A numerical approach to compensate for phase field interface effects in alloy solidification

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ABSTRACT

The use of a phase field approach to simulate solidification of metallic alloys has many computational advantages, but if obtaining quantitative results relies on the interface between phases being physically realistic, the computational advantage is much reduced. We propose here a method for compensating for a computationally convenient large interface width by simply transferring a numerically derived 1D steady state anti-trapping current to a general non-steady 2D simulation. The method proposed is not restricted to dilute or ideal materials and has a high degree of interface width independence, illustrated here with two models, illustrating a broad applicability for the approach.

1. Introduction

In phase-field modelling of alloy solidification, applying the variational principle to the Gibbs free energy results in equations for phase, solute and temperature, which optimally minimise the Gibbs free energy, see [1]. The principle is clear and elegant but suffers from the practical disadvantage that the length scale of the solid liquid boundary is far smaller than that associated with solute and temperature diffusion. Consequently, phase field modellers of solidification seek to use a larger than physical interface width to make the mathematical system computationally easier to solve. Simple adoption of a larger interface width, though, reveals that solutions are width dependent, see for example [2]. It is generally accepted that the approach to compensate for this is not to be found in a variational formulation, see [3,4] (though, see the discussion in Appendix C which postulates a variational formulation for including anti-trapping currents). Rather, in an approach initiated by [3], one provides an extra degree of freedom at the level of the partial differential equations post variation by typically matching the phase field equations model to a sharp interface model so that the resulting equations have an element of interface width independence. One feature of the application of matched asymptotic analysis to a sharp interface, e.g. [4,5], is that there is necessarily a degree of approximation used in order to simplify the free energy functional to a point where analysis and comparison with sharp interface models becomes tractable. For example, [6] extends [3,4] to use in multiphase models, but only for the simplest thermodynamics. It is of note that models that use physically realistic free energies for complex materials avoid this approach, e.g. [2,7].

The phase field technique for alloy solidification, as established in simpler form by [8] (WBM) is challenged by two phase modelling as described in [9]. This approach associates a unique concentration field, c_L or c_S, for the liquid or solid phases respectively, and the true concentration field is constructed as a weighted average using the phase field. The quantities c_L and c_S are determined through a concentration equation and, crucially, a constraint. The constraint can take the form of proportionality, using a partition coefficient, or by equating the chemical potential. The latter led [10] to unify the methodology using a grand-potential functional (GPF) in place of the usual free energy. This served also to show that the two phase approach was equivalent to a variational formulation, in particular the equal chemical potential constraint in the two-phase formulation is a natural consequence of the new variational technique based on a GPF.

The GPF methodology has been applied in [11] to dilute alloys, but it is notable here that the GPF approach still requires an anti-trapping current to compensate for interface width, and thus, by implication, the model of [9] would benefit from an anti-trapping current to alleviate interface width dependence. [12] argues that the two phase models, with the constant chemical potential across the interface, needs modification for rapid solidification, and suggest modifications that model this: namely, to replace the constant chemical potential constraint with equations for c_L and c_S. It is of note that [12] uses a physically realistic $\delta = 1.875$ nm in their 1D simulations (and so the method advocated in our paper naturally do not apply here). However, for 2D/3D simulations it is likely that larger interface widths will be computationally expedient and thus some method for compensating for artificial solute trapping will become necessary.

The GPF approach has also been extended to include non-dilute alloys in [13]. Here, central to the application of GPF is a quadratic approximation of the free energies about the equilibrium concentration values (in a multiphase setting), c_{ij}^E. An approach which does not require approximation to the data bases, for general alloy using an entropy functional is found in [14]. This latter approach is motivated by a general free boundary problem formulation and contains the equal diffusion chemical potentials of the two-phase method as a constraint. For more general thermodynamics the authors state that there is potentially a numerical bottle neck due to this constraint.

We choose to adopt and extend the method of [8] to allow quantitative simulation of more general alloys, without recourse to special cases and approximations. It is of note that the application of the WBM approach to general free energy models has only previously been its extension to multiphase models. Consequently, this work represents a first attempt at quantitative modelling, and modelling in itself, for solidification with arbitrary CALPHAD thermodynamics, whilst allowing a conveniently larger than physical interface width. In this sense the method may be seen as both an extension of [3,8], to allow rapid solidification modelling for arbitrary two phase binary alloys.

A constraint on the phase field approach is that the interface must have sufficient resolution to capture the finest curvature found at the solid-liquid boundary. But typically, tip radii, $\rho \gg d_0 \sim 1$ nm where d_0 is the chemical capillary length, being the same order of magnitude as physical interface width, this being the distance over which long-range atomic ordering is lost at the interface between a crystal and its parent melt and which is typically a few atomic radii. Of more concern is the effect of large interface width on solute partitioning where the maximum and minimum values for solute concentration found at the solid-liquid interface are very much interface width dependent. This effect is known as artificial solute trapping, since it is a model dependent effect that tends to drive the partition coefficient closer to unity. Solute trapping also arises naturally in systems where the velocity of growth is sufficiently high, see [15], which analyses three regimes from low to high growth velocity. We propose here an approach which compensates for artificial (interface width induced) solute trapping, for realistically modelled binary alloys at arbitrary concentration.

In outline, the method we propose consists in solving a 1D steady state problem where the solution not only depends on input values for tip speed and tip interface width (and given tanh profile), but also the strength of an *anti-trapping* current, **j**. We seek the strength of **j** in the steady state 1D problem such that the maximum and minimum values for solute, c, within the interface, coincide with the equilibrium values found from the free energy functions for liquid and solid by well known common tangent construction. Once **j** is found from the 1D problem we apply it to the full (non steady) 2D problem. New values for tip speed and width are extracted from the 2D simulation and used, intermittently, to solve the 1d problem, where the new value for **j** is applied thereon.

We find, for the PbSn alloy tested, and to a large extent model of [8], tested to make connection with a standard model, that this approach gives a high degree of interface independence across a range of measures at the crystal tip. The measures used are tip radius, ρ, tip speed, V, and measures for solute partitioning: $\Delta c \equiv c_L - c_S$ and $k \equiv c_S/c_L$, where c_S is the solid concentration near the tip and c_L is the liquid concentration near the tip.

2. Solute trapping in 1D

In this section we focus on a specific phase-field model for alloy solidification, Pb-Sn in this case, in order to introduce the method proposed to compensate for solute trapping. This is based upon looking at the dependence of solute partitioning on interface width in a steady state 1D scenario.

The phase equations governing the evolution of phase, ϕ, (where $\phi = 0,1$ is solid and liquid respectively) and solute concentration, c, on a
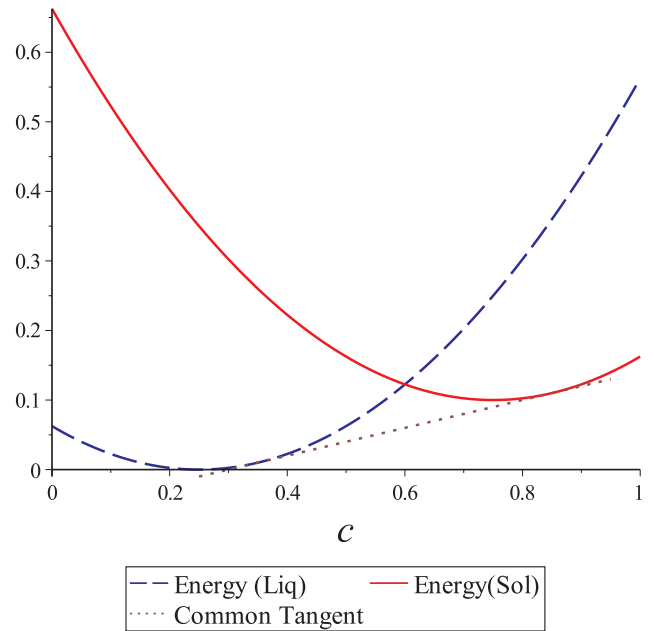

Fig. 1. A constructed example free energy curves, $f^L = (c-0.25)^2, f^S = (c-0.75)^2 + 0.1$ with the common tangent construction that give the equilibrium values of c in the two phases: $c_S^E = 0.85, c_L^E = 0.35$.

domain, Ω, are, respectively (see, for example, [1])

$$\dot{\phi} = -M\frac{\delta F}{\delta \phi},$$ (1)

and

$$\dot{c} = \nabla \cdot D \nabla \frac{\delta F}{\delta c}$$ (2)

where

$$F = \int_\Omega f(\phi, \nabla \phi, c) \, d^3x,$$ (3)

M is the mobility and $D = [\phi D_L + (1-\phi)D_S]c(1-c)/(RTv_m)$, with the liquid and solid diffusivities $D_L \gg D_S$, and R and v_m the molar gas constant and the molar volume respectively. The free energy density, f, is decomposed into a surface part, f_S, and bulk part f_B:

$$f = f_S(\phi, \nabla \phi, c) + f_B(\phi, c)$$ (4)

where f_B combines, by interpolation, the liquid and solid free energy curves, illustrated in Fig. 1 for a simple constructed example. The surface term is

$$f_S = W(c)\left(\frac{\delta^2}{8}\nabla \phi \cdot \nabla \phi + \phi^2(1-\phi)^2\right)$$ (5)

where W is the surface energy of the barrier height between the two phases and δ is a measure of the interface width.

In 1D and at equilibrium Eq. (1) becomes

$$0 = \frac{\delta F_S}{\delta \phi}$$ (6)

where

$$F_S = \int_\Omega f_S \, dx$$ (7)

The phase profile,[1]

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[1] more generally the interface width, and even the general shape is modified by the bulk driving term. Some of our tests imply that the resulting profile is well fitted by a continuous piecewise function using two tanh profiles defined on $\phi \in [0,0.5]$ and $\phi \in [0.5,1]$, respectively, though we cannot assert the generality of this result.