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## NUMERICAL MODELLING OF MACROSEGREGATION INDUCED BY SOLIDIFICATION SHRINKAGE IN A LEVEL SET APPROACH

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<b>Acronym</b>	<b>Standing for</b>
ALE	Arbitrary Lagrangian-Eulerian
CCEMLCC	Chill Cooling for the Electro-Magnetic Levitator in relation with Continuous Casting of steel
CEMEF	Center for Material Forming
DLR	Deutsches Zentrum für Luft- und Raumfahrt
EML	Electromagnetic levitation
ESA	European Space Agency
FEM	Finite Element Method
ISS	International Space Station
IWT	Institut für Werkstofftechnik
LHS	Left Hand Side
MAC	Marker-and-cell
PF	Phase field
RHS	Right Hand Side
RUB	Ruhr Universität Bochum
RVE	Representative Elementary Volume
VOF	Volume Of Fluid

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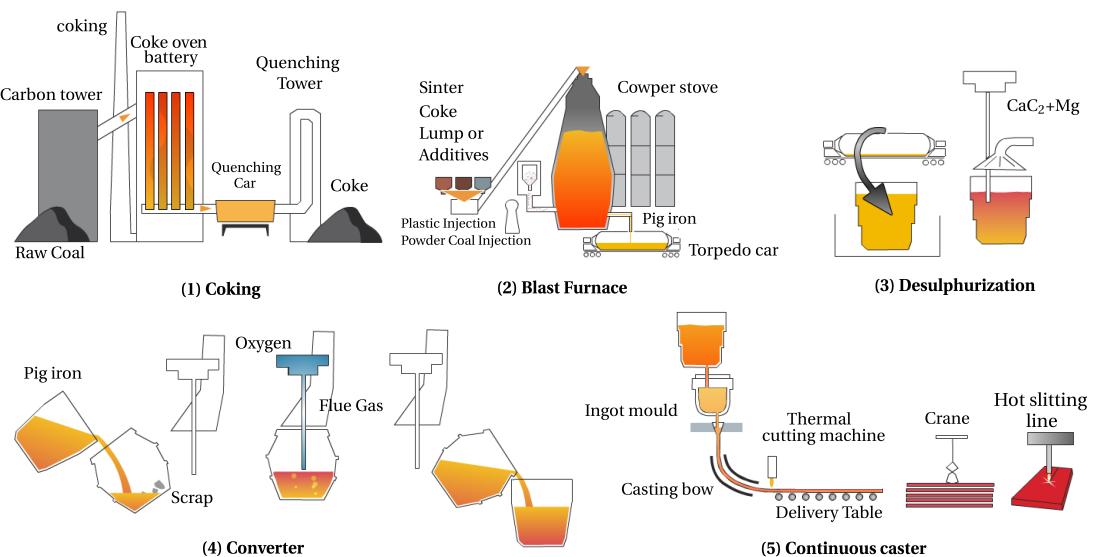
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# Chapter 1

## General Introduction

**Macrosegregation** is a very known defect to metallurgical processes. Despite a great evolution achieved by active research during the last 60 years, it remains partially understood. Macrosegregation is often the consequence of several factors at the scale of a casting, all related to *microsegregation* happening at the scale of dendrites. Today, research in metallurgy focuses on a deeper understanding of such a connection between the different physical scales. Solidification is not only a phase change, but also a complex transformation involving small scales like nucleation, medium scales like grains growth and large scales like convection in the melt. From the nucleation theory to the mechanical behavior of metals, intricate phenomena combine to form defects in the final product. This has been seen in casting processes, such as continuous casting (fig. 1.1) and ingot casting. Surface and volume porosity, hot tearing and composition heterogeneity are known defects to the casting community. After a brief introduction of these defects, macrosegregation will be the focus of this dissertation.



**Fig. 1.1 – Main steps in a continuous casting plant**

## 1.1 Casting defects

Undesired effects are inevitable in any industrial process. More importantly, a lot of defects in the casting industry can be disastrous in some situations where the cast product is not serviceable and hence rejected. This leads to a systematic product recycling, i.e. the product is ditched to be reheated, remelted and then cast again. From an economic point view, the operation is expensive timewise and profitwise. Understanding and preventing defects when possible, is thus crucial in the casting industry. We focus hereafter on the main encountered defects.

### Hot tearing

This defect, also denoted solidification cracking or hot cracking, occurs in the mushy zone at high solid fractions when a failure or crack appears at specific locations, the hot spots. The temperature range in which the steel is vulnerable to hot tearing is known as the brittleness temperature range (BTR). It corresponds to solid fractions greater than 90%, with the liquid phase forming a discontinuous film. Many factors can cause the failure, but the main origin is a lack of liquid feeding required to compensate for the solidification shrinkage, in the presence of thermal stresses in the mushy region. Therefore, a crack initiates then propagates in the casting, as shown in [fig. 1.2](#).

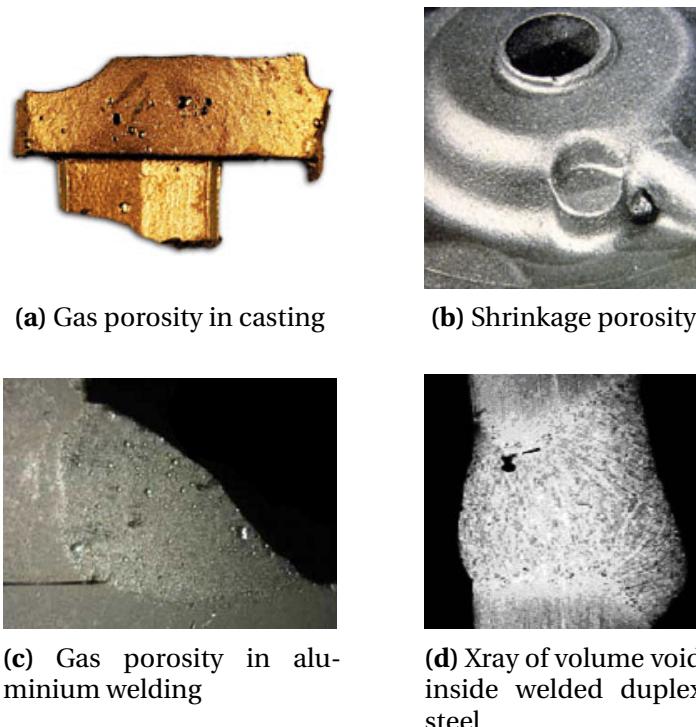


**Fig. 1.2 – Crack in an aluminium slab**

### Porosity

Porosity is a void defect formed inside the casting or at the outer surface. It may attributed to two different factors. Firstly, we speak of *shrinkage porosity*, when a void forms as a result of density differences between the interdendritic liquid and solid network, the latter being denser than the former ([figs. 1.3c](#) and [1.3d](#)). It is basically, the same reason that initiates hot cracks. The second factor is the presence of dissolved gaseous phases in the melt ([figs. 1.3a](#) and [1.3b](#)). According to [Dantzig et al. \[2009\]](#), these gases may be initially in the melt, or created by the reaction between the metal and water found in the air or at trapped in grooves at the moulds surface. If the

decreasing temperature and pressure drop in the liquid are large enough, the latter becomes supersaturated. Consequently, the nucleation of gaseous phase is triggered (just like when you open a cold bottle of coca-cola is opened!).



**Fig. 1.3 – Examples of porosity in casting and welding**

### Freckles or segregated channels

The origin of this defect is a combined effect of microsegregation and buoyancy forces. Upon solidification, solid forms while rejecting some solute in the liquid due to partitioning (steels have a partition coefficient less than unity). When segregated solute is the lighter species, an increasing concentration in the liquid phase produces a solutal driving force inside the mushy zone, generating unstable convection currents, with "plume" shapes as often reported in the literature [Sarazin et al. 1992; Schneider et al. 1997; Shevchenko et al. 2013]. When temperature gradient is an additional force of convection, the latter is hence qualified as "thermosolutal".

## 1.2 Macrosegregation

Macrosegregation generally stems from a solubility difference between a liquid phase and one or more solid phases, along with a relative velocity between these phases. While the former is responsible for local solute enrichment or depletion, the latter will propagate the composition heterogeneity on a scale much larger than just a few

## Chapter 1. General Introduction

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dendrites. This is why macrosegregation could be observed on the scale of a casting, up to several meters in length. While microsegregation can be healed by annealing the alloy to speed up the diffusion process and allow homogenization, heterogeneities spanning on larger distances cannot be treated after solidification. It is obvious that macrosegregation is irreversible defect. Failure to prevent it may lead to a substantial decline in the alloy's mechanical behavior, hence its serviceability.

### 1.2.1 Causes

Four main factors can (simultaneously) cause fluid flow leading to macrosegregation:

#### Liquid dynamics

During solidification, thermal and solutal gradients result in density gradients in the liquid phase:

$$\rho^l = \rho_{\text{ref}}(1 - \beta_T(T - T_{\text{ref}}) - \sum_i \beta_{w_i^l}(w_i^l - \langle w_i \rangle_{\text{ref}}^l)) \quad (1.1a)$$

$$\vec{\nabla} \rho^l = -\rho_{\text{ref}}(\beta_T \vec{\nabla} T + \sum_i \beta_{w_i^l} \vec{\nabla} w_i^l) \quad (1.1b)$$

In eq. (1.1a), density is assumed to vary linearly with temperature and phase composition for each chemical species (index  $i$ ). The slopes defining such variations are respectively the thermal expansion coefficient  $\beta_T$  and solutal expansion coefficient  $\beta_{w_i^l}$ , given by [Kohler 2008]:

$$\beta_T = -\frac{1}{\rho_{\text{ref}}} \left( \frac{\partial \rho^l}{\partial T} \right) \quad (1.2a)$$

$$\beta_{w_i^l} = -\frac{1}{\rho_{\text{ref}}} \left( \frac{\partial \rho^l}{\partial w_i^l} \right) \quad (1.2b)$$

The linear fit assumes also that the density takes a reference value,  $\rho_{\text{ref}}$ , when temperature and liquid composition reach reference values, respectively  $T_{\text{ref}}$  and  $\langle w_i \rangle_{\text{ref}}^l$ . However, in some situations, a suitable thermodynamic database providing accurate density values is far better than a linear fit, especially in the current context of macrosegregation. Such possibility will be discussed later in the manuscript (cf. SECTION TODO). In the presence of gravity, the density gradient in eq. (1.1b), causes thermosolutal convection in the liquid bulk and a subsequent macrosegregation.

#### Solidification shrinkage

Solid alloys have a greater density than the liquid phase ( $\rho^s > \rho^l$ ), thus occupy less volume. Upon solidification, the liquid moves towards the solidification front to compensate for the volume difference caused by the phase change, as well as the thermal

contraction. When macrosegregation is triggered by solidification shrinkage, we speak of *inverse segregation*. Theoretically, if solute mass is conserved, a decreasing volume results in a positive segregation. Shrinkage deforms the outer surface of a solidifying alloy, causing positive macrosegregation. While one would naturally expect negative macrosegregation in areas where solidification begins and positive inside the alloy, shrinkage promotes the opposite phenomenon, hence the term *inverse segregation*. In contrast to liquid convection, shrinkage flow may cause macrosegregation even without gravity.

### Movement of equiaxed grains

Equiaxed grains can grow in the liquid bulk where thermal gradients are weak, or in the presence of inoculants. Consequently, they are transported by the flow (floating or sedimenting, depending on their density [Beckermann 2002]) which leads to negative macrosegregation in their final position.

### Solid deformation

Stresses of thermal and mechanical nature are always found in casting processes (e.g. bulging between rolls in continuous casting). Deformation of the semi-solid in the mushy zone causes a relative solid-liquid flow in the inward (tensile stresses) or outward (compressive stresses) direction, causing macrosegregation.

#### 1.2.2 Types

##### In continuous casting

The semi-solid billet is carried through a series of rolls that exert a radial force to straighten it and get it to its horizontal position. As the mushy part of a slab enters through these rolls, interdendritic liquid is expelled backwards, i.e. regions with lower solid fraction. Since the boundaries solidify earlier than the centre, the enriched liquid accumulates halfway in thickness, forming a centreline macrosegregation as shown in [fig. 1.4](#). Other types of segregates (channels, A-segregates ...) can also be found but remain more specific to ingot casting.

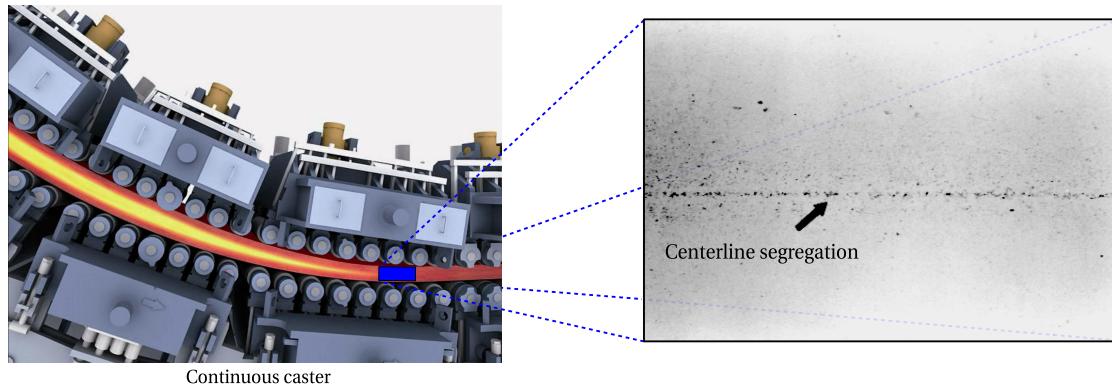
##### In ingot casting

A variety of segregation patterns can be encountered in heavy ingots:

- the lower part is characterized by a negative segregation cone promoted by the sedimentation of equiaxed crystals,
- positive segregation channels, known as A-segregates, form along the columnar dendritic zones, close to the vertical contact with the mould,
- positive V-segregates can be identified in the centre of the ingot,

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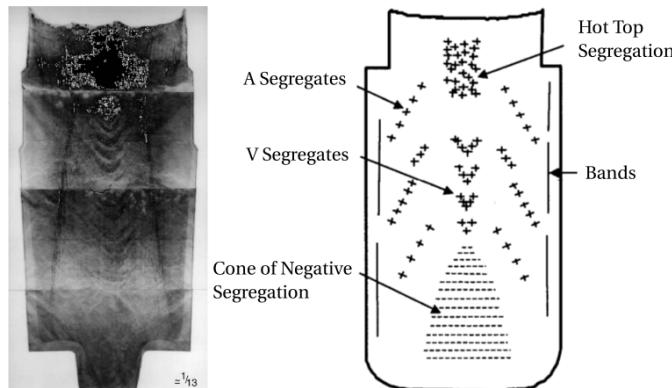
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**Fig. 1.4 – Centreline segregation in a steel slab [Beckermann 2002]**

- a positive macrosegregation in the upper zone where the last liquid solidifies, the so-called "hot-top", caused by solidification shrinkage (inverse segregation) and thermosolutal buoyancy forces.

Combeau et al. [2009] state that A-segregates and V-segregates formation is mainly attributed to local flow phenomena. As such, their scale is finer than macrosegregation, hence called "mesosegregates".



**Fig. 1.5 – Sulphur print (left) of a 65-ton steel ingot [Lesoult 2005] showing various patterns (right) of macrosegregation [Flemings 1974]**

### In investment casting

This process is widely used to cast single-crystal (SC) alloys for turbines and other applications that require excellent mechanical behavior [Giamei et al. 1970]. During directional solidification, thermosolutal forces thrust segregated species outside of the mushy zone into the liquid bulk. The segregation scale ranges from a few dendrites to a few hundreds of them, hence forming "long and narrow trails" (fig. 1.6a) as described by Felicelli et al. [1991]. Freckles are frequently formed by small equiaxed grains (fig. 1.6b), probably caused by a uniform temperature gradient that settles as

the channels become richer in solute. They can be observed on the ingot's surface, as well as in the volume.



(a) Small cylinder ingot showing freckles on the outer surface

(b) Zoomed image showing the microstructure in one freckle

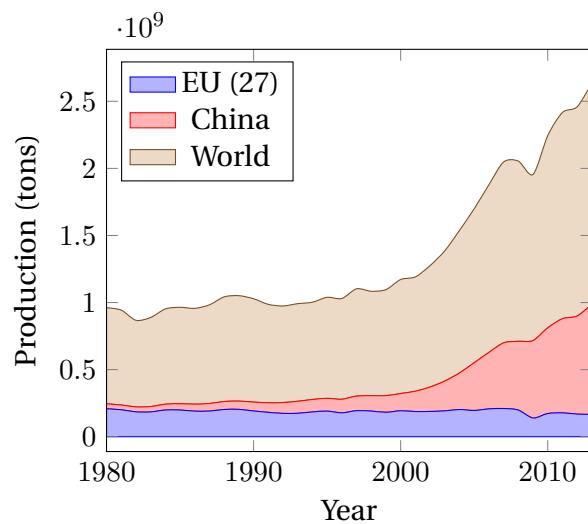
**Fig. 1.6 –** Freckles in directional casting of nickel-base superalloys

## 1.3 Industrial Worries

**Steel production** has continuously increased over the years to meet the industrial needs. [Figure 1.7](#) shows this increase between 1980 and 2013 with a clear dominance of the Chinese production. Quality constraints have also increased where specific grades of steel are needed in critical applications such as mega-structures in construction and heavy machinery. Therefore, alloys with defects are considered vulnerable and should be avoided as much as possible during the casting process. As such, steelmakers have been investing in research, with the aim of understanding better the phenomena leading to casting problems, and improve the processes when possible.

**Simulation software** dedicated to alloy casting is one of the main research investments undertaken by steelmakers. These tools coming from academic research are actively used to optimize the process. However, few are the tools that take into account the casting environment. For instance, the continuous casting process, in [fig. 1.1](#), is a chain process where the last steps involve rolls, water sprays and other components. A dedicated software is one that can provide the geometric requirements with suitable meshing capabilities, as well as respond to metallurgical and mechanical requirements, mainly:

- handling moulds and their interaction with the alloy (thermal resistances ...)
- handling alloy filling and predicting velocity in the liquid and mushy zone
- handling thermomechanical stresses in the solid



**Fig. 1.7 – Evolution curves of crude steel worldwide production from 1980 to 2013**

- handling multicomponent alloys and predicting macrosegregation
- handling finite solute diffusion in solid phases
- handling real alloy properties (not just constant thermophysical/thermomechanical properties)

## 1.4 Project context and objectives

### 1.4.1 Context

The European Space Agency (ESA) has been actively committed, since its foundation in 1975, in the research field. Its covers not only exclusive space applications, but also fundamental science like solidification. This thesis takes part of the ESA project entitled *CCEMLCC*, abbreviating "Chill Cooling for the Electro-Magnetic Levitator in relation with Continuous Casting of steel". The three-year contract from 2011 to late 2014 denoted *CCEMLCC II*, was preceded by an initial project phase, *CCEMLCC I*, from 2007 to 2009. The main focus is studying containerless solidification of steel under microgravity conditions. A chill plate is later used to extract heat from the alloy, simulating the contact effect with a mould in continuous casting or ingot casting. A partnership of 7 industrial and academic entities was formed in *CCEMLCC II*. A brief summary of each partner's commitment:

#### Academic partners

- CEMEF (France): numerical modelling of microgravity chill cooling experiments
- DLR (German Aerospace Centre) and RUB university (Germany): preparation of a chill cooling device for electromagnetic levitation (EML), microgravity testing

and investigation of growth kinetics in chill-cooled and undercooled steel alloys

- University of Alberta (Canada): impulse atomization of the D2 tool steel
- University of Bremen - IWT institute (Germany): study of melt solidification in atomization processing

### Industrial partners

- ARCELORMITTAL (France): elaboration of a series of steel grades used in microgravity and ground studies
- METSO Minerals Inc. (Finland): material production with D2 tool steel for spray forming
- TRANSVALOR (France): development and marketing of casting simulation software *Thercast<sup>®</sup>*

CEMEF as an academic partner, contributed to the work by proposing numerical models in view of predicting the chill cooling of steel droplets. A first model was developed by [Rivaux \[2011\]](#) whereas the present discusses a new model. The experimental work considered various facilities and environments to set a droplet of molten alloy in levitation: EML ([fig. 1.8](#)) for ground-based experiments, microgravity in parabolic flight or sounding rockets and last, microgravity condition on-board the International Space Station (ISS)



**Fig. 1.8 – Electromagnetic levitation**

### 1.4.2 Objectives and outline

The main focus of the present thesis is predicting macrosegregation with liquid dynamics assuming a fixed solid phase, i.e. no account of solid transport (e.g. equiaxed crystals sedimentation) and no account of solid deformation. In CEMEF, this scope has been adopted for previous work by [Gouttebroze \[2005\]](#), [Liu \[2005\]](#), [Mosbah \[2008\]](#), [Rivaux \[2011\]](#), and [Carozzani \[2012\]](#). Nevertheless, many modelling features evolved with time such as going from two-dimensional to three-dimensional modelling, resolution schemes for each of the conservation equations: energy, chemical species and liquid momentum, Eulerian or Lagrangian descriptions, modelling of grain structure and others. In this thesis, we propose a numerical model that takes into account i) the energy conservation in a temperature formulation based on a thermodynamic

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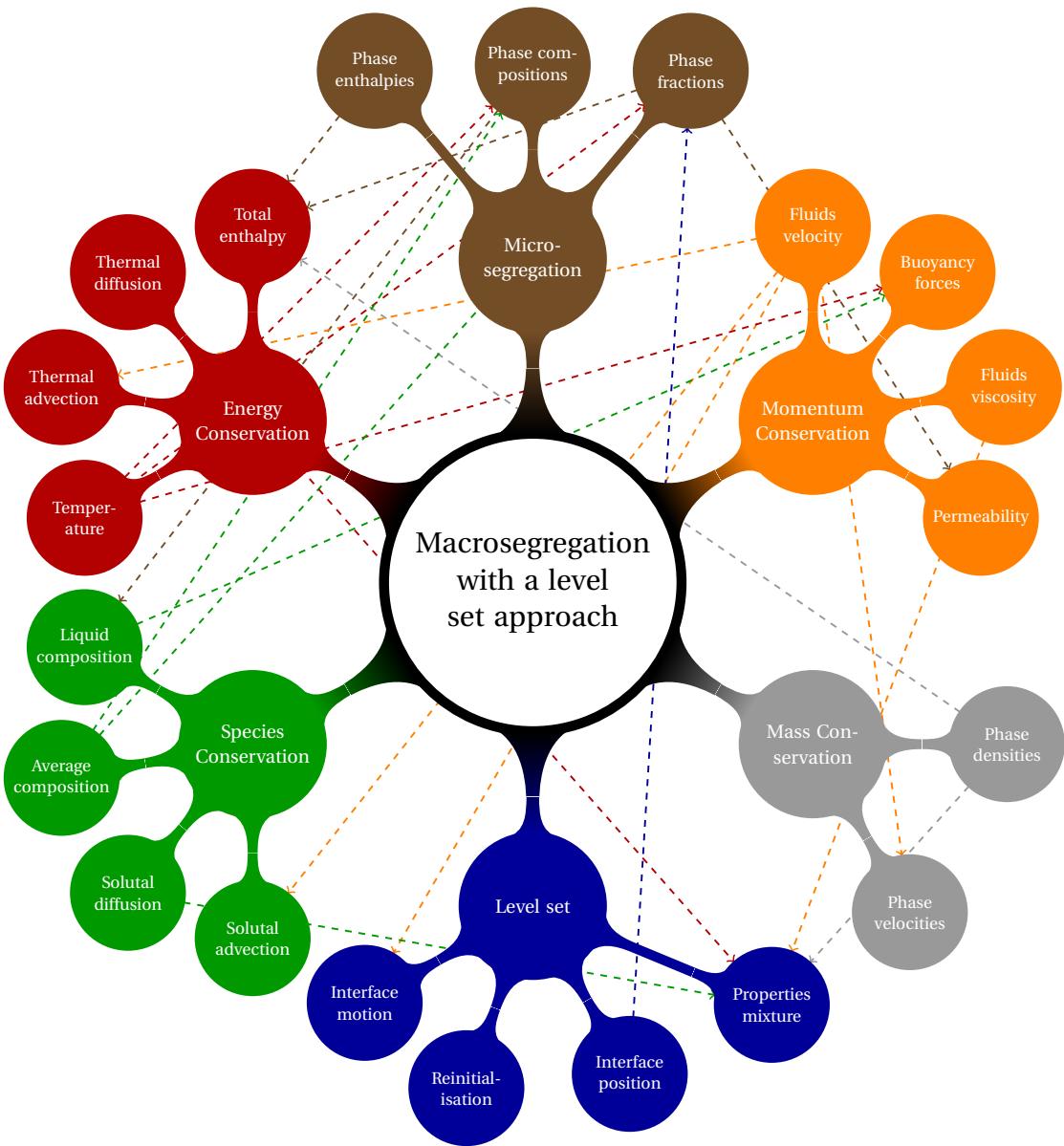
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database mapping, ii) the liquid momentum conservation with thermosolutal convection and solidification shrinkage as driving forces, iii) solute mass conservation and iv) solidification paths at full equilibrium for multicomponent alloys microsegregation. Moreover, all equations are formulated in a pure Eulerian description while using the Level Set method to keep implicitly track of the interface between the alloy and surrounding gas. To the author's knowledge, this work combining macrosegregation prediction using the level set methodology to track the metal-air interface during shrinkage has no precedent in casting and solidification literature.

Numerical tools: Cimlib relying on PETSc, parallelized with MPICH2, paraview and python as tools for postprocess and analysis.

The previously mentionned simulation requirements are not met in a single casting software package. Nevertheless, *Thercast*<sup>®</sup> is a promising tool that already handles a part of the above points. The current thesis developments are done using C++ language as a part of the in-house code, known as *CimLib* [Digonnet et al. 2007; Mesri et al. 2009]. This fully parallel library is the main academic research support for *Thercast*<sup>®</sup>

Outline: each chapter content ...



**Fig. 1.9** – A graphical representation of the main ingredients of the macrosegregation model with the level set methodology. The dashed lines represent the possible interaction between of the components.

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# Chapter 2

## Modelling Review

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## 2.1 Modelling macrosegregation

### 2.1.1 Dendritic growth

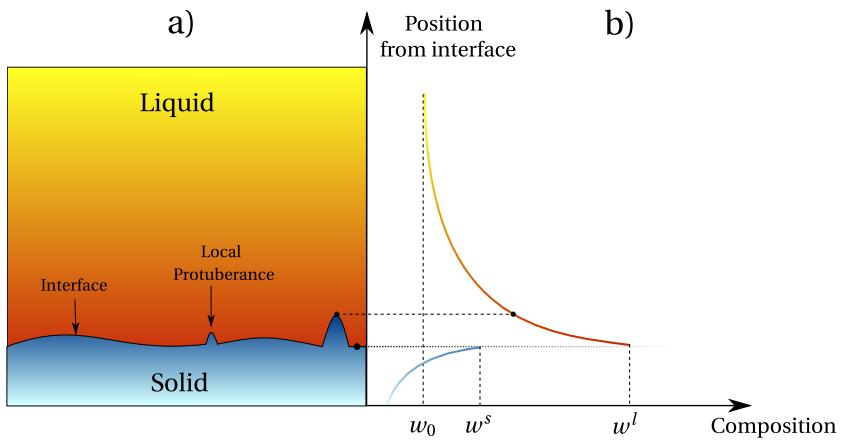
In a casting process, the chill surface i.e. the contact between the molten alloy and relatively cold moulds, is the first area to solidify. Thermal gradient,  $G$ , and cooling rate  $R$  are two crucial process parameters that define the interface speed  $\vec{v}^*$ , which in turn, affects the initial microstructure. Although it may be not easy to control them, it remains important to understand their implication in solidification.

The solid-liquid interface fluctuates when solidifying, thus perturbations may appear on the front, locally destabilizing it. Two outcome scenarios are possible. The first scenario is characterized by low values of  $\vec{v}^*$  where the interface maintains a planar shape, hence we speak of *planar growth*. With this kind of growth, a random protuberance appearing at the interface, has a low tip velocity (low driving force of solidification). As such, the rest of the interface catches up, keeping the planar geometry. In another scenario, where a real casting is considered, the interface speed is greater in general, due to high solidification rate. The protuberance tip will be pulled into a liquid less rich in solute than the interface. The zone ahead of the solid-liquid interface is constitutionally undercooled [Tiller et al. 1953], giving a greater driving force for the protuberance to grow in the direction of the thermal gradient. As it has a tree-like shape, we speak of *dendritic growth*. Near the chill surface, dendrites are columnar, with a favourable growth in the  $<100>$  direction for alloys with cubic lattices, but different orientations are also reported in the literature [see Dantzig et al. 2009, p. 289]. If temperature is uniform, which is the case usually far from mould walls, a similar dendritic growth phenomenon occurs, but with an equiaxed morphology.

Columnar dendrites are characterized by a primary spacing,  $\lambda_1$ , between the main trunks, and a secondary spacing,  $\lambda_2$ , for the arms that are perpendicular to the trunks. It should be noted that  $\lambda_2$ , together with the grain size, are two important microstructural parameters in the as-cast microstructure [Easton et al. 2011]. Further branching may occur but will not be discussed here.

### 2.1.2 Mush permeability

The dendritic geometry is crucial in solidification theory as it exhibits lower solid fraction compared to a microstructure formed by planar growth. This fact has consequences on the fluid-structure interaction in the mushy zone, namely the liquid flow through dendrites. At the chill surface, the solid grows gradually from dispersed growing nuclei to a permeable solid skeleton, until finally grains have fully grown at the end of phase change. The intermediate state where liquid can flow in and out of the mushy zone through the dendrites is a key phenomenon from a rheological perspective. The flow through the solid skeleton is damped by primary and secondary dendrites, resulting in momentum dissipation just like in saturated porous media. The famous Darcy [1856] law relates the pressure gradient ( $\vec{\nabla}p$ ) to the fluid velocity  $\vec{v}$ ,



**Fig. 2.1** – Schematic of a) a protuberance growing on the solid-liquid interface with b) the corresponding composition profiles (Reproduced and adapted from DoITPoMS [2000], © DoITPoMS, University of Cambridge).

through the following equation [Rappaz et al. 2003]:

$$\vec{v} = \frac{\mathbb{K}}{\mu^l} \vec{\nabla} p \quad (2.1)$$

where  $\mu^l$  is the liquid dynamic viscosity and  $\mathbb{K}$  is the permeability tensor. The latter parameter has been the subject of numerous studies that aimed to predict it from various microstructural or morphological parameters. Some of these studies have started even before the first attempts to model macrosegregation by Flemings et al. [1967], Flemings et al. [1968a], and Flemings et al. [1968b]. Basically, all models include the solid fraction,  $g^s$ , as input to predict mush permeability along with empirical data. An instance of such models is the work of Xu et al. [1991]. Some models rely additionally on the primary dendrite arm spacing  $\lambda_1$  like Blake-Kozeny [Ramirez et al. 2003], or the secondary dendrite arm spacing  $\lambda_2$  like Carman-Kozeny, as a meaningful parameter to determine an isotropic permeability. Other models like Poirier [1987] and Felicelli et al. [1991] derive an anisotropic permeability based on both  $\lambda_1$  and  $\lambda_2$ .

The present work uses Carman-Kozeny as a constitutive model for the isotropic permeability scalar (zero order tensor):

$$\mathbb{K} = \frac{\lambda_2^2 g^l{}^3}{180 (1 - g^l)^2} \quad (2.2)$$

### 2.1.3 Microsegregation

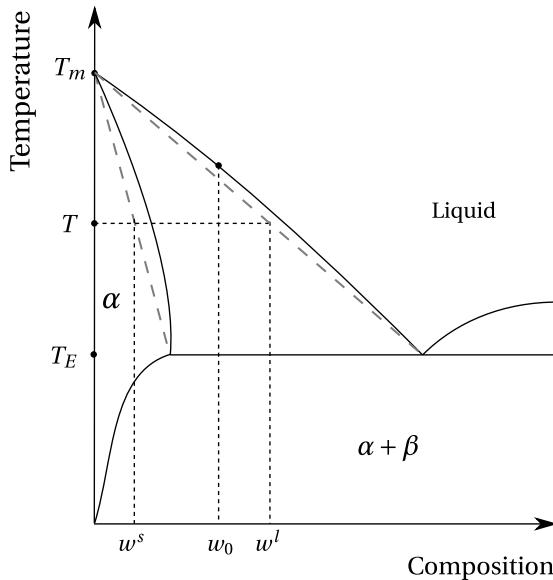
Microsegregation is a fundamental phenomenon in solidification. The simplest definition is an uneven distribution of solute between the liquid and the herein growing solid, at the microscopic scale of the interface separating these phases. If we consider a binary alloy, then the solubility limit is the key factor that dictates the composition

at which a primary solid phase exists in equilibrium. The segregation (or partition) coefficient  $k$  determines the extent of solute rejection into the liquid during solidification:

$$k = \frac{w^{s^*}}{w^{l^*}} \quad (2.3)$$

where  $w^{s^*}$  and  $w^{l^*}$  are the compositions of the solid and liquid respectively, at the interface. When the segregation coefficient is less than unity (such is the case for most alloys during dendritic solidification), the first solid forms with a composition  $w^{s^*} = kw^{l^*} = kw_0$  less than the liquid's composition  $w_0$ , the latter being initially at the nominal composition,  $w_0$ . Figure 2.2 illustrates a typical binary phase diagram where the real solidus and liquidus are represented by solid lines, while the corresponding linear approximations are in grey dashed lines. For most binary alloys, this linearisation simplifies derivation of microsegregation models, as  $k$  becomes independent of temperature.

For each phase, the relationship between the composition at the interface and that in the bulk depends on the chemical homogenisation ability of the phase. The more homogeneous a phase, the closer the concentrations between the interface and the bulk, hence closer to equilibrium. It is thus essential to study the effect of homogenisation on the segregation behaviour and the subsequent effect on solidification, which leads the formalism of microsegregation models.



**Fig. 2.2** – Typical eutectic phase diagram of a binary alloy showing the real solidus and liquidus at full equilibrium, with the corresponding linear approximations (grey dashed lines).  $T_m$  and  $T_E$  are respectively the melting point of the solvent and the eutectic temperature.

### **Microsegregation models**

Solid formation depends greatly on the ability of chemicals species to diffuse within each of the solid and liquid phases, but also across the solid-liquid interface. Furthermore, chemical diffusion like all other diffusional process, is a time-dependent phenomenon. One can thus conclude that two factors influence the amount of solid formation: cooling rate and diffusion coefficients. However, convection and other mechanical mixing sources, homogenise the composition much faster than atomic diffusion. As such, *complete mixing* in the liquid is always an acceptable assumption, regardless of the solidification time. We may speak of infinite diffusion in the liquid. Nevertheless, diffusion in the solid, also known as *back diffusion*, is the only transport mechanism with very low diffusion coefficients. Therefore, chemical species require a long time, i.e. low cooling rate, to completely diffuse within the solid. The difference in diffusional behaviour at the scale of a secondary dendrite arms, is summarized by two limiting segregation models of perfect equilibrium and nonequilibrium, which are the lever rule and Gulliver-Scheil models, respectively. Afterwards, models with finite back diffusion are presented.

#### **Lever rule**

The lever rule considers an ideal equilibrium in all phases, i.e. solidification is extremely slow, hence phase compositions are homogeneous ( $w^{l^*} = w^l$  and  $w^{s^*} = w^s$ ) at all times as a consequence of complete mixing. These compositions are given by:

$$w^l = w^{l^*} = kw^{s^*} = kw^s \quad (2.4)$$

$$w^s = w^{s^*} = \frac{w_0}{k(1 - f^s) + f^s} \quad (2.5)$$

At the end of solidification, the composition of the solid phase is equal to the nominal composition,  $w^s = w_0$

#### **Gulliver-Scheil**

The other limiting case is the absence of diffusion in the solid. That includes also the diffusion at the interface, so nothing diffuses in or out. The consequence is a steady increase of the homogeneous liquid composition while the solid composition remains non-uniform. Compared to a full equilibrium approach, higher fractions of liquid will remain until eutectic composition is reached, triggering a eutectic solidification. The phase compositions are given by:

$$w^l = w^{l^*} = kw^{s^*} \quad (2.6)$$

$$w^s = kw_0(1 - f^s)^{1-k} \quad (2.7)$$

### Finite back diffusion

It has been concluded that the assumption of a negligible back diffusion overestimates the liquid composition and the resulting eutectic fraction. Therefore, many models studied the limited diffusion in the solid. One of the earliest models is the Brody-Flemings models [Khan et al. 2014] that is based on a differential solute balance equation for a parabolic growth rate, as follows:

$$w^l = w^{l*} = kw^{s*} \quad (2.8)$$

$$w^s = kw_0 [1 - (1 - 2\text{Fo}^s k) f^s]^{\frac{k-1}{1-2\text{Fo}^s k}} \quad (2.9)$$

where  $\text{Fo}^s$  is the dimensionless *Fourier number* for diffusion in the solid [Dantzig et al. 2009]. It depends on the solid diffusion coefficient  $D^s$ , solidification time  $t_s$  and the secondary dendrite arm spacing, as follows:

$$\text{Fo}^s = \frac{D^s t_s}{(\lambda_2/2)^2} \quad (2.10)$$

Several other models were since suggested and used. The interested reader is referred to the following non exhaustive list of publications: Clyne et al. [1981], Kobayashi [1988], Ni et al. [1991], Wang et al. [1993], Combeau et al. [1996], Martorano et al. [2003], and Tourret et al. [2009]. It is noted that some of these publications consider also a finite diffusion in the liquid phase.

#### 2.1.4 Macroscopic solidification model: monodomain

In this section, we will present the macroscopic conservations equations that enable us to predict macrosegregation in the metal when the latter is the only domain in the system.

### Volume averaging

It is crucial for a solidification model to represent phenomena on the microscale, then scale up to predict macroscopic phenomena. Nevertheless, the characteristic length of a small scale in solidification may represent a dendrite arm spacing, for instance the mushy zone permeability, as it may also represent an atomic distance if one is interested, for instance, in the growth competition between diffusion and surface energy of the solid-liquid interface. Modelling infinitely small-scale phenomena could be prohibitively expensive in computation time, if we target industrial scales.

The volume averaging is a technique that allows bypassing this barrier by averaging small-scale variations on a so-called *representative volume element* (RVE) [Dantzig et al. 2009] of volume  $V_E$ , with the following dimensional constraints: the element should be large enough to "see" and average microscopic fluctuations whilst being smaller than the scale of macroscopic variations. Solid and liquid may exist simultaneously in

the RVE, but no gas phase is considered (volume saturation:  $V^s + V^l = V_E$ ). Moreover, temperature is assumed uniform and equal for all the phases. The formalism, introduced by Ni et al. [1991], is summarized by the following equations for any physical quantity  $\psi$ :

$$\langle \psi \rangle = \frac{1}{V_E} \int_{V_E} \psi \, d\Omega = \langle \psi^s \rangle + \langle \psi^l \rangle \quad (2.11)$$

where  $\langle \psi \rangle^s$  and  $\langle \psi \rangle^l$  are phase averages of  $\psi$ . Then, for any phase  $\phi$ , one can introduce the *phase intrinsic average* of  $\psi$ , denoted  $\langle \psi \rangle^\phi$ , by writing:

$$\langle \psi^\phi \rangle = \frac{1}{V_E} \int_{V^\phi} \psi \, d\Omega = g^\phi \langle \psi \rangle^\phi \quad (2.12)$$

where  $g^\phi$  is the volume fraction of the phase. To finalize, the averaging is applied to temporal and spatial derivation operators [Rivaux 2011]:

$$\langle \frac{\partial \psi^\phi}{\partial t} \rangle = \frac{\partial \langle \psi^\phi \rangle}{\partial t} - \int_{\Gamma^*} \psi^\phi \vec{v}^* \cdot \vec{n}^\phi \, dA \quad (2.13)$$

$$\langle \vec{\nabla} \psi^\phi \rangle = \vec{\nabla} \langle \psi^\phi \rangle + \int_{\Gamma^*} \psi^\phi \vec{n}^\phi \, dA \quad (2.14)$$

where  $\vec{v}^*$  is the local relative interface velocity and  $\Gamma^*$  is the solid-liquid interface, while  $\vec{n}^\phi$  is the normal to  $\Gamma^*$ , directed outwards. The surface integral term in eqs. (2.13) and (2.14) is an *interfacial average* that expresses exchanges between the phases across the interface. The previous equations will be used to derive a set macroscopic conservation equations. It is noted that the intrinsic average  $\langle \psi \rangle^\phi$  may be replaced by  $\psi^\phi$  for notation simplicity, whenever the averaging technique applies.

## Macroscopic equations

A monodomain macroscopic model relies on four main conservation equations to predict macrosegregation in a single alloy domain, i.e. the latter is considered without any interaction with another alloy or ambient air. The general form of a conservation equation of any physical quantity  $\psi$  is given by [Rappaz et al. 2003]:

$$\frac{\partial \psi}{\partial t} + \nabla \cdot (\psi \vec{v}) - \nabla \cdot \vec{j}_\psi = Q_\psi \quad (2.15)$$

The first LHS term in eq. (2.15) represents the time variation of  $\psi$ , the second term accounts for transport by advection while the third is the diffusive transport and the RHS term represents a volume source. The considered equations are mass, energy, liquid momentum and species conservation, all summarized in table 2.1. The solid momentum is not considered as we assume a fixed and rigid solid phase ( $\vec{v}^s = \vec{0}$ ). We

## Chapter 2. Modelling Review

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**Table 2.1** – Summary of conservation equations with their variables.

Conservation Equation	$\psi$	$\vec{j}_\psi$	$Q_\psi$
Mass	$\langle \rho \rangle$	—	—
Energy	$\langle \rho h \rangle$	$\langle \vec{q} \rangle$	—
Species	$\langle \rho w_i \rangle$	$\langle \vec{j}_i \rangle$	—
Liquid momentum	$\langle \rho \vec{v}^l \rangle$	$-\langle \overline{\vec{\sigma}^l} \rangle$	$\vec{F}_v^l$

develop the ingredients of these equations using the averaging technique, as follows:

$$\langle \rho \rangle = g^l \rho^l + g^s \rho^s \quad (2.16)$$

$$\langle \rho \vec{v} \rangle = g^l \rho^l \vec{v}^l + g^s \rho^s \vec{v}^s \quad (2.17)$$

$$\langle \rho h \rangle = g^l \rho^l h^l + g^s \rho^s h^s \quad (2.18)$$

$$\langle \rho h \vec{v} \rangle = g^l \rho^l h^l \vec{v}^l + g^s \rho^s h^s \vec{v}^s \quad (2.19)$$

$$\langle \rho w_i \rangle = g^l \rho^l w_i^l + g^s \rho^s w_i^s \quad (2.20)$$

$$\langle \rho w_i \vec{v} \rangle = g^l \rho^l w_i^l \vec{v}^l + g^s \rho^s w_i^s \vec{v}^s \quad (2.21)$$

Next, we define the average diffusive fluxes,  $\vec{q}$  for energy and  $\vec{j}_i$  for solutes, using Fourier's conduction law and Fick's first law, respectively:

$$\langle \vec{q} \rangle = -g^l \langle \kappa^l \rangle \vec{\nabla} T - g^s \langle \kappa^s \rangle \vec{\nabla} T \quad (2.22)$$

$$\langle \vec{j}_i \rangle = -g^l D^l \vec{\nabla} w_i^l - g^s D^s \vec{\nabla} w_i^s \quad (2.23)$$

In eq. (2.23), the solid diffusion coefficient is neglected, by considering that for macroscopic scales, the average composition of the alloy is much more influenced by advective and diffusive transport in the liquid. In eq. (2.22), we assumed that phases are at thermal equilibrium, that is, temperature is uniform in the RVE.

Now that the main conservation equations ingredients are properly defined, we may write each averaged conservation equations as the sum of two local conservation equations for each phase in the RVE, hence introducing also interfacial average terms. For instance, the local mass balance in each phase is given by:

$$\frac{\partial}{\partial t} (g^l \rho^l) + \nabla \cdot (g^l \rho^l \vec{v}^l) = S_V \langle \rho^l \vec{v}^{l*} \cdot \vec{n} \rangle^* - S_V \langle \rho^l \vec{v}^* \cdot \vec{n} \rangle^* \quad (2.24a)$$

$$\frac{\partial}{\partial t} (g^s \rho^s) + \nabla \cdot (g^s \rho^s \vec{v}^s) = -S_V \langle \rho^s \vec{v}^{s*} \cdot \vec{n} \rangle^* + S_V \langle \rho^s \vec{v}^* \cdot \vec{n} \rangle^* \quad (2.24b)$$

where  $S_V = A_{sl}/V_E$  is the specific surface area,  $\vec{v}^{l*}$  and  $\vec{v}^{s*}$  are respectively, the liquid and solid phase velocity at the interface and  $\vec{v}^*$  is the previously introduced solid-liquid interface velocity. For instance, the first interfacial exchange term in the RHS of

eq. (2.24a) is expanded as follows [Dantzig et al. 2009]:

$$S_V \langle \rho^l \vec{v}^{l*} \cdot \vec{n} \rangle^* = \frac{A_{sl}}{V_E} \left( \frac{1}{A_{sl}} \int_{A_{sl}} \rho^l \vec{v}^{l*} \cdot \vec{n} \, dA \right) \quad (2.25a)$$

$$= \frac{1}{V_E} \int_{A_{sl}} \rho^l \vec{v}^{l*} \cdot \vec{n} \, dA \quad (2.25b)$$

Summing equations (2.24a) and (2.24b), results in the overall mass balance in the RVE:

$$\begin{aligned} \frac{\partial}{\partial t} \left( g^l \rho^l + g^s \rho^s \right) + \nabla \cdot \left( g^l \rho^l \vec{v}^l + g^s \rho^s \vec{v}^s \right) = \\ S_V \langle \rho^l \left( \vec{v}^{l*} - \vec{v}^* \right) \cdot \vec{n} \rangle^* - S_V \langle \rho^s \left( \vec{v}^{s*} - \vec{v}^* \right) \cdot \vec{n} \rangle^* \end{aligned} \quad (2.26)$$

where the RHS cancels to zero as shown by Ni et al. [1991]. Moreover, the authors show that with their averaging technique, interfacial exchanges for energy, chemical species and momentum cancel out as they are equal in absolute value but opposite in sign. Using eqs. (2.16) to (2.23) and following the same procedure done in eq. (2.26), the averaged mass balance hence writes:

$$\frac{\partial \langle \rho \rangle}{\partial t} + \nabla \cdot \langle \rho \vec{v} \rangle = 0 \quad (2.27)$$

whereas the averaged energy balance writes:

$$\frac{\partial \langle \rho h \rangle}{\partial t} + \nabla \cdot \langle \rho h \vec{v} \rangle + \nabla \cdot \left( \langle \kappa \rangle \vec{\nabla} T \right) = 0 \quad (2.28)$$

and finally the species balance writes:

$$\frac{\partial \langle \rho w_i \rangle}{\partial t} + \nabla \cdot \langle \rho w_i \vec{v} \rangle + \nabla \cdot \left( g^l D^l \vec{\nabla} \left( \rho^l w_i^l \right) \right) = 0 \quad (2.29)$$

As stated previously, the momentum balance in the solid phase is not taken into consideration, hence we do not sum the corresponding local conservation equations. This has consequences on the advection terms in energy and species conservation, and later on we will show the consequences on the momentum conservation in the liquid. First, the advection terms in eqs. (2.28) and (2.29) shall be redefined by considering that the fluid is incompressible ( $\nabla \cdot \langle \vec{v}^l \rangle = 0$ ), which yields:

$$\nabla \cdot \langle \rho h \vec{v} \rangle = \langle \vec{v}^l \rangle \cdot \vec{\nabla} \left( \rho^l h^l \right) \quad (2.30)$$

$$\nabla \cdot \langle \rho w_i \vec{v} \rangle = \langle \vec{v}^l \rangle \cdot \vec{\nabla} \left( \rho^l w_i^l \right) \quad (2.31)$$

As for the liquid momentum balance, we write:

$$\frac{\partial}{\partial t} \left( \rho^l g^l \vec{v}^l \right) + \vec{\nabla} \cdot \left( \rho^l g^l \vec{v}^l \times \vec{v}^l \right) = \vec{\nabla} \cdot \left( g^l \bar{\bar{\sigma}}^l \right) + g^l \vec{F}_v^l + \vec{\Gamma}^l \quad (2.32)$$

where  $\vec{F}_v^l$  is the vector of external body forces exerted on the liquid phase. In our case, it accounts for the fluid's weight:

$$\vec{F}_v^l = \rho^l \vec{g} \quad (2.33)$$

The interfacial momentum transfer between the solid and liquid phases in [eq. \(2.32\)](#) is modelled by a momentum flux vector  $\vec{\Gamma}^l$ , consisting of hydrostatic and deviatoric parts, such that:

$$\vec{\Gamma}^l = \vec{\Gamma}_p^l + \vec{\Gamma}_{\mathbb{S}}^l \quad (2.34)$$

$$\vec{\Gamma}_p^l = p^{l*} \vec{\nabla} g^l = p^l \vec{\nabla} g^l \quad (2.35)$$

$$\vec{\Gamma}_{\mathbb{S}}^l = -g^{l2} \mu^l \mathbb{K}^{-1} \left( \vec{v}^l - \vec{x}^s \right) \quad (2.36)$$

where  $p^{l*}$  is the pressure at the interface, considered to be equal to the liquid hydrostatic pressure  $p^l$ ,  $\mathbb{K}$  is a permeability scalar (isotropic) computed using [eq. \(2.1\)](#) and  $\mu^l$  is the liquid's dynamic viscosity. The general form of the Cauchy liquid stress tensor in [eq. \(2.32\)](#) is decomposed as follows:

$$\langle \bar{\bar{\sigma}}^l \rangle = g^l \bar{\bar{\sigma}}^l = - \left( \langle p^l \rangle - \lambda \nabla \cdot \langle \vec{v}^l \rangle \right) \bar{\bar{\mathbf{I}}} + \langle \bar{\bar{\mathbb{S}}}^l \rangle \quad (2.37)$$

where  $\lambda$  is a dilatational viscosity [[Dantzig et al. 2009](#)] and  $\bar{\bar{\mathbb{S}}}^l$  is the liquid strain deviator tensor. In the literature, the coefficient  $\lambda$  is taken proportional to the viscosity:  $\lambda = \frac{2}{3} \mu^l$ . However, as we consider an incompressible flow, the divergence term vanishes, thus rewriting [eq. \(2.37\)](#) as follows:

$$\langle \bar{\bar{\sigma}}^l \rangle = -\langle p^l \rangle \bar{\bar{\mathbf{I}}} + \langle \bar{\bar{\mathbb{S}}}^l \rangle \quad (2.38a)$$

$$\langle \bar{\bar{\sigma}}^l \rangle = -\langle p^l \rangle \bar{\bar{\mathbf{I}}} + 2\mu^l \langle \dot{\bar{\bar{\mathbb{S}}}^l} \rangle \quad (2.38b)$$

where the transition from [eq. \(2.38a\)](#) to [eq. \(2.38b\)](#) is made possible by assuming a Newtonian behaviour for the liquid phase. The strain rate tensor,  $\langle \dot{\bar{\bar{\mathbb{S}}}^l} \rangle$ , depends on the average liquid velocity:

$$\langle \dot{\bar{\bar{\mathbb{S}}}^l} \rangle = \frac{1}{2} \left( \bar{\bar{\nabla}} \langle \vec{v}^l \rangle + \bar{\bar{\nabla}}^t \langle \vec{v}^l \rangle \right) \quad (2.39)$$

Finally, we obtain the final form of momentum conservation in the liquid phase coupled with the averaged mass balance, by injecting [eqs. \(2.33\), \(2.35\), \(2.36\), \(2.38b\)](#)

## 2.2. Eulerian and Lagrangian motion description

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and (2.39) in eq. (2.32):

$$\frac{\partial}{\partial t} \left( \rho^l \langle \vec{v}^l \rangle \right) + \frac{1}{g^l} \vec{\nabla} \cdot \left( \rho^l \langle \vec{v}^l \rangle \times \langle \vec{v}^l \rangle \right) = - g^l \vec{\nabla} p^l - 2\mu^l \vec{\nabla} \cdot \left( \overline{\overline{\nabla}} \langle \vec{v}^l \rangle + \overline{\overline{\nabla}}^t \langle \vec{v}^l \rangle \right) - g^l \mu^l \mathbb{K}^{-1} \langle \vec{v}^l \rangle + g^l \rho^l \vec{g} \quad (2.40)$$

where we intentionally employed the *superficial velocity*,  $\langle \vec{v}^l \rangle = g^l \vec{v}^l$ , as the main unknown, together with the liquid pressure  $p^l$ . This system, when modelled in 3D, has a total of 4 unknowns (velocity vector and pressure) and 3 equations ( $X, Y$  and  $Z$  projections for the velocity vector). A fourth equation is therefore added for closure, it's the mass balance for an incompressible fluid, derived from eq. (2.27), thus yields the so-called *Navier-Stokes* equations:

$$\begin{cases} \frac{\partial}{\partial t} \left( \rho^l \langle \vec{v}^l \rangle \right) + \frac{1}{g^l} \vec{\nabla} \cdot \left( \rho^l \langle \vec{v}^l \rangle \times \langle \vec{v}^l \rangle \right) = \\ - g^l \vec{\nabla} p^l - 2\mu^l \vec{\nabla} \cdot \left( \overline{\overline{\nabla}} \langle \vec{v}^l \rangle + \overline{\overline{\nabla}}^t \langle \vec{v}^l \rangle \right) - g^l \mu^l \mathbb{K}^{-1} \langle \vec{v}^l \rangle + g^l \rho^l \vec{g} \\ \nabla \cdot \langle \vec{v}^l \rangle = 0 \end{cases} \quad (2.41)$$

Since all conservation equations were presented and simplified by the main assumption of a static solid phase, we may include them in a graphical summary in section 2.1.4

Macro models: Rivaux ? Gu beckermann 1999

Micro macro: Tommy Carozzani2013, guo beckermann 2003, Combeau 2009, Miha Zaloznik 2010 (indirect), P. Thévoz, J.-L. Desbiolles, M. Rappaz, Metallurgical and Materials TransactionsA 20 (2) (1989) 311–322

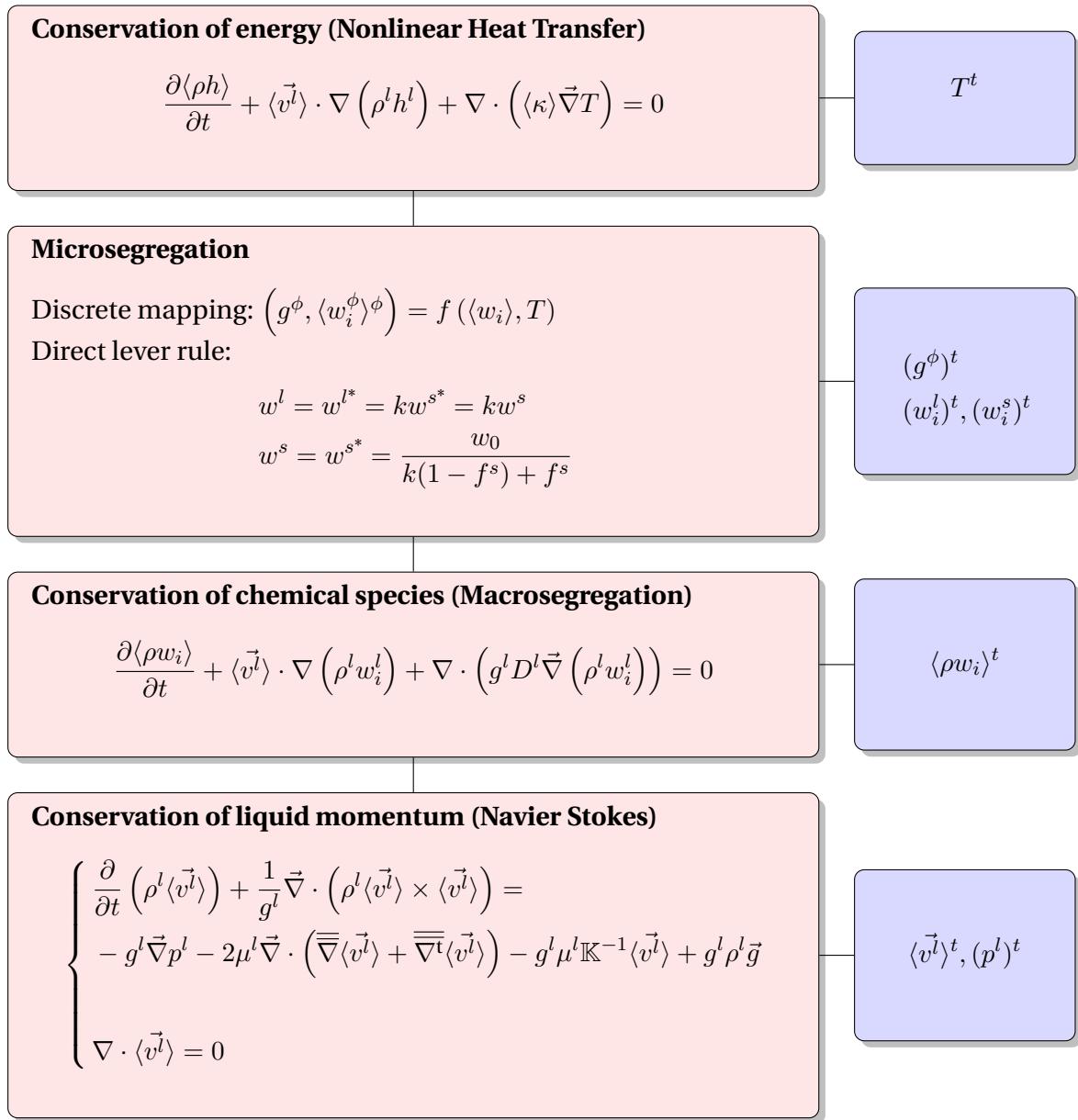
end by talking about taking air into account and the need for an interface capturing method

## 2.2 Eulerian and Lagrangian motion description

### 2.2.1 Overview

In mechanics, it is possible to describe motion using two well-known motion description: Eulerian and Lagrangian descriptions. To start with the latter, it describes the motion of a particle by attributing a reference frame that moves with the particle. In other words, the particle itself is the center of a reference frame moving at the same speed during time. The position vector, denoted by  $\vec{x}$ , is hence updated as follows:

$$\vec{x}^{(t+1)} = \vec{x}^{(t)} + \vec{v} \Delta t \quad (2.42)$$



**Fig. 2.3** – Graphical resolution algorithm of the conservation equations used in a monodomain macroscopic model to predict macrosegregation. The blue boxes represent the output of each equation at a time step  $t$ .

## 2.2. Eulerian and Lagrangian motion description

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As such, the total variation of any physical quantity  $\psi$  related to the particle can be found by deriving with respect to time,  $\frac{d\psi}{dt}$ . In contrast to the Lagrangian description, the Eulerian description considers a fixed reference frame and independent of the particle's trajectory. The total variation of  $\psi$  cannot be simply described by a temporal derivative, since the particle's velocity is not known to the reference frame, and thus the velocity effect, namely the advective transport of  $\psi$ , should also be considered as follows:

$$\frac{d\psi}{dt} = \frac{\partial\psi}{\partial t} + \underbrace{\vec{v} \cdot \vec{\nabla}\psi}_{\text{Advection}} \quad (2.43)$$

In this case, the LHS term is also known as *total* or *material derivative*. The importance of these motion descriptions is essential to solve mechanics, whether for fluids or solids, using a numerical method like the finite element method (FEM). One of the main steps of this method is to spatially discretise a continuum into a grid of points (nodes, vertices ...), where any physical field shall be accordingly discretized. Now, if we focus on a node where velocity has a non zero value and following the previously made analysis, two outcomes are possible: either the node would be fixed (Eulerian) or it would move by a distance proportional to the prescribed velocity (Lagrangian). In the latter case, points located on the boundaries constantly require an update of the imposed boundary conditions.

From these explanations, one can deduce that an Eulerian framework is suited for fluid mechanics problems where velocities are high and may distort the mesh points, whereas the Lagrangian framework is better suited for solid mechanics problems where deformation velocities are relatively low and should well behave when predicting strains.

Another motion description has emerged some decades ago, [Hirt \[1971\]](#) call it the Arbitrary Langrangian-Eulerian (ALE) method. ALE combines advantages from both previous descriptions as it dictates a Lagrangian behavior at "solid" nodes where solid is deforming, and an Eulerian behavior at "fluid" nodes.

### 2.2.2 Interface capturing

As no solid deformation is considered in this work, the Eulerian framework is a convenient choice. Although solidification shrinkage is to be considered in the current scope, it will deform the alloy's outer surface in contact with the air. We intend to track this interface and its motion over time via a numerical method. A wide variety of methods accomplish this task while they yield different advantages and disadvantages. Such methods fall into two main classes, either interface tracking or interface capturing, among which we cite: marker-and-cell (MAC) [[Harlow et al. 1965](#)], volume of fluid (VOF) [[Hirt et al. 1981](#)], phase field methods (PF), level set method (LSM) [[Osher et al. 1988](#)], coupled level set - VOF method and others. The interested reader may refer to

quick references by Prosperetti [2002] and Maitre [2006] about these methods.

In the past years, the level set method received a considerable attention in many computational fields, specifically in solidification. For this reason, we will focus on this method henceforth, giving a brief literature review and technical details in the next sections.

### 2.3 Solidification models with level set

In classic solidification problems, the need to track an interface occurs usually at the solid-liquid interface, that is why the phase field method [Karma et al. 1996; Boettinger et al. 2002] and the level set method [Chen et al. 1997; Gibou et al. 2003; Tan et al. 2007] were applied at a microscale to follow mainly the dendritic growth of a single crystal in an undercooled melt. In our case however, when we mention "solidification models using LSM", we do not mean the solid-liquid interface inside the alloy, but it is the alloy(liquid)-air interface that is tracked, assuming that microscale phenomena between the phases within the alloy, are averaged using the previously defined technique in section 2.1.4.

Very few models were found in the literature, combining solidification and level set as stated previously. Du et al. [2001] applied it to track the interface between two molten alloys in a double casting technique. Welding research, on another hand, has been more active adapting the level set methodology to corresponding applications. In CEMEF, two projects use the metal-air level set methodology in welding simulations and showed promising results.

two sentences not more about the contribution of OD and SC

Firstly, Desmaison et al. [2014] ... TODO

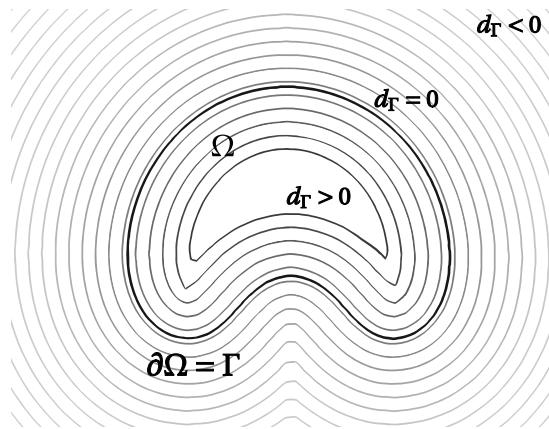
Later, Chen [2014] applied it to gas metal arc welding (GMAW) to predict the grain structure in the heat affected zone essentially. More recently, Courtois et al. [2014] used the same methodology but this time to predict keyhole defect formation in spot laser welding. The tracked interface in this case was that between the molten alloy and the corresponding vapor phase.

### 2.4 The level set method

Firstly introduced by Osher et al. [1988], this method became very popular in studying multiphase flows. It is reminded that the term *multiphase* in computational domains usually refers to multiple fluids, and thus should not be mixed with definition of a phase in the current solidification context. For disambiguation, we shall use *multifluid flow* when needed. The great advantage lies in the way the interface between two fluids,  $F_1$  and  $F_2$  is implicitly captured, unlike other methods where the exact interface position is needed. In a discrete domain, the concept is to assign for each mesh node of position vector  $\vec{x}$ , the minimum distance  $d_\Gamma(\vec{x})$  separating it from an interface  $\Gamma$ .

The distance function, denoted  $\alpha$  and defined in [eq. \(2.44\)](#), is then signed positive or negative, based on the fluid or domain to which the node belongs.

$$\alpha(\vec{x}) = \begin{cases} d_\Gamma(\vec{x}) & \text{if } \vec{x} \in F_1 \\ -d_\Gamma(\vec{x}) & \text{if } \vec{x} \in F_2 \\ 0 & \text{if } \vec{x} \in \Gamma_{F1,F2} \end{cases} \quad (2.44)$$



**Fig. 2.4** – Schematic of the interface  $\Gamma$  (thick black line) of a rising air bubble ( $\Omega$ ) in water. The other contours represent isovalues of the distance function around and inside the interface contour. Those outside are signed negative whereas inside they are signed positive.

### 2.4.1 Diffuse interface

The level set has many attractive properties that allows seamless implementation in 2D and 3D models. It is a continuously differentiable  $C^1$ -function. For instance, a *Heaviside* function, denoted  $H$ , can be obtained by first order derivation of the level set function. The Heaviside function is continuous but non differentiable, with an abrupt transition from 0 to 1 across the sharp interface, as follows:

$$H = H(\alpha(\vec{x})) = \begin{cases} 0 & \text{if } \alpha(\vec{x}) < 0 \\ 1 & \text{if } \alpha(\vec{x}) \geq 0 \end{cases} \quad (2.45)$$

With the help of [eq. \(2.45\)](#), we can define the geometric "presence" of a domain with respect to the interface. As such, material properties depend upon this function, which will be discussed later in [section 2.4.2](#). It is established that a steep transition can lead to numerical problems, so the Heaviside function should be smoothed in a fixed

thickness. Sinusoidal smoothing in [eq. \(2.46\)](#) is widely used with level set formulations.

$$H = \begin{cases} 0 & \text{if } \alpha(\vec{x}) < -\varepsilon \\ 1 & \text{if } \alpha(\vec{x}) > \varepsilon \\ \frac{1}{2} \left( 1 + \frac{\alpha(\vec{x})}{\varepsilon} + \frac{1}{\pi} \sin \left( \frac{\pi \alpha(\vec{x})}{\varepsilon} \right) \right) & \text{if } -\varepsilon \leq \alpha(\vec{x}) \leq \varepsilon \end{cases} \quad (2.46)$$

where the interval  $[-\varepsilon; +\varepsilon]$  is an artificial interface thickness around the zero distance. Defining a diffuse interface rather than a sharp one, is also a common approach in phase field methods [[Beckermann et al. 1999](#); [Sun et al. 2004](#)]. It is emphasized that the latter methods give physically meaningful analysis of a diffuse interface and the optimal thickness by thoroughly studying the intricate phenomena happening at the scale of the interface. However, for level set methods, there has not been a formal work leading the same type of analysis. For this reason, many aspects of the level set method lack physical meanings but still computationally useful. In a recent paper by [Gada et al. \[2009\]](#), the authors respond partially to this problem by analysing and deriving conservation equations using a level set in a more meaningful way, but do not discuss the diffuse interface aspect.

The dirac delta function is also an important property to convert surface integrals to volume terms, which could turn useful when modelling surface tension effects for instance, using the *continuum surface force* method (CSF) [[Brackbill et al. 1992](#)]. The dirac function, plotted in [fig. 2.5](#) along with the Heaviside function within an interface thickness of  $[-\varepsilon; +\varepsilon]$ , is derived from the Heaviside as follows:

$$\delta(\alpha) = \delta(\alpha(\vec{x})) = \frac{\partial H}{\partial \alpha(\vec{x})} = \begin{cases} \frac{1}{2\varepsilon} \left( 1 + \cos \left( \frac{\pi \alpha(\vec{x})}{\varepsilon} \right) \right) & \text{if } |\alpha(\vec{x})| \leq \varepsilon \\ 0 & \text{if } |\alpha(\vec{x})| > \varepsilon \end{cases} \quad (2.47)$$

The Heaviside and delta dirac functions can be readily processed to obtain other geometric properties from the level set, which are extremely useful. We mention the most relevant ones [[Peng et al. 1999](#)]:

$$\text{normal vector : } \vec{n} = \frac{\vec{\nabla} \alpha}{\|\vec{\nabla} \alpha\|} \quad (2.48)$$

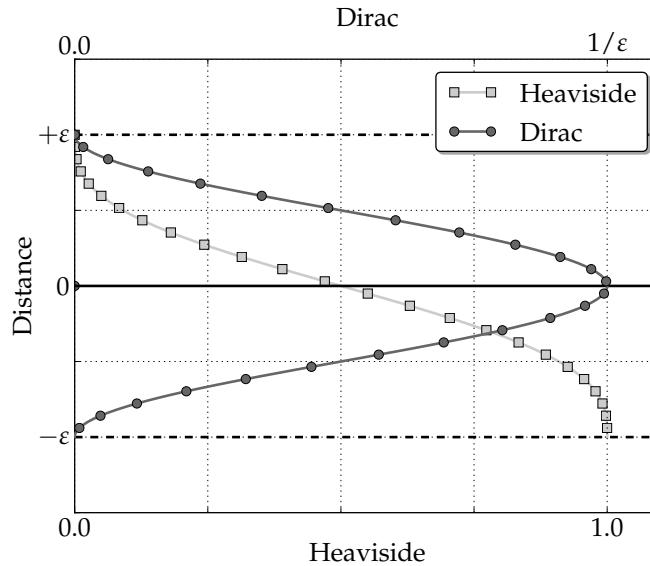
$$\text{curvature : } \zeta = -\nabla \cdot \vec{n} \quad (2.49)$$

$$\text{surface area of the air-metal interface : } A^\Gamma = \int_{\Omega} \delta(\alpha) \|\vec{\nabla} \alpha\| d\Omega \quad (2.50)$$

$$\text{metal volume : } V^M = \int_{\Omega} H^M d\Omega \quad (2.51)$$

where, for the last two equations, we considered a three-dimensional domain  $\Omega$  containing two subdomains, metal and air, separated by an interface  $\Gamma$ . It is reminded that for a 2D case, [eq. \(2.50\)](#) evaluates a length instead of the area while [eq. \(2.51\)](#) gives the area instead of volume. Finally, within the diffuse interface, fluids properties may vary

linearly or not, depending on the mixing law, which is presented in the next section.



**Fig. 2.5** – Schematic of two level properties inside the diffuse interface: Heaviside (lower x-axis) and Dirac delta (upper x-axis) functions. Note that the peak of the dirac function depends on the interface thickness to ensure a unity integral of the delta function over  $\Omega$ .

### 2.4.2 Mixing Laws

A *monolithic* resolution style, as opposed to a *partitioned* resolution, is based on solving a single set of equations for both fluids separated by an interface, as if a single fluid were considered. Level set is one many methods that use the monolithic style to derive a single set of conservation equations for both fluids. The switch from one material to the other is implicitly taken care of by using the Heaviside function as well mixing laws. These laws are crucial to define how properties vary across the diffuse interface in view of a more accurate resolution. The most frequently used mixing law in the literature is the arithmetic law. Other transitions are less known such as the harmonic and logarithmic mixing. The first law is maybe the most intuitive and most used for properties mixture as it emanates from VOF-based methods. If we consider any property  $\psi$ , for instance the fluid's dynamic viscosity  $\mu$ , then the arithmetic law will give a mixed property  $\hat{\psi}$  as follows:

$$\hat{\psi} = H^{F_1} \psi^{F_1} + H^{F_2} \psi^{F_2} \quad (2.52)$$

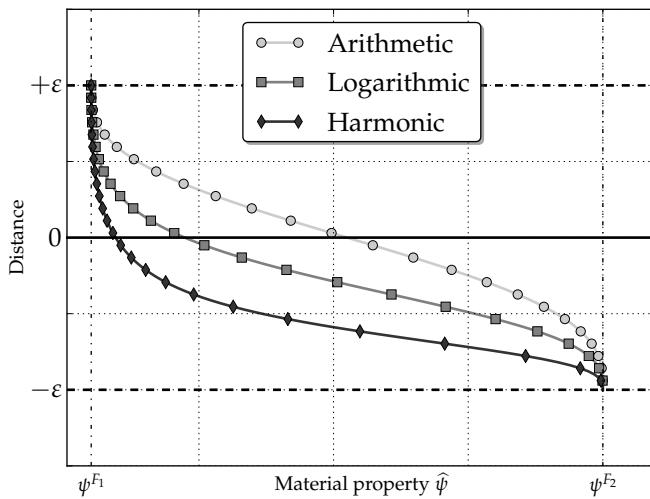
Basically, the result is an average property that follows the same trend as the Heaviside function. As for the harmonic law, it writes:

$$\hat{\psi} = \left( \frac{H^{F_1}}{\psi^{F_1}} + \frac{H^{F_2}}{\psi^{F_2}} \right)^{-1} \quad (2.53)$$

and last, the logarithmic law writes:

$$\hat{\psi} = n^{(H^{F_1} \log_n \psi^{F_1} + H^{F_2} \log_n \psi^{F_2})} \quad (2.54)$$

where  $n$  is any real number serving as a logarithm base, which often is either the exponential  $e$  or 10. The mixture result with this law is the same, regardless of the value of  $n$ . By looking to [fig. 2.6](#), we clearly see that the difference between all three approaches is the property weight given to each side of the level set in the mixture. The arithmetic law, being symmetric, has equal weights,  $\psi^{F_1}$  and  $\psi^{F_2}$ , in the final mixture. Nevertheless, the asymmetric harmonic mixing varies inside the diffuse interface with a dominant weight of one property over the other. As for the logarithmic mixture, it can be seen as an intermediate transition between the preceding laws. As long



**Fig. 2.6** – Three mixing laws, arithmetic, logarithmic and harmonic commonly used in monolithic formulations.

as the interface thickness is small enough, the choice of a mixing law should not drastically change the result, inasmuch as it depends on the discretisation resolution of the interface. This fact made the arithmetic mixing the most applied one, because it is symmetric and easy to implement (no handling of potential division problems like harmonic laws for instance). However, [Strotos et al. \[2008\]](#) claim that the harmonic law proves to conserve better diffusive fluxes at the interface. More recently, an interesting study made by [Ettrich et al. \[2014\]](#) focused on mixing thermal properties using a phase field method. They define a diffuse interface in which they separately mix the thermal conductivity,  $\kappa$ , and the heat capacity,  $C_v$ , then compute the thermal diffusivity as the ratio of these properties. Later, the authors compare the temperature field obtained by diffusion to a reference case in order to decide which combination of mixing laws gives the best result. Despite not being directly related to a level set method, this work gives an insight of the mixture possibilities and their effect on a pure thermal diffusion. Otherwise, little work has been found in the literature on the broad effects of mixture

types on simulation results in a level set context.

## 2.5 Interface motion

When a physical interface needs to have topology changes because of fluid structure interaction or surface tension for instance, the level set model can easily follow these changes by a transport step. The idea is to advect the signed distance function, its zero isovalue representing the interface and all other distant isovales, with the velocity field as input. The motion of the interface is thus expressed by:

$$\frac{d\alpha}{dt} = \frac{\partial\alpha}{\partial t} + \vec{v} \cdot \vec{\nabla}\alpha = 0 \quad (2.55)$$

Associated boundary conditions

### 2.5.1 Level set transport

The finite element method gives the fully discretised weak form of [eq. \(2.55\)](#) by using a convenient set of test functions  $\alpha^*$  belonging to the hilbertian *Sobolev* space:

$$\int_{\Omega} \alpha^* \frac{\partial\alpha}{\partial t} d\Omega + \int_{\Omega} \alpha^* \vec{v} \cdot \vec{\nabla}\alpha d\Omega = 0 \quad \forall \alpha^* \in \mathcal{H}^1(\Omega) \quad (2.56)$$

The spatial discretisation of  $\alpha$  assigns, for each of the total  $N$  nodes of a simplex, the following values:

$$\alpha = \sum_N P_j \alpha_j \quad (2.57)$$

Furthermore, with the Galerkin method, we replace test functions by the interpolation functions  $P_j$ , then we apply a temporal discretisation for the main unknowns by a forward finite difference in time. Consequently, [eq. \(2.56\)](#) can be recast as follows:

$$\frac{1}{\Delta t} (\alpha_j^t - \alpha_j^{t-\Delta t}) \int_{\Omega} P_i P_j d\Omega + \alpha_j^t \int_{\Omega} \vec{v}^t \cdot \vec{\nabla} P_j d\Omega = 0 \quad (2.58a)$$

$$\left[ \frac{1}{\Delta t} \int_{\Omega} P_i P_j d\Omega + \int_{\Omega} \vec{v}^t \cdot \vec{\nabla} P_j d\Omega \right] \alpha_j^t = \frac{1}{\Delta t} \int_{\Omega} \alpha^{t-\Delta t} P_i d\Omega \quad (2.58b)$$

$$[\mathcal{M}_{ij} + \mathcal{A}_{ij}] \alpha_j^t = \mathcal{F}_i \quad (2.58c)$$

where  $\mathcal{M}_{ij}$  and  $\mathcal{A}_{ij}$  are respectively the mass (or capacity) matrix and advection matrix, both written within a local finite element, whereas  $\mathcal{F}_i$  is a local vector of known quantities from the previous time step. The solution of the linear system in [eq. \(2.58c\)](#) is the transported distance function.

### 2.5.2 Level set regularisation

Upon transport the distance function field, a crucial property of the level set may be partially or totally lost over the domain, which is:

$$\begin{cases} \|\vec{\nabla}\alpha\| = 1 \\ \alpha(x, t) = 0 \end{cases} \quad \text{if } x \in \Gamma(t) \quad (2.59)$$

The closer this  $L^2$ -norm to one, the more regular the level set. An irregular distance function induces cumulative numerical errors in the transport step (eq. (2.55)) and thus results in non conservation of mass, because of spurious distance information. When the transport equation in eq. (2.55) is discretised in time then solved, a *regularisation* (also known as *reinitialisation*) is necessary to conserve as much as possible the property in eq. (2.59).

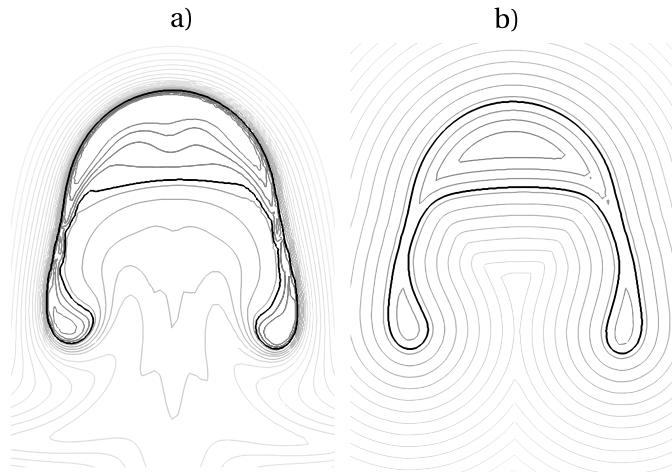
Figure 2.7 shows the need of regularisation in two different simulations of the same phenomenon: rising air bubble inside water. The importance of this well studied case [Sussman et al. 1994; Hysing et al. 2009] is that the interface between two fluids is highly deformable as the bubble rises because of buoyancy, and therefore the task of tracking the dynamic interface while maintaining an accurate distance function is a considerable numerical task. In the first simulation, the distance contours are squeezed against the zero-distance contour marked by the thick black line. A closer look to the interface reveals undesired distortions, with a "wavy" shape at some points. This effect is evidently an artefact of a level set transport lacking subsequent reinitialisation, inasmuch as the surface tension tends to minimise the total surface area and make it as smooth as possible. Nevertheless, the second simulation unveils much better results, especially how the interface shows no sign of destabilisation. We also note the regular spacing between contours, which is a consequence of conserving the property defined in eq. (2.59). We attribute this improvement to the regularization done at each time step after the transport. In the forthcoming sections, we present two regularisation methods, then show their strong and weak points.

#### Classic Hamilton-Jacobi reinitialisation

In order to repair a distance function impaired by convective transport, Sussman et al. [1994] proposed solving a classic *Hamilton-Jacobi* equation, given in its most general form:

$$\frac{\partial \alpha}{\partial t} + \mathbb{H}(\alpha, x, t) = 0 \quad x \in \Omega, t > 0 \quad (2.60)$$

where  $\alpha(x, t = 0) = \alpha_0$  is the initial value of the distance function. The term  $\mathbb{H}$  is known as the *Hamiltonian*. When the sign of the level set and its metric property



**Fig. 2.7** – Schematic of the influence of level set regularisation on the distance function at the same time frame: a) without any regularisation step, the isovalue contours are distorted in the wake of the rising air bubble while being squeezed ahead of it, b) in contrast to regularising the distance function, where the contours maintain their spacing and geometric properties with respect to the tracked interface.

( $\|\vec{\nabla}\alpha\| = 1$ ) are considered, eq. (2.60) reduces to:

$$\frac{\partial \alpha}{\partial t} + S(\alpha) (\|\vec{\nabla}\alpha\| - 1) = 0 \quad (2.61)$$

where  $S(\alpha)$  is a step function giving the sign of the level set as follows:

$$S(\alpha) = \frac{\alpha}{|\alpha|} = \begin{cases} -1 & \text{if } \alpha < 0 \\ 0 & \text{if } \alpha = 0 \\ +1 & \text{if } \alpha > 0 \end{cases} \quad (2.62)$$

The sign function defined in eq. (2.62) is often smoothed to avoid numerical problems, as proposed for instance by [Sussman et al. \[1994\]](#):

$$S(\alpha) = S_\varepsilon(\alpha) = \frac{\alpha}{\sqrt{\alpha^2 + \varepsilon^2}} \quad (2.63)$$

where  $\varepsilon$  is a smoothing parameter that depends on the mesh size around the interface. However, one should be aware that within the smoothing thickness, the regularised function may suffer from local oscillations because of the reciprocal reinitialisation taking place at each side of the level set. [Peng et al. \[1999\]](#) states that this problem is more likely to happen if the initial level set shows very weak or very steep gradients, and therefore is not regular enough. The authors eventually propose a new sign function which would reinitialise the distance function, as close as possible to the interface

without modifying the latter, as follows:

$$S(\alpha) = S_\varepsilon(\alpha) = \frac{\alpha}{\sqrt{\alpha^2 + \|\vec{\nabla}\alpha\|^2\varepsilon^2}} \quad (2.64)$$

### Convective reinitialization

A recent work by Ville et al. [2011] introduced another concept for reinitialisation called the *convective reinitialisation*. The idea lies in combining both level set advection and regularisation in a single equation, saving resolution time. The key components of their method starts by defining a pseudo time step,  $\Delta\tau$ , that is linked to the main time variable through a numerical parameter  $\lambda_\tau$ , as follows:

$$\lambda_\tau = \frac{\partial\tau}{\partial t} \quad (2.65)$$

The order of magnitude of  $\lambda_\tau$ , which can be seen as a relaxation parameter [see Vigneaux 2007, p. 89], is close to the ratio  $h/\Delta t$ . Then, the classic Hamilton-Jacobi eq. (2.60) is combined into the convection step by writing:

$$\frac{\partial\alpha}{\partial t} + (\vec{v} + \lambda_\tau \vec{U}) \cdot \vec{\nabla}\alpha = \lambda_\tau S(\alpha) \quad (2.66)$$

where  $\vec{U}$  is a velocity vector in the normal direction to the interface, defined by  $\vec{U} = S(\alpha)\vec{n}$ . the normal vector  $\vec{n}$  being previously defined in eq. (2.48). The obvious shortcoming of convective reinitialisation is that it depends on a numerical parameter  $\lambda_\tau$ . Another limitation of the method is the use of a sinusoidal filter to modify the distance function by truncating its values beyond a thickness threshold, which is also another parameter to calibrate the resolution. The drawback of truncating the level set is the loss of information far from the interface and the inability to fully reconstruct the distance function. If we denote this threshold by  $E$  and the modified level set by  $\tilde{\alpha}$  inside the thickness, then eq. (2.66) is recast as:

$$\frac{\partial\alpha}{\partial t} + (\vec{v} + \lambda_\tau \vec{U}) \cdot \vec{\nabla}\alpha = \lambda_\tau S(\alpha) \sqrt{1 - \left(\frac{\pi}{2E}\tilde{\alpha}\right)^2} \quad (2.67)$$

Equation (2.67) describes the transport and partial reconstruction of the distance function  $\alpha$ , knowing its value  $\tilde{\alpha}$  inside the thickness  $E$ .

### Geometric reinitialization

This category of methods go from the level set's basic geometric principle to construct a distance function, instead of solving a partial differential system of equations as in the classic Hamilton-Jacobi reinitialisation. A widely known instance of this category is the *fast marching method* developed by Sethian [1996] and influenced by the Dijkstra [1959]'s method to compute the shortest path in a network of nodes. The method aims

to solve the eikonal equation in [eq. \(2.59\)](#) to propagate the distance function in a single direction by *upwinding*, i.e. going from low to high values of the distance function, while preserving a unitary distance gradient.

*Direct reinitialisation* is another interesting method in the geometric reinitialise category. However, it has not gained noticeable attention in the literature given the terribly cost in terms of computation time and efficiency. The main idea is very simple: reconstruct the distance function over  $\Omega$  or a subset of  $\Omega$ , by computing the minimum distance between each mesh node and the interface. It means that, for any point  $\vec{x} \in \Omega$ , the following constraint should be satisfied [Osher et al. \[2003\]](#):

$$d_\Gamma(\vec{x}) = \min \|\vec{x} - \vec{x}_\Gamma\| \quad \forall \vec{x}_\Gamma \in \partial\Omega = \Gamma, \quad (2.68)$$

A efficient implementation of this method is done by [Shakoor et al. \[2015\]](#) making use of *k-d* trees to limit the search operations of elements and the subsequent distance evaluations in each of these elements. Moreover, the authors give a comparison of the previously stated methods on 2D and 3D cases, showing the great performance of direction reinitialisation when used with k-d trees algorithm, hence we use it in the present work.

### To do ?

Interface Remeshing: Importance when using a static level set and more importantly when LS is transported, influence of mixing area *thickness* and *resolution* (i.e. nb of nodes with the area), Isotropic or anisotropic ? the first is more important to composition calculation while the second is more relevant if we mean do thermohydraulics without macrosegregation



## Chapter 3

# Energy balance with thermodynamic tabulations

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### 3.1 State of the art

Use of enthalpy resolution in the majority of works  
motivation and advantages of TvsH without talking about resolution time  
use article's introduction to fill this section (or improvise new things)

TODO: replace wt pct by the macro bin or tern

When speaking about macrosegregation, one needs to know that the problem involves phase change. For that, a minimum of four conservation equations are necessary: conservation of mass, momentum, chemical species and energy. The phase change literature contains a wealth of numerical methods to solve energy conservation in solidifying alloys. A comprehensive overview of these methods is given by [Swaminathan. et al. \[1993\]](#). The corresponding equation associates the total average enthalpy to the temperature via intrinsic alloy properties, such as the heat capacity of the phases and the latent heat associated with the phase transformations. However, in the course of solidification and while macrosegregation is taking place, these properties may change because the average composition may vary significantly: the transformation paths are thus modified, as well as the phases' composition and heat capacity. Similarly, the latent heat of phase transformations is not a mere constant that could be distributed as a function of the phase fractions assuming only temperature-dependent phases' properties, as often found in the literature [\[Bellet et al. 2009\]](#). It is thus impossible to establish a priori the dependence of the enthalpy with respect to temperature when macrosegregation takes place, even in the case of full thermodynamic equilibrium between phases. In this chapter, we discuss a suitable numerical scheme based on an enthalpy method, already used in the literature to alleviate this macrosegregation-related problem [\[Swaminathan. et al. 1993; Carozzani et al. 2013\]](#). Later on, we introduce a modified formulation, using the effective heat capacity method that increases the original scheme's efficiency.

The current method is thus an enthalpy method that makes use of a temperature-based solver. Moreover, it uses tabulated thermodynamic quantities (solidification paths, phases' enthalpy and composition) in a range of average compositions and temperatures as found in the literature [\[Doré et al. 2000; Thuinet et al. 2004; Du et al. 2007\]](#), with the aim of evaluating the total average enthalpy as well as the effective heat capacity. The novelty of the modified method resides in the use of thermodynamic tabulations without losing the advantages of the previous method, thus yielding faster computation times while maintaining a good accuracy.

### 3.2 Thermodynamic considerations

#### 3.2.1 Volume averaging

the following paragraph will be deleted once the volume averaging has been introduced in chapter 1

A volume averaging technique was suggested to deal with the presence of multiple phases [Ni et al. 1991]. It locally considers a Representative Volume Element (RVE) that contains a single or several phases (these are not necessarily in thermodynamic equilibrium) at a mesoscopic scale. We represent, for each unknown  $\psi$ , an intrinsic volume average,  $\langle \psi \rangle^\phi$  (also denoted  $\langle \psi^\phi \rangle^\phi$  in the literature), corresponding to a phase  $\phi$ . The volume average  $\langle \psi \rangle$  for this unknown in the RVE, hence averaged over all the present phases writes:

$$\langle \psi \rangle = \sum_{\phi} g^{\phi} \langle \psi \rangle^{\phi} \quad (3.1)$$

where  $g^{\phi}$  denotes the volume fraction of phase  $\phi$  in the RVE. It should be emphasized that the averaging technique applies to virtually all thermodynamic variables (enthalpy, density ...). Among these variables, the temperature is also considered to be uniform in the RVE. Applying the volume averaging technique to the energy conservation principle along with interfacial balances between the phases, results in the following averaged equation [Rappaz et al. 2003]:

$$\frac{\partial \langle \rho h \rangle}{\partial t} + \vec{\nabla} \cdot \langle \rho h \vec{v} \rangle = \vec{\nabla} \cdot (\langle \kappa \rangle \vec{\nabla} T) + \langle \dot{Q}_V \rangle \quad (3.2)$$

where  $\rho$  stands for the density,  $h$  the mass enthalpy,  $\vec{v}$  the velocity field,  $\kappa$  the thermal conductivity,  $T$  the temperature and  $\dot{Q}_V$  a possible volume heat source. (3.2) is the standard averaged form of the energy conservation equation used in non-stationary phase change problems.

I could elaborate more in this paragraph by showing the possible equations for the explicit formulation and maybe a figure to show the AlSi7 computation that i did with a v small time step

Once the variational form has been discretized in space and time, two possible resolution schemes emerge: the first is an explicit forward Euler scheme which gives rise to a linear equation where the temperature is known at time  $t$ ,  $T^t$ . This requires very small time steps in the current context, which limits the solver's usability at the scale of industrial applications. The second scheme is the backward Euler or full implicit discretization where terms are function of  $T^{t+\Delta t}$ . It leads to a nonlinear equation with 2 interdependent unknowns,  $\langle \rho h \rangle^{t+\Delta t}$  and  $T^{t+\Delta t}$ . It is clear that the nature of the temperature-enthalpy relationship plays a central role when formulating the resolution strategy of this nonlinear equation. Generally, it is admitted that, depending on the resolution strategy, it is necessary to express enthalpy as a function of temperature or vice-versa, together with associated partial derivatives,  $\frac{d\langle \rho h \rangle}{dT}$  or  $\frac{dT}{d\langle \rho h \rangle}$ .

### 3.2.2 The temperature-enthalpy relationship

In solidification problems, additional variables are involved in (3.1) and (3.2), like the transformation path that defines the history of the phase fractions, as well as the

### Chapter 3. Energy balance with thermodynamic tabulations

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average chemical composition  $\langle w_i \rangle$ ,  $i$  being the index of the chemical species (only the solutes are considered). The temperature-enthalpy relation averaged over the phases in a given RVE writes:

$$\langle \rho h \rangle = \sum_{\phi} g_{(T, \langle w_i \rangle \dots)}^{\phi} \rho_{(T, \langle w_i \rangle^{\phi} \dots)}^{\phi} h_{(T, \langle w_i \rangle^{\phi} \dots)}^{\phi} \quad (3.3)$$

Note that the volume average enthalpy is approximated by the product  $\langle \rho h \rangle^{\phi} = \langle \rho \rangle^{\phi} \langle h \rangle^{\phi}$  in the current work. As stated in the introduction, it becomes clear from (3.3) that phase properties, i.e. average phase density,  $\rho^{\phi}$  and enthalpy,  $h^{\phi}$ , are temperature and composition dependent. This equation is the key to convert the average volume enthalpy to temperature (through a procedure named H2T) or vice-versa (T2H). The values of the different phase fractions  $g^{\phi}$  (solidification path) and phase enthalpies  $\langle \rho h \rangle^{\phi}$  are thus needed to close the relation.

#### 3.2.3 Tabulation of properties

The complexity of performing a thermodynamic conversion is directly linked to the simplicity of determining the alloy properties, namely the phase fractions and phase enthalpies. In the case of binary alloys and with several assumptions with respect to the system (e.g., linear monovariant temperature composition relationships, constant heat capacity of phases and constant latent heat of transformations, equilibrium approximations between phases) analytical calculations are often used to determine the properties. Nevertheless, analytical relations are more complex or even impossible to derive in the case of multicomponent alloys ( $i > 1$ ). To overcome this problem, one can resort to thermodynamic databases and phase equilibrium calculations to tabulate the transformation paths and the phase enthalpies for a given range of temperatures and average compositions. It is a handy solution for two main reasons: first, the conversion is merely a binary search in a table; secondly, it is a simple solution for coupling with macrosegregation. In this way, phase fractions  $g^{\phi}$  are tabulated as functions of temperature and average composition, while for each phase  $\phi$  the mass enthalpy,  $h^{\phi}$ , and the density,  $\rho^{\phi}$ , are tabulated as functions of temperature and phase intrinsic average compositions HERE, as well as other possible parameters. Figure 1 summarizes the steps in order to perform a temperature-to-enthalpy (T2H) conversion using the predefined tabulation approach. In step 1, the transformation path is acquired for each average composition and temperature to determine the list of phases, their volume fractions  $g^{\phi}$  and their intrinsic compositions  $\langle w_i \rangle^{\phi}$ . In step 2, the phase enthalpy  $h^{\phi}$  and density  $\rho^{\phi}$  are determined by searching for the temperature and the already known phase composition  $\langle w_i \rangle^{\phi}$ . In step 3, the average volume enthalpy is computed from the volume fraction, density and mass enthalpy of phases using (3.3).

Figure 3 goes here

The methodology to build the tabulations is straightforward. It is based on two main

scans. On the one hand, intervals for the variation of the average composition  $\langle w_i \rangle$  are chosen from the known alloy composition. These variations have to cover the extreme values adopted during the simulation, which are not known a priori. An interval is also selected for the variation of temperature. The latter is easier to determine as it usually starts from the initial melt temperature and goes down to the room temperature in a standard casting simulation. For these intervals, a systematic scan is made with chosen steps in each composition and T, during which a thermodynamic equilibrium is computed. The outputs are the number of phases encountered, together with their fraction and intrinsic composition. The minimum and maximum intrinsic composition for each phase could then be determined. On the other hand, for each phase, a scan of the intrinsic composition and temperature is made to compute the intrinsic properties. The same temperature interval and step as defined earlier are used.

below paragraph should be re written and maybe stress LESS on the speed effect

I should change the superscript  $k$  which may be confused with partition coefficient

Regarding the enthalpy-to-temperature conversion (H2T), a backward iterative T2H search is performed. For a known composition  $\langle w_i \rangle$ , denoting  $k$  the iteration index to convert the enthalpy  $\langle ph \rangle_{\text{input}}$ , we start with an initial guess for temperature  $T^{(k=0)}$  then convert it to an enthalpy  $\langle ph \rangle^{(k=0)}$  with the T2H conversion. Using an appropriate nonlinear algorithm (Brent is the most versatile in our case), we aim at minimizing the following residual:  $\text{Residu}_{\langle ph \rangle} = |\langle ph \rangle_{\text{input}} - \langle ph \rangle^{(k)}|$ . Once the algorithm has converged, the temperature  $T^{(k)}$  is the result of the H2T conversion. It is inferred that the first conversion (T2H) is a direct one whereas the latter (H2T) is indirect and requires a series of iterative steps; each step being a single T2H resolution. In other words, a H2T conversion is a backward search for a temperature, hence it's slower. This conversion's speed lag is exacerbated when tabulations increase in size (e.g. large number of temperature and composition steps) and complexity (e.g., multicomponent industrial alloys used in casting), since the search gets more complicated with the increasing number of input columns (one column for each alloying element).

## 3.3 Validation

### 3.3.1 Pure diffusion

The two solvers are first tested in a purely diffusive case for a one-dimensional solidification configuration. Predictions with a 1D front tracking model [Gandin 2000] is used as a benchmark. It provides solutions for the temperature and solid fraction during directional solidification of a 10 cm long Al – 7 wt.% Si ingot. The melt, with initial uniform temperature, is cooled with a heat exchange coefficient (assuming a Fourier boundary condition) from one side, the other side being adiabatic. All values for alloy properties, initial and boundary conditions and numerical parameters are listed in [Table 3.1](#). For this simple test case, we use linear temperature dependence of

### Chapter 3. Energy balance with thermodynamic tabulations

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Parameter	Symbol	Value	Unit
Nominal composition	$\langle w \rangle_0$	7	wt.%
Liquidus temperature	$T_L$	618	°C
Eutectic temperature	$T_E$	577	°C
Segregation coefficient	$k$	0.13	—
Liquidus slope	$m_L$	-6.5	K wt.% <sup>-1</sup>
Heat capacity (liquid and solid)	$\rho C_p$	$2.6 \times 10^6$	J m <sup>-3</sup> K <sup>-1</sup>
Enthalpy of fusion	$\rho L$	$9.5 \times 10^8$	J m <sup>-3</sup>
Thermal conductivity (liquid and solid)	$\kappa$	70	W m <sup>-1</sup> K <sup>-1</sup>
Heat transfer coefficient	$h_{\text{ext}}$	500	W m <sup>-2</sup> K <sup>-1</sup>
External temperature	$T_{\text{ext}}$	100	°C
Initial temperature	$T_0$	800	°C
Ingot length		0.1	m
FE mesh size		$10^{-3}$	m
Time step	$\Delta t$	0.1	s
Convergence criterion (residual)	$\varepsilon_R$	$10^{-6}$	—
Convergence criterion (temperature)	$\varepsilon_T$	$10^{-2}$	K

**Table 3.1** – Parameters for the pure diffusion test case with alloy Al – 7 wt.% Si presented in **FIGURE REF**

the intrinsic phase enthalpies, that is  $\langle \rho h \rangle^s = \langle \rho C_p \rangle T$  and  $\langle \rho h \rangle^l = \langle \rho C_p \rangle T + \rho L$ , where  $\langle \rho C_p \rangle$  is the heat capacity per unit volume and  $\rho L$  is the latent heat per unit volume. Values for  $\langle \rho C_p \rangle$  and  $\rho L$ , as well as for the thermal conductivities,  $\kappa = \langle \kappa^l \rangle = \langle \kappa^s \rangle$ , are taken constant. Moreover, a Gulliver Scheil approximation is used to compute a single temperature – fraction of solid relationship in the absence of macrosegregation. This is done assuming a linear binary phase diagram and thus requires using the properties listed in **Table 3.1**, i.e. the segregation coefficient,  $k$ , the liquidus slope,  $m_L$ , the liquidus temperature,  $T_L$ , and the eutectic temperature,  $T_E$ . **Figure REF** shows the comparison with the Hsolver and Tsolver. The results are found superimposed to the front tracking solution, thus giving validation of the implementation as well as the iterative schemes presented above to solve the energy conservation.

#### 3.3.2 Convection-diffusion with macrosegregation

Conservation equations in **Table 2** are for mass, momentum and chemical species. As for energy, they are presented after the volume averaging technique has been applied [Ni et al. 1991; Dantzig et al. 2009]. 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. This assumption also yields some other consequences on the mass balance and the liquid momentum conservation. In the latter, a Darcy term is added to take into account the dissipative interfacial stress in the porous-like mushy zone. Its main parameter is the permeability of the mushy zone,  $K$ . It is considered isotropic, hence reducing to a scalar which is

given by the Carman-Kozeny relation, based on the secondary dendrite arm spacing  $\lambda_2 : K = \frac{g^l^3 \lambda_2^2}{180(1-g^l)^2}$ . The liquid density being taken constant, its spatial variations as a function of temperature and average composition are still needed to compute thermosolutal convection forces. For that purpose, the Boussinesq approximation  $\langle \rho \rangle^l = \rho_{\text{ref}} \left( 1 - \beta_T (T - T_{\text{ref}}) - \beta_{\langle w \rangle^l} (\langle w \rangle^l - w_{\text{ref}}^l) \right)$  is used, considering the thermal  $\beta_T$  and solutal  $\beta_{\langle w \rangle^l}$  expansion coefficients and a reference density,  $\rho_{\text{ref}}$ , defined at a reference temperature  $T_{\text{ref}}$  and reference composition  $w_{\text{ref}}^l$ . Values for the references are taken at the liquidus temperature and the nominal composition of the alloy,  $\langle w \rangle_0$  [Carozzani et al. 2013]. More details about the FE formulation can be found in the Ph.D work of Rivaux [2011] and Carozzani [2012]. It should be noted that the macroscopic solute diffusion coefficient in the solid phase is neglected in **REF Eq. 15c**.

in this table I use directly the simplified equations, but this was done only for the article, now I have to go from the full conservation equations and state the hypothesis and methods that i used to reach this simplified form

$$\nabla \cdot (g^l \vec{v}^l) = 0 \quad (3.4a)$$

$$\frac{\partial}{\partial t} (g^l \rho_{\text{ref}} \vec{v}^l) + \vec{\nabla} \cdot (g^l \rho_{\text{ref}} \vec{v}^l \times \vec{v}^l) - \vec{\nabla} \cdot (\langle S \rangle^l - g^l \nabla p^l + \mu^l g^l K^{-1} \vec{v}^l - g^l \rho^l \vec{g}) = 0 \quad (3.4b)$$

$$\frac{\partial}{\partial t} (\langle w_i \rangle) + (g^l \vec{v}^l) \cdot \vec{\nabla} w_i^l + \nabla \cdot (g^l D^l \vec{\nabla} w_i^l) = 0 \quad (3.4c)$$

**Table 3.2** – Averaged conservation equations for the conservation of mass (a), momentum (b) and solute mass (c)

Figure SMACS: Computed unidirectional heat diffusion during solidification of an Al – 7 wt.% Si alloy using (orange) the enthalpy method and (black) the temperature method, comparison being made for (left) cooling curves and (right) time history of the liquid fraction. Each curve corresponds to a position along the sample, from 0 cm (cooling side) to 10 cm (insulated side), with 2 cm spacing between the positions.

The Tsolver's ability to be coupled with various physical phenomena like macrosegregation and fluid flow in porous medium is displayed in this test case. It consists of a solidification benchmark where a 10 cm width  $\times$  6 cm height  $\times$  1 cm thick cavity containing a Sn – 3 wt.% Pb melt 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 et al. [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 discretization, material properties and boundary conditions can be found in the latter reference.

I could develop more here giving additional details

### **Chapter 3. Energy balance with thermodynamic tabulations**

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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 **REF Figure 4** show a satisfactory agreement with the experimental temperature measurements recorded at mid heights of the cavity and uniformly distributed along its width [Carozzani et al. 2013]. In fact, simulation results with the Tsolver and the Hsolver were found to be almost superimposed, as in **REF Figure 4**. Regarding the computation, the Tsolver resolution proves to be faster than the Hsolver used by Carozzani et al. [2013]: a process time of 7000s 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%.

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