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NUMERICAL MODELLING OF MACROSEGREGATION FORMED DURING SOLIDIFICATION WITH SHRINKAGE USING A LEVEL SET APPROACH

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Acknowledgement

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List of Acronyms

Acronym	Standing for
ALE	Arbitrary Lagrangian-Eulerian
BTR	Brittle temperature range
CAFD	Cellular Automata Finite Difference
CAFE	Cellular Automata Finite Element
CBB	Circumventing Babuška-Brezzi
CCEMLCC	Chill Cooling for the Electro-Magnetic Levitator in relation with Continuous Casting of steel
CEMEF	Centre de Mise en Forme des Matériaux
CFL	Courant–Friedrichs–Lewy
C.FL.	Computing and FLuids
CSF	Continuum Surface Force
DLR	Deutsches Zentrum für Luft- und Raumfahrt
DSPG	Darcy-Stabilising/Petrov-Galerkin
EML	Electromagnetic levitation
ESA	European Space Agency
FEM	Finite Element Method
FVM	Finite Volume Method
GMAW	Gas Metal Arc Welding
ISS	International Space Station
IWT	Institut für Werkstofftechnik
LHS	Left-hand side
LSIC	Least squares on incompressibility constraint
LSM	Level set method
MAC	Marker-and-cell
PF	Phase field
PSPG	Pressure-Stabilising/Petrov-Galerkin
RHS	Right-hand side
RUB	Ruhr Universität Bochum
RVE	Representative Elementary Volume
SBB	Satisfying Babuška-Brezzi
SUPG	Streamline-Upwind/Petrov-Galerkin
VMS	Variational MultiScale
VOF	Volume Of Fluid

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Macrosegregation with liquid metal motion

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4.1 Introduction

Fluid flow is an important part in understanding the evolution of an alloy system undergoing phase change. It is attributed to the convective transport in fluids where the time scale is much smaller than other transport mechanisms (e.g. diffusive transport). To understand how fluid motion contributes to the heat and mass transfer, we have swiftly presented the momentum conservation equation in a solidifying liquid, [eq. \(2.42\)](#). In this chapter, we will first give a quick overview of the numerical treatment of this system of Navier-Stokes equations, then comment on some computational aspects such as the choice of a suitable time step and the conditions that impose minimum and maximum bounds on both time step and mesh size. Then, we shall present solidification applications where macrosegregation is mainly induced by thermosolutal convection.

4.2 Formulation stability

A wide array of numerical methods can be used to solve systems like [eq. \(2.42\)](#). When speaking about Navier-Stokes equations, the choice can be narrowed to two famous approaches with some similarities: stable mixed finite element method and Variational MultiScale (VMS) method. When two finite element spaces are introduced (e.g. one for velocity and another for pressure), the essential *inf-sup* condition (also known as stability condition) determined by [Babuška \[1971\]](#) and [Brezzi \[1974\]](#) should be fulfilled. It states that the formulation is ill-posed if the both spaces have the same interpolation order. For instance, a P1/P1 element (i.e. P1 for velocity / P1 for pressure) cannot guarantee the stability of the Navier-Stokes solution since velocity and pressure are both linearly interpolated at the simplex vertices. However, the major difference between the previously mentioned formulations is the way in which the inf-sup condition is accounted for. Stable mixed finite elements are stable because they directly respond to the stability condition by enriching the velocity space, hence they fall under the category of Satisfying Babuška-Brezzi (SBB) methods. In contrast, methods like VMS belong to the Circumventing Babuška-Brezzi (CBB) category [[Barbosa and Hughes 1991](#)]. CBB methods rely on equal-order interpolations with additional stabilisation that circumvents the need to satisfy the stability condition. Further details about both formulation types are given in the next subsections.

4.2.1 Stable mixed finite elements

First introduced by Arnold et al. [1984], the MINI element is the key ingredient of this approach. This type of element introduces an additional degree of freedom for the velocity field while keeping a linear interpolation for the pressure field, thus satisfying the Babuška-Brezzi condition with an enriched velocity space. The additional degrees of freedom are interpolated by means of the so-called *bubble* function and vanish on the element's boundary. We may therefore speak of a P1+/P1 finite element in a velocity-pressure formulation. This stable formulation has been the de facto standard for solving fluid and solid mechanics for many years at CEMEF. In *FORGE®* and *THERCAST®* codes, the P1+/P1 MINI element is used with a linear bubble function, leading to a decomposition of tetrahedra into four sub-elements to treat additional degrees of freedom for the velocity field.

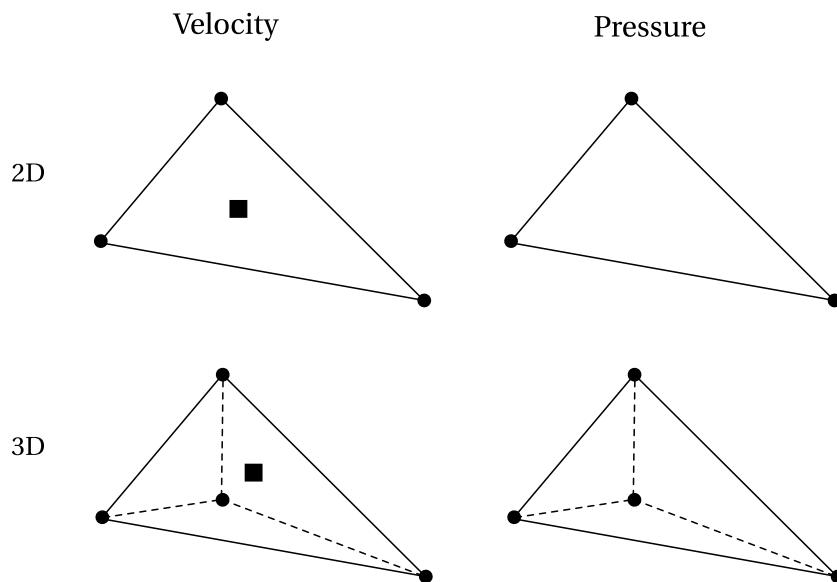


Fig. 4.1 – Schematic of 2D and 3D stable P1+/P1 finite elements, respectively triangle and tetrahedron, with velocity and pressure fields interpolation order. The dots represent the nodes while the squares represent the additional degrees, the *bubbles*.

4.2.2 Variational multiscale (VMS)

As the name indicates, this approach considers two scales of phenomena: the coarse and fine scales. Applied to our velocity-pressure formulation, these fields are decom-

posed according to these scales as follows:

$$\langle \mathbf{v}^l \rangle = \langle \mathbf{v}^l \rangle_h + \tilde{\mathbf{v}}^l \quad (4.1)$$

$$p = p_h + \tilde{p} \quad (4.2)$$

where $\langle \mathbf{v}^l \rangle_h$ and p_h are the coarse scale velocity and pressure discretised on the finite element mesh (hence the subscript h), while the remaining terms represent the fine scale velocity and pressure that cannot be captured at the scale of the FE grid. Instead of defining a finer grid to model the effect of these terms, one can solve the fine scale equations obtained once eqs. (4.1) and (4.2) are injected in eq. (2.42) then use the output in the coarse scale equations. Further technical details about the method and the equations are found in the PhD work of [Hachem \[2009\]](#).

The added value of the VMS method is the time gain that we get by incorporating the effect of the fine scale into the coarse scale physics without discretising on a finer grid, while maintaining the ability to predict localised fluid motion such as small vortices.

4.3 Navier-Stokes solver

In the present thesis, we chose to solve the fluid momentum conservation using a stabilised P1/P1 formulation with additional element-level integrals to add stability for convection-dominated terms, transient-dominated terms and pressure terms. The stabilisation techniques include the streamline upwind/Petrov-Galerkin (SUPG), pressure stabilising/Petrov-Galerkin (PSPG) and the least-squares on incompressibility constraint (LSIC) as a stabilisation framework introduced by [Tezduyar et al. \[1992\]](#). The global approach, more commonly known as SUPG-PSPG-LSIC, prevents the classical formulation instability coming from the linear equal-order interpolation functions.

It is important to note that the P1/P1 SUPG-PSPG-LSIC approach is slightly different than a VMS approach as the derivation of stabilising terms in the latter comes from a physical interpretation of two different length scales, a resolved coarse scale and an unresolved fine scale. The incorporation of the fine-scale equation within the coarse-scale one results in additional stabilising terms, while in the current approach these terms come from a mathematical analysis based on limiting cases of diffusion or advection. The final stabilising scheme is however very similar. The Navier-Stokes solver developed by [Hachem et al. \[2010\]](#) and [Rivaux \[2011\]](#) is a convenient choice to solve a stabilised Navier-Stokes system with Darcy terms.

4.3.1 Strong and weak formulations

The strong form of the fluid mechanics problem has been previously established in chapter 2 (section 2.1.1), where we obtained the following system of mass and liquid momentum conservation equations:

$$\begin{cases} \rho_0^l \left(\frac{\partial \langle v^l \rangle}{\partial t} + \frac{1}{g^l} \nabla \cdot (\langle v^l \rangle \times \langle v^l \rangle) \right) = \\ - g^l \nabla p^l - \nabla \cdot \left(2\mu^l \langle \bar{\dot{\varepsilon}}^l \rangle \right) - g^l \mu^l \mathbb{K}^{-1} \langle v^l \rangle + g^l \langle \rho \rangle^l \mathbf{g} \\ \nabla \cdot \langle v^l \rangle = 0 \end{cases} \quad (4.3)$$

The strong solution consists of finding the pair $(\langle v^l \rangle, p^l)$ of the previous system, when the following essential (Dirichlet type) and natural (Neumann type) boundary conditions are applied :

$$\langle v^l \rangle = \mathbf{v}_0 \text{ on } \partial\Omega_{\text{Dirichlet}} \quad (4.4)$$

$$\langle \bar{\sigma}^l \rangle \cdot \mathbf{n} = \mathbf{N} \text{ on } \partial\Omega_{\text{Neumann}} \quad (4.5)$$

$$\text{with } \partial\Omega_{\text{Dirichlet}} \cup \partial\Omega_{\text{Neumann}} = \partial\Omega \quad (4.6)$$

We can comment on the strong form with the following recap points:

1. the liquid metal is Newtonian with a dynamic viscosity denoted μ^l
2. the metal is incompressible, therefore the liquid and solid densities are constant and equal (hence ρ_0^l in the inertial term) and the mass balance reduces to $\nabla \cdot \langle v^l \rangle = 0$
3. the Boussinesq approximation is used to compute the thermosolutal buoyancy force in the melt via the term $g^l \langle \rho \rangle^l \mathbf{g} = g^l \langle \rho \rangle^l (T, \langle w_i \rangle^l) \mathbf{g}$, where $\langle \rho \rangle^l (T, \langle w_i \rangle^l)$ can be either tabulated as a function of temperature and liquid composition for each solute i , or directly approximated by:

$$\langle \rho \rangle^l = \rho_0^l \left(1 - \beta_T (T - T_0) - \sum_{i=1}^{\text{nb species}} \beta_{w_i^l} (\langle w_i \rangle^l - \langle w_i \rangle_0^l) \right) \quad (4.7)$$

where β_T and $\beta_{w_i^l}$ are respectively the thermal and solutal expansion coefficients, while T_0 and $\langle w_i \rangle_0^l$ represent a reference temperature and a reference liquid composition for each chemical species, respectively.

The weak form treated by the VMS solver derives from the strong form by multiplying by test functions for velocity and pressure belonging to these functional spaces:

$$\begin{aligned} v &= \left\{ \mathbf{u}, \quad \mathbf{u} \in (\mathcal{H}^1(\Omega))^d \mid \mathbf{u} = \mathbf{v}_0 \text{ on } \partial\Omega \right\} \\ v^0 &= \left\{ \mathbf{u}, \quad \mathbf{u} \in (\mathcal{H}^1(\Omega))^d \mid \mathbf{u} = \mathbf{0} \text{ on } \partial\Omega \right\} \\ \varrho &= \{q, \quad q \in L^2(\Omega)\} \end{aligned}$$

where d stands for the space dimension. Then, based on these definitions, we write the advective upwinding stabilised test function for the velocity, \mathbf{U} :

$$\mathbf{U} = \mathbf{u} + \tau_{\text{SUPG}} \bar{\nabla} \mathbf{u} \cdot \langle \mathbf{v}^l \rangle_{\Omega_E} \quad (4.8)$$

τ_{SUPG} is an elemental stabilising parameter for advection-dominated terms and $\langle \mathbf{v}^l \rangle_{\Omega_E}$ is the superficial velocity in the element Ω_E , calculated by regular P1 interpolation:

$$\langle \mathbf{v}^l \rangle_{\Omega_E} = \frac{\sum_{i=1}^D \langle \mathbf{v}^l \rangle_i}{D} \quad (4.9)$$

Moreover, we need the following operators in order to simplify the notation of element-based variational integrals:

$$[a, b] = \int_{\Omega_E} ab \, d\Omega \quad (4.10)$$

$$[c, d]^* = \int_{\partial\Omega_E} cd \, d\Gamma \quad (4.11)$$

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Finally, the SUPG-PSPG-LSIC stabilised weak formulation writes:

$$\left\{ \begin{array}{l} \forall \mathbf{u} \in v^0 \\ \left[\left(\frac{\rho_0^l}{g^l} \frac{\partial \langle \mathbf{v}^l \rangle}{\partial t} \right), \mathbf{U} \right] + \left[\left(\frac{\rho_0^l}{g^{l2}} (\bar{\nabla} \langle \mathbf{v}^l \rangle) \langle \mathbf{v}^l \rangle \right), \mathbf{U} \right] + \left[\left(\frac{2\mu^l}{g^l} \right), \bar{\bar{\epsilon}}(\langle \mathbf{v}^l \rangle) : \bar{\bar{\epsilon}}(\mathbf{U}) \right] \\ + \left[(\mu^l \mathbb{K}^{-1} \langle \mathbf{v}^l \rangle), \mathbf{U} \right] - \left[(\langle \rho \rangle^l \mathbf{g}), \mathbf{U} \right] - [p, \nabla \cdot \mathbf{U}] - \left[\frac{\mathbf{N}}{g^l}, \mathbf{U} \right]^* \\ + [\tau_{\text{LSIC}}, (\rho_0^l \nabla \cdot \langle \mathbf{v}^l \rangle \nabla \cdot \mathbf{u})] = 0 \\ \\ \forall q \in \varrho \\ \left[\nabla \cdot \langle \mathbf{v}^l \rangle, q \right] + \\ \left[\tau_{\text{PSPG}} \frac{\nabla q}{\rho_0^l}, \left(\frac{\rho_0^l}{g^l} \frac{\partial \langle \mathbf{v}^l \rangle}{\partial t} + \frac{\rho_0^l}{g^{l2}} (\bar{\nabla} \langle \mathbf{v}^l \rangle) \langle \mathbf{v}^l \rangle + \mu^l \mathbb{K}^{-1} \langle \mathbf{v}^l \rangle - \langle \rho \rangle^l \mathbf{g} \right) \right] = 0 \end{array} \right. \quad (4.12)$$

Replacing eq. (4.8) in eq. (4.12), we get the final weak form:

$$\left\{ \begin{array}{l} \forall \mathbf{u} \in v^0 \\ \left[\left(\frac{\rho_0^l}{g^l} \frac{\partial \langle \mathbf{v}^l \rangle}{\partial t} \right), \mathbf{u} \right] + \left[\left(\frac{\rho_0^l}{g^{l2}} (\bar{\nabla} \langle \mathbf{v}^l \rangle) \langle \mathbf{v}^l \rangle \right), \mathbf{u} \right] + \left[\left(\frac{2\mu^l}{g^l} \right), \bar{\bar{\epsilon}}(\langle \mathbf{v}^l \rangle) : \bar{\bar{\epsilon}}(\mathbf{u}) \right] \\ + \left[(\mu^l \mathbb{K}^{-1} \langle \mathbf{v}^l \rangle), \mathbf{u} \right] - \left[(\langle \rho \rangle^l \mathbf{g}), \mathbf{u} \right] - [p, \nabla \cdot \mathbf{u}] - \left[\frac{\mathbf{N}}{g^l}, \mathbf{u} \right]^* \\ + \left[\tau_{\text{SUPG}} \bar{\nabla} \mathbf{u} \cdot \langle \mathbf{v}^l \rangle_{\Omega_E}, \left(\frac{\rho_0^l}{g^l} \frac{\partial \langle \mathbf{v}^l \rangle}{\partial t} + \frac{\rho_0^l}{g^{l2}} (\bar{\nabla} \langle \mathbf{v}^l \rangle) \langle \mathbf{v}^l \rangle + \mu^l \mathbb{K}^{-1} \langle \mathbf{v}^l \rangle + \nabla p^l - \langle \rho \rangle^l \mathbf{g} \right) \right] \\ + [\tau_{\text{LSIC}}, (\rho_0^l \nabla \cdot \langle \mathbf{v}^l \rangle \nabla \cdot \mathbf{u})] = 0 \\ \\ \forall q \in \varrho \\ \left[\nabla \cdot \langle \mathbf{v}^l \rangle, q \right] + \\ \left[\tau_{\text{PSPG}} \frac{\nabla q}{\rho_0^l}, \left(\frac{\rho_0^l}{g^l} \frac{\partial \langle \mathbf{v}^l \rangle}{\partial t} + \frac{\rho_0^l}{g^{l2}} (\bar{\nabla} \langle \mathbf{v}^l \rangle) \langle \mathbf{v}^l \rangle + \mu^l \mathbb{K}^{-1} \langle \mathbf{v}^l \rangle + \nabla p^l - \langle \rho \rangle^l \mathbf{g} \right) \right] = 0 \end{array} \right. \quad (4.13)$$

4.3.2 Stabilisation parameters

Several expressions for τ_{SUPG} were derived by Tezduyar et al. [1992] and Tezduyar and Osawa [2000], from which we retain the following:

$$\tau_{\text{SUPG}} = \left(\frac{1}{\tau_{\text{advec}}^2} + \frac{1}{\tau_{\text{diff}}^2} + \frac{1}{\tau_{\text{trans}}^2} \right)^{-1/2} \quad (4.14)$$

where we use three parameters τ_{advec} , τ_{diff} and τ_{trans} having time as unit (*s*) that stabilise respectively advection-dominated, diffusion-dominated and transient-dominated regimes, given by:

$$\tau_{\text{advec}} = \frac{h_{\text{stream}}}{2\|\langle \mathbf{v}^l \rangle_{\Omega_E}\|} \quad (4.15)$$

$$\tau_{\text{diff}} = \frac{h_{\text{stream}}^2}{4\nu^l} \quad (4.16)$$

$$\tau_{\text{trans}} = \frac{\Delta t}{2} \quad (4.17)$$

where $h_{\text{stream}} = 2\|\langle \mathbf{v}^l \rangle_{\Omega_E}\| (\langle \mathbf{v}^l \rangle_{\Omega_E} \cdot \nabla \mathcal{P})$ is the element length in the stream direction, computed using the local superficial velocity and the interpolation functions \mathcal{P}_j relative to each local node j , ν^l is the liquid's kinematic viscosity ($\text{m}^2 \text{s}^{-1}$) and Δt is the time step. The transient term stabilisation was initially derived for Navier-Stokes equations without Darcy term. As the latter has a significant role in the weak form eq. (4.13), it needs to be stabilised. The current thesis is based on several past projects that either considered eq. (4.17) like Liu [2005] or modified it like Gouttebroze [2005] and Rivaux [2011] to take account the Darcy term, giving the term:

$$\tau_{\text{trans-darcy}} = \frac{\Delta t}{2 \left(1 + \Delta t \frac{\mu^l}{\rho_0^l \mathbb{K}} \right)} \quad (4.18)$$

In the literature, no substantial references were found to backup the formulation of eq. (4.18), it will be used in the current work though. It is worth mentioning that Zabaras and Samanta [2004] has invoked the necessity to stabilise Darcy terms in a generalised Navier-Stokes/Darcy P1/P1 formulation, and introduced what they call Darcy-Stabilising/Petrov-Galerkin (DSPG), using local non-dimensional numbers of Darcy (Da) and Prandtl (Pr) numbers expressing respectively the ratio of local permeability to a characteristic length L_c and the ratio of momentum diffusivity, ν^l , to heat

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diffusivity in the liquid, α^l :

$$\tau_{\text{darcy}} = \frac{Da}{Pr} \left(\frac{g^l}{1 - g^l} \right)^2 = \frac{\alpha^l \mathbb{K}}{\nu^l L_c^2} \left(\frac{g^l}{1 - g^l} \right)^2 \quad (4.19)$$

Last, the definitions of the remaining stabilisation parameters are given as follows:

$$\tau_{\text{PSPG}} = \tau_{\text{SUPG}} = \left(\left(\frac{2 \|\langle \mathbf{v}^l \rangle_{\Omega_E} \|}{h_{\text{stream}}} \right)^2 + \left(\frac{4 \nu^l}{h_{\text{stream}}^2} \right)^2 + \left(\frac{2 \left(1 + \Delta t \frac{\mu^l}{\rho_0' \mathbb{K}} \right)}{\Delta t} \right)^2 \right) \quad (4.20)$$

$$\tau_{\text{LSIC}} = \frac{h_{\text{stream}}}{2} \|\langle \mathbf{v}^l \rangle_{\Omega_E} \| Z(Re_{\Omega_E}) \quad (4.21)$$

with $Z(Re_{\Omega_E})$ being a local Reynolds-dependant function that evaluates to:

$$Z(Re_{\Omega_E}) = \begin{cases} Re_{\Omega_E}/3 & \text{if } Re_{\Omega_E} \leq 3 \\ 1 & \text{if } Re_{\Omega_E} > 3 \end{cases} \quad (4.22)$$

and

$$Re_{\Omega_E} = \frac{\|\langle \mathbf{v}^l \rangle_{\Omega_E} \| h_{\text{stream}}}{2 \nu^l} \quad (4.23)$$

4.3.3 Implementation

Time step

One solving a convective transport problem, the time step may lead to a transport inaccuracy if the so-called Courant–Friedrichs–Lowy (CFL) condition is not satisfied. The latter imposes that the optimal time step value be determined from the mesh size, h , and the velocity, $\langle \mathbf{v}^l \rangle$, over the domain as follows:

$$\Delta t \leq \Delta t_{\text{CFL}} = \min_{\Omega} \frac{h}{\langle \mathbf{v}^l \rangle} \quad (4.24)$$

In this work, we chose to compute an initial optimal time step using eq. (4.24), but keep it constant for all simulations.

Integration order

The quadratic integrals terms in eq. (4.13) are interpolated at predefined integration points. The number of these points needed to evaluate the integrals depends on the

form of finite elements as well as on the polynomial degree that we want to integrate. The higher the degree, the more points are needed to get the most accurate results. Therefore special care should be given choosing an integrator degree to solve the previous Navier-Stokes system.

4.4 Application to multicomponent alloys

4.4.1 *Tsolver* validation with fluid flow

To validate the *Tsolver* with fluid flow, we consider the following set of equations already defined in the flowchart of [section 2.1.1](#). Moreover, an assumption of a static and non deformable solid phase is made. Consequently, the mechanical model is reduced to the conservation of momentum in the liquid phase.

The *Tsolver*'s ability to be coupled with various physical phenomena like macrosegregation and fluid flow in porous medium is validated using an experimental solidification benchmark. The validation case consists of a 10 cm width \times 6 cm height \times 1 cm thick crucible containing a Sn-3 wt.% Pb melt. The alloy is cooled down from its two narrowest vertical sides using heat exchangers (LHE: left heat exchanger, RHE: right heat exchanger). The experiment, inspired by [Hebditch and Hunt \[1974\]](#) similar set up, has been revisited by [Hachani et al. \[2012\]](#) who performed the solidification with better controlled conditions and using an increased number of samples for composition analysis. Recently, a successful attempt to simulate the experiment was carried out by [Carozzani et al. \[2013\]](#) relying on an enthalpy resolution. All details regarding geometry, finite element discretisation, material properties and boundary conditions can be found in the latter reference.

For this computation, solidification paths, phase compositions and phase enthalpies were determined by a thermodynamic module dedicated to equilibrium calculations for binary alloys. The 3D simulation results in [fig. 4.2](#) show a satisfactory agreement with the experimental temperature measurements recorded at mid-heights of the cavity and uniformly distributed along its width.

Furthermore, simulation results with the *Tsolver* and the *Hsolver* previously obtained by [Carozzani et al. \[2013\]](#) were found to be almost superimposed. This is confirmed by a comparison made between both solvers, as shows [fig. 4.3](#), where the average composition, liquid fraction and temperature fields are extracted from a cut plane halfway through the ingot. On the same figure, if we compare the composition, we notice that the solidified part on the RHE side has basically the same segregation pattern, while the convected liquid has a slightly different solute distribution.

As for the extent of the mushy zone, we observe that liquid fraction contours are very close, indicating that temperature distributions and interdendritic segregation are also close between both solvers predictions.

Regarding the computation, the *Tsolver* resolution proves to be faster than the *Hsolver* used by [Carozzani et al. \[2013\]](#): a process time of 7000 s required a computation time of 90 hours 13 minutes compared to 114 hours 21 minutes spent by the enthalpy resolution with 32 cores on the same cluster. The gain factor is about 20%.

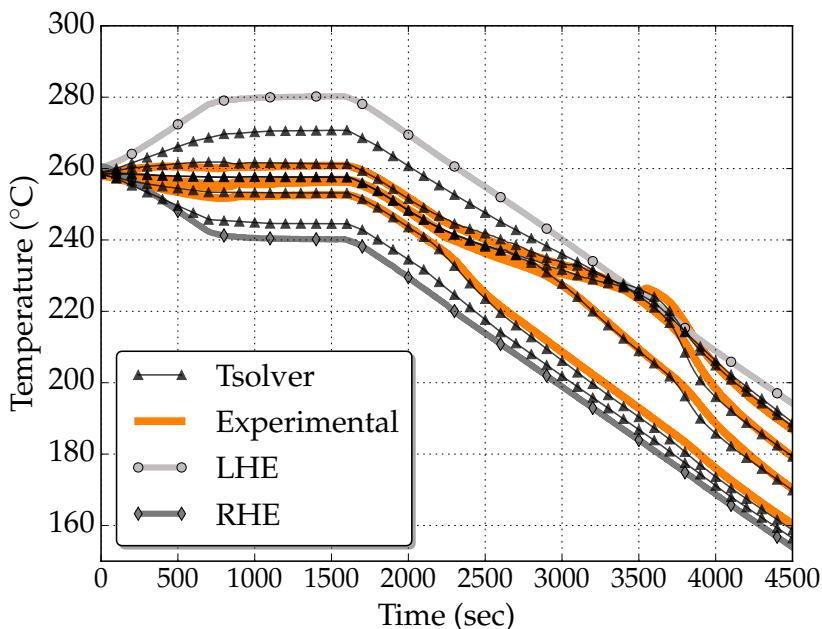


Fig. 4.2 – Results of the 3D FE convection-diffusion simulation, overlapping with the experimental cooling curves. The left (LHE) and right (RHE) heat exchangers impose the boundary temperature in the experiment.

In the previous chapter, we have considered a static melt upon solidification of multi-component alloy. This artificial assumption is dropped in this chapter, hence taking into account solute transport caused by fluid motion, using realistic values of thermal and solutal expansion coefficients given in [table 3.3](#). In such conditions, the melt is in constant motion and knowing that the carbon and chromium solutes have lightening effects on the liquid at nominal composition, the density inversion resulting from the composition gradient in the interdendritic liquid, may cause flow instability (segregation plumes) at the solidification front. While the selected alloy is a steel, this application is also representative of directional cooling in a single crystal casting, e.g. for nickel-base superalloys [[Beckermann et al. 2000](#)]. Solidification of this class of alloys is carefully controlled so as to prevent any freckle-type defect to exist in the as-cast state. In this section, we consider the same simulation parameters defined

4.4. Application to multicomponent alloys

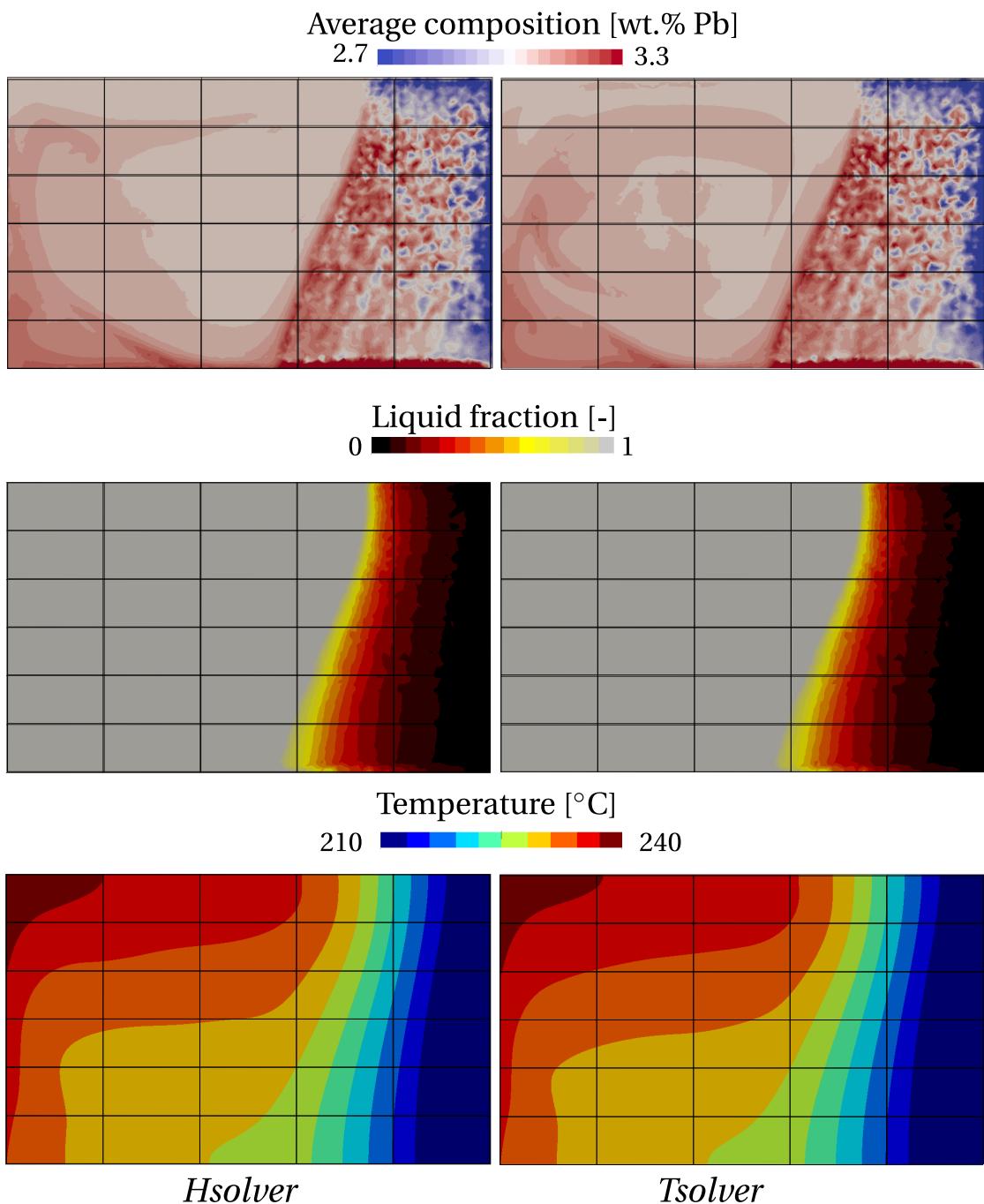


Fig. 4.3 – Comparison of 3D simulation results: average composition, liquid fraction and temperature at $t=3000$ s for *Tsolver* and *Hsolver*.

in [table 3.3](#) as well as the cylindrical geometry and thermal boundary conditions previously defined in [fig. 3.6](#). Moreover, we solve the liquid momentum conservation equation, with non-slip boundary conditions on all external sides of the cylinder.

4.4.2 Results

Solidification starts at 407 s when the cylinder's bottom base temperature reaches the liquidus temperature of the alloy. In fact, the solidification onset is the same as in the pure diffusion case in [fig. 3.9](#), since the average composition remains unchanged for an entirely liquid domain (assuming an initially infinite solute mixing in the melt).

As shown in [fig. 4.5](#) at 600 s, the first solid phase to form remains ferrite. We can also see solute-rich channels forming in the mushy zone and solute plumes rising in the melt above the mushy zone due to a subsequent upward flow. It is actually caused by the thermosolutal buoyancy force created by the carbon and chromium solutes. Such phenomenon delays solidification inside the liquid-rich channels and could result in freckling defects [Felicelli et al. 1991] on the surface of the cylinder as well as inside, as shown later in this section. As solidification proceeds, the liquid becomes more enriched with solute and the peritectic reaction forming the austenite phase is reached. However, for very large enriched melt, it can also be observed that primary solidification proceeds with the austenite phase rather than the ferrite phase. The carbide phase can form with the austenite phase at some locations. These observations correspond to a simulation time of 2000 s in [fig. 4.5](#).

Solidification ends at around 2475 s, the last liquid solidifying at the cylinder's top surface, where the average composition reaches a maximum of Fe-2.151 wt.% C-30.633 wt.% Cr, i.e. a relative positive macrosegregation, $(\langle w_i \rangle - \langle w_i \rangle_0) / \langle w_i \rangle_0$, of 7.5% for carbon and 2.1% for chromium. The fact that the maximum average composition is observed at the top, is verified in [fig. 4.4](#) which shows the composition map in a 2D vertical slice through the longitudinal axis of the cylinder. We can also see it in [fig. 4.6](#) where the relative composition profile are plotted at the end of the cooling process along the longitudinal cylinder axis Z-Z' and along the axis of the segregated channel, F-F'. Segregation becomes more and more negative up to 1 cm from the chill, corresponding to solute depletion caused by the first solid formation. Subsequent solidification enriches further the liquid; hence the solid composition also increases.

The composition evolution trend for both solutes is similar: an overall rise until positive segregation is achieved above 5 cm from the chill on ZZ' in [fig. 4.6](#). The positive macrosegregation intensifies when the profile is chosen at the center of the segregated channel, negative segregation then becoming less pronounced.

4.4. Application to multicomponent alloys

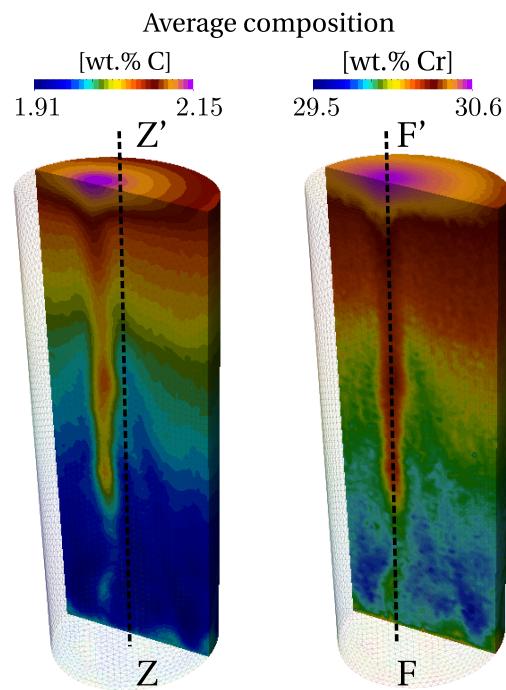


Fig. 4.4 – Average composition map on a vertical section inside the sample, along two vertical lines: the cylinder revolution axis and another axis parallel to the latter but passing through a segregated solidified channel.

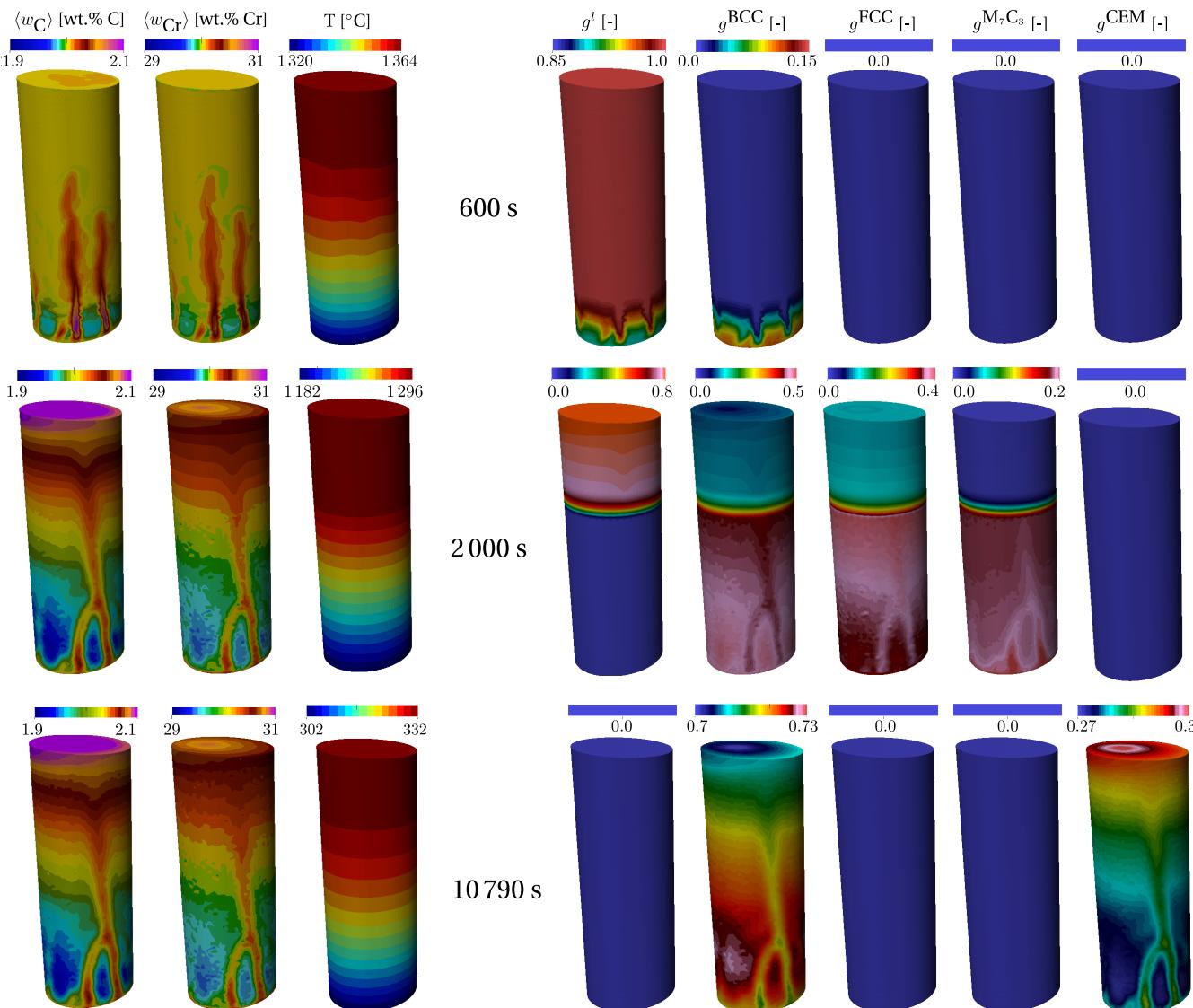


Fig. 4.5 – Upward solidification of a cylinder rod at 3 stages showing the metallurgical consequences of macrosegregation Fe-2 wt.% C-30 wt.% Cr. The left columns show the average composition and temperature distribution, while the right columns show the phase fractions.

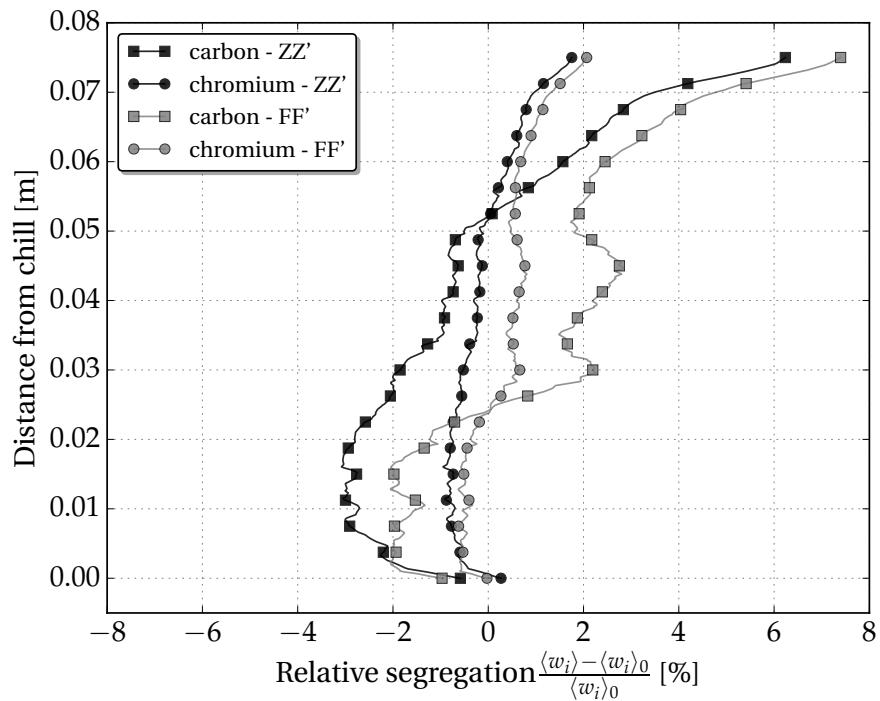


Fig. 4.6 – Relative macrosegregation profiles along the vertical revolution axis.

Beyond 2475 seconds, no variations of the average composition maps are observed since solidification is complete. Nonetheless, as temperature decreases, solid-state transformations are still possible as for the case with no macrosegregation. The formation of a cementite phase begins at the cylinder base at 8843 s with a temperature of 496.9 °C. At about 9293 s, the isotherm 488.5 °C reaches the top surface. This temperature value is the local cementite solvus temperature. The difference in the solvus temperature between the bottom and top surfaces is due to macrosegregation. Macrosegregation also explains the variation in the cementite content. The solid state transformation ends shortly before 10 500 s.

Comparing figs. 3.9 and 4.5, the influence of the solidification process is clear on the final macrosegregation pattern, hence the final phase distribution. This is better illustrated by drawing the time evolution of phase fractions at the center of the bottom and top surfaces of the cylinder in fig. 4.7. With no macrosegregation, in fig. 4.7a, the final distribution of the phases is the same at time 12 000 s, while in the presence of macrosegregation, as shown in fig. 4.7b, variations of the cementite and ferrite are revealed. The segregated channels inside the cylinder and on the boundary, often lead to *freckles*, where they consist of visible equiaxed grains [Copley et al. 1970]. This defect is marked by a noticeable gradient of composition and phase fractions, possibly changing the mechanical properties in the channels, hence the overall mechanical be-

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haviour of the cast part. The coupling of the *Tsolver* with thermodynamic tabulations is thus demonstrated. It shows the ability to predict complex transformation paths, even if only at equilibrium. As for the computation time, the *Tsolver* resolution performed better: 500 seconds of solidification required 6 hours 14 minutes compared to 8 hours 6 minutes spent by the enthalpy resolution with 12 cores on the same machine. The gain factor is about 22%.

4.4. Application to multicomponent alloys

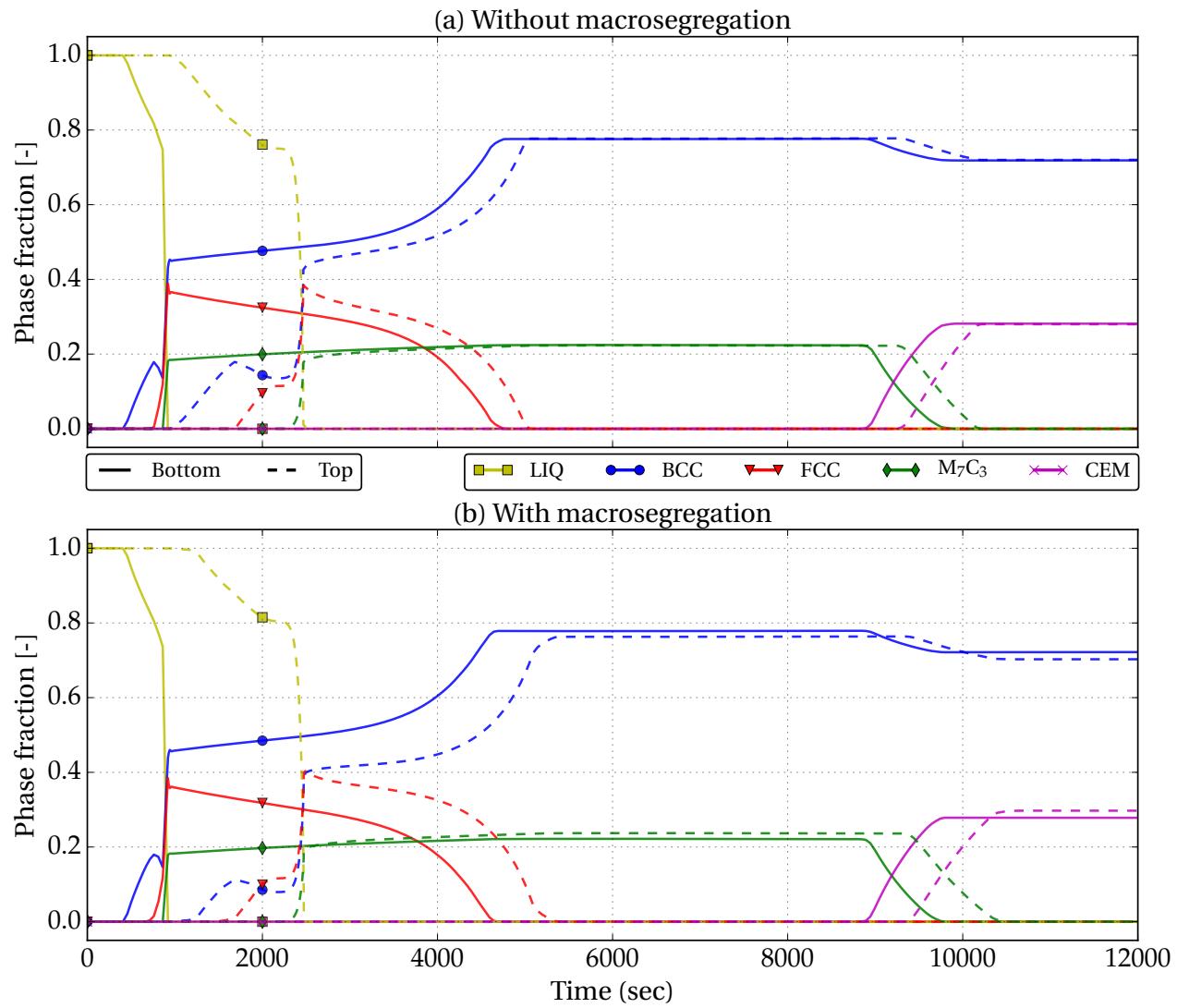


Fig. 4.7 – History of phase fraction (a) without macrosegregation and (b) with macrosegregation at the center of the bottom (solid lines) and top (dashed lines) of the cylinder surfaces. These plots are extracted from simulations displayed in (a) [fig. 3.9](#) and (b) [fig. 4.5](#).

4.5 Macroscopic prediction of channel segregates

4.5.1 Introduction

We have seen in the previous multicomponent solidification test case, a formation of segregated channels in the cylinder. This defect manifests itself as a composition inhomogeneity that is highly non-isotropic. A typical description of its morphology would consider a channel with a diameter proportional to few primary dendrite arm spacing and a length that could vary from millimeters to centimeters. These “worm”-like shapes may form during directional solidification of cast parts designed for engine applications, particularly in nickel-base superalloys [Giamei and Kear 1970; Beckermann et al. 2000; Genereux and Borg 2000; Schneider et al. 1997]. In the latter situation, the channels are often filled with a chain of small equiaxed crystals, thus referring to the term “freckle”. In large steel ingots, these channel defects are also related to A- and V-segregates [Pickering 2013].

Considering a binary alloy with a partition coefficient less than unity and having a negative liquidus slope, channel segregates may form by the following mechanisms: i) solute partitioning occurs at the scale of dendrite arms and solute is rejected in the melt, ii) local composition gradients are intensified resulting in an increase of the solutal buoyancy force in the mushy zone, iii) solute-rich pools are formed, causing segregation chimneys and convective plumes in the melt, iv) which lead to partial remelting and transport of dendrites, continuous solute feeding and locally delayed solidification, and finally v) accumulation of fragments and/or equiaxed crystals in the chimneys before the end of solidification.

Because it is of prime importance to control the occurrence of channel segregation, several attempts have been made from the late 1960's [Flemings and Nereo 1967; Flemings et al. 1968; Flemings and Nereo 1968] to the early 2000's [Ramirez and Becker-
mann 2003] to understand it and characterise it by deriving freckling criteria. These studies are summarized in [Auburtin 1998]. One of the reasons for only considering freckling criteria is that direct realistic simulations of the formation of freckles in a casting geometry are still difficult. It is indeed not crucial for industrial applications, as freckles cannot be cured and lead systematically to part rejection, except for casting ingots, where channel segregation cannot be avoided.

Experimental observations show that it requires a satisfying description of the microstructure together with the 3D convective flow controlled by the cooling conditions of the complete cast part [Shevchenko et al. 2013]. Such information is not accessible yet. Only simulations in representative simple cuboid or cylindrical domains are usually achieved [Felicelli et al. 1991; Felicelli et al. 1998; Kohler 2008; Guo and Beck-

ermann 2003], except when considering small volume casting [Desbiolles et al. 2003]. They are usually limited to unstable thermosolutal convection without or with little regard to the microstructural features. Considering the spatial resolution of the defect, being for example of the order of the primary dendrite arm spacing, a fluid flow computation in the 3D casting part is also very demanding and not common in the literature. Among other criteria, the dimensionless Rayleigh number has been identified as a good indicator for the occurrence of segregation channels and freckle defects. The dependence of freckling tendency on the Rayleigh number has been studied numerically and compared to experimental observations, as done by [Ramirez and Beckermann 2003].

4.5.2 Experimental work

An interesting experimental work on directional solidification of In-75 wt.% Ga featuring in-situ X-ray monitoring has been recently carried out by Shevchenko et al. [2013] at Dresden's Helmholtz institute. We give here a detailed description of their experimental setup, as this experimental work has been used as a reference to test the performance of our numerical simulations. This was done through a collaboration resulting in the joint publication [Saad et al. 2015]. The comparison with numerical modelling is paramount for two main reasons: firstly, the in-situ technique allows to follow solidification in real-time and offers visual description of the system behaviour: grain morphology, composition evolution, effect on fluid flow in the mushy zone and chimney initiation, as well as other modelling input data such as dendritic and eutectic nucleation undercooling; secondly, an indium-gallium system is more representative of metallic alloy solidification than the widely used organic systems, e.g. the succinonitrile-acetone mixture that exhibits alloy-like dendritic formation in its growth stage. Further information with respect to the experimental hardware, procedure and data analysis can be found in [Boden et al. 2008; Shevchenko et al. 2013].

4.5.3 Macroscopic scale simulations

Configuration

The focus of this section is on qualitative comparison between numerical simulation and the previously mentioned experiment. The experimental cell geometry shown in fig. 1.7c is hexagonal. With adiabatic lateral sides, it results in a bending of the isotherm surfaces as shown in the experiment. The metallic cooling plates shown in

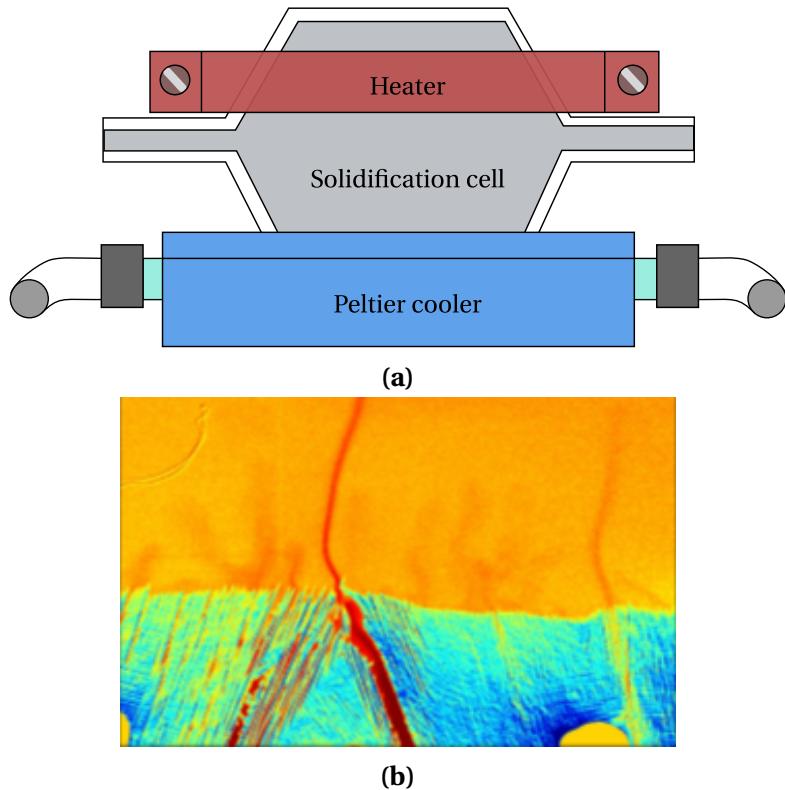


Fig. 4.8 – Illustration of the benchmark experiments for in-situ observation of segregated channels formation using X-Ray radiography with (a) a schematic of the cell and (b) a typical image of the microstructure formed during directional solidification of an In-75 wt.% Ga alloy.

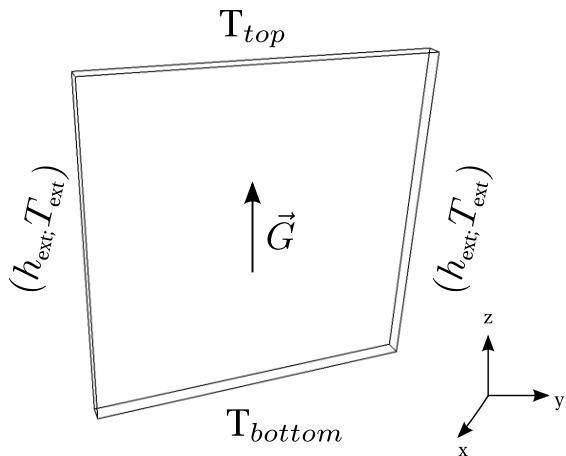


Fig. 4.9 – Computational geometry used for simulating channel segregation phenomena, along with top and bottom imposed boundary temperatures, T_{Top} and T_{Bottom} , and lateral Fourier cooling fluxes using a heat transfer coefficient and an external temperature, h_{ext} and T_{ext} . The resulting temperature gradient, G , is parallel to the vertical z-axis.

4.5. Macroscopic prediction of channel segregates

[fig. 4.8a](#) partly compensate for this effect. However, a residual horizontal component of the temperature gradient remains.

To qualitatively replicate this effect while simplifying the cell geometry, a cuboid cell measuring $22\text{ mm} \times 22\text{ mm} \times 1\text{ mm}$ is considered. On the lateral vertical side surfaces, small cooling fluxes are imposed, after being computed from a constant heat transfer coefficient, h_{ext} , and a constant external temperature, T_{ext} , as shown in [fig. 4.9](#). On the same figure, temperatures at the bottom and top surfaces, respectively T_{Top} and T_{Bottom} , are imposed in a way to maintain a constant vertical gradient, G , thus linearly decreasing over time with the same cooling rate R . Both square faces of the geometry, having an area of $22\text{ mm} \times 22\text{ mm}$, are adiabatic. In spite of taking cell dimensions similar to benchmark experiments presented above, the cell thickness is increased from $150\text{ }\mu\text{m}$ to 1 mm . One of the main obstacles of simulating a $150\text{ }\mu\text{m}$ -thick cell is the number of elements in the thickness. An insufficient number may lead to incorrect velocity computation in the cell thickness, resulting in inaccurate channel segregation prediction. In general, a number around 5 to 8 elements is needed to get a better representation of the fluid motion. When the domain thickness is $150\text{ }\mu\text{m}$, imposing 8 elements in the thickness results in a huge homogeneous isotropic mesh containing about 10^9 elements, the mesh size being equal to 0.018 mm . For now, this would only lead to slower simulations and therefore more complicated analysis with the large data files. Instead, we choose to keep the same number of thickness elements, but increase the thickness instead. That way, the resulting isotropic mesh would reduce to 2.5×10^4 elements. Therefore, we keep the latter thickness as the current study is limited to qualitative comparisons, but the mesh size is imposed to about $2\lambda_2=140\text{ }\mu\text{m}$, and the fixed finite element grid contains around 2×10^6 elements and 3×10^5 degrees of freedom.

Table 4.1 – Summary of the simulations and the corresponding parameters for the FE cases, where a purely macroscopic model is used. Parameters are varied from (G1) low to (G2) high gradient and (L0) no, to (L1) low lateral cooling.

Case	Vertical gradient $\ G\ $ [Kmm $^{-1}$]	Cooling rate R [Ks $^{-1}$]	Lateral cooling L ($h_{\text{ext}}, T_{\text{ext}}$) [W m $^{-2}$ K $^{-1}$, °C]	Initial temperature $(T_{\text{top}}, T_{\text{bottom}})$ [°C]
FE-G1R1L0	G1:0.2	R1:-0.01	L0:(0,0)	(29.75, 25.25)
FE-G1R1L1	G1:0.2	R1:-0.01	L1:(20,0)	(29.75, 25.25)
FE-G2R1L1	G2:1.5	R1:-0.01	L1:(20,0)	(58.25, 25.25)

Materials properties are provided in [table 4.2](#) while initial and boundary conditions are given in [table 4.1](#). A series of computations is performed to understand the influence of process parameters on the final macrosegregation pattern. In directional growth,

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the main parameters are the vertical temperature gradient, G , and the cooling rate, R , since they control the isotherms speed. However, the effect of a higher lateral cooling is also considered below by increasing the heat transfer coefficient, h_{ext} . Finally, the grain structure is another crucial parameter that drastically changes the analysis inasmuch as growth undercooling is fundamental to determine the onset of solidification.

The computation cases used in this study are presented in [table 4.1](#). The label of each case allows direct access to the simulation parameters as explained in the caption. Values for these parameters are inspired from the above experiments ([section 4.5.2](#)). Initial conditions consider a quiescent liquid at uniform composition given by the nominal alloy composition $\langle w_0 \rangle$. The temperature field is also initially uniform at a temperature averaged between the top and bottom initial values provided in [table 4.1](#). It has been checked that a uniform temperature gradient is swiftly reached, and that the unsteady regime to settle a vertical temperature gradient does not affect the phenomena studied. For simulations with grain structures, boundary conditions for nucleation at the bottom horizontal $22 \text{ mm} \times 1 \text{ mm}$ surface are kept constant as given in [table 4.1](#).

The macroscale approach employs the finite element method to compute the temperature and composition fields at FE nodes. The liquid fraction is then determined directly from the former fields, assuming a linear phase diagram, i.e. linear liquidus with full thermodynamic equilibrium between phases or lever rule approximation. This linear approximation is made available by the dotted line provided in [fig. 4.10](#). Note that this line defines a phase diagram that seems very different from the correct one. However, this linear fit is only used in a composition region located around the nominal composition of the alloy. It is also worth noticing that the eutectic microstructure is expected to appear at 15.3°C . Nevertheless, experimental observations revealed that large eutectic nucleation undercooling was reached, so the eutectic solidification was not reported in the experiments studied by [Shevchenko et al. \[2013\]](#). Consequently, the solidification path is computed without accounting for the eutectic microstructure in the present simulations, thus extending the liquidus and solidus lines below the eutectic temperature as sketched with the linear approximations in [fig. 4.10](#).

Results

The first case labeled FE-G1R1L0 is a reference case that features a low gradient (G1), low cooling rate (R1), and without any lateral cooling (L0), ensuring that isotherms retain a planar shape. These simulation parameters defined in [table 4.2](#), result in a negligible fluid flow reaching a maximum velocity of $4 \times 10^{-8} \text{ mm s}^{-1}$ in the bulk.

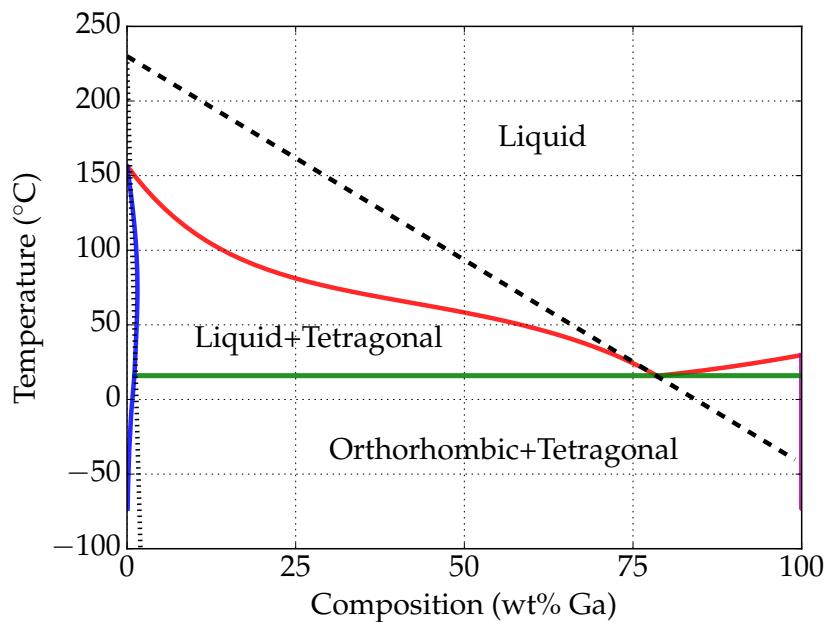


Fig. 4.10 – Binary phase diagram of the In-Ga system [Andersson et al. 2002; TCBIN 2006] and its approximation for solidification studies with an In-75 wt.% Ga alloy. The dashed and dotted lines are linear liquidus and solidus approximations near the nominal composition.

Accordingly, the solidification front remains stable and follows the planar isotherms; no convective plumes are observed. The average composition field is thus only little modified in the mushy zone as shown in fig. 4.11 (mind the values of the scale limits). It is concluded that velocity in the bulk is not high enough to initiate instabilities. In the next case, FE-G1R1L1, a cooling flux with a constant and very low value of the heat transfer coefficient is imposed on both vertical lateral surfaces to initiate a downward fluid flow due to thermal buoyancy. Once solidification starts, solute-rich regions start to appear on the sides of the domain. Despite the visible concentration difference between these lateral regions and the central mush seen in fig. 4.11, their diffuse and uniform aspect indicates no resemblance to channel segregations. We keep the same configuration but increase the vertical gradient from 0.2 K mm^{-1} (G1) to 1.5 K mm^{-1} (G2) in the case FE-G2R1L1.

The isotherms become closer to each other hence reducing the depth of the mushy zone for the same time increment compared to the preceding case. The rejected gallium solute locally accumulates at several different positions in the mushy zone, stemming from the base of the cell, with a maximum of 0.7 wt.%Ga above nominal composition. This is the consequence of segregation of gallium rich liquid being lighter than the above liquid bulk and creating an upward buoyancy force. A positive segregation and subsequent Ga-rich chimneys then rise up with an upward velocity component

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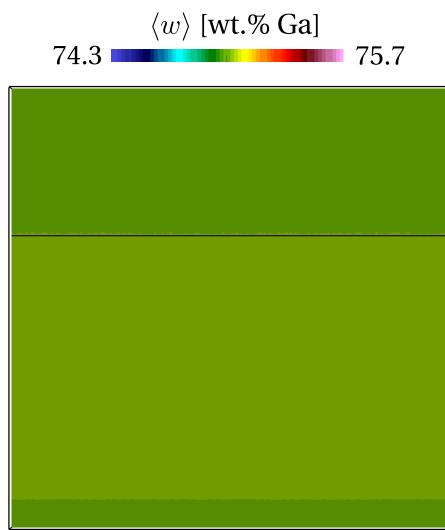
Table 4.2 – Material parameters for In-75 wt.% Ga and numerical parameters.

Parameter	Symbol	Value	Unit
Nominal composition	$\langle w_0 \rangle$	75	wt.%
Liquidus temperature	T_l	25.25	°C
Segregation coefficient	k	0.0165	
Liquidus slope	m_l	-2.73	K wt.% ⁻¹
Gibbs-Thomson coefficient	Γ_{GT}	2×10^{-7}	K m ⁻¹
Heat capacity	C_p	380.74	J kg ⁻¹ K ⁻¹
Enthalpy of fusion	L	8.02×10^{-4}	J kg ⁻¹
Diffusion coefficient of Ga in liquid In	$\langle D \rangle^l$	1.525×10^{-9}	m ² s ⁻¹
Dynamic viscosity	μ^l	2×10^{-3}	Pa s
Thermal expansion coefficient	β_T	0.0978×10^{-3}	K ⁻¹
Solutal expansion coefficient	$\beta_{\langle w \rangle^l}$	1.44×10^{-3}	wt.% ⁻¹
Thermal conductivity in the solid	$\langle \kappa \rangle^s$	40	W m ⁻¹ K ⁻¹
Thermal conductivity in the liquid	$\langle \kappa \rangle^l$	28	W m ⁻¹ K ⁻¹
Dendrite arm spacing	λ	60×10^{-6}	m
Density	ρ_0^l	6725	kg m ⁻³
Reference composition	w_0^l	75	wt.%
Reference temperature	T_0	25.25	°C
CA cell size		30×10^{-6}	m
FE mesh size		140×10^{-6}	m
Time step	Δt	0.1	s

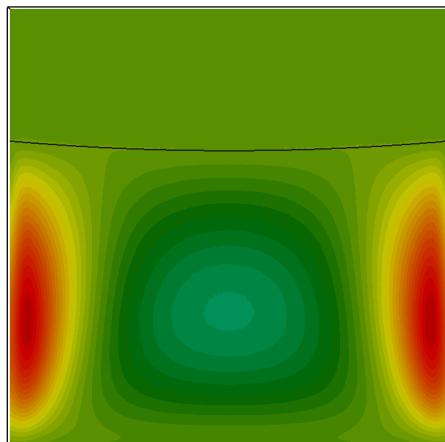
slightly greater than 1 mm s⁻¹.

Figure 4.12 gives a series of snapshots for case FE-G2R1L1 at three different times. Among the two clear distinct plumes that are visible at 250 s in fig. 4.11, only one has led to the formation of a segregated channel that remains in fig. 4.12 at 500 s. In fact, an animation between 250 s and 500 s (not shown here) reveals that one plume vanishes, thus permitting the first one to further develop. A second segregated channel is also seen on the left hand side of the cell. These two channels are stable for a long time since they remain at time 1000 s. However, the left side channel develops further to become the main one at 1500 s, while the mid-width channel decreases in intensity, changes orientation and subsequently disappears (not shown here). Thus, the birth and death of very few channels is observed in this simulation, mainly due to solutal instability, as the temperature field shown in fig. 4.12 clearly remains stable despite the low lateral heat flux. As shown in fig. 4.11, instability is yet required to create these chemical plumes and channels. Here, it is created by a very small lateral heat flow but other sources of instability could be involved, as shown with the grain structure in the

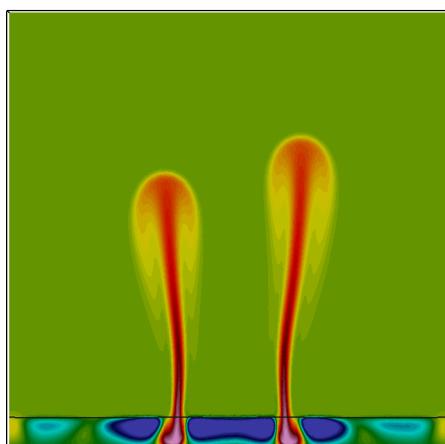
4.5. Macroscopic prediction of channel segregates



(a) FE-G1R1L0



(b) FE-G1R1L1



(c) FE-G2R1L1

Fig. 4.11 – Average Ga composition field at 250 s for the 3 FE cases showing the influence of process parameters on the freckling tendency. The black line represents the liquidus isotherm given in [table 4.2](#).

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next section.

4.5. Macroscopic prediction of channel segregates

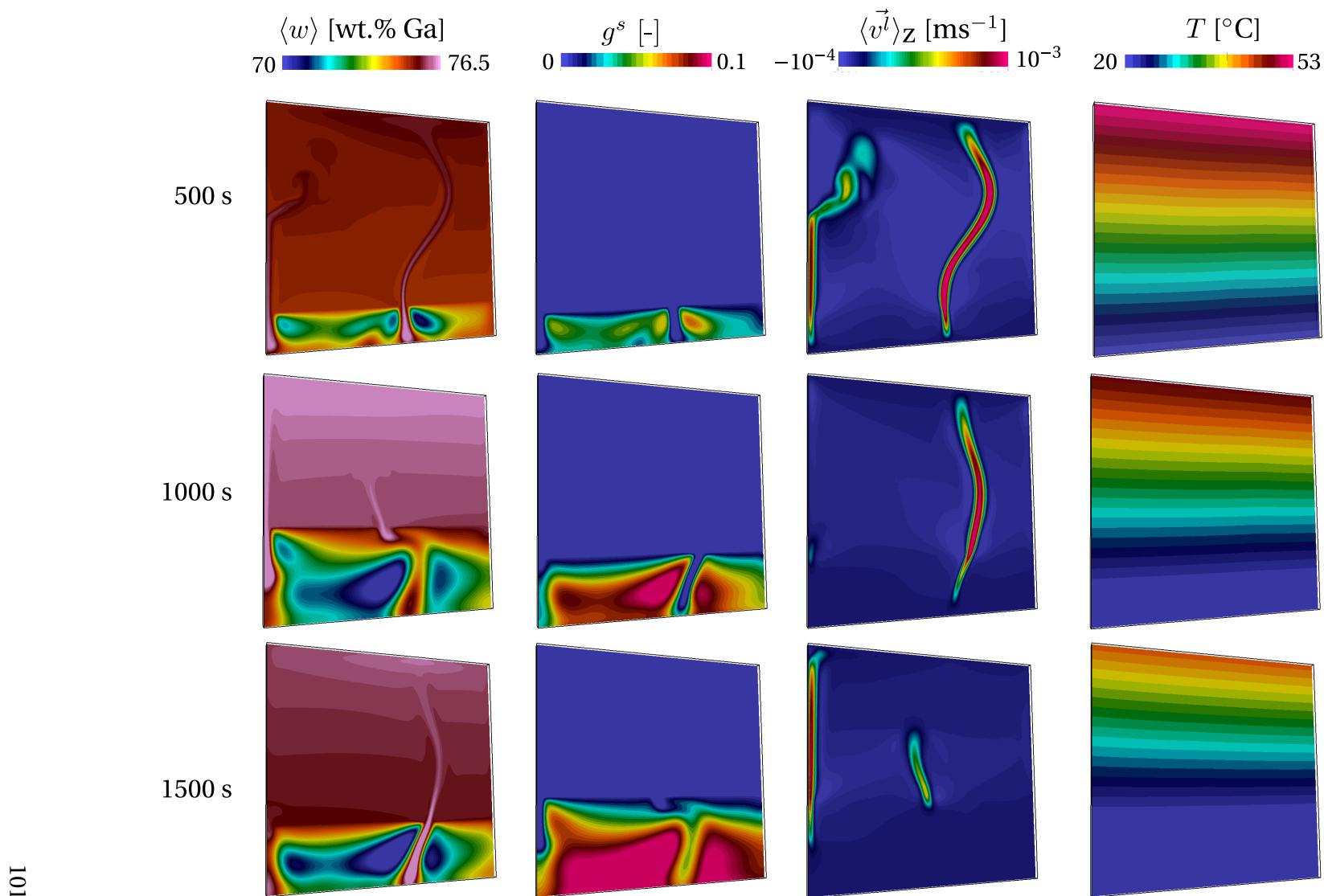


Fig. 4.12 – Simulation results for case FE-G2R1L1 showing maps of the average composition in gallium, the solid fraction, the vertical component (z-axis) of the superficial velocity field and the temperature, on a cut plane at the center of the cell at 500 s, 1000 s and 1500 s.

Discussion

In section 4.5.1, we have introduced some successful attempts of freckle predictions. The authors tackled the problem from an qualitative perspective. To our knowledge, the closest work to quantitative freckling analysis in solidification literature was done by Ramirez and Beckermann [2003]. They attempted to draw a correlation (freckling criterion) between the process parameters and the occurrence of freckles, without any size or shape constraints, i.e. any flow instability that may appear and form the smallest freckle is considered.

To accomplish this, they took a number of experiments done independently by Pollock and Murphy [1996] and Auburtin et al. [2000] where the casting parameters vary one at a time: casting speed (R), thermal gradient (G), angle (θ) with respect to vertical orientation and nominal composition ($\langle w_0 \rangle$), giving a database for 6 different superalloys. The experimental results were compared to a modified Rayleigh number that accounts for the various parameters. It allowed them to define a threshold for freckle formation in Nickel-base superalloys, as well as Pb-Sn alloys.

Other contributions by Yuan and Lee [2012] (Pb-Sn alloy) and Karagadde et al. [2014] (In-Ga alloy) relied on a Cellular Automata Finite Difference (CAFD) model developed by Lee et al. [2002], which solves the dendrite tip growth kinetics at the solid-liquid interface together with macroscopic conservation equations. The authors compared the simulated formation of freckles with the results obtained by Shevchenko et al. [2013]. However, these simulations follow solidification in a small volume that contains a few dendrites with interdendritic liquid, therefore limited as far as to predict the liquid behaviour outside the mushy zone. On another hand, experimental observations reveal a great deal of information regarding solute redistribution, first in the chimneys that wash the dendrites in their way and then convective plumes that expel chemical species outside the mush, resulting in a global complex phenomenon.

In order to capture simultaneously the interaction between the mushy zone and the free liquid, we use the Cellular Automata Finite Element (CAFE) method to combine the macroscopic and mesoscopic length scales and predict more realistic channel segregation.

4.6 Meso-Macro prediction of channel segregates

4.6.1 Numerical method

Microscopic scale

The CAFE model introduces a grid of regular and structured cubic cells, with a constant size in all space directions, referred to as the cellular automaton (CA) grid. It is different from the unstructured finite element mesh previously mentioned for the solution of the average conservation equations. A typical CA step dimension is smaller than the smallest FE mesh size. The CA grid serves to represent solidification phenomena including nucleation, growth and remelting of the envelope of the primary dendritic grains. Details about the CAFE model can be found in [Carozzani et al. 2012; Carozzani et al. 2013; Carozzani et al. 2014]. Cell information, such as the temperature, the average composition or the velocity of the liquid phase, is interpolated from the nodes of the FE mesh. State indices are also defined for each CA cell, providing the presence of liquid or solid phases.

Nucleation

Initially, cells are in a fully liquid state. In the present situation, random nucleation sites are chosen based on a nucleation density, n_{\max} (expressed in surface density inverse m⁻²), at the bottom surface of the geometry in contact with the cooler. Nucleation occurs in a cell only if the latter contains a nucleation site, and when the local undercooling of the cell reaches the critical nucleation undercooling given as input by a Gaussian distribution of mean undercooling ΔT_N with a standard deviation ΔT_σ . The crystallographic orientation of each grain newly nucleated is also randomly chosen using values of the Euler angles to fully define the three rotations that transform the reference frame to the $\langle 100 \rangle$ directions that define the main growth axes of the dendrite trunks and arms. Grain selection is therefore solely controlled by growth competition.

Growth

Dendrite growth is driven by the chemical supersaturation, Ω_{sat} , which is a dimensionless number proportional to the difference between the liquid composition at the dendrite tip and the melt composition far away from the tip. The higher the supersaturation, the faster the dendrite tip velocity. However, in the presence of a convective fluid, the chemical supersaturation is highly influenced by the intensity and the direc-

tion of the flow with respect to the growth direction of the dendrites. In the current model, convection is central in studying the formation of channel segregation. Therefore, the purely diffusive Ivantsov relation used to determine the Peclet number Pe as function of the supersaturation, is replaced by a modified relation using a boundary layer correlation model that accounts for both the intensity and the misorientation of the liquid velocity with respect to the growth direction of the dendrites [Gandin et al. 2003]. The main parameters for this growth kinetics models are the Gibbs Thomson coefficient, Γ_{GT} , and the diffusion coefficient for Ga in In, $\langle D \rangle^l$.

Solidification path

The CA model gives the presence of the grains in the liquid as well as its growth undercooling. For coupling with macroscopic scale modelling, the fraction of phases needs to be fed back to the FE model. This is now done by accounting for the information provided by the CA model. Thus, the fraction of solid is no longer the consequence of a simple conversion of the temperature and composition assuming thermodynamic equilibrium. It also includes the solidification delay due to the kinetics of the development of the grains as detailed elsewhere in the work of Carozzani et al. [2013].

Numerical method

Both the finite element mesh and the cellular automaton grid play a role in predicting channel segregation inasmuch as this type of defect originate from interplays between hydrodynamic instabilities on the scale of the dendrites and macroscopic flows defined by the geometry of the experimental cell [Shevchenko et al. 2013]. One has to respect a small maximum FE mesh size, comparable to the dendrite arm spacing. With such an element size, composition gradients giving rise to solutal buoyancy forces can be captured. This limits consequently the CA cell size, as a minimum number of cells is required in each finite element. In the array of simulations that will be presented in the next section, the value of λ_2 was considered. We have chosen a fixed mesh element size of $2\lambda_2$ and a CA cell size of $\lambda_2/2$. An average of 4 CA cells per unit length of a finite element is enough to accurately compute the development of the grain envelopes together with the solutal, thermal and mechanical interactions.

4.6.2 Configuration

Knowing that the configuration in FE-G2R1L1 produces segregated channels, the same set of parameters is first used for case CAFE-G2R1L1 by adding the effect of the grain structure using the CAFE model. Results are accessible in fig. 4.14 for comparison with

[fig. 4.12](#). A striking difference is seen: the composition maps become more perturbed as shown by the formation of numerous plumes when coupling with grain structure is active. The growing front displayed on the grain structure at the right most column of [fig. 4.14](#) dictates the leading position of the mushy zone shown in the third column. Note that each color corresponds to one grain, with 17 grains having nucleated at the cell's bottom surface. However, comparison of the solid fraction maps between [fig. 4.12](#) and [fig. 4.14](#) at the same times reveals a delay in the growing front position. Values of the nucleation parameters in [table 4.3](#) are such that few grains rapidly form below the nominal liquidus isotherm. The delay is therefore not due to the nucleation undercooling but to the growth undercooling of the dendrite tips. It should be noticed that, the growth front driven by undercooling in [fig. 4.14](#) also forms with a higher initial solid fraction and hence larger solute segregation occurs at the front. This effect, together with instabilities of the composition field, is caused by a more perturbed fluid flow and more plumes as observed in CAFE-G2R1L1 compared to FE-G2R1L1. Such observations fit to the complicated fluid and solute flow patterns typically occurring in the experiments as shown in [fig. 4.15](#). It becomes obvious that the consideration of grain structure and growth undercooling are vital to accurately simulate chimney formation in these experiments. The reasons for the instabilities are discussed hereinafter.

In the present 3D CAFE simulation, each grain shown in [fig. 4.13](#) is associated with a crystallographic orientation. The growth kinetics is only given for the $\langle 100 \rangle$ crystallographic directions at the grain boundaries with the liquid. The CA growth model is based on the hypothesis that, in a quiescent liquid of uniform temperature distribution and composition, the grain envelop should reproduce an octahedral grain shape with main directions given by the six $\langle 100 \rangle$ directions.

In the present situation where complicated fields are present for temperature, composition and liquid velocity, each grain envelope with different crystallographic orientation adapts differently to its local environment. Thus, the local undercooling of the front varies everywhere. Such variations are within few degrees here, but this is sufficient to create irregularities on the growth front, as seen on the grain structure in [fig. 4.13](#). Apart from that, these variations are linked to the position of the instabilities for the chemical and liquid velocity fields, thus demonstrating the full coupling between the CA and FE models.

Chapter 4. Macrosegregation with liquid metal motion

Table 4.3 – Summary of the simulations and the corresponding parameters for the CAFE cases, coupling macroscopic model with the grain structure model. Parameters are varied from (G1) low to (G2) high gradient, (R1) low to (R2) high cooling rate and (L0) no, (L1) low and (L2) high lateral cooling.

Case	Vertical gradient $\ G\ $ [K mm ⁻¹]	Cooling rate R [K s ⁻¹]	Lateral cooling L (h _{ext} , T _{ext}) [W m ⁻² K ⁻¹ , °C]	Initial temperature (T _{top} , T _{bottom}) [°C]	Nucleation (n _{max} , ΔT _N , ΔT _σ) [m ⁻² , °C, °C]
CAFE-G2R1L1	G2:1.5	R1:-0.01	L1:(20,0)	(58.25, 25.25)	(10 ⁶ , 1, 0.2)
CAFE-G1R1L1	G1:0.2	R1:-0.01	L1:(20,0)	(29.75, 25.25)	(10 ⁶ , 1, 0.2)
CAFE-G1R1L2	G1:0.2	R1:-0.01	L2:(500,0)	(29.75, 25.25)	(10 ⁶ , 1, 0.2)
CAFE-G2R2L1	G1:0.2	R2:-0.05	L1:(20,0)	(29.75, 25.25)	(10 ⁶ , 1, 0.2)

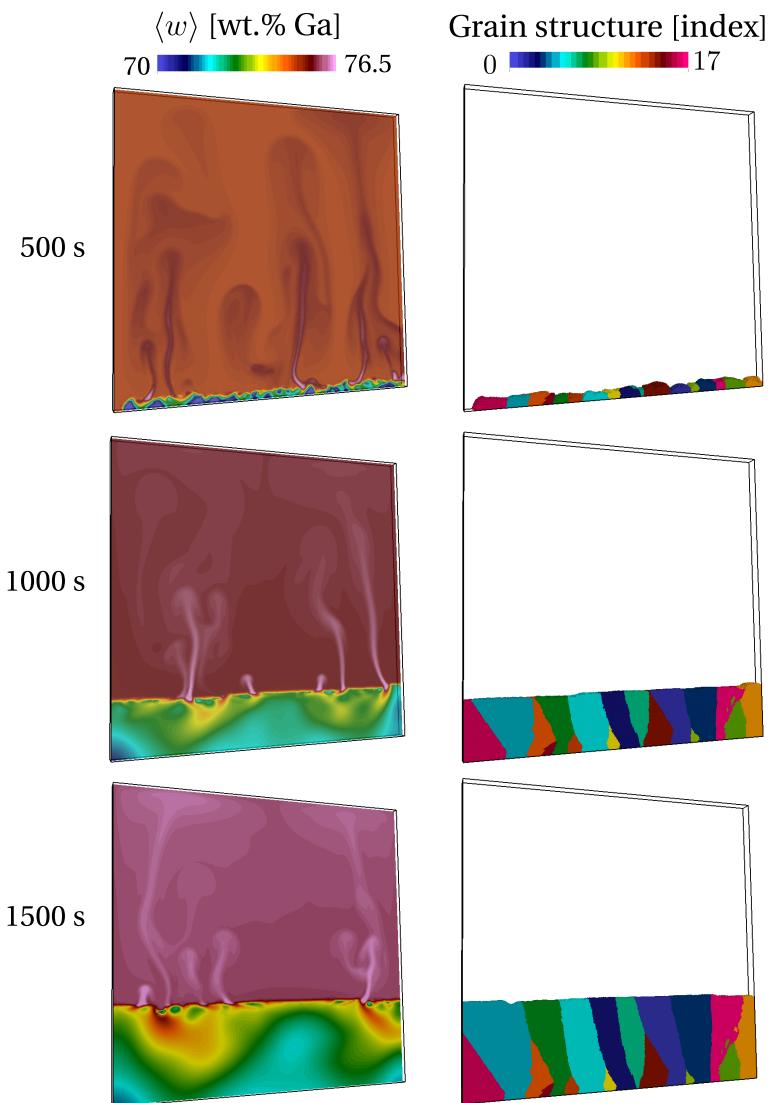


Fig. 4.13 – Simulation results the predicted of mushy grain structure with the corresponding composition maps, at 500 s, 1000 s and 1500 s.

4.6. Meso-Macro prediction of channel segregates

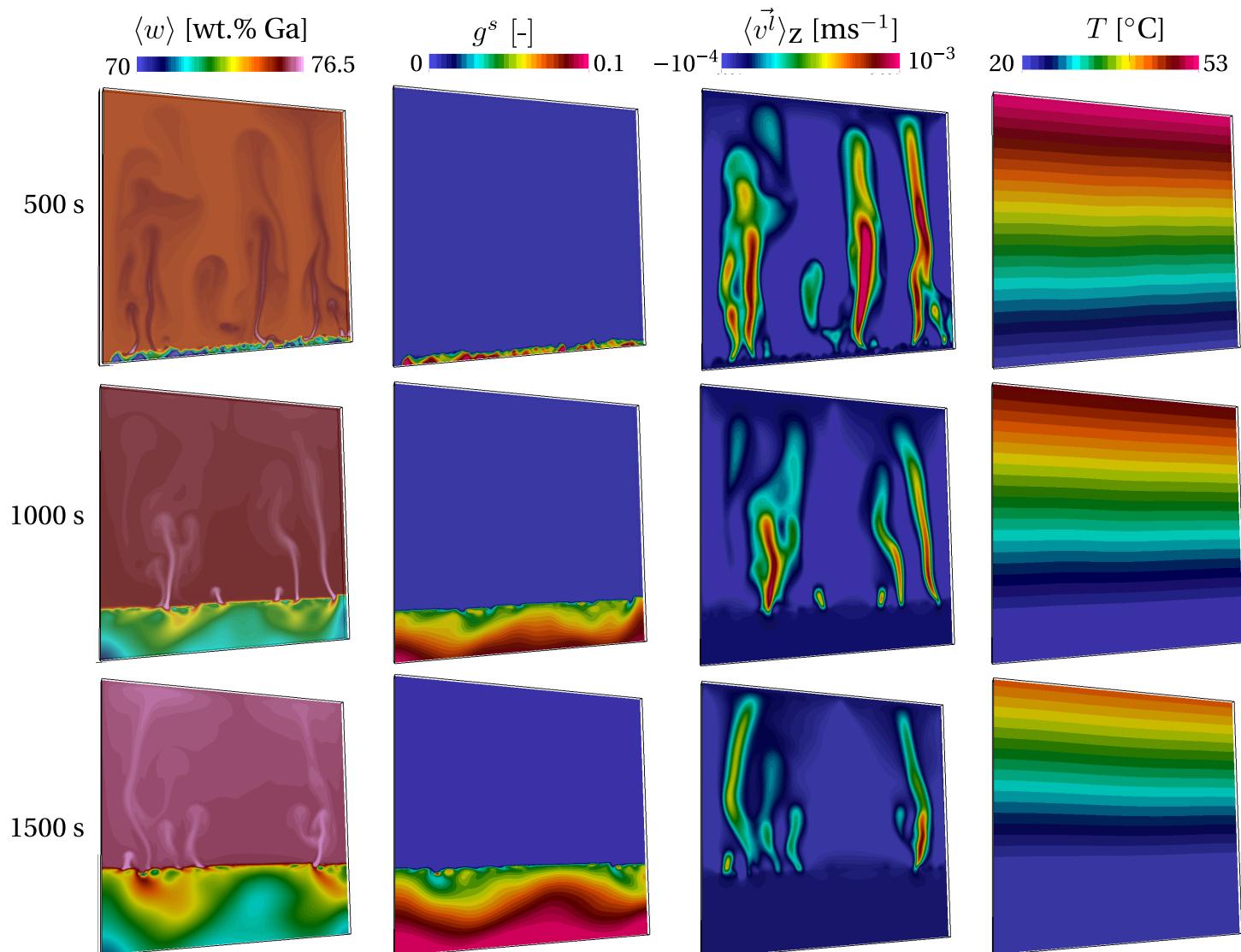


Fig. 4.14 – Simulation results for case CAFE-G2R1L1 showing maps of the average composition in gallium, the solid fraction, the vertical component (z-axis) of the superficial velocity field and the temperature, on a cut plane at the center of the cell at 500 s, 1000 s and 1500 s.

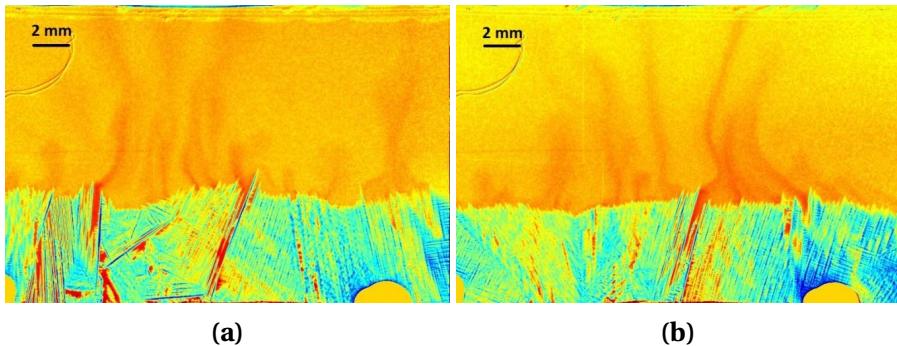


Fig. 4.15 – Snapshots of dendritic structure and composition field obtained from two solidification experiments at a cooling rate $R=-0.01 \text{ K s}^{-1}$ and temperature gradients of (a) $\|G\|=1.1 \text{ K mm}^{-1}$ and (b) $\|G\|=1.3 \text{ K mm}^{-1}$.

4.6.3 Effect of vertical temperature gradient

The influence of diverse process parameters can now be considered in context of the grain structure. The effect of the vertical temperature gradient is shown by comparing the previous case CAFE-G2R1L1 with case CAFE-G1R1L1. The temperature gradient is decreased about 7 times here, from $G_2=1.5 \text{ K mm}^{-1}$ to $G_1=0.2 \text{ K mm}^{-1}$. In fact, both cases share almost all traits with respect to flow patterns and velocity magnitude in the bulk. Main differences are yet seen regarding the dynamics of the plumes shown in fig. 4.16.

In the case of a low temperature gradient (G_1), the solidification front cannot maintain a shape as smooth as for the case of a large temperature gradient (G_2): the solute gradient in the liquid of the mushy zone (basically following the lever rule approximation for a given temperature) decreases, leading to a lower gradient of the solutal buoyancy force. In turn, more solute accumulates close to the front and locally reduces the growth velocity, thus creating larger “valleys” or steps with higher solute content. The irregular geometry of the front is also influenced by the dendrite tip growth kinetics model. The velocity of the isotherms is the ratio of the cooling rate, R , to the temperature gradient, G . Consequently, the isotherm velocity in case G1 is larger than in G2, since cooling rate, R_1 , is the same in both cases. Moreover, because the dendrite tip velocity is a monotonously increasing function with the undercooling [Gandin et al. 2003], the latter for CAFE-G1R1L1 is larger than for CAFE-G2R1L1. Height differences of the growth front are proportional to the variations of the undercooling by the temperature gradient. Therefore, this forms larger steps on the growth front for case G1 compared to G2.

The chimney extends deeper in the mushy zone when the temperature gradient in-

creases. This is confirmed by both the simulation results shown in [fig. 4.16](#) as well as the experimental observations. Another remarkable phenomenon is also observed in the low gradient case: a “pulsing” mechanism in CAFE-G1R1L1 where a series of solute rich liquid pockets are observed one above the other. This corresponds to a repeated and localized strong spatial variation of the liquid velocity field outside the mushy zone, regularly thrusting away small plumes. These pulses are roughly similar to each other in size and exit speed, creating thus a very regular pattern during some time.

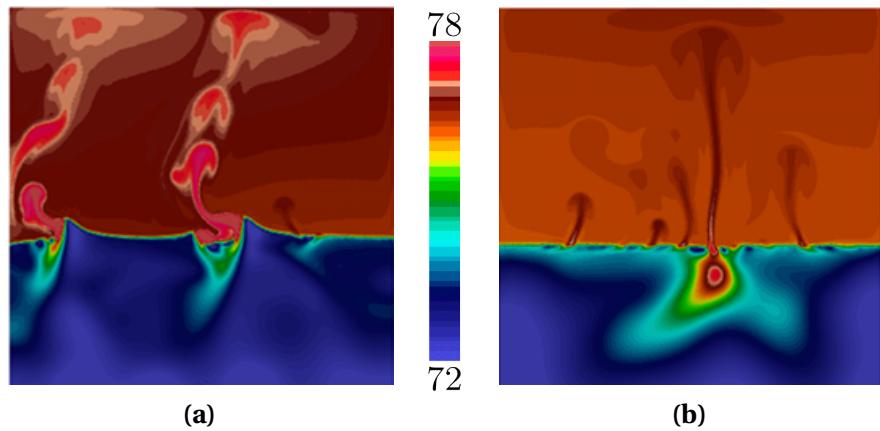


Fig. 4.16 – Average composition maps for CAFE-G1R1L1 at time 1060 s and CAFE-G2R1L1 at time 1845 s.



Fig. 4.17 – Snapshot of the pulsing mechanism coming from a groove shape in the mushy (check animation in the PDF file).

In the case of a high temperature gradient (case CAFE-G2R1L1) this phenomenon is barely seen. In fact, the pattern shown in [fig. 4.16](#) is more typical, with continuous plume rising from the mushy zone and reaching the top of the domain. However, such regular plume is the initial and final pattern seen for low gradient before the pulsing regime. Similar observations have been made in the experiments too. [Figure 4.18a](#)

displays the phenomenon of the “pulsing” plumes, which could be explained by the following mechanisms. The permeability of the mushy zone and the narrow gap of the solidification cell obstruct the feeding of the plumes by solute. A critical solute concentration has to be accumulated at a specific location in order to trigger the formation of a rising plume. An interim drop of the solute concentration below such a threshold would interrupt the plume. Flow instabilities can be another reason for the peculiar shape of the plumes. [Figure 4.18b](#) shows a pronounced continuous plume. The same plume can be seen a few seconds later in [fig. 4.18c](#). The plume structure becomes unstable; one can observe an indentation of streamlines followed by a mixing of rising solute-rich liquid with descending In-rich fluid. This mechanism also causes a non-continuous structure of the plumes.

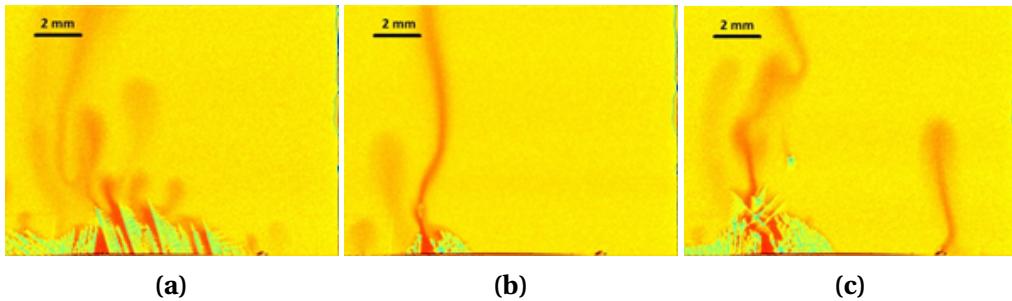


Fig. 4.18 – Snapshots of dendrite structure and composition field from two solidification experiments conducted at a cooling rate $R=-0.01 \text{ K s}^{-1}$ and a temperature gradient $\|G\|=1 \text{ K mm}^{-1}$: (a) “pulsing” plumes, (b) continuous plume, (c) upcoming plume instability.

4.6.4 Effect of cooling rate

The next parameter studied is the cooling rate, corresponding to case CAFE-G1R2L1. A snapshot of the composition map and the corresponding vertical component of the velocity field are given in [fig. 4.19](#). We see a similarity with case CAFE-G1R1L1 in [fig. 4.16a](#) with respect to the buckled interface between the liquid and the mushy zone as well a plume pulsing effect when a low temperature gradient is applied. On the other hand, segregation inside the mush is more irregular with more pronounced patterns reaching a larger depth.

One could distinguish alternating V and A shapes patterns in the mushy zone. As for case CAFE-G1R1L1, these patterns are created by a network of pulsing plumes formed by the steps created on the delocalized growth front due to the low temperature gradient. However, these considerations are not sufficient to explain the shape of the growth front. The reason for the protuberances created at the tips of the V shape is the presence of a descending bulk liquid with a low composition seen by the growth

front. It infers that favorable growth conditions are created for a higher working temperature since the dendrite tip undercooling decreases for facing liquid flow and a lower composition; the growth rate is given by the isotherm velocity. The growth front thus adjusts its position to catch up with the corresponding isotherm, the latter being located at the tips of the V shape, i.e. the outmost advanced position of the growth front. It also means that the V shape angle depends on the size and intensity of the convection loops above the front. When the steps are formed on the growth front, the plumes exiting the mushy zone follow a direction normal to the front. They are inclined towards each other above the V shape. As a result, they may join and form a larger plume as seen in CAFE-G1R1L1 ([fig. 4.16a](#)), thus forming larger and more stable chimneys. The other observation in [fig. 4.19](#) is the existence of stable regions of the growth front. For instance, this is seen in between the two V shape forming or on the right hand side of the cell.

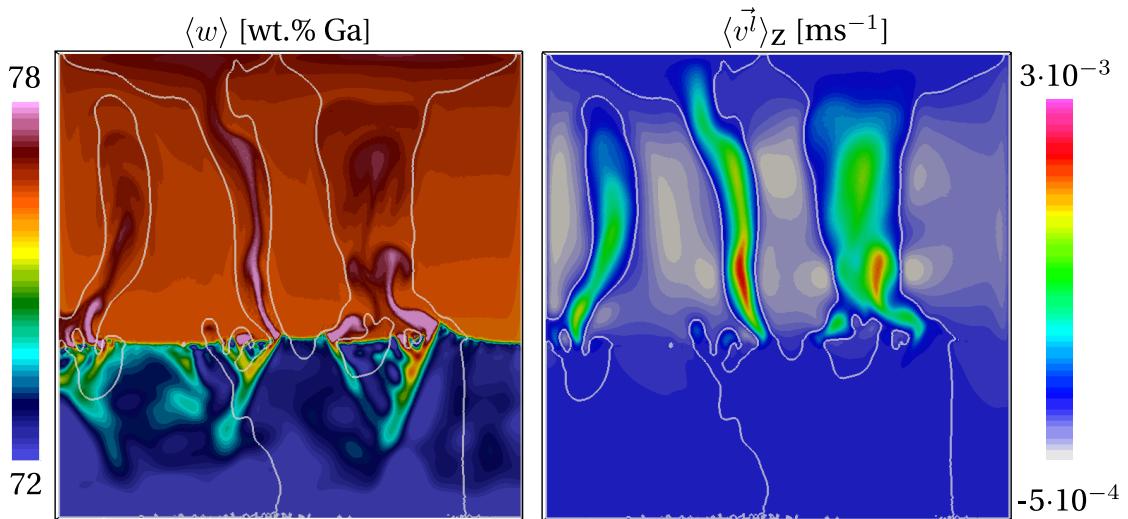


Fig. 4.19 – Average fields inside the cell for case CAFE-G1R2L1 at 350 s. The white contour identifies the zero velocity limit for the vertical component (z-axis) of the velocity field .

The reason for this stability is the inversion of the composition gradient located ahead. Animation shows that solute coming from the top of the cell is responsible for this accumulation, creating a layering that provides a stabilization effect above the mushy zone. This is verified by the vertical component of the average velocity also made available in [fig. 4.19](#). It is negative outside the path of the plumes. A resulting concurrent effect is the formation of the A shape segregates in between the V shape patterns seen in [fig. 4.19](#). Finally, it can be observed that these patterns are sustained longer compared to [fig. 4.16](#) CAFE-G1R1L1 because, at high cooling rate, the flow in the mushy zone is decreased due to a faster solidification. This is the same effect as described for

the large gradient configuration in CAFE-G2R1L1 ([fig. 4.16b](#)).

It is not clear how these observations could be compared with the A and V shapes segregates reported for steel ingots [Pickering 2013]. Despite the fact that macrosegregation is the main phenomenon leading to these patterns, there has not been a clear explanation yet in the literature for their formation. However, for steel casting, the A and V patterns are believed to form concomitantly. Further investigations would thus be required to quantify the consequences of thermosolutal instabilities simulated here for an In-75 wt.% Ga alloy and check their possible correlation with experimental observations in steel casting.

4.6.5 Effect of lateral temperature gradient

The previous simulations show the effect of cooling rate and temperature gradient on the survival of segregation patterns deep in the mushy zone. Another simulation is performed by increasing the cooling rate using higher heat flux extracted from the vertical side boundaries. This is achieved in case CAFE-G1R1L2 where the heat transfer coefficient reaches $500 \text{ W m}^{-2} \text{ K}^{-1}$. As a consequence of the large cooling from the sides, the temperature gradient is no longer vertical. A distinct flow due to thermal buoyancy is created, driving a cold liquid downwards near the sides of the cell. Under the influence of these two main convection loops, all segregation plumes tend to regroup in the middle of the domain, forming a larger central plume, as seen in the composition map at 450 s in [fig. 4.20](#). However, this regime occurs at times earlier than 500 s, where the effect of thermally induced buoyancy forces is prevailing, feeding the convection loops. Approximately 500 s later, the mushy zone has extended, favouring the segregation mechanical forces i.e. $\rho_{\text{ref}} \left(1 - \beta_{\langle w \rangle^l} \Delta \langle w \rangle^l \right) g$, rather than the thermal mechanical forces, $\rho_{\text{ref}} (1 - \beta_T \Delta T) g$.

[Figure 4.20](#) shows the corresponding composition maps with stable segregated channels at about 1000 s that also remain at 1500 s. The solidification front then tends to form a concave shape at the center of the cell, thus partially revealing the form of the isotherms toward the cell center. The stable pattern in the center is similar to the plateau seen at the center, between the A-shapes in [fig. 4.16](#) and [fig. 4.19](#). As stated before, it is an inactive region with respect to plume initiation due to the inversion of the solute composition gradient. In other words, the high gallium concentration at the top of cell causes indium, which is the heavier species, to accumulate and be partially trapped between the mushy walls, thus creating a stable flow configuration.

Outside of the plateau, two plumes are observed from the prominent instabilities of the growth front, adopting diverging directions. This is also observed at the center of the cell in [fig. 4.19](#) on each side of the A-shape segregate. These plumes in [fig. 4.20](#)

lead to the formation of two stable channels. The corresponding situation in the experiment is shown in [fig. 4.21](#). The chimneys on both sides and the plateau in between can be clearly recognised.

The additional cooling at the side walls produces two flow vortices between the side wall and the strong convective plumes above the chimneys. The central part of the sample remains almost unaffected by the additionally driven thermal convection. This area is characterised by the occurrence of a number of smaller convective plumes.

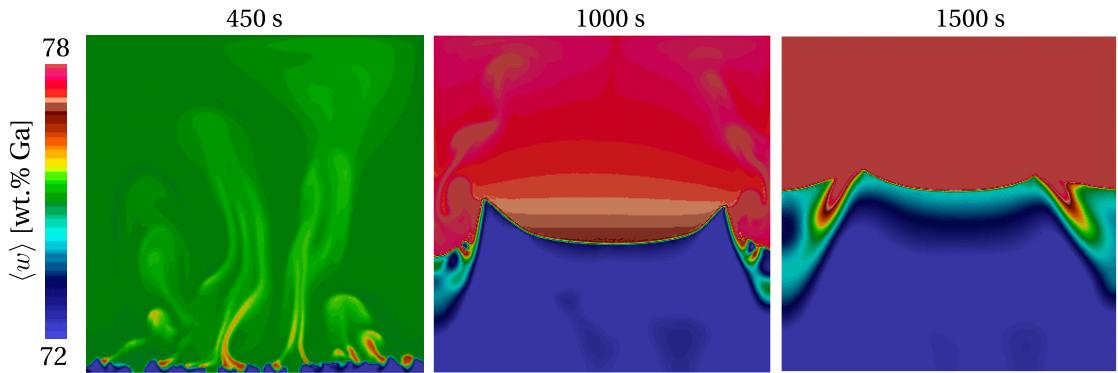


Fig. 4.20 – 2D cut plane of the average composition inside the cell for case CAFE-G1R1L2 at the following time increments: 450 s, 1000 s and 1500 s.

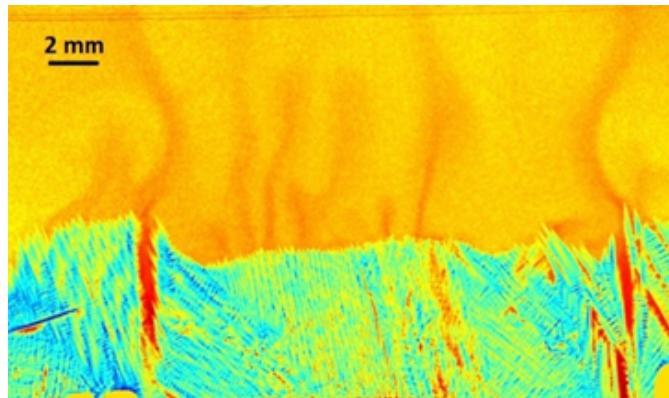


Fig. 4.21 – Snapshot of dendritic structure and composition field from a solidification experiment recorded at 1000 s for a cooling rate $R = -0.01 \text{ K s}^{-1}$ and a temperature gradient $\|G\| = -1 \text{ K mm}^{-1}$.

4.6.6 Mono-grain freckles

All CAFE simulations were performed considering a normal classic heterogeneous nucleation at the surface of the mould where the metal is cooled down. However, as the number of grains is not negligible, the subsequent fluid-structure interaction between

dendrites, simply represented by an isotropic permeability and the interdendritic flow cannot be easily interpreted. Therefore, one may consider simpler solidification cases like a mono-grain growth. This ideal situation is not always experimentally viable: either the mould surface is not perfectly smooth, and therefore nucleation can be triggered by a wetting mechanism, or the metal contains a certain level of impurity which can trigger nucleation heterogeneously in the liquid bulk. Regardless of this experimental limitation, this type of simulations allows simpler understanding as the grain growth is not disturbed by another neighbouring grain, it grows while exclusively interacting with the liquid phase.

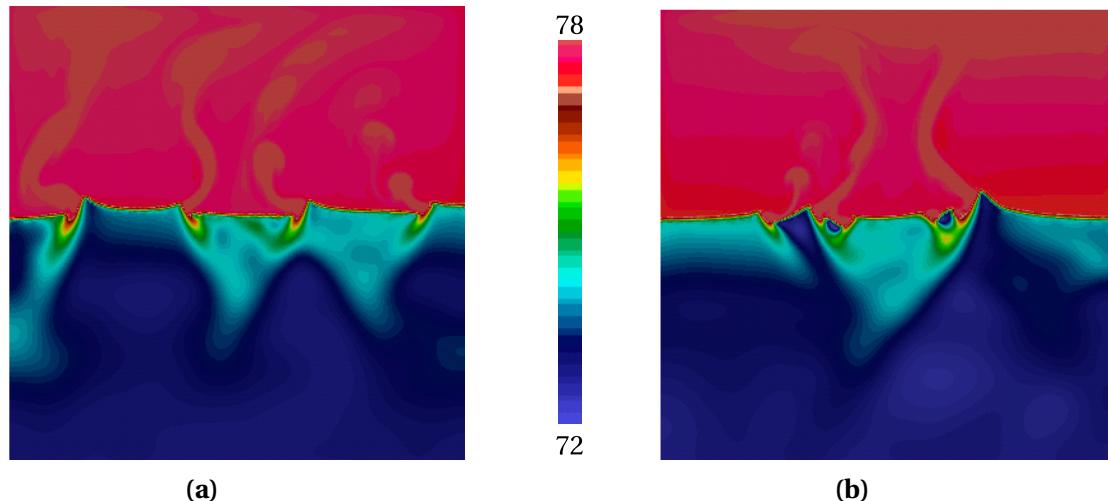


Fig. 4.22 – Snapshots of mono-grain solidification showing the average composition in gallium predicted by the CAFE approach. Two orientation scenarios are considered where a) the grain is upright with a Euler angle of $(90^\circ, 0^\circ, 0^\circ)$ or b) the grain is tilted with a Euler angle of $(90^\circ, 30^\circ, 0^\circ)$ (check animation in the PDF file).

Résumé chapitre 4

Ce 4^e chapitre est dédié à la macroségrégation induite par le mouvement de la phase liquide par convection thermosolutale, à solide fixe et en absence de retrait à la solidification ($\langle \rho \rangle^s = \langle \rho \rangle^l$). Pour cela, nous introduisons dans un premier temps les principaux schémas de résolution des équations Navier-Stokes selon la façon dont ils répondent aux critères de stabilité de Babuška-Brezzi : les éléments finis mixtes et la méthode multi-échelles variationnelle (Variational MultiScale).

En choisissant la seconde méthode, nous donnons les détails de la formulation éléments finis correspondante qui régit les écoulements dans la phase liquide loin du front de solidification, ainsi qu'au sein de la zone dendritique pâteuse. Le principal moteur de mouvement liquide est la convection thermosolutale. Celle-ci est générée par la densité du liquide qui varie à la fois avec la température et la composition intrinsèque de la phase liquide, contribuant ainsi à la redistribution des éléments d'alliage. On s'intéresse à ce type de méso-macroségrégation en montrant une application de solidification dirigée, traitée dans le chapitre 3 en diffusion pure. Nous montrons qu'en fin de solidification, les écoulements créent des canaux à forte ségrégation positive en peau et dans le cœur de la pièce.

L'investigation de ce défaut fait ensuite l'objet d'une confrontation qualitative entre la simulation et une expérience de solidification. Cette dernière consiste en un banc de solidification dirigée d'un alliage d'indium-gallium à bas point de fusion. Un suivi en caméra rapide permet de suivre la formation de la microstructure en fonction du temps. Par le biais de la simulation, on teste d'abord la performance du modèle purement macroscopique, i.e. avec suivi indirect des structures et phases via leur fraction volumique. Les résultats montrent que les canaux de ségrégation sont visibles mais sont moins nombreux et moins stables que l'on prédit expérimentalement.

Ensuite, on rajoute au modèle précédent une couche de modélisation à l'échelle mésoscopique pour suivre directement les enveloppes des grains. Cette fois, la comparaison avec l'expérience montre que nous prédisons mieux qualitativement l'interaction complexe entre structure de solidification, l'écoulement au sein de la zone pâteuse et la ségrégation conséquente. Une étude paramétrique permet après d'étudier la sensibilité de l'occurrence et la forme des canaux ségrégés par rapport aux différents paramètres de contrôle du procédé.

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Modélisation par Level Set des macroségrégations induites par le retrait à la solidification

RESUME : La macroségrégation est un défaut connu dans les procédés de coulées industrielles. La genèse de ce défaut est la conséquence de l'interaction complexe entre la microségrégation ou la distribution des espèces chimiques à l'échelle de la microstructure et les mouvements des phases liquide et solides. Les hétérogénéités de concentration en solutés à l'échelle de la pièce peuvent être rédhibitoires vis-à-vis de la qualité du produit. Dans ce travail, on propose un modèle numérique pour simuler et prédire la formation des macroségrégations en cœur des pièces d'alliages multi-constitués, induites par des variations thermiques et solutales dans la phase liquide. Dans un premier temps, on considère que le métal solidifie à volume constant. Dans ce contexte, la convection thermosolutale est étudiée ainsi que son influence sur la formation des canaux ségrégés à différentes échelles de modélisation. Dans un deuxième temps, le modèle vise à prédire les macroségrégations en présence de changement de volume du métal, dont la cause principale est le retrait à la solidification, pouvant être à l'origine du phénomène de ségrégation inverse. La surface entre le métal et le gaz environnant au cours du retrait évolue pendant le retrait en fonction du chemin de solidification qui varie avec la macroségrégation. Cette évolution d'interface est suivie par la méthode Level set. Des prédictions de concentration moyenne, couplées aux bases de données thermodynamiques pour mieux prédire les chemins de solidification des alliages multi-constitués, sont analysées et comparées avec des résultats expérimentaux. Finalement, des calculs de solidification en microgravité sont présentées, simulant un essai expérimental dans le contexte du projet CCEMLCC lancé par l'Agence Spatiale Européenne. Les résultats en fin de solidification montrent un accord acceptable quant à la forme et l'elongation des échantillons solidifiés. Ces calculs sont faits avec des approximations binaire, ternaire et quaternaire d'une même nuance d'acier utilisée dans les essais en microgravité.

Mots clés : modélisation, solidification, ségrégation, Level Set, éléments finis, métallurgie

Numerical modeling of macrosegregation formed during solidification with shrinkage using a Level Set Approach

ABSTRACT : Macrosegregation is key defect in industrial casting processes. During solidification, solute redistribution at the scale of microstructure, also known as microsegregation, take place with complex interactions, in order to form one or more solid phases. These interactions between microsegregation and movements of liquid and solid phases may lead to macrosegregations. These solute heterogeneities spanning on a larger scale, may result in a bad casting quality. In this thesis, we propose a numerical model to simulate and predict macrosegregations occurring in the centre of multicomponent alloys, caused by thermal and solutal variations in the liquid phase. First, we assume that the metallic alloy solidifies with a constant volume. In this context, we study the influence of thermosolutal convection on the formation of channel segregations, at different modelling scales. The second part of this modelling work consider solidification while the metallic alloy's volume is decreasing, mainly due to overall density variation, also known as solidification shrinkage, possibly leading to the so-called inverse segregation phenomenon, appearing on the alloy's skin. In the context of solidification shrinkage, the shape of the metal's boundary with surrounding gases varies according to a constantly changing solidification path due to macrosegregation. The Level Set method is therefore used to track its evolution with time. Composition predictions, coupled with thermodynamic database mappings for more accurate multicomponent solidification paths, are analysed and compared to existing experimental setups. Finally, simulations of a reduced-gravity solidification cases are performed, mocking an experimental benchmark from the CCEMLCC project launched by the European Space Agency. The results show acceptable agreement for the final shape, compared to experimental results. These computations were performed with binary, ternary and quaternary approximations of the same steel grade which was used in reduced-gravity experiments.

Keywords : modélisation, solidification, segregation, Level Set, finite elements, metallurgy

