A phase field model for brine channels in sea ice

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Abstract

In this paper, we present a phenomenological mathematical model for describing the features of the brine channels in sea ice. The differential system is composed by the Ginzburg-Landau and Cahn-Hilliard equations, in addition to the heat equation, that controls the ice-liquid phase transition by the temperature and hence the establishment of brine channels. The compatibility of this system with the thermodynamic laws and a maximum theorem are proved.

Keywords: Brine channels; phase field; Cahn-Hilliard; solute separation

1. Introduction

When sea water freezes, the dissolved salts are not incorporated in the crystal lattice of the ice, which consists of pure water, but rather they are kept in the residual liquid solution and tend to increase its salinity, especially if the interstitial solution remains quite isolated from the rest of the sea water. This turns out in a lowering of the freezing point of the residual liquid (cryoscopic effect), which hinders the further freezing process. These enclaves of highly

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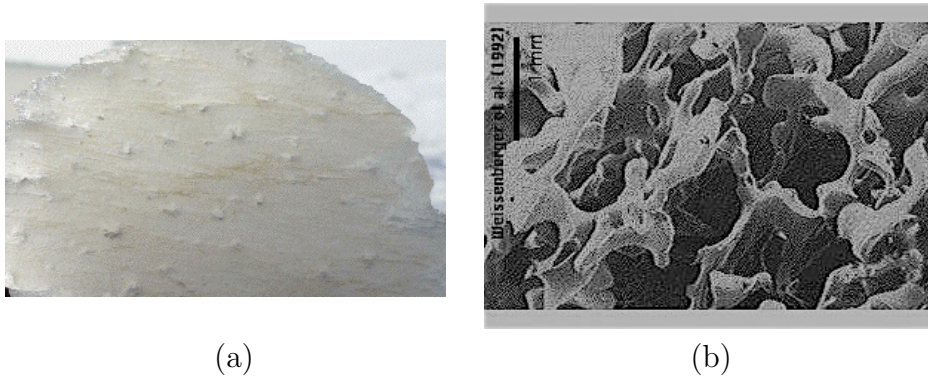`mauro.fabrizio@unibo.it` (Mauro Fabrizio), `diego.grandi@unibo.it` (Diego Grandi)

Figure 1: Image of: (a) microstructural "holes" visible in the ice core, corresponding to the termination points of brine channels; (b) a brine channel system within the sea ice.

concentrated solution among the ice crystals form the brine channels (Fig. 1). In fact, it is widely recorded that sea ice is a mixture of ice crystals and salt channels [1]. Some of these channels are very small with an average diameter of 100 μm. However, they may increase to millimetre sized tubes. Many of the physical features of the brine channels depend on the bulk salinity. In some cases these properties are influenced by the sea ice porosity [3]. Under suitable conditions, these small channels in the ice, filled with a salty solution, form a unique habitat for some bacteria and microscopic plants [2].

In this paper we adopt the phase-field approach of [4, 5] to describe the ice-water phase transition by a suitable form of the Ginzburg-Landau equation for first order transitions. The concomitant separation between salt and (frozen) water is accounted by a differential equation of the Cahn-Hilliard type. An alternative approach by a morphogenetic field is presented in [6]. It is well known that the phase field method is a convenient tool in describing phase transitions under many respects, in particular for computer simulations of phase changes. In fact, it allows to overcome the difficulties typical of sharp interface models, especially in regard to the topological changes during the interface tracking. At the same time, the sharp interface (macroscopic) description can be obtained from the phase field description as a limit case. This issue is widely discussed in literature; we refer for example to [7, 8] for the solidification phenomena and solid-solid transformations and to [9] for the non-miscibility regime in Cahn-Hilliard theory .

In our model, we neglect any macroscopic motion of the fluid, considering only time and spatial variations of the salinity and of the ice concentra-

tion. Moreover, we take into account the non-isothermal effects related to the phase transitions by adding a suitable form of the heat equation deduced by the First Law of Thermodynamics. In this framework it is crucial to consider the internal power related with the ice sea-water phase transition and the structural power associated with the Cahn-Hilliard equation, leading to a non-classical expression for the *local* energy balance. Then, we prove that the differential model is compatible with the Second Law of Thermodynamics expressed by the local Clausius-Duhem inequality. Finally, we obtain a maximum theorem for the system consisting of the Ginzburg-Landau and Cahn-Hilliard equations.

2. Differential Model

We denote with $c(x,t)$ the mass concentration field of the solute at point x and time t:

$$c = \frac{\rho_s}{\rho}, \tag{1}$$

where ρ_s is the density of the salt and ρ is the total density of the solution (brine). The physically meaningful domain of the variable c is the interval $[0,1]$. Actually, in the liquid phase the salt concentration reaches a maximum saturation value $c_s < 1$ ($c_s = 0.26$ for a pure sodium chloride solution). Beyond this value, salt precipitates are formed; however this phenomenon should not take place in the environmental conditions considered in this paper.

As a consequence of the mass conservation, the evolution of the solute density obeys a continuity law

$$\dot{\rho}_s + \nabla \cdot \mathbf{j}_s = 0, \tag{2}$$

where $\mathbf{j}_s$ denotes the solute current. As we are considering a fluid at rest, the superimposed dot actually represents a partial time derivative: $\dot{\rho}_s = \partial \rho_s(x,t)/\partial t$; at the same time, the solute current $\mathbf{j}_s$ has a *diffusive* character. For this reason, we do not choose a convective definition, such as $\mathbf{j}_s = \rho_s \mathbf{v}_s$ where $\mathbf{v}_s$ is a macroscopic velocity. Rather, the constitutive law for $\mathbf{j}_s$ should ultimately depend on the gradient of c (as will be explained later on). In a more general model, accounting for macroscopic fluid motion, convective solute transport would also appear with the introduction of the material time derivative.

Under the well-justified incompressibility approximation (and according our discarding of the macroscopic motion), the overall density ρ of the solution is

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a well-defined function $\hat{\rho}(c)$. Here, we make the approximation of a constant overall density $\hat{\rho}(c) = \rho_0$: the small density variations induced by salinity changes are completely inessential in the static framework of our model. So the solute density is expressed by $\rho_s = \rho_0 c$ and the diffusive equation takes the form

$$\rho_0 c_t(x, t) + \nabla \cdot \mathbf{j}_s = 0, \tag{3}$$

where ρ_0 is a constant which can always be set to the value 1.

To describe the solid-liquid phase transition we introduce, according to the phase-field approach, a scalar field f with physical variation range $[0, 1]$ discriminating the two aggregation states of water: we associate the value $f = 0$ to the liquid phase and $f = 1$ to the solid one.

We propose a differential evolutive system for the fields c and f so to respect the basic features of the solidification (and fusion) process in salty water:

  i) the *cryoscopic effect* has to be accounted for, so the freezing temperature θ_* is a decreasing function of the concentration c :

  $$\theta_*(c) = \theta_T - \alpha(c), \tag{4}$$

  where $\theta_T = 273.15°K$ is the pure water freezing point. The function $\alpha(c)$ is, to a good approximation, linear increasing, giving a freezing point depression of about one Kelvin degree for every percent of mass concentration c;

  ii) the *separation effect*: the salt is not (microscopically) miscible in ice, while it is miscible in water (below the saturation concentration $c_s \simeq 0.26$).

As regards the last condition, we point out that the *Cahn-Hilliard equation* is suitable to describe the diffusive dynamics of some chemical species into another one when both mixing and separation phenomena can take place. We then find convenient to describe the solute dynamics by means of a Cahn-Hilliard-type equation. The current is accordingly assumed in the form

$$\mathbf{j_s} = -M(c)\nabla\mu, \tag{5}$$

where $M(c) \geq 0$ is named the *mobility* and μ the *chemical potential*, defined by

$$\mu = -\nu\Delta c + \frac{\partial W(c, f, \theta)}{\partial c} \tag{6}$$

The function $W(c, f, \theta)$ is the local part of the free energy potential (to be discussed in the next section).

On the other hand as in [4], the equation for the liquid-solid phase transition will be defined by the well-known (time-dependent) Ginzburg-Landau equation

$$\tau \dot{f} = \gamma \Delta f - \frac{\partial W(c, f, \theta)}{\partial f} \tag{7}$$

The appearance of the same potential W is essential to satisfy the thermodynamic compatibility of the Cahn-Hilliard and the Ginzburg-Landau equations, as it will be apparent in the next section.

The form of the potential W as a function of f is dictated by the first order character of the water-ice transition with its concentration-dependent transition temperature $\theta_*(c)$, for which the following equation is appropriate [4]

$$\tau f_t = \gamma \Delta f - \theta_T F'(f) + (\theta - \theta_*(c) + \theta_T) G'(f), \tag{8}$$

with the potentials $F(f)$ and $G(f)$ defined by

$$F(f) = 3f^4 - 4f^3, \quad G(f) = \begin{cases} 3f^4 - 8f^3 + 6f^2 & \text{if } 0 \le f \le 1 \\ 0 & \text{if } f < 0 \\ 1 & \text{if } f > 1 \end{cases} \tag{9}$$

As it is customary in phase field models, the transition is described through a (possibly) double-well potential

$$V(f) = \theta_T F(f) + u G(f), \tag{10}$$

where the parameter u is the transition driving parameter: for $u = \theta_T$ the potential V has two equal minima at $f = 0$ and $f = 1$, corresponding to liquid-solid equilibrium; for $u > \theta_T$ the $f = 0$ phase (liquid) is stable, while for $u < \theta_T$ the solid phase $f = 1$ is the stable one. The dependence of u on c should describe the cryoscopic effect.

With regard to the expression of the chemical potential, that is the full dependence of W on c, we assume the standard Cahn-Hillard fourth order polynomial; taking into account the already described contribution depending on f, we put

$$W(c, f, \theta) = \theta_T F(f) + (\theta + \alpha(c)) G(f) + \theta_C H(c). \tag{11}$$

with

$$H(c, \theta) = c^2 (c - c_0(\theta))^2. \tag{12}$$

The function H is a double well potential with equal minima in $c = 0$ and $c = c_0$. The dynamics of the Cahn-Hilliard equation is such that for concentrations $c > c_0$ the mixed homogeneous state is stable, while, for $0 < c < c_0$ a separation process takes place in which some regions are filled with the $c = 0$ mixture (pure water) and the other ones with $c = c_0$ brine. This suggests to take c_0 as the critical salinity at any given temperature θ:

$$c_0(\theta) = \frac{(\theta_T - \theta)_+}{\alpha_0}, \tag{13}$$

where, for any real number a, $a_+ := \max\{a, 0\}$.

In this way, at a given temperature $\theta < \theta_T$ and for initial brine concentration $c < c_0$, the Cahn-Hilliard potential H favors the salt separation in a salt-free zone $c = 0$ (which will freeze) and a brine solution with critic concentration c_0, which will remain in the liquid state. If $c > c_0$ the salt will not separate and all the solution will be in the liquid state, as a consequence of the Ginzburg-Landau equation. Finally, for $\theta > \theta_T$, the potential reduces to $H(c) = c^4$, which is a single well (that is mixing) potential: obviously solidification and separation do not take place. We point out that the dimensional parameter θ_C in (11) has not the meaning of a critical temperature, since it does not trigger the bifurcation of $H(c)$ from single well to double well potential. So it does not influence the equilibrium properties, but rather the dynamics of the Cahn-Hilliard equation relative to the Ginzburg-Landau equation.

To sum up, we collect the phase fields equations (for $\rho = 1$)

$$\begin{cases} \tau f_t = \gamma \Delta f - [\theta_T F'(f) + (\theta + \alpha(c))G'(f)], \\ c_t = -\nabla \cdot \{ M(c)\nabla[-\nu\Delta c + \alpha'(c)G(f) + \theta_C \partial H/\partial c] \}. \end{cases} \tag{14}$$

3. Heat Equation and Thermodynamic Consistence

We aim to extend the system to include non-isothermal situations with a suitable evolution equation for the temperature field (up to now considered as a parameter inside W). To this end we have to add an energy balance equation. We express it in terms of internal energy variation and internal power expenditures [4, 5, 10]

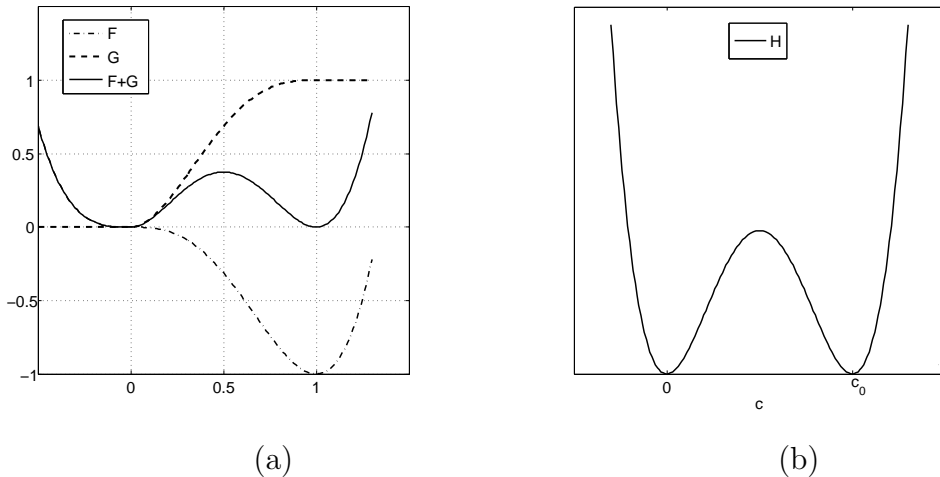$$\dot{e} = \mathcal{P}_f^i + \mathcal{P}_c^i + h, \tag{15}$$

Figure 2: (a) the functions F and G and the double well $F + G$; (b) the double well potential H (for $\theta < \theta_T$).

Here, h is the internal heat power satisfying the classical heat balance law

$$h = -\nabla \cdot \mathbf{q} + r, \tag{16}$$

with $\mathbf{q}$ the heat flux and r the external heat supply. $\mathcal{P}_f^i$ and $\mathcal{P}_c^i$ are the internal powers of suitable power balances equations associated to the differential system (14), which we resume here for convenience in the synthetic form

$$\begin{cases} \tau f_t = \gamma \Delta f - W_f(c, f), \\ c_t = \nabla \cdot [M \nabla(-\nu \Delta c + W_c(c, f))]. \end{cases} \tag{17}$$

The essential fact is that we consider the evolutive equations for f and c as balances equations [4, 5, 12, 13] associated with their own power balance laws. As a consequence, the first laws assumes a non classical form, with extra power terms, compared to the usual energy balance of simple materials. Nevertheless, we will verify that the system will satisfy the second law of Thermodynamics in the classical form of Clausius-Duhem inequality.

The power balances laws associated to the Ginzburg-Landau and Cahn-Hilliard equations are discussed at length for example in [10]. We remind it concisely. We multiply the first equation by f_t and the second one by $\mu = -\gamma \Delta c + W_c$ and simply manipulate the gradients term to obtain the

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power balances equations:

$$\begin{cases} \tau f_t^2 + \gamma \nabla f \cdot \nabla f_t + W_f f_t = \nabla \cdot (\gamma f_t \nabla f), \\[2mm] M|\nabla \mu|^2 + \nu \nabla c \cdot \nabla c_t + W_c c_t = \nabla \cdot (M\mu\nabla\mu + \nu c_t \nabla c). \end{cases} \tag{18}$$

The first members of these two equations define, respectively, the internal powers $\mathcal{P}_f^i$ and $\mathcal{P}_c^i$, while the second members define the external powers $\mathcal{P}_f^e$ and $\mathcal{P}_c^e$. So, the First Law (15) can be written as

$$e_t = \mathcal{P}_f^i + \mathcal{P}_c^i + h = \tau f_t^2 + M|\nabla\mu|^2 + \frac{d}{dt}\left(\frac{\gamma}{2}|\nabla f|^2 + \frac{\nu}{2}|\nabla c|^2 + W\right) - \theta_t W_\theta + h. \tag{19}$$

Then, defining the free energy ψ, as

$$\psi = \psi_0(\theta) + W + \frac{\gamma}{2}|\nabla f|^2 + \frac{\nu}{2}|\nabla c|^2, \tag{20}$$

and the entropy η and the internal energy e according to the usual thermodynamic relations

$$\eta = -\psi_\theta = -(\psi_0)_\theta - W_\theta, \tag{21}$$
$$e = \psi + \theta\eta = e_0(\theta) + W - \theta W_\theta + \frac{\gamma}{2}|\nabla f|^2 + \frac{\nu}{2}|\nabla c|^2 \tag{22}$$

with $e_0(\theta) = \psi_0 - \theta(\psi_0)_\theta$, the heat balance equation (19) assumes the form

$$e_0'(\theta)\theta_t - \theta\frac{d}{dt}(W_\theta) - \tau f_t^2 - M|\nabla\mu|^2 = -\nabla \cdot \mathbf{q} + r. \tag{23}$$

To verify the second law of Thermodynamics, it is useful to express (23) in terms of the entropy function η, obtaining

$$\theta\eta_t - \tau f_t^2 - M|\nabla\mu|^2 = -\nabla \cdot \mathbf{q} + r,$$

or

$$\eta_t = \frac{1}{\theta}\left[\tau f_t^2 + M|\nabla\mu|^2 - \frac{\mathbf{q}\cdot\nabla\theta}{\theta}\right] - \nabla\cdot\left(\frac{\mathbf{q}}{\theta}\right) + \frac{r}{\theta} \tag{24}$$

If the quantity in square brackets is non-negative definite, the Clausius-Duhem inequality

$$\eta_t \geq -\nabla\cdot\left(\frac{\mathbf{q}}{\theta}\right) + \frac{r}{\theta} \tag{25}$$

is satisfied. A sufficient condition is to assume a Fourier-like constitutive equation for the heat flux:

$$\mathbf{q} = -k(\theta)\nabla\theta, \quad k(\theta) > 0, \tag{26}$$

and the natural sign-conditions

$$\tau \geq 0, \quad M(c) \geq 0. \tag{27}$$

From the heat equation (23), we can identify

$$C(\theta) = e_0'(\theta) \tag{28}$$

with the specific heat, while the latent heat is given by (see equations (9), (11), (21))

$$L(\theta) = \theta[\eta(\theta, c, f = 0) - \eta(\theta, c, f = 1)] = -\theta[G(1) - G(0)] = \theta. \tag{29}$$

Of course, this means that the temperature θ is a rescaled temperature expressed in energy units, for sake of simplicity. We could as well use the physical units for temperature and put a suitable energy-scale constant multiplying the functions $F(f)$ and $G(f)$.
We collect the equations of the model

$$\begin{cases} \tau f_t = \gamma\Delta f - [\theta_T F'(f) + (\theta + \alpha(c))G'(f)], \\[2mm] c_t = \nabla \cdot \{ M\nabla[-\nu\Delta c + \theta_C\, \partial H/\partial c + \alpha'(c)G(f)]\}, \\[2mm] e_0'(\theta)\theta_t - \theta G'(f)f_t - \theta\dfrac{d}{dt}\left(\dfrac{\partial H}{\partial\theta}\right) = \tau f_t^2 + M|\nabla\mu|^2 + \nabla\cdot(k\nabla\theta) + r \end{cases} \tag{30}$$

and the suitable boundary conditions

$$\begin{cases} \nabla f \cdot \boldsymbol{\nu}|_{\partial\Omega} = 0, \quad \nabla c \cdot \boldsymbol{\nu}|_{\partial\Omega} = 0, \\[2mm] \nabla\mu \cdot \boldsymbol{\nu}|_{\partial\Omega} = 0, \; \theta|_{\partial\Omega_1} = \theta_1, \\[2mm] \nabla\theta \cdot \boldsymbol{\nu}|_{\partial\Omega_2} = g, \quad \nabla\theta \cdot \boldsymbol{\nu}|_{\partial\Omega_3} = h(\theta|_{\partial\Omega_3} - \theta_3) \end{cases} \tag{31}$$

where $\partial\Omega_i$ are the components of a partition of the boundary. The boundary conditions on the temperature depend on the physical conditions of the

experiment. Here, we have collected the most used ones: the fixed boundary temperature, the fixed heat flux and the convective heat transfer conditions. The equation $\nabla\mu\cdot\boldsymbol{\nu}|_{\partial\Omega} = 0$, that is $\mathbf{j}\cdot\boldsymbol{\nu}|_{\partial\Omega} = 0$ represents the isolation condition for the solute. The other homogeneous Neumann boundary conditions on c and f are the natural ones from a thermodynamic point of view, as they guarantee

$$\int_\Omega \mathcal{P}_f^e dx = 0, \quad \int_\Omega \mathcal{P}_c^e dx = 0, \tag{32}$$

where $\mathcal{P}_f^e$ and $\mathcal{P}_c^e$ represent the external powers (that is right members) in the balance equations (18). As a consequence, the energy balance referred to the whole system involves only the classical energy fluxes (in this case, the heat flux).

4. Maximum Theorem

Since the phase f and the concentration c do not make sense for $f \notin [0,1]$ or $c \notin [0,1]$, we need to prove a maximum theorem, that is, if (f, c, θ) is a solution of (30)-(31) with initial data satisfying $0 \leq f(x,0) \leq 1$ and $0 \leq c(x,0) \leq 1$, a.e. $x \in \Omega$, then we have to ensure that

$$0 \leq f(x,t) \leq 1 \text{ and } 0 \leq c(x,t) \leq 1,$$

a.e. $x \in \Omega$ and for all $t > 0$.

The proof that, for our problem, $0 \leq f(x,t) \leq 1$ is completely analogous to the Theorem 2, given in [5]. Therefore, it will be sufficient only to prove the result related with c. For the proof of this theorem we follow the method presented in the paper [11] that is based on the following assumptions:

i) $M = M_0 c^2 (c-1)^2$, $M_0 > 0$;

ii) H is strictly convex in $]-\infty, 0]$ and in $[1, +\infty]$;

iii) $\alpha'(c) = 0 \quad \forall c \leq 0$ or $c \geq 1$.

The potential H given in equation (12) satisfies the hypothesis, while for the function $\alpha(c)$, which is approximately linear from the empirical point of view, one has to (continuously) truncate it to a constant function outside the interval $[0,1]$ and a the same time to regularize it in 0 and 1 to obtain a continuous derivative. The further hypothesis of isothermal processes

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iv) θ is constant

is needed, since the potential H depends on θ.
Under those assumptions, we consider the initial value problem

$$c_t = \nabla \cdot [M(c)\nabla\mu(c)] \qquad c(\mathbf{x}, 0) = c_0(\mathbf{x}) \qquad \mathbf{x} \in \Omega \tag{33}$$

where

$$\mu(c) = -\nu\Delta c + \alpha'(c)G(f) + \theta_C H'(c), \tag{34}$$

with boundary conditions

$$\nabla c \cdot \boldsymbol{\nu}|_{\partial\Omega} = 0, \quad \nabla\mu \cdot \boldsymbol{\nu}|_{\partial\Omega} = 0. \tag{35}$$

Then the following theorem holds.

Theorem 1. *Let $c_0(\mathbf{x}) \in [0, 1]$ for each $\mathbf{x} \in \Omega$, then the solution $c(\mathbf{x}, t)$ of equation (33) takes value in $[0, 1]$ a.e $\mathbf{x} \in \Omega$ and for each $t \in \mathbb{R}^+$.*

Proof. We will explicitly prove only the lower bound $c \geq 0$. The proof of the upper bound is completely analogous thanks to the symmetry of the key assumptions i)-iii) with respect to the end points 0 and 1. Let c_- be the function defined by

$$c_-(\mathbf{x}, t) := \min\{c(\mathbf{x}, t), 0\}$$

and put

$$\Omega_+(t) = \{\mathbf{x} : c(\mathbf{x}, t) > 0\}, \quad \Omega_-(t) = \{\mathbf{x} : c(\mathbf{x}, t) < 0\} = \Omega \setminus \overline{\Omega}_+. \tag{36}$$

Accordingly,

$$c_-(\mathbf{x}, 0) = 0, \quad H(c_-(\mathbf{x}, 0)) = H(0) \quad \forall \mathbf{x} \in \Omega. \tag{37}$$

Using hypothesis iii), we can write

$$\int_\Omega c_t\, \mu(c_-)\, dv = \int_\Omega c_t\, (\theta_C H'(c_-) - \nu\Delta c_-) dv. \tag{38}$$

In fact, the left hand side integrand and the right hand side integrand are identical for any $\mathbf{x} \in \Omega_- \cup \Omega_+$, where Δc_- is well defined. However, since ∇c_- is in general discontinuous in $\partial\Omega_+$, $\mu(c_-)$ is not well-defined for $c = 0$; in

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a distributional sense, Δc_- has a δ-like behaviour. Nevertheless, the multiplication by c_t makes the contribution of integration over $\overline{\Omega}_+ \setminus \Omega_-$ irrelevant, because the chosen quadratic form of $M(c)$ in assumption i) guarantees that c_t is proportional to c.

By multiplying the differential equation in (33) by $\mu(c_-)$ and integrating over Ω (using the divergence theorem), we obtain

$$\int_\Omega c_t \, \mu\,(c_-) \, dv = - \int_\Omega M(c) \nabla \mu\,(c) \cdot \nabla \mu\,(c_-) \, dv. \tag{39}$$

Again, we remark that the integrability is a consequence of the factor c^2 in the expression of M. By the same reason, the integral can be restricted to Ω_-:

$$\int_\Omega M(c) \nabla \mu\,(c) \cdot \nabla \mu\,(c_-) \, dv = \int_{\Omega_-} M(c) |\nabla \mu\,(c_-)|^2 dv \geq 0.$$

So, combining equations (38) and (39),

$$\int_\Omega [c_t \, \theta_C H'\,(c_-) - \nu c_t \, \Delta c_-] \, dv \leq 0. \tag{40}$$

We will need the following

Lemma 1. *The following identities hold:*

$$- \int_\Omega c_t \Delta c_- \, dv = \frac{d}{dt} \int_\Omega \frac{1}{2} |\nabla c_-|^2 dv, \tag{41}$$

$$\int_\Omega c_t H'(c_-) dv = \frac{d}{dt} \int_\Omega \left(H(c_-) - H'(0) c_- \right) dv. \tag{42}$$

Proof. Consider the first equation; using the divergence theorem and the boundary conditions, we have

$$- \int_\Omega c_t \Delta c_- \, dv = - \int_\Omega c_- \Delta c_t dv = - \int_\Omega c_- \Delta (c_-)_t =$$
$$= \int_\Omega \nabla (c_-)_t \nabla c_- \, dv = \frac{d}{dt} \int_\Omega \frac{1}{2} |\nabla (c_-)|^2 dv.$$

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The second equality is grounded on the regularizing effect of the multiplication by c_-.

As for the second equation, we break up the integration domain to obtain

$$\int_\Omega c_t H'(c_-) dv = \int_{\overline{\Omega}_+} H'(0) c_t dv + \int_{\Omega_-} (c_-)_t H'(c_-) dv. \qquad (43)$$

Due to the mass conservation, $\int_\Omega c_t dv = 0$ (a consequence of (5) and the boundary conditions); hence

$$\int_{\overline{\Omega}_+} c_t dv = \int_\Omega c_t dv - \int_{\Omega_-} c_t dv = -\int_{\Omega_-} (c_-)_t dv. \qquad (44)$$

By substituting in (43), we obtain the statement

$$\int_\Omega c_t H'(c_-) dv = \int_{\Omega_-} [H'(c_-) - H'(0)](c_-)_t dv =$$

$$= \int_\Omega [H'(c_-) - H'(0)](c_-)_t dv = \frac{d}{dt} \int_\Omega [H(c_-) - H'(0) c_-] dv.$$

In the last equality hypothesis iv) has been used.

Continuation of the main proof. Combining inequality (40) with the previous Lemma, we get

$$\frac{d}{dt} I(t) \le 0, \qquad I(t) := \int_\Omega \left[\frac{\nu}{2} |\nabla c_-|^2 + \theta_C \big(H(c_-) - H'(0) c_- \big) \right] dv. \qquad (45)$$

Because of the non negative initial condition (37), the initial value of $I(t)$ is

$$I(0) = |\Omega| \theta_C H(0), \qquad (46)$$

and because of (45),

$$0 \ge I(t) - I(0) = \int_\Omega \left[\frac{\nu}{2} |\nabla c_-|^2 + \theta_C \big(H(c_-) - H'(0) c_- - H(0) \big) \right] dv \qquad (47)$$

By the hypothesis ii), we have

$$H(c_-) - H'(0) c_- - H(0) > 0 \quad \forall c_- < 0. \qquad (48)$$

Then, inequality (47) easily implies that $c_-(\mathbf{x}, t) = 0$ a.e., which proves the theorem.

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5. Acknowledgements

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