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

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Acronym	Standing for
ALE	Arbitrary Lagrangian-Eulerian
BTR	Brittleness temperature range
CAFD	Cellular Automata Finite Difference
CAFE	Cellular Automata Finite Element
CBB	Circumventing Babuška-Brezzi
CCEMLCC	Chill Cooling for the Electro-Magnetic Levitator in relation with Continuous Casting of steel
CEMEF	Center for Material Forming
CSF	Continuum Surface Force
DLR	Deutsches Zentrum für Luft- und Raumfahrt
EML	Electromagnetic levitation
ESA	European Space Agency
FEM	Finite Element Method
GMAW	Gas Metal Arc Welding
ISS	International Space Station
IWT	Institut für Werkstofftechnik
LHS	Left Hand Side
LSM	Level set method
MAC	Marker-and-cell
MIN	Mini-element
PF	Phase field
RHS	Right Hand Side
RUB	Ruhr Universität Bochum
RVE	Representative Elementary Volume
SBB	Satisfying Babuška-Brezzi
SCPG	Shock Capturing Petrov-Galerkin
SUPG	Streamline Upwind Petrov-Galerkin
VMS	Variational MultiScale
VOF	Volume Of Fluid

Contents

Chapter 1

General Introduction

Casting is one the earliest production techniques created by human civilisation since the Bronze Age, dated to circa 3000 BC. From ancient swords to nowadays toys, the need for alloys has never decreased. The key phenomenon behind this technique is solidification, or the transformation of matter from liquid to solid state. With this phase change, many phenomena, not visible to the naked eye, take place with a very complex interaction, in order to form a solid. However, the combination of thermal phenomena, like release of latent heat of solidification, and chemical phenomena like redistribution of chemical element atoms (also known as solute) with other atoms, often lead to *segregation*. The origins of this word from Latin, *segregatus*, has a social meaning of "separating a group from the dominant majority", while in metallurgy it means a non uniform distribution of chemical species. Depending on the scale, we may speak of *microsegregation* when the heterogeneity spans some few hundred microns, whereas the term *macrosegregation* refers to a much coarser length scale, ranging from some millimetres to some meters ! The final solid structure has intrinsic thermophysical and thermomechanical properties directly influenced by the segregation pattern. In casting processes, such as continuous casting (fig. 1.7) and ingot casting, it is crucial to apprehend these intricate phenomena leading to macrosegregation as well as the influence on the final product, at each step of the process.

In this introductory chapter, we give a quick overview of solidification phenomena and microstructure, then present the factors which promote segregation, on both microscopic and macroscopic scales. Aside from macrosegregation, others defects are also briefly presented. In a continuous casting process (fig. 1.1a), the partially solidified slab is carried through a series of rolls that exert contact forces to straighten it. As the mushy part of a slab enters through these rolls, interdendritic liquid is expelled backwards, i.e. in regions with lower solid fraction. Since the slab edges solidify earlier than the centre, the enriched liquid accumulates halfway in thickness, forming a centreline macrosegregation as shown in fig. 1.2. Other types of segregates (channels, A-segregates ...) can also be found but remain more specific to ingot casting. A variety

Chapter 1. General Introduction

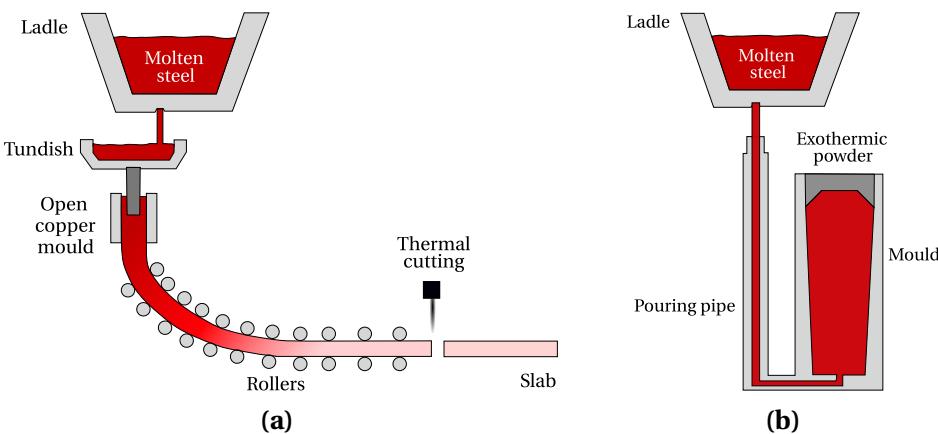


Fig. 1.1 – Main steelmaking processes by (a) continuous casting or (b) ingot casting

of segregation patterns can be encountered while casting heavy ingots, as in [fig. 1.1b](#):

- the lower part is characterized by a negative segregation cone promoted by the sedimentation of equiaxed crystals and settling of dendrite fragments,
- 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,
- a positive "hot-top" macrosegregation in the upper zone where the last rich liquid solidifies, caused by solidification shrinkage 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".

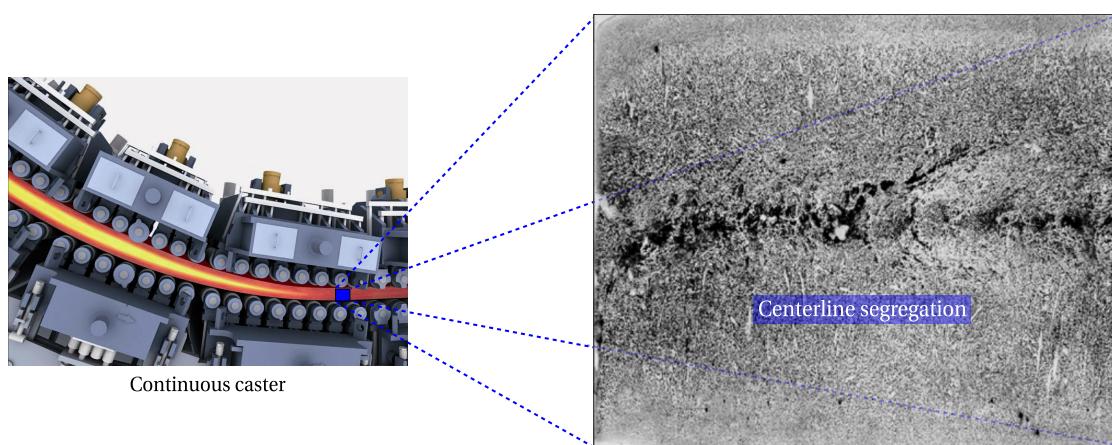


Fig. 1.2 – Zoom on a sulphur print of a continuously cast high carbon steel billet at a longitudinal section, showing high positive centreline segregation [[Choudhary et al. 2007](#)].

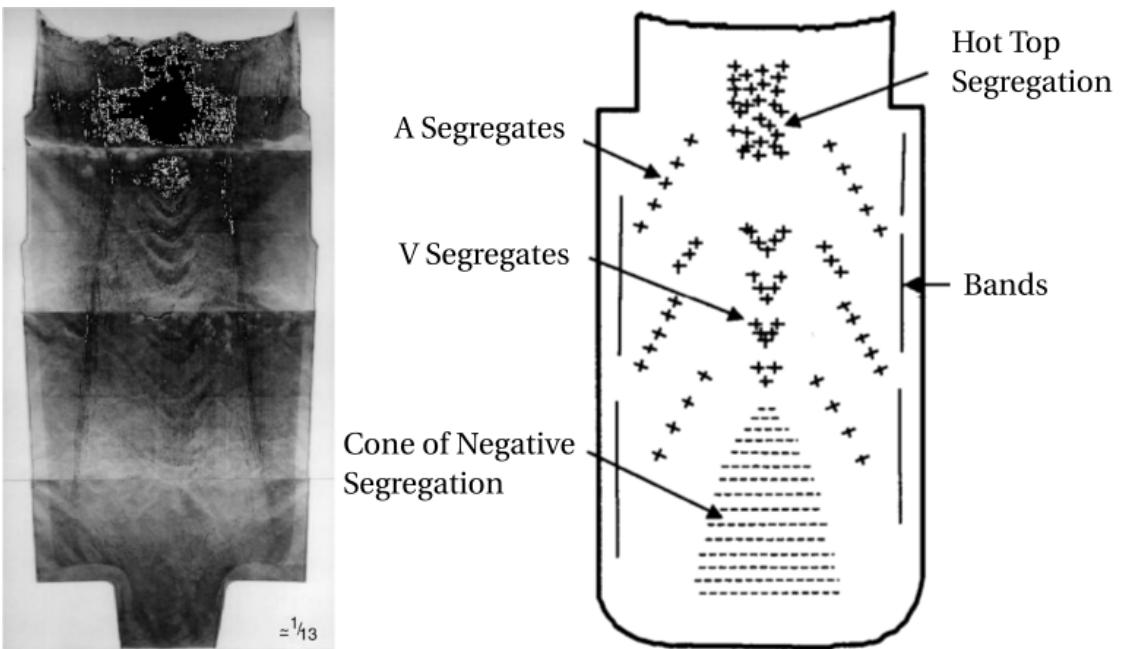


Fig. 1.3 – Sulphur print (left) of a 65-ton steel ingot [Lesoult 2005] showing various patterns (right) of macrosegregation [Flemings 1974]

1.1 Solidification notions

1.1.1 Solute partitioning

The simplest definition of this phenomenon is an uneven distribution of solute between the liquid and the 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 at equilibrium. The segregation (or partition) coefficient k determines the extent of solute rejection into the liquid during solidification:

$$k = \frac{w^{s*}}{w^{l*}} \quad (1.1)$$

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 at the liquidus temperature, T , 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 1.4 illustrates a typical binary phase diagram where the real solidus and liquidus are represented by solid lines, while the possible 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 (i.e. solute diffusivity) 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 is seen by a non-uniform composition through the cast product on a microscopic scale, better known as microsegregation. This phenomenon is essential in a casting process inasmuch as it affects the microstructure and grain morphology, hence the final mechanical properties of the alloy.

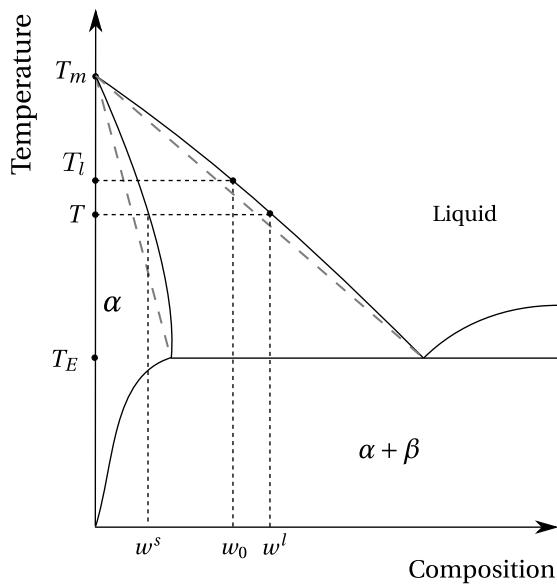


Fig. 1.4 – Typical eutectic phase diagram of a binary alloy showing the real solidus and liquidus at full equilibrium, with the corresponding linear approximations (grey dashed lines). T_m and T_E are respectively the melting point of the solvent and the eutectic temperature.

1.1.2 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 solid-liquid interface velocity, \vec{v}^* , which in turn affects the initial microstructure. Although it may be not easy to control them, their crucial role in solidification is well established.

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

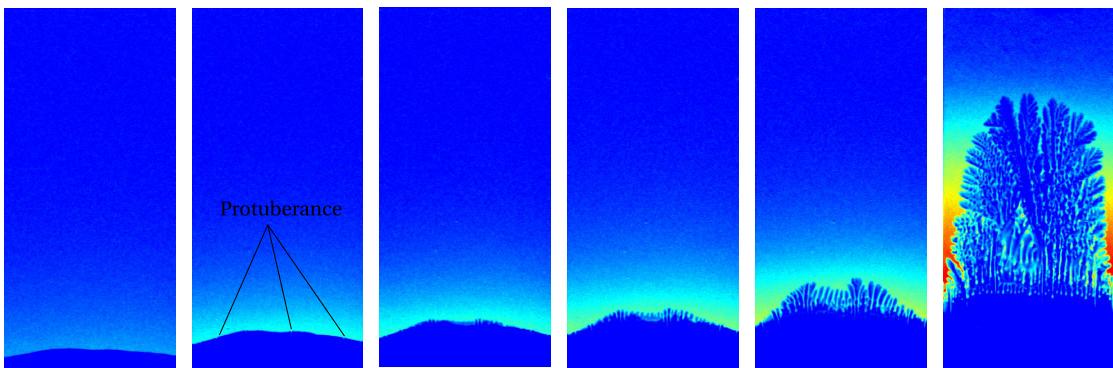


Fig. 1.5 – Time evolution of a solidifying Al-4 wt.% Cu sample, showing interface destabilisation and subsequent dendritic solidification [Buffet et al. 2010]. The liquid far from the interface and having a blue color is at nominal composition, while the one near the dendritic structure with the yellow and red colors, is richer in solute.

solidification). As such, the rest of the interface catches up, maintaining the planar geometry. In another scenario more representative of a real casting, 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, as shown in the time frames of fig. 1.5. 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 the solid-liquid interface adopts a tree-like shape, we speak of *dendritic growth*. Near the chill surface, dendrites are columnar, with a favourable growth in the $\langle 100 \rangle$ direction for alloys with cubic lattices, but different orientations are also reported in the literature [see Dantzig et al. 2009, p. 289]. Far from mould walls, a similar dendritic growth phenomenon occurs where temperature is uniform, but with an equiaxed morphology. Figure 1.6 shows both columnar and equiaxed morphologies. Columnar dendrites are characterised by a primary spacing, λ_1 , between the main trunks, and a secondary spacing, λ_2 , for the arms that are perpendicular to the trunks. It should be noted that λ_1 and λ_2 , together with the grain size, are three important microstructural parameters in the as-cast microstructure [Easton et al. 2011]. Further branching may occur but will not be discussed here.

1.1.3 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. In the intermediate state, the liquid flow in and out of the mushy zone through the dendrites is a key phenomenon from a rheological perspective. The

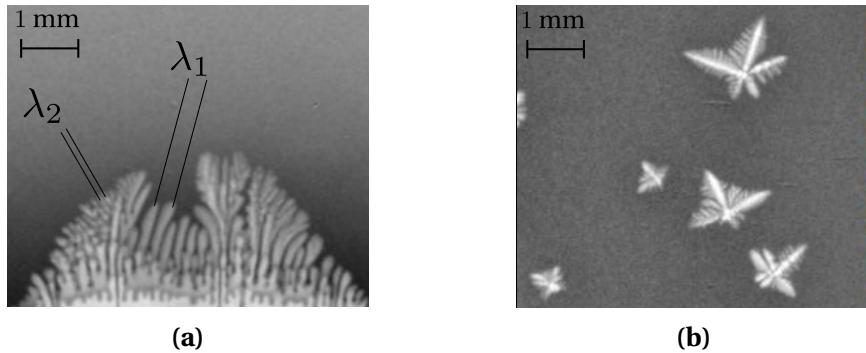


Fig. 1.6 – In situ observation by X-ray radiography of the (a) columnar microstructure for Al-4 wt.% Cu alloy [Buffet et al. 2010] and (b) equiaxed microstructure during solidification of Al-10 wt.% Cu alloy [Bogno et al. 2013].

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} (assuming a the solid phase is fixed), through the following equation [[Rappaz et al. 2003](#)]:

$$\vec{v} = \frac{\mathbb{K}}{\mu^l} \vec{\nabla}p \quad (1.2)$$

where μ^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 λ_1 like Blake-Kozeny [[Ramirez et al. 2003](#)], or the secondary dendrite arm spacing λ_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 λ_1 and λ_2 .

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

$$\mathbb{K} = \frac{\lambda_2^2 g^l}{180 (1 - g^l)^2} \quad (1.3)$$

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 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 heat treatments, 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 an irreversible defect. Failure to prevent it, may lead to a substantial decline in the alloy's mechanical behaviour and its serviceability. Experimental investigations of macrosegregation in steels were the subject of numerous studies in the past, like the work of Suzuki et al. [1973], Shah et al. [1989], and Lesoult [2005]. They were mainly motivated by industrial research that aimed at reducing defective production caused by macrosegregation. However, steel was not the only alloy that was investigated, probably because of its high melting point. Instead, many studies in the past relied on other metallic alloys as a replacement recourse, for their low melting points, also for their well-known thermophysical and mechanical properties, such as aluminium-based alloys (aluminium-copper or aluminium-silicon) [Lesoult et al. 2001; Ferreira et al. 2004; Ferreira et al. 2009] and lead-tin/tin-lead alloys[Hebditch et al. 1974; Prescott et al. 1994; Hachani et al. 2012]. Investigating macrosegregation was not only limited to metallic alloys, but also to organic compounds like the well-known sodium chloride [Wanqi et al. 1989]. Four main factors can (simultaneously) cause fluid flow leading to macrosegregation:

1.2.1 Liquid thermosolutal convection

During solidification, the liquid density undergoes changes due to temperature gradients. Generally for steels, an increasing temperature results in a lighter liquid phase and vice-versa. These variations create a driving force of thermal convection in the melt, during which chemical species are redistributed by convective transport. While a uniform composition is usually maintained throughout the liquid phase, solute gradients may appear in zones where thermal convection currents are not sufficient to homogenise the liquid solution. Similarly to thermal gradient, a solute gradient is behind liquid density variations. Nevertheless, the relationship between the liquid's composition and its density is a characteristic of the alloy. While for some alloys, a positive solute gradient creates a positive density gradient, for others the opposite is true, due to a heavier solute. Whether convection is solute or temperature dominated, convection currents are important in the formation of macrosegregation, especially that phase densities vary considerably upon cooling the superheated alloy to room temperature.

1.2.2 Solidification shrinkage

Solid alloys generally have a greater density than the liquid phase ($\rho^s > \rho^l$), thus occupy less volume, with the exception of silicon where the opposite is true. 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*: while one would naturally expect negative macrosegregation near the contact with the chill, shrinkage-induced flow promotes the opposite phenomenon, by bringing solute-richer liquid towards the solidifying areas, thus raising their solute content, and resulting in a positively segregated solid. In contrast to liquid convection, shrinkage flow may cause macrosegregation even without gravity.

1.2.3 Movement of equiaxed grains

Globular and equiaxed dendritic grains nucleate and grow in the liquid bulk where thermal gradients are weak, or in the presence of inoculants. Consequently, they are transported by the flow, thus floating or sedimenting depending on their density [Beckermann 2002]. During their movement, the nearby liquid is driven away before the grains finally settle in the end of solidification. Since solute-rich liquid is expelled, a negative macrosegregation zone (often referred to as *negative segregation cone* in cast ingots) is detected in their final position.

1.2.4 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.

1.3 Other defects

Apart from macrosegregation, other defects are also encountered in industrial processes. In the casting industry, defects like pores and cracks can be disastrous causing the cast product to be rejected. If possible, the product is reheated, remelted and then cast again. Otherwise it is scrapped. From an economic point view, these operations are very expensive.

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

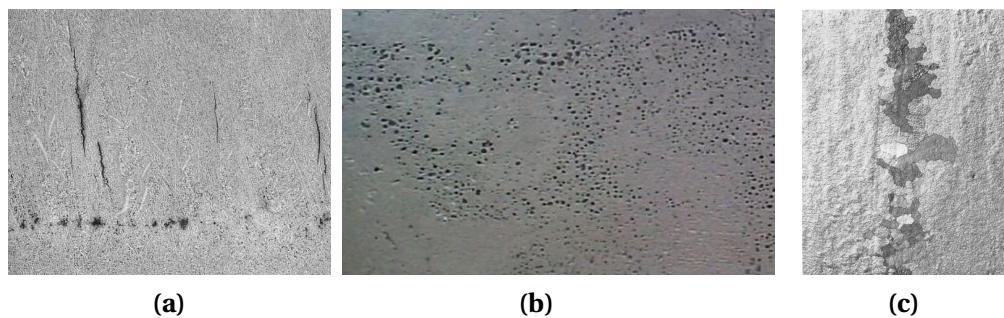


Fig. 1.7 – Three instances of solidification-related defects found in cast products: (a) internal cracking [Arcelormittal] (b) surface cavities [Carlson et al. 2002] and (c) freckle in the form of a chain of equiaxed crystals [Giamei et al. 1970].

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%, where the liquid phase starts to form a discontinuous film. Many factors can initiate 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 and then propagates in the casting.

Porosity

Porosity is a void defect formed inside the casting or at the outer surface. It may be attributed to two different factors. Firstly, we speak of *shrinkage porosity*, when a void forms as a result of density differences between the liquid and its surrounding dendritic solid network, the latter being generally denser than the former. The second factor is the presence of dissolved gaseous phases in the melt. 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 trapped in grooves at the moulds surface. Providing sufficient cooling and pressure drop in the liquid, the latter becomes supersaturated. The nucleation of a gaseous phase is then triggered.

Freckles or segregated channels

The origin of this defect is a combined effect of microsegregation and buoyancy forces. Upon solidification, solid forms while exchanging solutes with the liquid due to partitioning. For a solute species that preferentially segregates into the liquid (partition coefficient less than unity) and locally reduce the liquid density, a solutal driving force is created inside the mushy zone, generating convection currents, with "plume" shapes as often reported in the literature [Sarazin et al. 1992; Schneider et al. 1997; Shevchenko et al. 2013]. Temperature gradient is often an additional force of convection as the liquid density depends also temperature-dependent, the resulting driving

force is then qualified as "thermosolutal".

1.4 Industrial Worries

Steel production has continuously increased over the years to meet the industrial needs. [Figure 1.8](#) 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.

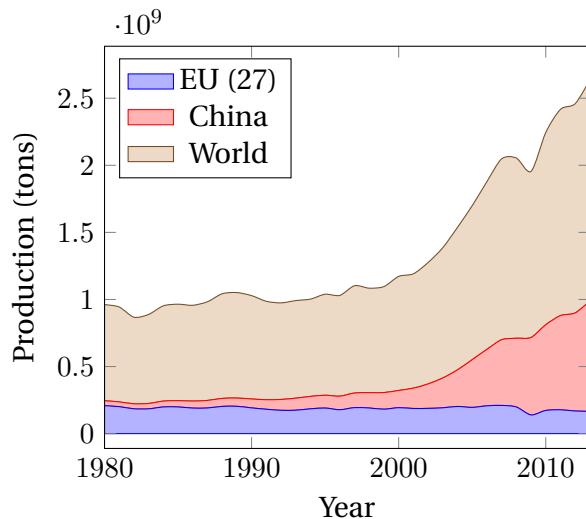


Fig. 1.8 – Evolution curves of crude steel worldwide production from 1980 to 2013 [[WSA 2014](#)].

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. 1.7](#), 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 by handling:

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

- microstructure and phases
- finite solute diffusion in solid phases
- real alloy properties (not just constant thermophysical/thermomechanical properties)

1.5 Project context and objectives

1.5.1 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 in 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 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*. Here is a brief summary of each partner's commitment:

Academic partners

- Center for Material Forming (CEMEF) - France: numerical modelling of microgravity chill cooling experiments
- Deutsches Zentrum für Luft- und Raumfahrt (DLR or German Aerospace Centre) and Ruhr Universität Bochum (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 and spray deposition of the D2 tool steel
- University of Bremen - Institut für Werkstofftechnik (IWT) institute - Germany: study of D2 tool steel melt solidification in atomization processing

Industrial partners

- ARCELORMITTAL (France): elaboration of a series of steel grades used in microgravity and ground-based studies
- METSO Minerals Inc. (Finland): material production with D2 tool steel for spray forming

- TRANSVALOR (France): development and marketing of the casting simulation software *Thercast*®

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]. The experimental work by DLR considered various facilities and environments to set a droplet of molten alloy in levitation: EML (fig. 1.9) for ground-based experiments, microgravity during parabolic flight or sounding rockets and last, microgravity condition on-board the International Space Station (ISS).



Fig. 1.9 – Electromagnetic levitation [DLR 2014].

1.5.2 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. At CEMEF, this scope has been adopted in previous studies 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 database mapping, ii) the liquid momentum conservation with thermosolutal convection and solidification shrinkage as driving forces, iii) solute mass conservation with solidification shrinkage (to predict inverse segregation) and iv) solidification paths at full equilibrium for multicomponent alloys. 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 the 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. The model couples in a weak fashion, all four conservation equations presented in fig. 1.10, showing on the one hand, that microsegregation is an essential common link between these equations, while on the

other hand, the level set interacts with conservations equations by giving the interface position.

Numerical tools

The current thesis developments are done using C++ language as a part of the in-house code, known as *CimLib* [Mesri et al. 2009]. It's a finite element library with an object-oriented hierarchy consisting of global finite element solvers for physical differential equations, also point-wise and element-wise solvers for preprocessing and postprocessing the finite element solution. It is also highly parallelised with MPICH2, which makes it possible to handle large meshes with heavy operations like mesh partitioning and adaptation. Side utilities for plotting and analysis are also developed, using python language.

Content outline

This manuscript consists of 5 chapters. In **chapter 2**, a short review introduces the state-of-art in the literature of macroscopic modelling of solidification with transport of energy, species, mass and momentum. **Chapter 3** provides details of the energy conservation equation then introduces a new temperature-based solver compatible with thermodynamic databases **Chapter 4** focuses on the mass and momentum conservation equations and their derivation in the context of a single metallic domain (i.e. without the level set method) where the solid is assumed a fixed and rigid body. As an application, this chapter shows a breakthrough comparison between purely macroscopic and mesoscopic-macroscopic approaches in predicting the hydrodynamic instabilities that lead to channel segregation. The derived energy, mass and momentum equations for the monodomain model are revisited in **chapter 5** in the context of the level set method to predict solidification shrinkage. The influence of shrinkage on macrosegregation is then studied in two applications.

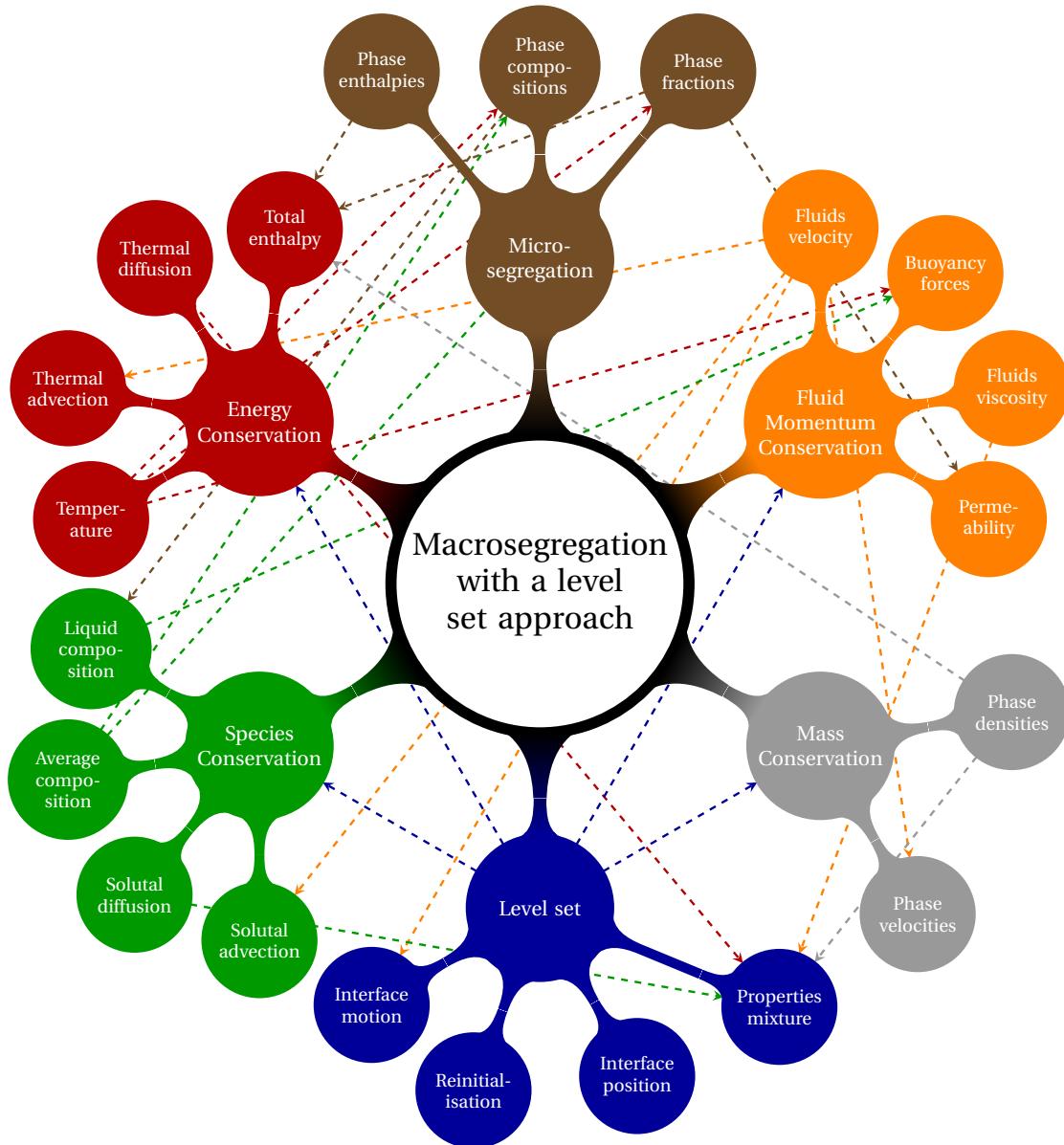


Fig. 1.10 – A graphical representation of the main elements of the numerical approach, for a macrosegregation model with the level set methodology, when no solid deformation or movement are considered. The dashed lines represent the possible interaction between the components.

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