamic interface shape, and (2) handling both the length scales.

3. Mathematical and computational modelling

In our present work, we aim to outline the modelling and computer simulation for prediction of transport phenomena associated with binary alloy solidification, using a generalized approach. The development of a general-purpose algorithm satisfying the above requirements starts with the formulation of the problem in terms of the governing differential equations along with the initial and boundary conditions. Most significantly, it is observed that all the dependent variables of interest seem to obey a generalized conservation principle. If ϕ is a general scalar variable for which we write a governing differential equation arising out of a conservation law, the general differential equation can be cast in the following form:

$$\frac{\partial}{\partial t}(\rho\phi) + \nabla\cdot(\rho \vec{u}\phi) = \nabla\cdot(\Gamma\nabla\phi) + S, \quad (1)$$

where ρ is the density of the medium, u is the velocity, Γ is the diffusion coefficient and S is the source term. Both Γ and S are specific to the particular meaning of ϕ used in eq.

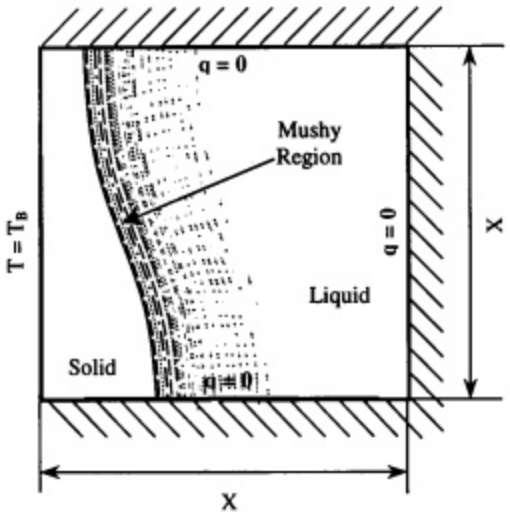

Figure 1. Schematic diagram of the model problem.

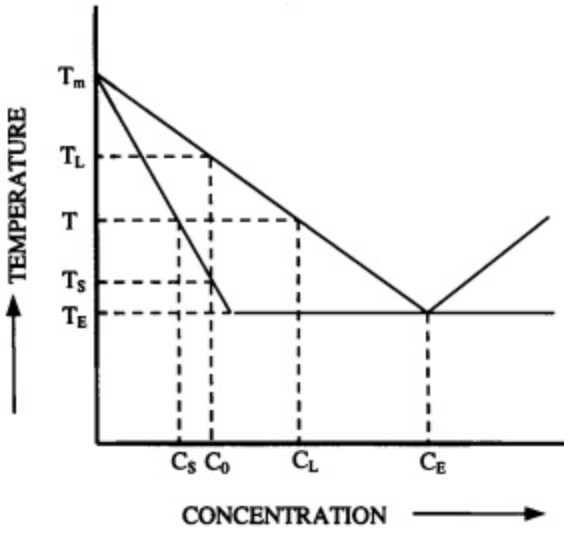

Figure 2. A typical linearized phase diagram.

(1) as well as the physics of the problem. Substituting for the appropriate ϕ in eq. (1), we can write equations for the conservation of mass, momentum, energy and species. As a consequence, we need to consider the numerical solution of equations only in the form of eq. (1). Special algorithms may need to be developed to model phase change phenomena and incorporate microscopic issues in our macroscopic framework. We first present below a method of solution of the general transport equations, followed by special issues related to alloy solidification problems.

3.1 *Solution of transport equations*

In the construction of a general purpose computer code for numerical simulation of eq. (1), it is sufficient to write a sequence of instructions for solving eq. (1), which can be repeatedly used for different meanings of ϕ along with appropriate expressions for Γ and S. The task then reduces to the development of suitably coupled discretization equations along with their solutions.

In the applications that are relevant for this study, it is important that the conservation laws in their integral form are represented accurately. The most natural method to accomplish this is to discretize the integral form of the conservation equations rather than the differential form, in order to satisfy the overall conservation. This is the basis of the finite volume method (FVM). By decoupling of volumes and computational cells, the freedom in the determination of the functional representation of the field variables in the FVM is much larger than in the case of finite element method (FEM) or finite difference method (FDM). Since the discretization is performed using an integral form of the governing equations, conservation is always preserved, unlike in the case of FDM which involves truncation error in the discretization of derivatives. Moreover, the discretization process in FVM is directly related to the physics of the problem, instead of using variational formulation or functionals (as employed in FEM) which have no easy physical interpretation in problems involving fluid flow and diffusion. It is this particular combination of formulation of a flow problem over control volumes with the geometric flexibility in the choice of grids as well as the flexibility in defining the discrete flow variables, that makes FVM suitable for solution of such physical problems involving fluid flow, heat and mass transfer. Details of the method can be found in textbooks such as Patankar[3].

Consistent with the choice of our computational technique as FVM, it is important to choose a solution scheme accordingly. Since the equations relevant to this class of problems are highly nonlinear, direct methods for solution of the algebraic equations (arising out of discretization) would be difficult and would require a large amount of computer storage and time. Hence, for two- and three-dimensional problems, an efficient iterative scheme, popularly known as 'line-by-line tridiagonal matrix algorithm' is employed. With this method, the boundary information is quickly transmitted to the interior of the domain by direction alteration of 'sweeping', resulting in quick convergence. Also, as an aid for handling nonlinearities, controlled convergence can be achieved by the introduction of suitable under-relaxation and over-relaxation parameters in the iterative scheme.

3.2 *Special issues related to alloy solidification*

Once the broad issue of selection of the computational method and solution scheme is settled, the next task is to look into special issues relevant to the particular class of problems and seek a formulation that is compatible with the general framework of the broad computational technique. In the context of phase change problems in materials processing, two such techniques parallely exist, namely the moving grid techniques and the enthalpy-based fixed grid techniques. In the moving grid technique, an explicit tracking of the interface boundary is a necessity. Since the shape of the interface can be highly complex in the presence of convective melt flow even for the case of a pure substance undergoing isothermal phase change, such a front-tracking technique is computationally too involved (requiring extremely fine grids and very small time-steps). Moreover, in the case of binary or higher order mixtures, the phase change does not take place isothermally, leading to the formation of a two-phase (or multiple phase) zone instead of a distinct interface. It would be virtually impossible to track such a morphologically complex zone in a macroscopic framework using any moving grid technique. Since most materials processing activities involve more than one substance, a fixed grid enthalpy-based formulation seems to be most suitable for the present scope of work. The detailed development of such a model is quite involved, and can be found in the literature[1,4]. The basic principle is to volume-average individual phase equations based on a classical continuum mixture theory so that we can arrive at an equivalent single phase equation in the form of eq. (1).

As discussed earlier, one of the major difficulties in the modelling of alloy solidification is the presence of two distinct length scales (microscopic and macroscopic) for the entire physical problem. An effective way of tackling both these scales in a single macroscopic framework is to model the two-phase mushy region as a porous medium, as described below.

In order to model the velocity field, ϕ in eq. (1) is replaced by u or v, for horizontal and vertical momentum balance, respectively. The source terms in these equations contain the following additional term (according to Darcy's model):

$$S_i = -\frac{\mu_l u_i}{K\rho_l}, \tag{2}$$

where u_l represents viscosity of the liquid, u_i is the respective velocity component along x_i, and K represents the permeability of the medium. The permeability is assumed to vary in the domain according to the local liquid fraction in the following manner:

$$K = K_0 \frac{g_l^3 + b}{(1-g_l)^2}. \tag{3}$$

Equation (3) represents the well-known Carman–Kozeny relation[1], in which K_0 stands for the isotropic permeability coefficient (a small number ~ 10^{-8}) of the medium, g_l is the local liquid volume fraction, and b is a small number to avoid division by zero. Effectively, g_l is the porosity of the medium, and is evaluated as the ratio, $\Delta H/L$ where ΔH is the latent heat content of the computational cell under consideration and L is the latent heat of fusion of the substance. In the fully liquid region, the value of g_l is 1, making K in eq. (3) very large, thus rendering S_i in eq. (2) zero (i.e. no porous medium-like resistance is offered). On the other hand, in the fully solid region ($g_l = 0$), K becomes a small number, thus forcing S_i in eq. (2) to be very large. This large source term offers a high flow resistance, making the velocities in the entire solid region effectively zero. In the mushy region ($0 < g_l < 1$), however, the porous medium-resistance varies smoothly from zero at the liquidus to a high value at the solidus, thus making the velocities vary accordingly.

Another potential complication is the treatment of the phase diagram of any general shape in the framework. This is taken care of in our model by the introduction of four mathematical functions, namely solidus, liquidus, inverse-solidus and inverse-liquidus functions. These functions can be derived analytically using thermo-physical considerations, or can be constructed from available experimental data. This enables us to perform suitable functional manipulation and the final result is of the form:

$$k_P = \frac{F_S^{-1} F_L(C_l^*)}{C_l^*}, \tag{4}$$

where k_P is the partition coefficient, F_S and F_L are the solidus and liquidus functions, respectively, C_l is the liquid composition, and the superscript * denotes equilibrium condition. Equation (4) helps to numerically calculate the partition coefficient inside the iteration loops using the current value of C_l^*. Additionally, one must note that the partition coefficient derived using eq. (4) is based on diffusion transport alone. Hence, the above partition coefficient has to be corrected appropriately to incorporate the effects of convection in the mushy region. The convection correction can be made using Burton–Fleming's equation[5], which requires numerical estimation of the interface speed and the diffusion boundary layer thickness.

Probably the most computationally involved portion of our numerical algorithm is the rationalization of the latent heat function which appears in our iterative procedure. The current value of nodal latent heat has to be adjusted such that the difference between the latest predicted value of nodal enthalpy from the energy equation and that determined from phase change considerations is nullified. The latent heat function, which gives the functional variation of nodal latent heat with temperature, can be very complex and nonlinear. To suit our generalized framework, we follow an iterative procedure for updating the nodal latent heat according to the following equation:

$$\left[\Delta H_P\right]_{n+1} = \left[\Delta H_P\right]_n + \frac{a_P}{a_P^0}\left[\left\{h_P\right\}_n - F^{-1}\left\{\Delta H_P\right\}_n\right], \tag{5}$$

where a_p and a_p^0 are the coefficients of the finite volume discretization equation, h_p is the enthalpy at node P, F is the latent heat function, ΔH is the latent heat, λ is a

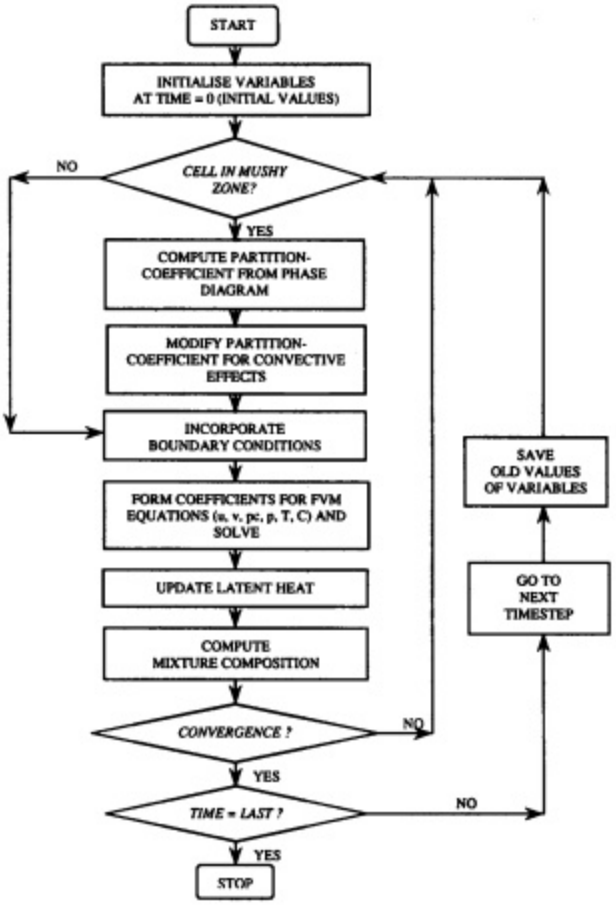

Figure 3. Flowchart of the numerical algorithm.

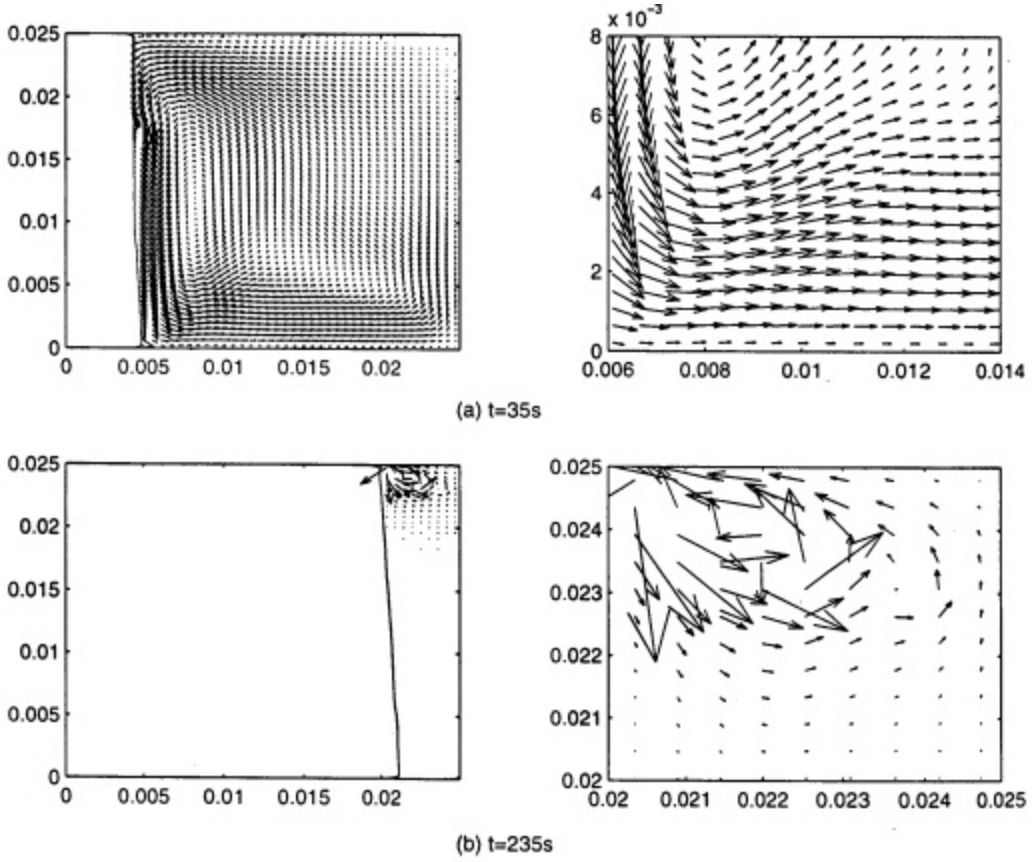

Figure 4. Velocity distribution during alloy solidification. Zoomed boxes on the right indicate macro-segregation.

relaxation factor to prevent possible numerical oscillations and the subscript n denotes the iteration stage. The key factor in successful implementation of such a latent heat update is to devise an algorithm suitable to generate the function F^{-1} for a given class of phase change problems. However, there is a basic requirement to ensure that the mathematically generated functions remain consistent with the physics of the phase change. A flow chart for the overall algorithm is presented in Figure 3.

3.3 *Simulation of solidification of NH_4Cl–H_2O solution in a cavity*

As an illustration, we present here a simulation of solidification in a square cavity with the boundary conditions as shown in Figure 1. The solution is initially in a fully liquid state with the left boundary maintained at a temperature less than the solidus temperature corresponding to the initial liquid composition. Figure 4 shows a typical velocity distribution during solidification of the binary mixture (ammonium chloride solution in water). It is evident from the figure that convection plays a significant role in the solidification process. The model incorporates non-equilibrium effects and convection-correction of the partition coefficient. Such effects play a significant role in enhancing macro-segregation effects, as observed in the zoomed boxes on the right in Figure 4.

4. Concluding remarks

A generalized algorithm for modelling phase change problems is presented. A single domain enthalpy-based method with porosity formulation for the mushy region is found to be suitable and flexible enough to accommodate a large variety of problems. A finite volume-based iterative algorithm is chosen for the solution of fluid flow and associated transport phenomena. Special algorithms are developed to address realistic issues such as non-equilibrium effects and convection-correction of partition coefficient.

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