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A phase-field model for the isothermal solidification process of a binary alloy

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We study a phase-field model for the isothermal solidification of a binary alloy which involves the relative concentration and the order parameter. We prove the existence of weak solutions as well as regularity and uniqueness results under Lipschitz and boundeness assumptions for the nonlinearities. A maximum principle holds that justifies these assumptions. A numerical approximation and some numerical results are also presented.

1. Introduction

Phase-field models have become of standard use to describe the solidification of both pure metals [1, 2] and more recently, alloys [11, 12]. One of their main goals is to obtain a regularized description of the phenomena of dendritic growth, which was earlier described by sharp-interface Stefan-like problems [10]. Most past works deal with thermal dendritic growth on pure metals, while we are interested in the description of solutal dendritic growth during the isothermal solidification process of binary alloys. The model we study is very similar to the Warren and Boettinger model [11]. This model involves the relative concentration c and an order parameter ϕ which accounts for the solidification state of the alloy by being equal to 0 if the system is in a solid phase and equal to 1 if it is in a liquid phase. For mathematical analysis, we study an isotropic model. The time evolution of c and ϕ is governed by the following equations:

$$(P)\begin{cases} \alpha\dfrac{\partial\phi}{\partial t} = \varepsilon^2\Delta\phi + F_1(\phi) + cF_2(\phi) & \text{in } \Omega\times(0,+\infty), \quad (1)\\ \dfrac{\partial c}{\partial t} = \operatorname{div}\Big(D_1(\phi)\nabla c + D_2(c,\phi)\nabla\phi\Big) & \text{in } \Omega\times(0,+\infty), \quad (2)\\ \dfrac{\partial\phi}{\partial n} = \dfrac{\partial c}{\partial n} = 0 & \text{on } \partial\Omega\times(0,+\infty), \quad (3)\\ \phi(0) = \phi_0, \quad c(0) = c_0 & \text{in } \Omega, \quad (4)\end{cases}$$

where Ω is an open subset of $\mathbb{R}^d$ with $1 \le d \le 3$ and with boundary $\partial\Omega$, n is the unit normal to $\partial\Omega$ and α, ε are given positive constants. This model is isotopic in that ε is taken as a constant. For physically meaningful numerical simulations, ε is replaced by a function of the local angle θ between the vector $\nabla\phi$ and an arbitrary axis.

The functions F_1, F_2 appearing in (P) are given and satisfy $F_i(0) = F_i(1) = 0$ for $i = 1, 2$, function D_1 is bounded from below by a positive constant and function D_2 is such that $D_2(0,\phi) = D_2(1,\phi) = 0$ for $\phi \in [0,1]$.

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Moreover, initial physical data ϕ_0 and c_0 are given with values between 0 and 1. The solutions c and ϕ of Problem (P) must be found with the same property.

In this paper, we present a number of mathematical and numerical results for Problem (P). First, in Section 2 we begin with a short description of the modelling leading to Problem (P). Then, in Section 3 we investigate the well-posedness of this problem. Section 4 is devoted to the numerical approximation of (P) by means of a finite element method. Finally, in Section 5 we present some numerical results for the isotropic problem (P) as well as for an anisotropic version of Problem (P).

2. Model

We consider a mixture of two pure elements A and B, present both in liquid and solid states inside a domain Ω. The system can be characterized by a relative concentration $c = c(x,t)$ of element B with respect to the mixture, for $x \in \Omega$ and at time t. The relative concentration of element A will then be $1-c$, and the variable c will take values in the interval $[0,1]$. We further characterize the system with an order parameter for solidification, the phase field $\phi = \phi(x,t)$, which also takes values in the interval $[0,1]$. A value of $\phi = 0$ corresponds to a solid region, and a value of $\phi = 1$ to a liquid region. Since we do not want to account for thermal effects, we assume the temperature T to be constant.

To have a thermodynamical description of the evolution of the variables c and ϕ, we introduce a Ginzburg–Landau free energy functional (see [3, 8])

$$F(T,c,\phi) = \int_\Omega \left(f(T,c,\phi) + \frac{\varepsilon^2}{2}|\nabla\phi|^2 \right) \mathrm{d}x, \tag{5}$$

where f is a free energy density and ε a small parameter (in the case of the isotropic model).

The evolution of the quantities c and ϕ for the isotropic model can then be described by the following equations:

$$\begin{cases} \alpha \dfrac{\partial\phi}{\partial t} = -\dfrac{\delta F}{\delta\phi} = \varepsilon^2\triangle\phi - \dfrac{\partial f}{\partial\phi}, & (6) \\ \dfrac{\partial c}{\partial t} = \operatorname{div}\left(\mu\nabla\dfrac{\delta F}{\delta c}\right) = \operatorname{div}\left(\mu\nabla\dfrac{\partial f}{\partial c}\right), & (7) \end{cases}$$

where $(\delta F/\delta\phi)$ and $(\delta F/\delta c)$ stand for the functional derivatives of F with respect to ϕ and c respectively.

Under the hypothesis that the coefficient α is positive and μ is a positive function of T, c and ϕ, the previous equations guarantee a locally time decreasing free energy (second principle of thermodynamics) as well as the conservation of matter (see [11, 12, 5]).

The general form (6)–(7) of solutal phase-field evolution equations is used both by Wheeler, Boettinger and McFadden [12] and Warren and Boettinger [11]. To ensure that we have a thermodynamically consistent model, we shall choose a free energy density $f(T,c,\phi)$ alike to that used by WB [11].

Warren and Boettinger's thermodynamically consistent free energy density [11] is given by

$$f(T,c,\phi) = (1-c)f^A(T,\phi) + cf^B(T,\phi) + \frac{RT}{v_m}\left[(1-c)\ln(1-c) + c\ln(c)\right], \tag{8}$$

where T is a constant, R is the Boltzman constant, v_m is the molar volume and $f^A(T,\phi)$ and $f^B(T,\phi)$ are the free energy densities for the pure elements ($c \equiv 0$ and $c \equiv 1$ respectively). We construct the functions f^A and f^B in a similar way to Warren and Boettinger [11] in order to ensure thermodynamic consistency. We assume that at the melting temperatures T_m^A and T_m^B, functions $f^A(T_m^A,\cdot)$ and $f^B(T_m^B,\cdot)$ are of double-well potential type and that at a given temperature T between T_m^A and T_m^B, functions $f^A(T,\cdot)$ and $f^B(T,\cdot)$ have each only two minima for $\phi \in [0,1]$, namely at

$\phi = 0$ and $\phi = 1$. Using some basic thermodynamical principles, we can obtain a general form for f^A, f^B and then for f at any temperature T. A particular choice for f is a polynomial function in ϕ of at least 5th degree when T is different from the melting temperatures T_m^A and T_m^B. We refer to [5] and [11] for the details.

Thus, we can infer that

$$-\frac{\partial f}{\partial \phi} = F_1(\phi) + cF_2(\phi), \tag{9}$$

where F_1, F_2 are functions of ϕ which vanish for $\phi = 0$ and $\phi = 1$, and

$$\nabla \frac{\partial f}{\partial c} = -F_2(\phi)\nabla\phi + \frac{RT}{v_m}\frac{1}{c(1-c)}\nabla c. \tag{10}$$

Hence, Eq. (6) with (9) becomes

$$\alpha \frac{\partial \phi}{\partial t} = \varepsilon^2 \Delta\phi + F_1(\phi) + cF_2(\phi). \tag{11}$$

Moreover, in order to recover a classical diffusion equation for c whenever $\phi \equiv 0$ or $\phi \equiv 1$ i.e. in a totally solid or liquid phase, we choose

$$\mu = \frac{v_m}{RT}\, c(1-c)\, D_1(\phi), \tag{12}$$

where D_1 is an increasing smooth function such that $D_s \equiv D_1(0) > 0$ and $D_l \equiv D_1(1) > 0$ are respectively the solid and liquid diffusion coefficients, and D_1 is bounded above and below by two positive constants. Thus, equation (7) with (10) and (12) leads to

$$\frac{\partial c}{\partial t} = \operatorname{div}\left(D_1(\phi)\nabla c + D_2(c,\phi)\nabla\phi\right), \tag{13}$$

where

$$D_2(c,\phi) = -\frac{v_m}{RT}\, c(1-c)\, D_1(\phi) F_2(\phi)$$

is a smooth function vanishing for $c = 0$ and $c = 1$.

The evolution equations (11) and (13) can be coupled to homogeneous Neumann boundary conditions on c and ϕ, ensuring that both matter and non-thermal free energy are confined on the domain Ω. Furthermore, considering initial conditions this leads to Problem (P).

We point out that the possible choice of a polynomial function of 5th degree in ϕ for f entails through (9), that F_1 and F_2 can be choosen as polynomial functions of 4th degree (see [11]). However, functions F_1, F_2, D_1 and D_2 have no physical meaning outside the sets $\{0 \le \phi \le 1\}$ and $\{0 \le c \le 1\}$. So, the functions F_1 and F_2 we consider subsequently are truncated outside the interval $[0,1]$ to 0 as shown in Fig. 1. We also truncate D_2 to 0 beyond the set $\{0 < c < 1\}$.

These truncations are fully justified in the next section in which we establish a maximum principle guaranteeing that the solution (ϕ, c) remains between 0 and 1 provided the initial data (ϕ_0, c_0) does. Moreover, this truncation procedure will enable us to prove existence, regularity and uniqueness results.

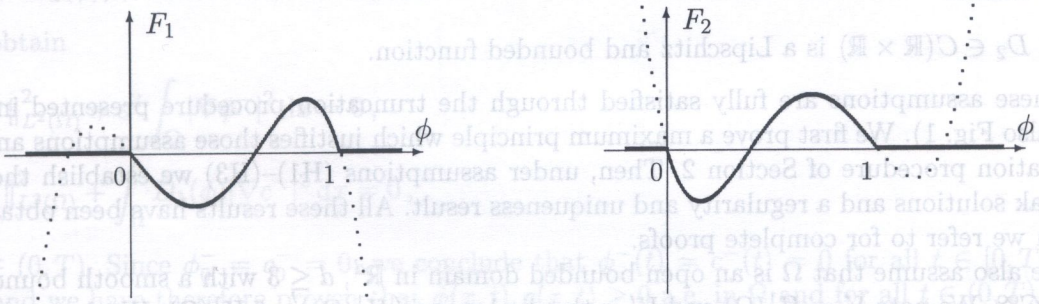

Fig. 1. Source terms F_1 and F_2 obtained by truncation to 0 outside the interval $[0,1]$

Sharp-interface limits as $\varepsilon \to 0$

The parameters α and ε can be related to physical parameters by exhibiting a special solution for the phase-field in a one-dimensional case for a pure element ($c \equiv 0$ or $c \equiv 1$) and performing an asymptotic analysis as ε tends towards 0 (see [11, 5, 4]). In that way, ε^2 is proportional to the liquid-solid interface thickness δ. Furthermore, it is possible to study general asymptotic limits of the model when $\delta \to 0$. Sharp interface Stefan-like problems are then found. The sharp-interface $\Gamma(t)$ splits Ω into two regions $\Omega_s(t)$ (solid) and $\Omega_l(t)$ (liquid). The concentration c and the interface $\Gamma(t)$ satisfy the following general form (see [4]):

$$\begin{cases} \dfrac{\partial c}{\partial t} = D_l \Delta c & \text{in} \quad \Omega_l(t), \\ \dfrac{\partial c}{\partial t} = D_s \Delta c & \text{in} \quad \Omega_s(t), \\ -V_n \left[c\right]_l^s = \left[D_1 \dfrac{\partial c}{\partial n} \right]_l^s & \text{on} \quad \Gamma(t), \\ \left[\dfrac{\partial f}{\partial c} \right]_l^s = 0 & \text{on} \quad \Gamma(t), \\ \left[f - c \dfrac{\partial f}{\partial c} \right]_l^s = \mathcal{F}(V_n, \kappa) & \text{on} \quad \Gamma(t), \end{cases}$$

where V_n and κ are respectively the normal velocity and the curvature of the interface $\Gamma(t)$ and $[q]_l^s$ denotes the difference of the quantity q between the solid and liquid sides of the interface. According to the dependency of α and other parameters inside F_1 and F_2 with respect to ε, the function $\mathcal{F}$ will linearly depend either on V_n , on κ, on both or on neither of them. The three interface conditions can be easily interpreted as the conservation of matter through the interface (first condition) and the eventual shifting of the equilibrium phase diagram due to interface curvature and normal velocity (second and third conditions, whose solving give the limit values of c on either side of the interface, as functions of κ and V_n).

Without loss of generality, we may choose $\alpha = 1$ for the rest of the article.

3. Mathematical analysis

In this section, we investigate the well-posedness of the isotropic solutal phase-field model (P). We suppose that the nonlinear functions F_i and D_i , $i = 1, 2$, satisfy Lipschitz and boundeness assumptions. More precisely, we assume that

(H1) F_1, $F_2 \in C(\mathbb{R})$ are Lipschitz and bounded functions.

(H2) $D_1 \in C(\mathbb{R})$ is a Lipschitz positive function, bounded above and below by two positive constants.

(H3) $D_2 \in C(\mathbb{R} \times \mathbb{R})$ is a Lipschitz and bounded function.

These assumptions are fully satisfied through the truncation procedure presented in Section 2 (see also Fig. 1). We first prove a maximum principle which justifies those assumptions and then the truncation procedure of Section 2. Then, under assumptions (H1)–(H3) we establish the existence of weak solutions and a regularity and uniqueness result. All these results have been obtained in [9], which we refer to for complete proofs.

We also assume that Ω is an open bounded domain in $\mathbb{R}^d$, $d \leq 3$ with a smooth boundary $\partial\Omega$ of class C^∞. We note $V = H^1(\Omega)$ and V' is the dual space $\left(H^1(\Omega)\right)'$ of $H^1(\Omega)$. We denote by $\langle \cdot, \cdot \rangle_{V',V}$ the duality product between V' and V. Finally, for $T > 0$ we note $Q_T = \Omega \times (0, T)$.

To begin with, let us define a weak solution of Problem (P).

Definition 1. *Let $(\phi_0, c_0) \in L^2(\Omega) \times L^2(\Omega)$ and $T > 0$. A couple of functions (ϕ, c) is a* weak solution *to Problem (P) if ϕ, $c \in L^2(0,T;H^1(\Omega)) \cap H^1(0,T;V')$, with $\phi(0) = \phi_0$, $c(0) = c_0$ and*

$$\left\langle \frac{\partial \phi}{\partial t}, v \right\rangle_{V',V} + \varepsilon^2 \int_\Omega \nabla\phi \cdot \nabla v \, \mathrm{d}x = \int_\Omega (F_1(\phi) + cF_2(\phi))\, v \, \mathrm{d}x,$$
$$\left\langle \frac{\partial c}{\partial t}, w \right\rangle_{V',V} + \int_\Omega (D_1(\phi)\nabla c + D_2(c,\phi)\nabla\phi) \cdot \nabla w \, \mathrm{d}x = 0, \tag{14}$$

for all v, $w \in H^1(\Omega)$ and a.e. in $(0,T)$.

Remark: Since ϕ, $c \in L^2(0,T;H^1(\Omega)) \cap H^1(0,T;V')$, it follows that ϕ, $c \in C([0,T];L^2(\Omega))$.

3.1. Maximum principle

In addition to (H1), (H2) and (H3), we suppose that the nonlinear terms F_1, F_2, and D_2 satisfy the following extra assumptions:

(H4) $F_1 \equiv F_2 \equiv 0$ in $]-\infty, 0] \cup [1, +\infty[$.

(H5) $D_2(\cdot, r_2) \equiv 0$ in $]-\infty, 0] \cup [1, +\infty[$ and for all $r_2 \in \mathbb{R}$.

These assumptions correspond to the truncation procedure of Section 2. The following result allows to justify the extensions of F_1, F_2 and D_2 to 0.

Theorem 1. (see [9]) *Let assumptions (H1)–(H5) be fulfilled.*
Suppose that the initial data $(\phi_0, c_0) \in L^2(\Omega) \times L^2(\Omega)$ is such that

$$0 \le \phi_0(x), \quad c_0(x) \le 1 \qquad \text{for a.e. } x \in \Omega.$$

Then for any $T > 0$, every weak solution $(\phi, c) \in \left(L^2(0,T;H^1(\Omega))\right)^2 \cap \left(H^1(0,T;V')\right)^2$ satisfies for all $t \in [0,T]$

$$0 \le \phi(x,t), \quad c(x,t) \le 1 \qquad \text{for a.e. } x \in \Omega.$$

Proof: Let $\phi^- = \max(-\phi, 0)$ and $c^- = \max(-c, 0)$. We have $(\phi^-, c^-) \in L^2(0,T;H^1(\Omega))^2$ and

$$-\left\langle \frac{\partial \phi}{\partial t}, \phi^- \right\rangle_{V',V} + \varepsilon^2 \int_\Omega \left|\nabla\phi^-\right|^2 \mathrm{d}x = -\int_\Omega (F_1(\phi) + cF_2(\phi))\, \phi^- \, \mathrm{d}x,$$
$$-\left\langle \frac{\partial c}{\partial t}, c^- \right\rangle_{V',V} + \int_\Omega D_1(\phi) \left|\nabla c^-\right|^2 = \int_\Omega D_2(c,\phi)\nabla\phi \cdot \nabla c^- \, \mathrm{d}x.$$

Using assumptions (H4) and (H5), we infer that

$$(F_1(\phi) + cF_2(\phi))\, \phi^- = 0 \quad \text{and} \quad D_2(c,\phi) = 0 \qquad \text{if } c < 0, \text{a.e. in } Q_T.$$

Thus, we obtain

$$\frac{1}{2}\frac{\mathrm{d}}{\mathrm{d}t}\|\phi^-\|^2_{L^2(\Omega)} + \varepsilon^2 \int_\Omega |\nabla\phi^-|^2 \, \mathrm{d}x = 0,$$
$$\frac{1}{2}\frac{\mathrm{d}}{\mathrm{d}t}\|c^-\|^2_{L^2(\Omega)} + \int_\Omega D_1(\phi)|\nabla c^-|^2 \, \mathrm{d}x = 0,$$

for a.e. $t \in (0,T)$. Since $\phi_0^- = c_0^- = 0$, we conclude that $\phi^-(t) = c^-(t) = 0$ for all $t \in [0,T]$ and a.e. in Ω, and we have therefore proven that $\phi(x,t), c(x,t) \ge 0$ a.e. in Ω and for all $t \in (0,T)$. It is sufficient to take $(\phi - 1)^+$ and $(c-1)^+$ instead of ϕ^- and c^- to prove that $(\phi - 1)^+ = (c-1)^+ = 0$ and consequently that $\phi(x,t), c(x,t) \le 1$ a.e. in Ω and for all $t \in (0,T)$.

3.2. Existence of weak solutions

Under the Lipschitz and boundeness assumptions on the nonlinear functions F_i and D_i, $i = 1, 2$, the following existence result holds.

Theorem 2. (see [9]) *Let assumptions (H1)–(H3) be fulfilled.*
For any $(\phi_0, c_0) \in L^2(\Omega) \times L^2(\Omega)$ and $T > 0$, there exists a weak solution to Problem (P).

Sketch of the proof: We employ a Faedo–Galerkin method. We consider the sequence $0 = \lambda_1 < \lambda_2 \leq \lambda_3 \ldots$ of the eigenvalues of the operator $(-\Delta)$ with homogeneous Neumann boundary conditions and $v_1, v_2, v_3, \ldots$ the corresponding eigenfunctions such that $(v_k, v_j)_{L^2(\Omega)} = \delta_{kj}$. We set $V_m = \operatorname{span}(v_1, v_2, \ldots, v_m)$ and we solve the finite dimensional problem:

$$(P_m)\begin{cases} \displaystyle\int_\Omega \frac{\partial \phi_m}{\partial t} v \,\mathrm{d}x + \varepsilon^2 \int_\Omega \nabla \phi_m \cdot \nabla v \,\mathrm{d}x = \int_\Omega \Big(F_1(\phi_m) + c_m F_2(\phi_m)\Big) v \,\mathrm{d}x, \\ \displaystyle\int_\Omega \frac{\partial c_m}{\partial t} w \,\mathrm{d}x + \int_\Omega (D_1(\phi_m)\nabla c_m + D_2(c_m, \phi_m)\nabla \phi_m) \cdot \nabla w \,\mathrm{d}x = 0, \\ \qquad\qquad \text{for all } v,\ w \in V_m \text{ and for a.e. } t \in (0, T), \\ \phi_m(0) = \phi_{0m} \in V_m, \quad c_m(0) = c_{0m} \in V_m, \end{cases}$$

where ϕ_{0m} and c_{0m} are L^2-projections of ϕ_0 and c_0 onto V_m.

By using assumptions (H1)–(H3), we prove there exists a unique maximal solution (ϕ_m, c_m) on the interval $[0, T_m[$ with $T_m > 0$. Moreover we can establish the following a priori estimate

$$\|(\phi_m, c_m)\|_{L^\infty(0,T_m;L^2(\Omega))^2} \leq C,$$

where C is a constant independent of m. This result proves that $T_m = +\infty$. It is not difficult to obtain two other a priori estimates:

$$\|(\phi_m, c_m)\|_{L^2(0,T;H^1(\Omega))^2} \leq C$$

and

$$\left\| \left(\frac{\partial \phi_m}{\partial t}, \frac{\partial c_m}{\partial t} \right) \right\|_{L^2(0,T;V')^2} \leq C.$$

Now, we recall the following well-known fact that

$$W_1 \equiv \left\{ u \in L^2(0,T;H^1(\Omega)),\ \frac{\partial u}{\partial t} \in L^2(0,T;V') \right\}$$

is compactly embedded in $L^2(Q_T)$ and

$$W_2 \equiv \left\{ u \in L^\infty(0,T;L^2(\Omega)),\ \frac{\partial u}{\partial t} \in L^2(0,T;V') \right\}$$

is compactly embedded in $C^0([0,T],V')$. From this result we can deduce that

$$(\phi_m, c_m) \to (\phi, c) \qquad \text{strongly in } \left(L^2(Q_T)\right)^2 \cap \left(C^0([0,T],V')\right)^2,$$

$$(\phi_m, c_m) \rightharpoonup (\phi, c) \qquad \text{weakly in } \left(L^2(0,T;H^1(\Omega))\right)^2,$$

$$\left(\frac{\partial \phi_m}{\partial t}, \frac{\partial c_m}{\partial t} \right) \rightharpoonup \left(\frac{\partial \phi}{\partial t}, \frac{\partial c}{\partial t} \right) \qquad \text{weakly in } \left(L^2(0,T,V')\right)^2.$$

It follows that

$$F_1(\phi_m), F_2(\phi_m), D_1(\phi_m) \to F_1(\phi), F_2(\phi), D_1(\phi) \quad \text{in } L^p(Q_T),\ \forall p \in [1, +\infty),$$

$$c_m F_2(\phi_m) \to c F_2(\phi) \quad \text{in } L^q(Q_T),\ \forall q \in [1, 2),$$

$$D_1(\phi_m)\nabla c_m \rightharpoonup D_1(\phi)\nabla c \quad \text{weakly in } L^q(Q_T),\ \forall q \in [1, 2),$$

$$D_2(c_m, \phi_m)\nabla \phi_m \rightharpoonup D_2(c, \phi)\nabla \phi \quad \text{weakly in } L^q(Q_T),\ \forall q \in [1, 2).$$

Taking the limit in Problem (P_m), we conclude that (c, ϕ) is a weak solution of Problem (P).

3.3. Regularity and uniqueness

Under the additional assumption that the initial data are smooth enough, we have the following regularity and uniqueness result.

Theorem 3. (see [9]) *Let assumptions (H1)–(H3) be fulfilled. Let* $\phi_0 \in H^2(\Omega)$ *such that* $\dfrac{\partial \phi_0}{\partial n} = 0$ *on* $\partial\Omega$ *and* $c_0 \in H^1(\Omega)$. *Then for any* $T > 0$, *there exists a unique couple of functions* (ϕ, c) *satisfying*

$$\phi \in L^2(0, T; H^3(\Omega)) \cap H^1(0, T; H^1(\Omega)) \quad \text{and} \quad c \in L^2(0, T; H^2(\Omega)) \cap H^1(0, T; L^2(\Omega)),$$

such that $\phi(0) = \phi_0$, $c(0) = c_0$ *and*

$$\frac{\partial \phi}{\partial t} - \varepsilon^2 \Delta \phi = F_1(\phi) + c F_2(\phi) \quad \text{a.e. in } \Omega \times (0, T),$$

$$\frac{\partial c}{\partial t} = div\left(D_1(\phi)\nabla c + D_2(c, \phi)\nabla \phi\right) \quad \text{a.e. in } \Omega \times (0, T), \tag{15}$$

$$\frac{\partial \phi}{\partial n} = \frac{\partial c}{\partial n} = 0 \quad \text{a.e. on } \partial\Omega \times (0, T).$$

Remark: We infer from the regularity of the solution that $\phi \in C^0([0, T]; H^2(\Omega))$ and $c \in C^0([0, T]; H^1(\Omega))$.

Theorem 3 is proved by establishing further a priori estimates in the Faedo–Galerkin procedure. These estimates are obtained thanks to Gagliardo–Nirenberg inequalities. We refer to [9] for the proof.

4. Numerical approximation

In order to approximate Problem (P), we use a semi-implicit scheme in time. Let us denote the time step by $\tau = T/N$, for $N \geq 1$ and the current time by $t^n = n\tau$, for $n = 0, \ldots, N$. We consider the approximations ϕ^n and c^n of $\phi(\cdot, t^n)$ and $c(\cdot, t^n)$ respectively, which are defined by the following scheme:

$$\begin{cases} \dfrac{\phi^{n+1} - \phi^n}{\tau} - \varepsilon^2 \Delta \phi^{n+1} = F_1(\phi^n) + c^n F_2(\phi^n), & (16) \\ \dfrac{c^{n+1} - c^n}{\tau} = \operatorname{div}\left(D_1(\phi^{n+1})\nabla c^{n+1} + D_2(c^n, \phi^{n+1})\nabla \phi^{n+1}\right). & (17) \end{cases}$$

The space discretization is made by means of a $\mathbb{P}_1$-finite element method. This provides the approximations ϕ_h^n and c_h^n of the exact solutions $\phi(\cdot, t^n)$ and $c(\cdot, t^n)$ at time t^n. Suppose that ϕ_h^n and c_h^n are known. Then we determine ϕ_h^{n+1} by first solving space discretized equation (4) and afterwards the resolution of equation (4) gives c_h^{n+1}.

We have the following convergence result for the above scheme.

Theorem 4. *(see [6]) Suppose that assumptions (H1)–(H3) are fulfilled and that the solution (ϕ, c) of (P) is smooth enough. Then there exists a constant $C > 0$ such that*

$$\max_{1 \le n \le N} \left(\|\phi_h^n - \phi(t^n)\|_{L^2(\Omega)} + \|c_h^n - c(t^n)\|_{L^2(\Omega)} \right) \le C(h + \tau). \tag{18}$$

The proof is based on the use of a the H^1-projectors of ϕ and c. We refer to [6] for the proof. This error estimate is not optimal as shown by numerical simulations. However, it is possible to improve the order of convergence in $\mathcal{O}(h^2 + \tau)$ by introducing a generalized elliptic projector for ϕ and c (see [6]). Let us remark finally that the constant C appearing in estimate (18) unfortunately depends on $1/\varepsilon^2$.

5. Numerical results

Problem (P) previously presented, corresponds to an isotropic situation, that is, there is no preferential solidification direction. Nevertheless, in this isotropic case we numerically observe that the mesh has an influence on the growth direction of the solidification front (see Fig. 2 (left) for a structured mesh of Fig. 3 type with 50×50 nodes). When the mesh is very fine the numerical simulation leads to a good symmetry of the solution (see Fig. 2 (right) for a structured mesh of Fig. 3 type with 160×160 nodes). These two figures were made for the same set of physical parameters and initial condition, the latter beeing a small isotropic solid germ in the middle of a liquid region.

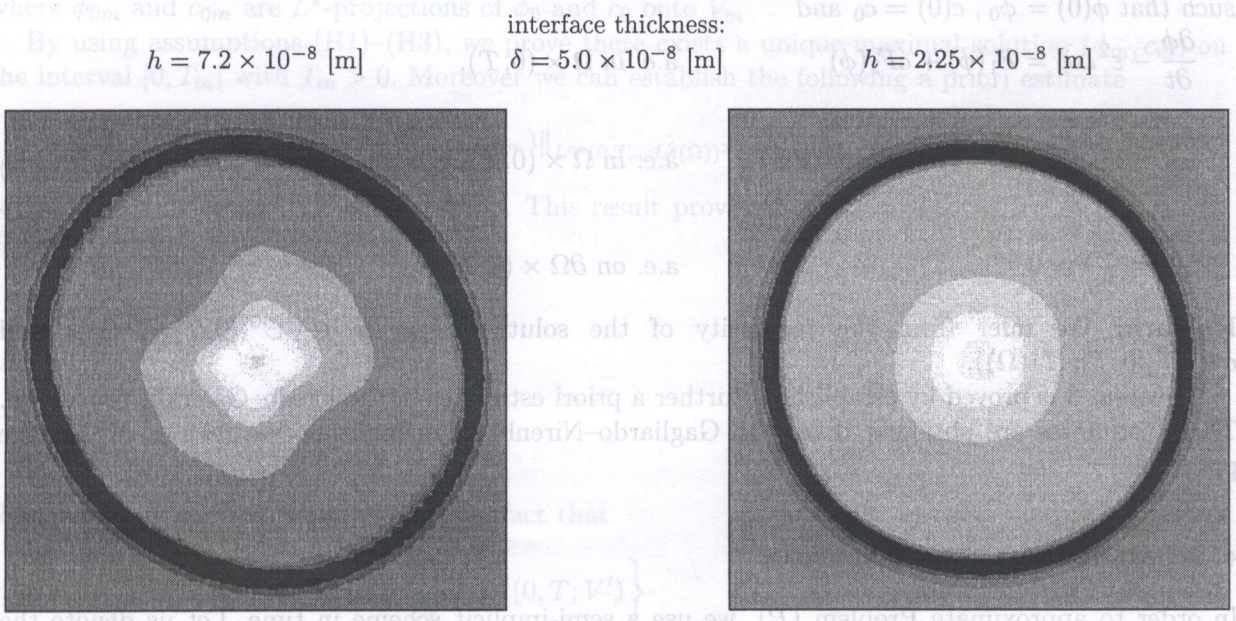

Fig. 2. Concentration in the isotropic case with a fixed and structured mesh of 50×50 nodes (left) and 160×160 nodes (right)

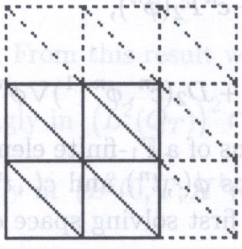

Fig. 3. Structured mesh

For the finite element method, we also use an adaptive mesh strategy. At each time t^n, we compute an estimator η_K on each triangle K based on the jump of the normal derivative of c_h^{n-1} and c_h^n across the edges of triangle K. The mesh is computed at time t^n in order to satisfy:

$$(1-\alpha)^2 TOL^2 \leq \sum_K {\eta_K}^2 \leq (1+\alpha)^2 TOL^2,$$

where TOL is a tolerance for the error and α is a variance. The new mesh is then generated by a Delaunay algorithm. For a complete description of this strategy we refer to [7].

Using the adaptive mesh procedure we obtain a perfectly circular numerical isotropic solution, with a mesh composed by less than 2500 nodes.

Anisotropy

From a physical point of view, it is interesting to consider anisotropic situations where preferential solidification directions are assigned. The anisotropic version of Problem (P) is obtained by choosing a function $\varepsilon = \varepsilon(\nabla\phi)$ in the expression of the Ginzburg–Landau free energy (5), more precisely by making ε depend on the angle between the gradient of ϕ and the horizontal axis in (5) (see [11]). Thus, in place of the isotropic evolution equation (1) for ϕ in (P), we obtain the following anisotropic equation:

$$\frac{\partial\phi}{\partial t} = \operatorname{div}(A(\nabla\phi)\nabla\phi) + F_1(\phi) + cF_2(\phi), \tag{19}$$

where $A(\nabla\phi)$ is the anisotropy matrix given by

$$A(\nabla\phi) = \begin{pmatrix} \varepsilon^2(\theta(\nabla\phi)) & -\varepsilon(\theta(\nabla\phi))\,\varepsilon'(\theta(\nabla\phi)) \\ \varepsilon(\theta(\nabla\phi))\,\varepsilon'(\theta(\nabla\phi)) & \varepsilon^2(\theta(\nabla\phi)) \end{pmatrix},$$

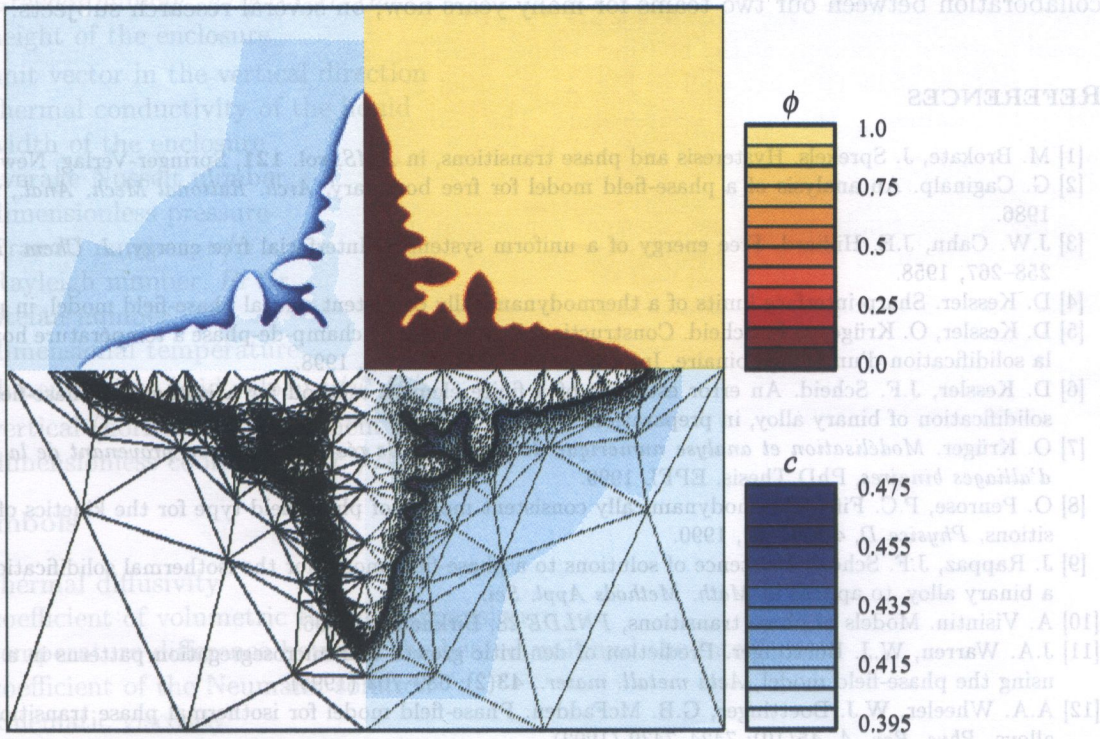

Fig. 4. Dendrite in the anisotropic case

where $\theta(\nabla\phi)$ is defined by

$$\nabla\phi \equiv \|\nabla\phi\| \begin{pmatrix} \cos\theta(\nabla\phi) \\ \sin\theta(\nabla\phi) \end{pmatrix}.$$

Then, Eq. (19) is discretized in time as follows:

$$\frac{\phi^{n+1} - \phi^n}{\tau} = \operatorname{div}\left(A(\nabla\phi^n)\nabla\phi^{n+1}\right) + F_1(\phi^n) + c^n F_2(\phi^n).$$

In Fig. 4 is shown an example of an anisotropic situation where the 4 cardinal directions are assigned to be preferential solidification directions. For this example, we have chosen $\varepsilon(\theta) = 1 + \bar{\varepsilon}\cos(4\theta)$, where $\bar{\varepsilon}$ is the amplitude of the anisotropy, and ε' is the derivative of ε with respect to θ. Figure 4 is obtained by solving the discretized problem with an adaptive mesh strategy.

In this case, the initial data corresponds to a small spherical grain at the center of the box.

6. Conclusion

We have worked with an isotopic solutal phase-field model closely based on Warren and Boettinger's model [11]. The peculiarity of this models' equations is having higher degree polynomial source terms and more complex cross-terms than the more standard pure element thermal phase-field models.

We have successfully established existence and uniqueness results, as well as asymptotic sharp-interface limits for the exact problem, and we have also established a working numerical aproximation. A paper on a priori error estimates on the proposed discrete scheme is currently in preparation.

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References

[1] M. Brokate, J. Sprekels. Hysteresis and phase transitions, in *AMS*, vol. **121**, Springer-Verlag, New York, 1996.

[2] G. Caginalp. An analysis of a phase-field model for free boundary, *Arch. Rational Mech. Anal.*, **92**: 205–245, 1986.

[3] J.W. Cahn, J.E. Hilliard. Free energy of a uniform system. I. Interfacial free energy, *J. Chem. Phys.*, **28**(2): 258–267, 1958.

[4] D. Kessler. Sharp-interface limits of a thermodynamically consistent solutal phase-field model, in preparation.

[5] D. Kessler, O. Krüger, J.F. Scheid. Construction d'un modèle de champ-de-phase à température homogène pour la solidification d'un alliage binaire. Internal report DMA-EPFL, 1998.

[6] D. Kessler, J.F. Scheid. An error estimate of a finite element method for a isothermal phase-field model for solidification of binary alloy, in preparation.

[7] O. Krüger. *Modélisation et analyse numérique de problčmes de réaction-diffusion provenant de la solidification d'alliages binaires*, PhD Thesis. EPFL 1999.

[8] O. Penrose, P.C. Fife. Thermodynamically consistent models of phase-field type for the kinetics of phase transitions. *Physica D*, **43**: 44–62, 1990.

[9] J. Rappaz, J.F. Scheid. Existence of solutions to a phase-field model for the isothermal solidification process of a binary alloy, to appear in *Math. Methods Appl. Sci.*

[10] A. Visintin. Models of phase transitions, *PNLDE* 28, Birkhaüser, 1996.

[11] J.A. Warren, W.J. Boettinger. Prediction of dendritic growth and microsegregation patterns in a binary alloy using the phase-field model, *Acta metall. mater.*, **43**(2): 689–703 (1995).

[12] A.A. Wheeler, W.J. Boettinger, G.B. McFadden. Phase-field model for isothermal phase transitions in binary alloys, *Phys. Rev. A*, **45**(10): 7424–7439 (1992).