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Grain growth under the influence of Smith-Zener pinning: multiscale modeling and application to nickel-based superalloys

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# Introduction

The first successful flight with a powered and controlled airplane was made by the Wright brothers in 1903 [1, 2, 3]. They used a wooden frame and a small gasoline engine integrated into a chain drive mechanism. This first biplane was small and lightweight, flying at a low speed compared to the present airplanes.

With world war I, new aircraft appeared, especially for military purposes, which boosted aviation. Larger airplanes were created and with them, the use of various engines.

One of the most critical developments in aviation was to implement a metallic structure [4, 5] allowing the creation of larger aircraft for commercial flights. These new airplanes presented changes in their forms and in the materials used to build them.

New technical advances were made to reach more power, durability, and efficiency. For this reason, the initial four-cylinder in-line engine used by the Wright brothers was replaced by the V-type piston engine and the radial and rotary piston engines. Then World War II came and with it, the development of new engines for military aircraft, where very high speeds were required. Thus was born the idea of coupling a turbine engine with a propeller, and the first airplanes with gas turbines were designed.

The turbojet (jet engines) [6, 7, 8] is the simplest of the gas turbine engines and provides higher speed than previous engines. The maximum speed of a military aircraft equipped with them was at least 100 miles per hour faster than that of the piston-powered aircraft. Since the 1950s, there have been fewer changes in aircraft design. Jet engines have remained but have been improved in power and efficiency thanks to new materials used in the airline industry.

The engine is one of the most complex parts of an aircraft and determines fuel efficiency and ultimately economic and environmental costs. So, optimizing its operation and performance has always been vital but is even more nowadays with the necessary drastic limitation of greenhouse gas emissions in order to reach the ACARE (Advisory Council for Aeronautical Research in Europe) objectives. The temperature of some of these motors can go up to  $2000^{\circ}C$ . Then, nowadays, the challenge remains to create or find new materials capable of resisting higher temperatures while preserving their mechanical properties to improve engine fuel consumption and efficiency.

Heat-resistant superalloys such as nickel alloys, titanium alloys, or non-metallic

materials such as ceramics are now used to respond to the demanding requirements of the aerospace industry. These materials have extraordinary thermomechanical properties but are more difficult to machine than traditional alloys. However, new material-forming processes have also been proposed in addition to the traditional forging process, for example, powder metallurgy, which has opened up the possibility of developing and producing new superalloy materials.

To further improve the efficiency and performance of the engine, it is essential to understand the microstructural evolutions and the different mechanisms occurring during metal forming processes to increase properties such as mechanical strength, fatigue limit, crack resistance, and corrosion resistance.

The nickel base superalloys contain different kinds of  $\gamma'$  particles that can be differentiated according to their size and function [9, 10, 11]. One can find the primary precipitates, i.e. the biggest ones (few microns); they are used to limit grain growth (GG). We also find the secondary and tertiary precipitates, which are smaller (a few to a few tens of nanometers), and control the mechanical behavior. After forging operations, polycrystalline  $\gamma/\gamma'$  superalloys are submitted to partial solution treatments below the  $T_{solvus}$ , where primary  $\gamma'$  precipitates remain to avoid excessive GG. In contrast, the fine hardening precipitates are dissolved to be formed again with optimized size distribution during the subsequent controlled cooling.

In this work, we will concentrate on the study of the population of primary particles in  $\gamma/\gamma'$  alloys to understand their behavior, and to analyze and quantify their impact on microstructural evolution. This analysis will be carried out by combining experimental and numerical approaches to propose a global understanding of the behavior of these particles and their impact on the matrix grain size.

As already established, in many polycrystalline materials, the grain size is controlled by pinning mechanisms by second phase particles (SPPs) that hinder the grain boundaries (GBs). The Smith-Zener pinning model (SZP) describes the physical interaction between SPPs and GBs [12, 13, 14, 15]. Both can evolve by applying a heat treatment to the material. As the industrial forging processes involve hot deformation steps close to the solvus temperature, it is of great importance to characterize both the mechanisms and kinetics by which SPP evolve, since they impact the final microstructure, especially, the grain size (as illustrated in Figure 1).

The modeling of grain growth (GG) and recrystallization (ReX) in polycrystalline materials during thermomechanical treatments has been the focus of numerous studies in the recent decades. Mean-field [16, 17, 18, 19, 20] and full-field models have been developed to treat this subject. The former predict average quantities (for instance, the mean grain size) or distributions of these quantities and are based on physical or phenomenological laws. These types of models have been largely studied and improved, so they can offer a good compromise in terms of accuracy and computational times. However, they are unable to predict poly-



Figure 1: Evolution of the  $\gamma'$  particle surface fraction and grain size of the AD730 superalloy under a thermal treatment of one hour at different temperatures [Safran].

crystal topology and to account for heterogeneous microstructures. Therefore, it is often necessary to consider the full-field approach to be able to reproduce heterogeneous properties and to consider concomitant mechanisms like the ones observed in the real manufacturing processes.

Numerical simulations of microstructural changes during hot metal forming processes are the aim of the DIGIMU<sup>®</sup> software developed at CEMEF and the Transvalor company in collaboration with several industrial partners: Safran, Aubert & Duval, Framatome, CEA, Timet, ArcelorMittal, Constellium and Ascometal. DIGIMU<sup>®</sup> software is based on a Finite Element (FE) approach combined with Level-Set (LS) description. An advantage of using a FE formulation lies in the capability to model large deformation thanks to meshing/remeshing algorithms; second, the LS method provides flexibility and robustness to reproduce numerous metallurgical mechanisms.

Today DIGIMU<sup>®</sup> can reproduce 2D and 3D simulations of microstructural evolutions, such as grain growth (GG), dynamic (DRX), post-dynamic (PDRX), and Smith-Zener pinning mechanism (SZP) under the influence of SPPs.

However, for the SZP phenomenon, SPPs are considered inert and represented as static holes in the simulation domain. It is possible to introduce a spherical population of SPPs and also to generate particles with the actual morphology extracted from Scanning Electron Microscopy (SEM) micrographs as illustrated in Figure 2. However, this approach has some limitations, mainly because SPPs are described as static holes with fine FE mesh at the SPP interfaces. Indeed, this strategy drastically increases the simulation time, especially in 3D, due to the remeshing operations around the particles. Moreover, considering the SPP behavior and their possible evolution during thermomechanical treatments is at yet, not possible.

Our main objective is to simulate GG in LS framework considering a realistic evolution of the SPPs in the nickel base superalloys to reproduce the real impact of precipitates on the microstructural evolution, to predict the final mean grain size and to enrich and extend the applications of DIGIMU<sup>®</sup> using all the advantages of the LS method.

More precisely, the grain and precipitates evolution for treatments close to the  $\gamma'$  phase solvus temperature will be established. Then, full-field simulations will be performed and the numerical results will be compared to the experimental data.



Figure 2: Particle description in DIGIMU<sup>®</sup> software: a. Monodisperse circular particles. b. Multidisperse circular particles. c. Real particle morphology obtained from a SEM micrograph in 2D section of the AD730 alloy. The particles are described as holes in the domain with a refined mesh around the particles to well describe the interfaces.

The evolution of the  $\gamma'$  precipitates as a function of temperature will be determined by doing experiments on two  $\gamma/\gamma'$  nickel-based superalloys, the N19 and AD730 alloys. The N19 manufacturing process is based on a powder metallurgical (PM) route, limiting the problems of chemical segregation. It is then possible to obtain a more homogeneous microstructure than the one obtained by conventional cast-and-wrough route. On the other hand, the cast-and-wrough AD730 has a more complex distribution of  $\gamma'$  particles than the N19 material, associated with heterogeneities in its microstructure. In this way, the impact of  $\gamma'$  precipitates will be studied for both materials, one with a homogeneous and the other with a heterogeneous microstructure that are related to their manufacturing processes.

This study will allow us to identify the various mechanisms (and related kinetics) occurring when particles evolve, to model them and to identify the relevant model parameters from experiments designed on purpose. Based on, a full-field simulation framework will be developed to describe the precipitate and grain evolutions observed experimentally.

Layout of the thesis A part of the work presented in this manuscript has been published in [21, 22]. The manuscript is organized as follows.

Chapter 1 presents the basis of GG modeling at the mesoscopic scale, as well as the description of the SZP mechanism and the other main mechanisms that precipitates may experience during heat treatment.

Chapter 2 offers a detailed description of the existing *full-field* methodologies used to reproduce microstructural evolutions, their advantages, weaknesses, and especially how they introduce and take into account the presence of SPPs in the simulation domain.

Chapter 3 introduces the new methodology proposed in this PhD to consider the evolution of particles in the LS numerical framework and its validation through academic and realistic polycrystalline configurations in 2D and 3D.

In Chapter 4, the description and characterization of the materials used in this work are presented, together with the manufacturing process of the different materials and the comparison of the two studied material microstructures.

Chapter 5 presents a new methodology to obtain the surface particle evolution as a function of temperature and time for different  $\gamma/\gamma'$  nickel base superalloys. This relationship was established using *Thermo-Calc* simulations together with a Johnson-Mehl-Avrami-Kolmogorov (JMAK) model. Thus, the implementation of a phenomenological model was used to describe the interface particles velocity. This velocity was then implemented in the full-field simulations to reproduce the evolution of the particles. The experimental data is also detailed.

Chapter 6 summarizes the main achievements of this PhD work and some perspectives for future work and potential applications considering the evolution of particles.

# Oral and written communications

These works have lead or contributed to the following written and oral communications.

# Articles

• Karen Alvarado, Sebastian Florez, Baptiste Flipon, Nathalie Bozzolo and Marc Bernacki, A level set approach to simulate grain growth with an evolving population of second phase particles, modeling and Simulation in Materials Science and Engineering 29 (3) (2021) 035009.

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# Presentations

- Karen Alvarado, Nathalie Bozzolo and Marc Bernacki. Influence of Zener pinning phenomena on the grain growth behavior: multiscale approach and application to nickel-base superalloys. La Métallurgie, Quel Avenir!, April 2019, Nancy, France.
- Karen Alvarado, Sebastian Florez, Baptiste Flipon, Nathalie Bozzolo and Marc Bernacki. Influence of smith-zener pinning on grain growth when second phase particles evolve: multiscale approach and application to nickelbase superalloys. 14th World Congress in Computational Mechanics (WCCM), January 2021, virtual conference.

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# Chapter 1 Literature Review

This chapter presents an overview of the  $\gamma/\gamma'$  superalloys microstructure. The definition of the matrix, particle phases with their specific characteristics and the description of the GBs are presented. Additionally, the equations governing the boundary migration due to capillarity and stored energy are detailed.

The Smith-Zener pinning phenomenon is introduced as it describes the impact of the particle phase in the microstructural evolution. The original hypotheses are presented as well as the different modifications that many authors have implemented in the last decades to dispel some of the initial hypotheses. Thanks to works focused on this topic, it is now possible to consider a more precise and accurate description of the particles, considering the size, morphology, coherency, and position of the particles in the microstructure.

Dissolution and Ostwald ripening of second phase particles are also introduced.

# 1.1 Microstructural definition and evolutions of superalloys materials

The  $\gamma/\gamma'$  nickel base superalloys are polycrystalline materials. These materials are composed of randomly oriented crystallites<sup>1</sup>. Polycrystals are formed when the material rapidly solidifies; then, small crystals begin to grow until contact, creating interfaces between crystals of different crystallographic orientations; these interfaces are known as the grain boundaries (GB). The  $\gamma/\gamma'$  alloys are composed of a  $\gamma$  matrix and  $\gamma'$  particles; their description will be presented below, along with the definition of GB migration.

# 1.1.1 $\gamma/\gamma'$ phases

The  $\gamma$  phase composes the matrix of the alloys. It is a solid solution with a facecentered cubic (FCC) austenitic phase of the alloying elements as presented in Figure 1.1.a. Example of such alloys with this matrix are *AD*730, *Rene*65, *N*19, *PER*72<sup>®</sup>. (See Table 1.1 for the chemical composition of these alloys).

The principal elements of the nickel base superalloys are Co, Cr, Mo, W, Nb, Fe, Ti, Al, where each of them is essential and brings specific characteristics to the material. For instance, chromium is used for oxidation and corrosion resistance; the aluminum and titanium are the principal  $\gamma'$  former, which provides creep resistance and, the molybdenum gives additional strengthening thanks to the carbides particles [23].



Figure 1.1: Crystal structure. a. $\gamma$  phase face-centered cubic (FCC) and b.  $\gamma'$  phase FCC L1<sub>2</sub> structure [24].

the  $\gamma'$  phase is an intermetallic phase with an ordered FCC L1<sub>2</sub> structure based on a  $Ni_3(Ti, Al)$ , where the Nickel atoms are placed at the cube faces, and

 $<sup>^1\</sup>mathrm{A}$  crystallite is a small crystal, and a crystal is a solid where the atoms form a periodic arrangement.

Superalloy	Ni	Cr	Co	Ti	Al	Mo	W	Fe	Nb	Zr	В	С	Hf
$AD730^{TM}$	Bal.	15.7	8.5	3.4	2.25	3.1	2.7	4	1.1	0.03	0.01	0.015	-
René 65	Bal.	16	13	3.7	2.1	4	4	1	0.7	0.05	0.016	0.01	-
N19	Bal.	12.9	11.8	3.8	2.5	4.7	3.2	-	1.6	0.06	0.015	0.022	0.3
PER72 <sup>6</sup>	Bal.	16.06	14.5	5.07	2.57	2.88	1.21	0.138	-	-	0.016	0.017	-

Table 1.1: Chemical compositions of AD730[25], Rene65 [26], N19 [27] and PER72<sup>®</sup> [28] superalloys (wt.%).

either Al or Ti atoms at the cube edges as shown in Figure 1.1.b.

This phase is composed of particles sought in order to make the alloys strong and useful at high temperatures. One can find three types of  $\gamma'$  particles: the primary, secondary, and tertiary  $\gamma'$ . As presented in Figure 1.5, each type has a different size and function in the microstructure behavior.



Figure 1.2: Phases of a  $\gamma/\gamma'$  nickel based superalloy. Images of the N19 superalloy at the initial state.

The primary  $\gamma'$  corresponds to the biggest particles, with an equivalent diameter around 1 to 3 [ $\mu m$ ] (at the initial state); they are formed during the material cooling process through the solvus temperature. These particles are mainly located at the GBs; they are incoherent with the matrix ( $\gamma$  phase), which implies a mismatch of the two lattices across the interface particle/matrix. These particles block the GB during GG, which helps to improve the material properties. They evolve during the material forming process as a function of the temperature. These precipitates can dissolve when the thermal treatments are near the primary  $\gamma'$  solvus temperature and grow or coarse when the material is treated at subsolvus temperatures. The secondary and tertiary  $\gamma'$  have an equivalent diameter of around 50-300 [nm] and 7-30 [nm] respectively; they are coherent with the matrix (cristallographic match at the interface) and are used to strengthen the alloy. These precipitates appear during the slow cooling processes and have lower solvus temperature than the primary  $\gamma'$  particles. These particles also evolve with the temperature. This evolution induces several changes in their morphology; for instance, in particle growth, the particles suffer a sequence of successive morphological changes: from sphere to cube to finally create octocubes [9]. A reverse shape evolution is observed when the material is submitted to a cooling treatment (dissolution).

In these materials, one can find small proportions of different other phases, such as carbides  $(M_X C_Y)$  and borides  $M_3 B_2$ , which tend to appear on the grain boundaries and therefore participates in the pinning of GBs. These particles have an FCC crystal structure, and they increase the rupture strength at high temperatures. These phases have higher dissolution temperatures than the  $\gamma'$ particles, remaining at supersolvus temperatures. Even if these particles can block the grain at the supersolvus domain, their volume fraction is very low and does not significantly impact the global grain evolution. For this reason, they will not be considered in this work. Only the impact of primary  $\gamma'$  particles will be taken into account.

Superalloys phases									
Phase	$\gamma'$	Carbie	Carbides		Oxides				
Thase	Primary	Secondary	Tertiary	$(M_X C$	Y)	$(M_X B_Y)$	Oxicarbides		
Shape	Oval and irregular	Spherical or cuboidal		Coarse, random, globular	Films, globules, platelets	Blocky, half-moon	Spherical		
$Size(D_{spp})$	ize(D <sub>spp</sub> ) 1-3 [μm] 50-300 [nm] 7		7-30 [nm]	1-2 [µ	m]	100-400[nm]	100-200[nm]		
Location	GB	Intragranular	GB or intragranular	GB or intragranular	GB	GB	GB of powder grains		

Table1.2 presents a summary of all the particle phases present in the considered nickel base superalloys with their size, morphology, and location.

Table 1.2: Common particle phases in a  $\gamma/\gamma'$  nickel base superalloy and their characteristics.

## 1.1.2 Grains Boundaries

A grain boundary is the interface between two regions of different crystallographic orientations [29].

A boundary  $\Gamma_{lw}$  between two grains,  $G_l$  and  $G_w$ , is typically defined by five macroscopic degrees of freedom (DOF) that describe its geometry and crystallographic, three of them are determined by the 3 Euler angles (i.e. by the misorientation tensor  $M_{lw}$  as detailed in [29, 30, 31]), defining the rotation of the crystallographic lattice from one side of the boundary to the other. Since a crystal lattice presents multiple symmetries, multiple  $M_{lw}$  tensors can be defined, however, only the tensor presenting the lowest misorientation angle (called disorientation) is relevant. The two remaining DOF correspond to the unitary output normal  $\vec{n}$  of the boundary, more frequently introduced as its *inclination*. These five DOF can be finally defined by the tuple  $(M_{lw}, \vec{n})$  as in [31] for the computation of anisotropic grain boundary properties.

The GBs are usually divided into two groups based on the misorientation angle between the two grains. Those whose misorientation angle is less than 15° are considered as Low-angle grain boundaries (LAGB), and those with a greater angle are defined as High-angle grain boundaries (HAGB); both are described in the following.

#### Low-angle grain boundaries (LAGB)

A low-angle boundary is composed of an array of dislocations [32]. The simplest boundary of this type is known as the tilt boundary, where a misorientation angle associates the grains at each side of the boundary, and the rotation axis is parallel to the boundary plane (see Figure 1.3.a.). Read and Shockley [33] established the energy of this type of boundary. They postulated that the energy of the tilt boundary increases with the degree of misorientation angle until the transition from low to high-angle boundary takes place.



Figure 1.3: Two types of grain boundaries. a. A symmetrical tilt boundary. b. A coincident site lattice ( $\sum 5$ ) formed with two cubic lattices rotated by 36.9° to the < 001 > axis [29]. The purple points are the common sites for both lattices.

### High-angle grain boundaries (HAGB)

The structure of a HAGB is more complex and disordered than the LAGB. These grain boundaries are defined as a function of the misorientation angle and the interface plane. The concept of the coincidence site lattice (CSL) [34] is often used to describe HAGB, where different forms are created with the coincident points of two misoriented lattices (see Figure 1.3.b.).

The  $\sum$  value describes the degree of fit between two crystals in the CSL theory. More precisely the  $\sum$  value corresponds to the inverse of the fraction of coincident sites when two neighboring grains are fictitiously extended beyond their common interface.

Thus, when no particular orientation relationship or fit is presented between two neighboring grains,  $\sum$  is significant, and the boundary is considered random. On the contrary, when there is a good fit between the lattices,  $\sum$  is small.

## 1.1.3 Boundary migration

Dislocations and grain boundaries are considered as crystallographic defects which induce the material to a thermodynamically unstable state. So, when the temperature increases and the defects become mobile, the system will evolve to decrease its global energy. The migration of grain boundary is one of the phenomena capable of reducing the global system energy [29]. It is classically well accepted that at the mesoscopic scale, the GB velocity can be described with the following equation:

$$\overrightarrow{\nu} = MP\overrightarrow{n} \tag{1.1}$$

with M the mobility,  $\vec{n}$  the outward unitary normal vector to the interface and P the sum of the involved driving pressures. During hot metal forming and annealing, GB migration is mainly caused by two driving pressures  $P = p_c + p_e$ , where the capillarity generates  $p_c$  due to the mean local curvature and the interfacial energy, and  $p_e$  is linked to the stored energy through dislocations, they can be approximated as follows:

$$p_c = -\gamma \kappa, \tag{1.2}$$

where  $\gamma$  is the grain boundary energy (material parameter) and  $\kappa = \nabla \cdot \vec{n}$  the local mean curvature (i.e. the curvature in 2D and the sum of the principal curvatures in 3D), and,

$$p_e = \frac{1}{2}Gb^2[\rho],$$
 (1.3)

with G the shear modulus, b the Burgers vector, and  $[\rho]$  the dislocation density jump at the GB.

For this work, the material will be considered isotropic, with M as temperature dependent and  $\gamma$  constant [35]. Of course, it exists full-field formulations, even in LS framework, enabling it to take into account anisotropic GB properties. We can cite heterogeneous formulations [30, 36, 37, 38] or anisotropic ones [39]. However, this topic will not be discussed here.

Then, for an isotropic material, the mobility can be approximated by an Arrhenius law as follows:

$$M = M_0 exp\left(\frac{-Q_m}{RT}\right),\tag{1.4}$$

with  $M_0$  a constant pre-exponential factor,  $Q_m$  the activation energy, R the gas constant, and T the absolute temperature.

Some phenomena can delay the migration of GB, such as the Smith-Zener pinning, where the particle population blocks grain growth; this phenomenon will be detailed below.

# **1.2** Classical equation of Smith-Zener

The Smith-Zener equation, proposed by Zener and published by Smith [12], is an analytical model that relates the average radius of grains  $\overline{R}$  of a given microstructure with the average radius  $\overline{r}_{spp}$  of particles and their volume fraction  $f_{spp}$  [12, 13].

This model is based on the following hypotheses:

- particles are assumed spherical, monodisperse  $(r_{spp} = \overline{r}_{spp})$ , and randomly distributed,
- the surface tension between grains and particles is isotropic ( $\gamma_p^1 = \gamma_p^2$  in Figure 1.4), which is equivalent to consider incoherent particles with the GB in motion,
- each particle exerts the maximum dragging force at the same time,
- the particles do not deform the grain boundaries macroscopically (i.e. r<sub>spp</sub> << R),</li>
- the microstructure is assumed free of stored energy (i.e.  $p_e = 0$ ).

This model gives an equation that predicts the critical or stationary grain radius, which neither lets decrease nor increase because of the balance of driving pressures. This means that we can estimate the final mean grain size but not the evolution of the grain size distribution or the evolution of individual grain.

Furthermore, without being explicitly stated by Smith in their original articles [12], the grain boundary thickness is assumed negligible comparatively to the particle size. However, it can be found in the literature that the Smith-Zener formalism is sometimes used to treat nanoparticles, which seems quite inappropriate regarding the original hypotheses.



Figure 1.4: Illustration of the interaction between a particle and a GB in motion (dashed lines between Grain 1 and Grain 2).  $\vec{n}$  corresponds to the normal to the particle interface, and  $\alpha$  to the angle established by the balance of surface tensions [29].

Considering these hypotheses, the dragging force of a particle on the GB can be calculated as the total length in contact with the GB in 3D ( $2\pi r_{spp}cos\theta$ , i.e. the perimeter of the blue circle Figure 1.4) multiplied by the vertical component of the surface tension ( $\gamma sin(\theta + \alpha)$ ).

$$F = 2\pi \overline{r}_{spp} \gamma cos\theta sin(\theta + \alpha), \tag{1.5}$$

Applying the hypothesis  $\gamma_p^1 \simeq \gamma_p^2$  (i.e  $\alpha \simeq 0$ , incoherent particles with the GB), the maximum pinning force is then obtained for  $\theta = 45^{\circ}$  and verifies:

$$F_{MAX} = \pi \overline{r}_{spp} \gamma, \tag{1.6}$$

The following relationship gives the number of particles per unit volume,

$$n_{spp} = \frac{3f_{spp}}{4\pi\bar{r}_{spp}^3},\tag{1.7}$$

with  $f_{spp}$  and  $\overline{r}_{spp}$  the fraction and radius of the particles. However, not all the particles block the grains; only those interacting with the GBs affect the GB migration. Thus, only the particles within a distance  $\pm \overline{r}_{spp}$  of the boundary are considered, so the number of precipitates per unit surface of GB can be evaluated:

$$n_{GBs} = n_{spp} \cdot 2\overline{r}_{spp} = \frac{3f_{spp}}{2\pi\overline{r}_{spp}^2},\tag{1.8}$$

then, the maximum pinning pressure applied by all the particles on the grain boundaries can be approximated as follows:

$$P_Z = F_{MAX} \cdot n_{GBs} = \frac{3f_{spp}\gamma}{2\overline{r}_{spp}}.$$
(1.9)

On the other hand, the driving pressure for grain growth due to curvature, which is expressed as  $P_G = \gamma \kappa$  can be approximated by considering the mean radius as:

$$P_G = \frac{2\gamma}{\overline{R}}.\tag{1.10}$$

At the steady-state, the two pressures cancel each other out  $(|P_Z| = |P_G|)$ . Then, the limit radius is expressed as follows [14]:

$$\overline{R} = K \frac{\overline{r}_{spp}}{f_{spp}{}^m},\tag{1.11}$$

With K = 4/3 and m = 1. Smith and Zener were the first authors to demonstrate that an increase in the volume fraction (for a constant  $\bar{r}_{spp}$ ) or a decrease in particle size (for a constant  $f_{spp}$ ) produces a decrease in the limit grain size for a given material containing particles. For more details of this demonstration see [12, 13, 14].

#### **1.2.1** Modifications of the Smith-Zener equation

Many modifications of these equations (Smit-Zener pressure and limiting grain size) have been proposed to improve the grain size prediction comparatively to experimental observations.

Indeed, it is well known that the original equation overestimates the limiting grain size [14] due to an overestimation of the driving pressure  $P_G$  or an underestimation of the pinning pressure  $P_Z$ , which rely on numerous hypotheses. These pressures depend on several factors like particle morphology and particle/matrix interaction (coherency). Grain and particle size distributions also have an impact on the actual values of these pressures.

In [29, 14, 40, 41, 42], we can find some of the modifications of the original hypotheses that several authors have made to reach a better approximation of the pressure and impact coming from SPPs in real materials. The work present in [40] introduced the possibility of considering different particle shapes. The effect of particle coherency was treated in [41] and the effect of particle position was studied in [29], where the particles are not random, but preferably in the GBs.

Among these modifications, one concern the extension of the initial SZP framework to the case of coherent particles.

In this context  $\gamma_p^1 \neq \gamma_p^2$ ,  $\sin \alpha = \frac{\gamma_p^2 - \gamma_p^1}{\gamma}$  and the pinning force can be defined as  $F = 2\pi \bar{r}_{spp} \cos\theta \times \gamma \sin(\theta + \alpha) = \pi \bar{r}_{spp} (\sin \alpha + \sin(2\theta + \alpha))$  and the maximal force is then obtained for  $\theta = 45^\circ - \frac{\alpha}{2}$ :

$$F = \pi \overline{r}_{spp} \alpha \left( 1 + \frac{\gamma_p^2 - \gamma_p^1}{\gamma} \right).$$
(1.12)

Assuming that the particle is coherent with the grain 1,  $\gamma_p^2 >> \gamma_p^1$  and  $\frac{\gamma_p^2}{\gamma} \sim 1$ , therefore the maximal force exerted by this coherent particle can be approximated as:

$$F_{MAX} = 2 \times \pi \overline{r}_{spp} \gamma. \tag{1.13}$$

The maximal force can then be two times higher than the one of incoherent particles; this proves the strong influence of initial hypothesis in the original Smith-Zener equations.

Then, the Smith-Zener equation can be generalized as presented in Eq. 1.11, where K and m are fitted parameters that are generally assumed constants. Thus, these values vary depending on the authors and the assumptions made to obtain  $P_Z$  [14, 43, 42, 44].

As established in the previous sections, the particles have a crucial influence on the microstructure evolution; thus, it is of prime importance to understand their behavior. The following section will study some of the different mechanisms acting on SPPs when submitted to a particular thermal treatment.

# **1.3** Particles mechanisms

Different mechanisms can occur concerning particle evolutions. These mechanisms are classically divided into two types: the conservative mechanisms because there is no volume change of the different phases, and the non-conservatives otherwise. This classification will be explained below and details in Table 1.3.

# 1.3.1 Conservative mechanisms

Here we consider mechanisms occurring with a stable volume fraction of the different phases present in the considered physical domain. The conservative mechanisms can be divided into surface and volume diffusion mechanisms as follows:

#### Surface diffusion

In the surface diffusion, the phases are locally reorganized to minimize the system's surface energy. Coalescence, spheroidisation, and splitting are among these mechanisms.

• **Coalescence:** In this process, two or more particles merge during contact to form a larger particle [29, 45], changing the morphology and the distribution of the particles. This means that the coalescence has an impact on the mechanical properties of the superalloy.

This mechanism is a local phenomenon affected by the concentration and the microstructure between the two encountering particles. Many factors

# CHAPTER 1. LITERATURE REVIEW

	$\begin{array}{c} Particle \\ fraction \end{array}$	Category	Mechanism	$Driving \\force$	Assumptions
			Coalescence	Interfacial energy	•Local phenomenon •Lattice misfit
s mechanisms		Surface diffusion	Spheroidisation	Interfacial energy	<ul> <li>Incoherent particles</li> <li>Surface energy minimization</li> </ul>
- conservative.	Constant volume fraction $f = cte$		Agglomeration	Elastic energy	•Particles are rigid •Incoherent particles
ves and non -			Splitting	Interfacial energy and elastic energy	<ul> <li>Coherent particles</li> <li>Morphological changes of particles from a cube to an octect</li> </ul>
Conservati			Ostwald ripening	Interfacial energy	<ul> <li>Spherical particles</li> <li>Particles centers are fixed</li> <li>No interaction among the particles</li> </ul>
	Variable volume fraction $f \neq cte$	Diffusive solid-solid phase transformation "SSPT"	Precipitation and dissolution	Chemical potential gradient	<ul> <li>Phase boundary moves.</li> <li>The material changes its composition or structure.</li> </ul>

Table 1.3: Different mechanisms that occur in the evolution of second phase particles (SPPs) [45, 46, 47, 48, 49].

can affect the coalescence rate, like the lattice misfit in coherent SPP, interface roughness, and diffusion mobility. For materials with a high volume fraction of precipitates, like the AD730 and N19 alloys where impingement of particles is frequent, the probability of coalescence rises.

- Spheroidization: The precipitate tends to take spherical or quasi-spherical shapes to minimize the interfacial energy of the microstructure [29, 50].
- Agglomeration The Precipitate Agglomeration Mechanism (PAM) [48, 49] relies on the existence of an attractive force between nearby particles when there is strain energy present in the matrix; the misfit generates this force at the interface matrix/precipitate.

Knowing that the strain energy can evolve as well as the interface because of the lattice misfit, the system will try to reduce the total strain energy merging adjacent particles.

Then, the agglomeration of the small particles with the large ones takes place. This mechanism can be interpreted as the coalescence's first steps because the interface between the two precipitates will evolve, and the two precipitates will merge.

• Splitting: Driving pressures tend always to minimize the energy, and sometimes the way of reducing the surface energy is to split the precipitate into two or more smaller particles. The splitting process starts from the edges of the initial precipitate advancing towards its center [51, 52].



Figure 1.5: Steps of the splitting mechanism [51].

Particle splitting is related to internal stress in two-phase systems with coherent precipitates due to the lattice mismatch and small interface energy. The balance of these two parameters determinates the morphology and splitting of the precipitate.

#### Volume diffusion

The diffusion is caused by the concentration gradients around the particles, allowing the large particles to grow to the expense of small ones to compensate the concentration gradient.

• Ostwald ripening [46, 47, 53, 9] or coarsening is a diffusion-controlled process present in two phases systems, driven by the reduction of the interfacial energy and thanks to the concentration gradients around the particles. Knowing as the Gibbs Thomson effect, the concentration around small particles (greater curvature) is higher than in the big ones so, the solute atoms move from the interface of small particles to the matrix and from the matrix to the surface of big particles [46]. In the end, the surface energy is reduced by coarsening; the large particle grows at the expense of the smaller ones, which are dissolved.

Ostwald ripening can, in fact, be seen as the natural evolution of a multimodal second phase particle population through the seek for an equilibrium between diffusive solid/solid phase transformation pressure and Gibbs Thomson effect.

### **1.3.2** Non-Conservative mechanisms

#### Diffusive solid state phase transformation

• Precipitation and dissolution of particles in a matrix is a diffusive transformation, characterized by the migration of atoms from the grain boundary to the atomic spaces of the new phase [54].

When precipitation occurs in a solid solution from an initial supersaturated matrix  $\gamma$  phase, the matrix evolves to reach the same crystallographic structure but a different composition of the original supersaturated phase.

Precipitates form as coherent particles with different structure, composition, and lattice misfit at the interface matrix/precipitate. The mechanical behavior of the two-phase material depends on the distribution, size, and shape of the precipitates. Hence it is crucial to understand and predict the influence of the precipitates in the matrix to improve the material properties.

The dissolution and precipitation phenomena play an important role in the control of grain size. The particle precipitation will increase the pressure produce by the Smith-Zener pinning effect, thus stopping the grain growth. On the other hand, the precipitate dissolution will release this effect, letting abnormal grain growth occur in some cases, where few grains grow much faster than the average grain, leading to a bimodal microstructure and eventually to a population of very coarse grains.

# 1.4 Analytical models

## 1.4.1 Coarsening or Ostwald ripening

#### • Model for low volume fractions of particles

Lifshitz, Slyozov and Wagner (LSW) Theory was the first model describing the Ostwald ripening. Nowadays, this model is known as the classical LSW theory [53, 55]. This theory was established under the following hypotheses.

- second phase particles are spherical  $(r = \overline{r}_{spp})$ ,
- particles centers are fixed in space,
- no interaction among the particles is considered (low volume fraction).

This coarsening law can be expressed as [46, 53]:

$$\overline{r}_{spp}^{3}(t) - \overline{r}_{spp}^{3}(0) = K_{LSW}t,$$
 (1.14)

where  $\overline{r}_{spp}(t)$  is the average radius of particles at time t,  $\overline{r}_{spp}(0)$  is the initial particle size and K is given by:

$$K_{LSW} = \frac{8\Psi_{int}\Omega^2 DC_0}{9RT},\tag{1.15}$$

with  $\Psi_{int}$  the surface energy per unit area of the matrix/particle boundary  $[J/m^2]$ ,  $\Omega$  the molar volume of the precipitate  $[m^3/mol]$ , D the particle diffusion  $[m^2/s]$ ,  $C_0$  the matrix concentration in equilibrium  $[mol/m^3]$ , R the gas constant  $[T/mol \cdot K]$  and T the absolute temperature [K].

This theory of diffusion-controlled particle coarsening (LSW) is accurate when the precipitate volume fraction is low, as assumed in the hypotheses presented above. This is not the case for Nickel based superalloys, where  $\gamma'$  volume fraction can go up to 50% or 70% for some of them. In the section below, some modifications of the LSW theory, proposed in the literature to consider the Ostwald ripening at high volume fraction, are described.

#### • Model for high volume fractions of particles

Extensions of the LSW theory to higher volume fraction has been studied by different authors. Some of them are summarized in the Table 1.4.

The value of  $A(f_{spp})$  depends on the volume fraction of precipitates and they change from one theory to another (see Table 1.5).

Model	Assumptions	Equations	Comments
The Ardell (MLSW)	<ul> <li>spherical particles,</li> <li>particle volume fraction ≠ 0,</li> <li>the particles are dispersed randomly,</li> <li>solute diffusion will depend on a distance characteristic between particles.</li> </ul>	$\overline{r}_{spp}^{3}(t) - \overline{r}_{spp}^{3}(0) = K(f_{spp})t$ $K(f_{spp}) = \frac{A(f_{spp})\Psi_{int}\Omega^{2}DC_{e}}{RT}$	The coarsening rate increases with increasing volume fraction.
The Brailsford and Wynbladt (BW)	<ul> <li>spherical particles,</li> <li>the particle growth rate is a function of the distribution and the concentration gradient around the particle.</li> </ul>	$A(f_{spp})$ dimensionless constant $\psi_{int}$ surface energy matrix/particle $\Omega$ molar volume of the precipitate D diffusion coefficient	The growth rate obtained is less sensitive to volume fraction than the MLSW theory but more than the LSEM theory.
The Lifshitz-Slyozov encounter modifed (LSEM)	<ul> <li>spherical particles,</li> <li>encounters between growing particles.</li> </ul>	Ce     matrix concentration in equilibrium       R     gas constant       T     absolute temperature	The diffusion field of the larger particle dominates over that of the smaller one, giving a rapid disolution and coalescence.

Table 1.4: Some theories of coarsening for high volume fractions [46, 56, 57, 58].

		$A(f_{spp})$	,)	
$f_{spp}(\%)$	LSW	MLSW	LSEM	BW
2	8/9 = 0.89	2.37	1.02	1.16
5	0.89	3.49	1.09	1.44
10	0.89	5.06	1.17	1.75
20	0.89	7.90	1.30	2.19
40	0.89	13.25	1.54	2.98
60	0.89	18.44	1.84	3.69

Table 1.5: Values of  $A(f_{spp})$  for the LSW, MLSW, LSEM, and BW theories as a function of the volume fraction of precipitates [46].

Numerous models describing the evolution of the distribution of the SPP sizes were also developed in [56, 57, 58, 59, 60, 61, 62, 63, 64] all based on the LSW theory.

The main conclusions of these models are:

- Particle distribution can be obtained independently of the nature of the initial distribution of particles.
- A linear relationship between the cube of the average particle size and time was obtained, where the slope is the growth rate constant,  $K(f_{spp})$ . The obtained temporal power law is similar to the initial theory (LSW).
- As the particle volume fraction  $f_{spp}$  increase, the distribution become broader.

Some of these theories are only valid up to a certain fraction or in 2D or 3D. It is difficult, a priori, to determine which of these theories best describes coarsening at high volume fractions. Therefore, more experimental and analytical studies are necessary to settle the differences between the various existing Ostwald ripening theories.

## 1.4.2 Precipitation

The theory of the growth of precipitates from solid solution was established by Zener [13] under the following hypotheses:

- the precipitates are spherical,
- no interaction between the particles is considered,
- the mechanism responsible for the transformation process is the mass diffusion.



Figure 1.6: Schematic of a) a  $\beta$  precipitate with planar boundary growing into a supersaturated  $\alpha$  matrix. b)  $\beta$  precipitate dissolving in an undersaturated matrix.

The growth of particles can be expressed according to the equation:

$$\overline{r}_{spp} = K_p \sqrt{Dt},\tag{1.16}$$

where the concentration is assumed to change at a constant rate from  $c^{\alpha}$  to  $c_0$ , assuming that the solute atoms remain constant (see figure 1.6.a), with r the radius of the particle, D the diffusion coefficient, and  $K_p$  a constant that depends on the concentrations and can be expressed as:

$$K_p = \sqrt{2\left(\frac{c_0 - c^{\alpha}}{c^{\beta} - c^{\alpha}}\right)},\tag{1.17}$$

with  $c^{\beta}$ ,  $c^{\alpha}$ , the concentration of the solute atoms in the precipitate and the matrix, respectively, which are in equilibrium, and  $c_0$  is the concentration in the matrix far from the interface [13, 65].

## 1.4.3 Particle dissolution

#### a. Model for a single precipitate

Thomas and Whelan [66] were the firsts to propose a solution to the dissolution problem (see Figure 1.6.b). They proposed to treat it as the reverse of the growing process (precipitation) based on the work of Zener [13] from equation 1.16.

$$\frac{d(\overline{r}_{spp}^{2})}{dt} = -K_d D \tag{1.18}$$

being  $\overline{r}_{spp}$  and  $K_d$  expressed as follows:

$$\overline{r}_{spp}^2 = r_0^2 - K_d Dt \tag{1.19}$$

$$K_d = 2\left(\frac{c^0 - c_\alpha}{c^\beta - c^\alpha}\right) \tag{1.20}$$

This theory assumes:

- Spherical and small precipitates (i.e.  $\bar{r}_{spp} \ll R$ ),
- 3D diffusion obtained essentially from the steady-state part of the diffusion field around a spherical precipitate.
- the dissolution rate is supposed to be low.

One year later, the same author, Whelan [67] obtained a model based on the resolution of the diffusion laws to describe the dissolution phenomenon of spherical particles taking into account the concentration gradients presented below.

#### b. Dissolution of a spherical particle

Based on the analytical models exposed before and on the equations governing diffusion processes (Fick's laws) [54], the evolution of the particle radius gives the dissolution kinetics of a spherical particle with time as follows:

$$\frac{d\overline{r}_{spp}}{dt} = \frac{-K_d D}{2\overline{r}_{spp}} - \frac{K_d}{2} \sqrt{\frac{D}{\pi t}}.$$
(1.21)

This equation includes the steady and transient states. If we neglect the transient part (the term including  $t^{-1/2}$ ), we obtain the equation given by Thomas and Whelan [66]:

$$\frac{d(\bar{r}_{spp}^2)}{dt} = -K_d D \tag{1.22}$$

This shows that the equation proposed by Thomas and Whelan is derived from the steady-state of the diffusion field in spherical diffusion. Therefore, equation 1.21 gives a better approximation of the actual dissolution process, taking into account the transient and steady-state parts.

The equation 1.21 do not have an analytical solution, but we can obtain an approximation assuming short times. In this context, the solution for the equation 1.21 can be expressed as:

$$\overline{r}_{spp} = \overline{r}_{spp}(0) - \frac{K_d Dt}{2\overline{r}_{spp}(0)} - \frac{K_d}{\sqrt{\pi}}\sqrt{Dt},$$
(1.23)

with  $\overline{r}_{spp}(0)$  the initial particle radius, and  $K_d$  is defined in equation 1.20.

# 1.5 Summary and discussion

This chapter provides some of the basic notions concerning the microstructure of the  $\gamma/\gamma'$  superalloys. The different particles population are described. In this work, we have focused our developments on the primary  $\gamma'$  phase due to their fundamental role in GB pinning during grain growth.

The phenomenon that predicts the impact of particles in the microstructure is given by the Smith-Zener pinning, which was described from its initial formalism until the different modifications developed by some authors to consider more realistic hypotheses.

We have also presented some of the mechanisms that lead to particles evolution. These mechanisms do not always occur isolated; some may be concomitant, increasing the complexity to identify them experimentally. The coarsening of particles is affected by the nearest particles. The LSW theory needs to be modified to take into account the high volume fraction of precipitates in the considered nickel base superalloys materials.

The kinematics of dissolution depends on different factors like the size and distribution of particles. Small particles dissolve easier than large ones; hence the beginning of the dissolution process is faster [45, 68, 11, 69, 70]. However, when the holding time is extended, dissolution becomes slower. Another factor that can affect the dissolution is, of course, the chemical composition of the material, including the  $\gamma$  phase and the  $\gamma'$  precipitates, and it will mainly depend on the elements with the lowest diffusion coefficient D, even if the diffusion in a multi-component alloy is much more complex.

In the next chapter, we will present the different existing numerical methods used to simulate microstructural evolutions at the mesoscopic scale, and especially how the presence of particles can be considered in each one of them.

# Résumé en Français du Chapitre 1

Ce chapitre introduit les notions de base liées à la microstructure, les phases et l'équation de migration des joints des grains. L'ancrage de Smith-Zener est également détaillé pour comprendre l'impact des particules de seconde phase sur la microstructure. L'idée étant de comprendre l'impact de l'évolution des particules afin d'améliorer les propriétés thermomécaniques des superalliages base nickel considérés.

Les mécanismes prépondérants d'évolution des particules de seconde phase ont été introduits.

CHAPTER 1. LITERATURE REVIEW
# Chapter 2 Numerical methods

For forty years, numerous full-field numerical methods have been developed to reproduce, at the mesoscopic scale, the observed experimental mechanisms that occur in polycrystalline materials, such as GG, ReX, and SSPT. From statistics to deterministic approaches with low and high complexity, these methods have proved their efficiency in reproducing some of these microstructural mechanisms.

In this chapter, some of these methods in the context of the GG and ReX, will be described. A brief description of the different formalisms will be proposed by focusing on the possibility of dealing or not with SPPs. Finally, the current strengths and weaknesses of these approaches will be highlighted.

# 2.1 Classical Full-Field Models for Microstructural Evolutions

The most used numerical methods to simulate microstructural evolutions are often categorized into three types:

- Stochastic techniques such as Monte Carlo (MC) and some Cellular Automata (CA) formulations.
- Deterministic front-tracking models such as vertex methodologies or Lagrangian formulations.
- Deterministic front-capturing methods such as the phase-field (PF) and level-set (LS).

In the following, a description of these models is proposed with a particular focus on their application to simulate the Smith-Zener pinning phenomenon and the possibility of reproducing particle evolution when the material is submitted to a thermal treatment. We will discuss some of the results available in the literature.

# 2.1.1 Monte Carlo models

The first Monte Carlo or Potts model for GG was proposed by Anderson et al. [71] in 2D and 3D [72] and a second phase particles population was first introduced in [73, 74]. The MC method is also used to reproduce other mechanisms such as recrystallization or abnormal grain growth.

In this model, the microstructure is represented by a discrete and regular lattice (see Figure 2.1). The structure comprises polyhedral shaped sites such as triangular, squared, or honeycomb shapes, forming pixels in 2D and voxels in 3D.



Figure 2.1: Microstructure representation in the MC models [29].

Each site has an attribute such as the orientation, which is given by an Id between 1 and Q, where Q is the total number of grains orientations that must be chosen large enough to avoid coalescence of two grains with the same orientation.

In MC simulation, a reorientation attempt consists to select a site at random and reorientating it to neighboring grain orientations. Then the energy of the new state is calculated. If the energy variation is negative, then the reorientation is accepted; for positive variations, a probability function can be used rather than systematically rejecting the swapping.

For GG, the energy of the grain boundary is described by the Hamiltonian as follows.

$$H = \sum -J(\delta_{S_i S_j} - 1), \qquad (2.1)$$

The sum is carried out for all closest neighbors sites j of site i and  $\delta_{ah}$  is the Kronecker delta symbol, and J is a boundary energy term given by the unlikeness of two consecutive sites.

Reorientation within a grain is not possible due to the elevated energy change, but the transition at the grain boundaries allows the boundary to migrate.

Boundary migration kinetics is modeled through *Monte Carlo Step* (MCS), corresponding to N reorientation attempts, where N is the total number of sites in the domain. This is considered as a drawback of the MC model because of the lack of physical time in basic formulations.

However, the MC method was the most used because it is the easiest to implement and to parallelize. It can also describe many features of the GG process, including some aspects of Zener drag.

### SPPs simulations

The presence of particles in the MC model was introduced in [74], where the precipitates were simulated in the domain by assigning them a constant Id which can not evolve during GB migration. Indeed, the corresponding sites are not allowed to switch their orientation during the simulation; the particles can be reproduced by one or several neighboring sites as presented in Figure 2.2.

The energy of the interface particles is considered the same as for the grain boundary. Then the total energy of the configuration when a particle is within a grain is more significant than when the particle is at the grain boundary. An attraction between particles and grain boundaries similar to the one predicted by the Smith-Zener pinning is observed. However, a correct physical description of the interaction between grain interfaces and SPPs is not considered.

Several studies related to the impact of the particles on the microstructure have been carried out with this model. For example, Gao. et al. [74] studied the effect of the initial location of the second phase particles and their size in 2D.



Figure 2.2: Particle representation in the MC and CA models,  $\lambda$  is the lattice site spacing [74].

Figure 2.3 shows the initial microstructure for the cases where the particles are randomly distributed (a.) and mainly located in the grain boundary (b.) and the results obtained (c.). They observed that the initial location of the particle population does not have a significant effect on the grain size evolution when the particle surface fraction  $f_{SPP} < 10\%$ . However, the particles considered spherical in 2D microstructures are equivalent to fibers in 3D, which means that the pinning forces in the 2D case are overestimated. Then, several studies have been developed in 3D cases; see for instance [72, 43, 75].



Figure 2.3: Initial numerical microstructures with  $f_{spp} = 1\%$ : a. Particles with a random distribution. b. Particles only on the GBs. c. Results of the GG simulations, effect of size and initial location of particles [74].

MC method had been adapted to reproduce some of the mechanisms presented in the previous chapter, such as the particle coarsening [76, 77] or Ostwald ripening in 2D, and 3D [78, 79, 80] and the agglomeration mechanism [81].

For the Ostwald ripening mechanism [74], the MC method has been modified to represent two components, the grains that are represented by a number (from 1 to Q) and the particles that are represented by a specific number for the entire second phase population. Two interfacial energies are defined, one for the matrix/matrix ( $E_A$ ) and the second for the matrix/particle ( $E_B$ ) boundaries.

The microstructural evolution by this mechanism must allow the material transfer from the matrix to the second phase. This mechanism was simulated by choosing a site i at random and then choosing a neighboring site j. If one site is an A-site and the other a B-site, then the two are temporarily exchanged with the A-site assuming one of the possible states at random. The energy change for this exchange is evaluated to determine if the exchange is accepted or rejected.

In [82] the authors presented an experimental study of the  $\gamma'$  particle evolution of the NASAIR 100 superalloy under applied stress at high temperatures. The  $\gamma'$  precipitates are usually discrete particles. However, once the stress is applied and the temperature increases, coalescence and coarsening of the particles occur, as shown in Figure 2.4.a and b. Then the MC model was used to reproduce the experimental evolution of these particles [76], and the results were quite similar, as can be observed in Figure 2.4. The experimental and the numerical results are in good agreement. This study shows the possibility of considering the MC method to reproduce complex evolutions of the particle phase. For further specifications see [82, 76].

The possibility of considering aggregate structures by the MC method to simulate the agglomeration mechanism was presented in [81], where different random accommodations of aggregates were tested to reproduce the experimental particle morphology. In these works, the particles grow thanks to the effect of attaching particles, but the growth of the particles themselves is not considered.

Generally speaking, MC models allow simulating the impact of particle population (Smith-Zener phenomenon) in 2D and 3D microstructures as well as some of the mechanisms presented in the previous chapter due to its easy implementation. However, some aspects are still discussed in the literature, such as the validity of the probabilistic laws and the time scale unit that is not straightforward to calibrate as mechanism dependant [83]. Moreover, the curvature effects are classically poorly estimated due to the pixelized/voxelized description of the grain interface.



Figure 2.4: Experimental and numerical comparison for the NASAIR 100 superalloy using the MC model, the material was submitted to a tensile stress of 500 MPa in the y-direction [82, 76].

# 2.1.2 Cellular Automaton models

The CA method was initially proposed by Von Neumann [84] and then further developed by Wolfram [85, 86].

Most used Cellular Automata formulations describe the microstructural evolution thanks to probabilistic laws as in the MC model. The domain is discretised by the cells of a regular (or non-regular) lattice.

This grid of cells are usually characterize by several parameters such as [29]:

- Cell geometry as their number, arrangement, size, and shape.
- The types of possible states for the cells (e.g. recrystallized or not).
- The definition of the neighborhood of a cell.
- The transitioning rules for a cell, describing the state of a cell as a function of its previous state and the state of the neighboring cells.

Cellular Automata implement a time discretization. After each time step, the values of the state variables are updated for all lattice points and the new values are assigned thanks to the transformation rules.

This discrete method opens the possibility of simulating the evolution of complex mechanisms. The microstructural evolutions are reproduced by the repeated and synchronous application of pre-established rules to all lattice nodes. These rules can come by implementing a phenomenological equation [87], or it can be derived with FE formulations [88].

CA model has been successfully used for several phenomena as the GG, the recrystallization in 2D, and 3D [87, 88]. We can also cite a comparative study between the MC and CA models for SRX modeling [89]. A new numerical model based on CA was proposed in [90, 91] for the simulation of dynamic recrystallization (DRX). CA simulation of the post-dynamic recrystallization (PDRX) was described in [92]. The presence of second phase particles has also been considered in [93] and the Ostwald ripening mechanism presented in [94].

### **SPPs** simulations

To consider the particle phase in the CA model, the definition of three new variables is required; the grain boundary energy per area, the volume fraction, and the size of the particles. Implementing these parameters and how forces are computed in the model open the possibility of representing the particle population [93].

The volume fraction of particles is fixed and is an attribute of each cell. For small SPPs (comparatively to the voxel size), some variants exist where an equivalent pinning pressure is introduced for voxels containing SPPs. It is also possible to consider an heterogeneous distribution of particles.

The simulation of particle evolution, such as the Ostwald ripening mechanism, has been described by the CA model in [94]. In this work, the author used the curvature-driven interface equations together with the GB migration and the CA rules to reproduce the Ostwald ripening mechanism.

To ensure the preservation of the  $\gamma'$  phase volume fraction, the author proposed two steps to update the cells. The first step consists to consider the curvature driven interface mechanism to describe the states of the  $\gamma'$  cells that are located in the  $\gamma/\gamma'$  and  $\gamma/\gamma$  interfaces; this step is in charge of the  $\gamma \rightarrow \gamma'$  transformation, which leads to the shrinkage of small particles which have the more significant curvature. The second step consists to identify the  $\gamma'$  located at the interface and forces them to change from  $\gamma'$  phase to  $\gamma$  phase. This second step allows the growth of the big particles given by a random selection operation. In the end, the Ostwald mechanism is well represented, where the big particles grow at the detriment of the small ones, and the particle volume fraction is conserved.

Figure 2.5 shows the results of the Ostwald ripening simulation with this CA model for two different volume fractions of particles  $(f_{spp})$ . The first case considers a  $f_{spp} = 10\%$ , and it can be observed that the small particles disappear and the big ones get bigger and bigger. For the second case, a  $f_{spp} = 50\%$  was considered, here the coarsening of the  $\gamma'$  is observed, and the impinge with adjacent particles. In this work, particle coalescence was not permitted.

In general, MC and CA methods can simulate particle-microstructure interactions through a simplified description of the SPP. However, their ability to reproduce boundary migration from curvature flow is still an open topic. This is partly attributed to their pixelized/voxelized interfaces, limiting the morphology definition of particles. Modeling large deformations in CA through mixed CAFE approaches is also an active research field.



Figure 2.5: Microstructure evolution given by a CA model simulating Ostwald ripening for different volume fractions  $f_{spp} = 10\%$  and  $f_{spp} = 50\%$ .

# 2.1.3 Front-tracking methodologies: Vertex and full Lagrangian models

One of the first vertex methods was proposed by Kawasaki et al. in [95] and then further developed by Weygand et al. [96]. The microstructure is represented with a set of actual vertices (multiple junctions) and virtual vertices that help to discretised the grain boundary. In [96], the boundary migration is given by the reduction of the surface energy of the grain boundary (V) as a result of the dissipation caused by the viscous friction that resists to the movement of the boundary (R). An equation is used to determine the velocity of the vertices in the domain as follows:

$$\frac{V}{\partial \vec{r}} + \frac{\partial R}{\partial \vec{v}} = 0, \qquad (2.2)$$

Where  $\vec{r}$  represents the position and  $\vec{v}$  the velocity of the vertex, this equation can be described as the amount of free-energy dissipated following the movement of the vertices with a velocity  $\vec{v}$  that is equal to the potential energy change following a change in the position of vertices  $\vec{r}$ .

The vertex method had been used to simulate several microstructural phenomena as the recrystallization in [97], or simulation of particles in 2D and 3D [96, 98, 42].

Recently, a highly performing Lagrangian model (ToRealMotion) for the simulation of evolving multi-domains problems has been introduced in [99] for GG, further developed in a parallel context [100] and extended to different microstructural mechanisms such as the dynamic and post-dynamic recrystallization [101]. The main difference with classical vertex methods is that the grain surfaces and volumes are fully meshing/remeshing through classical FE mesh algorithms.

#### SPPs simulations

In vertex algorithms, the SPPs are described as "pinning centers" where the pinning pressure must be overcome to liberate pinning GBs. These pinning centers are described by the maximum force  $(F_{max})$  that they can tolerate (see equation 1.13).

The particles are divided into two types: actives particles, which are place on the GB blocking the grain growth, and inactive particles, those inside the grains. Then, pinning and unpinning events switch the state of particles (active/inactive) during the simulation. For this reason, both types of particles are treated as a new kind of vertex [96].

The interaction particle/grain boundary gives the movement of the vertices considering the particle population. The vertices move without considering the pinning centers, which is finally accepted if the GB had not passed any particle. Hence, for each moved vertex, the presence of particles has to be searched in the swept area by the GBs. The final position of the vertices is chosen such that at most one particle is present in the triangular area swept.

Finally, the condition for unpinning is tested. In the simplest case, if the force acting on the particles is higher than  $(F_{max})$ , the GB can pass the pinning center and be unpinning. Several rules are considered for unpinning. These rules depend on the particle's position on the boundary as presented in Figure 2.6.



Figure 2.6: Interaction of a GB and a particle for different configurations [96].

A finite-element method coupled with the vertex philosophy was proposed by Couturier et al. in [102, 42]. They simulated a single grain boundary motion and its interaction with spherical and incoherent particles under the curvaturedriven force of grain growth. In their simulations, the interaction particle/grain boundary is given by the kinematic conditions applied to the particle/boundary interface nodes to guarantee that their velocities remain tangential to a meridian of the particles. Figure 2.7 shows the typical interaction of the boundary with a particle and its velocity changes during the contact. When the grain interacts with the particles, the velocity increases rapidly but then slows as the boundary moves ahead of the particle's "equator".

The authors have observed that the particles exert a pinning force almost twice smaller than the predicted value of the original Smith-Zener equation (Eq. 1.9). The methodology used here can also be applied to reproduce the recrystallization mechanism considering the particle phase; in this case, the driving force will be the stored energy in the grains, but the equations of motion will be very similar.

While pure vertex approaches are practical and enable an accurate calculation of grain boundary curvature, they drastically limit the SPPs representativity (shape, evolution, size). To our knowledge, there are no simulations that consider particle evolution with these approaches. Finally, a weakness of this method is the complexity of reproducing 3D simulations of polycrystals due to the different topological treatments that must be done to track explicitly the grain interfaces.

FE front-tracking approaches seem to be promising methodologies even if only one GB was described in the works of Couturier et al. [102] and if SPP description is currently not taken into account in the ToRealMotion code developed by Florez et al. [99, 100, 101, 103, 31].



Figure 2.7: Boundary/particle interaction, a) grain boundary evolution, b) normalized velocity changes during the particle/boundary contact.  $V_o$  is the velocity the boundary would have in the absence of particles and  $V_g$  the velocity of the GB geometric barycentre [102].

### 2.1.4 Phase-field model

Multi phase-field and phase-field approaches are front-capturing methods, i.e. where the interfaces are implicitly described. The method was proposed in [104] and further developed to consider microstructural evolution during solidification [105, 106]. In the last decades, the model had been adapted and developed to simulate different microstructural mechanisms such as GG [107], Rex [108, 109, 110], Smith-Zener phenomenon [44, 111, 112, 113, 114, 115, 116, 117, 118], SSPT, etc.

The PF approach is a thermodynamic model. The grains are represented by phase variables  $\eta_1, \eta_2, ..., \eta_p$  to distinguish the different grain orientations. The set of variables is allowed to evolve with time to represent the microstructural evolution. These variables vary continuously from one grain to another to form the diffusive GBs [113]. For a grain  $G_i$  identified by  $\eta_i$ , the  $\eta_i$  takes a value of 1 inside the grain, while all other phase-field variables equal zero. All  $\eta_i$  vary continuously between 0 to 1 with the neighboring grains at a grain boundary.

The evolution of the microstructure is given by the minimization of the free energy of the system F, which can be described by the Landau equation as follows

[104]:

$$F = \int_{V} \left[ f(c_1, c_2, \dots c_n, \eta_1, \eta_2, \dots \eta_p) + \sum_{i=1}^{n} \alpha_i (\nabla c_i)^2 + \sum_{i=1}^{3} \sum_{j=1}^{3} \sum_{k=1}^{p} \beta_{ij} \nabla_i \eta_k \nabla_j \eta_k \right] dV,$$
(2.3)

where f is the local free energy density,  $c_i$ , and  $\eta_i$  correspond to the conserved and non-conserved field variables, and  $\alpha_i$  and  $\beta_{ij}$  are gradient energy coefficients. The form of f depends on the simulated mechanisms.

Once the expression for free energy density f is set for specific phenomena, the total free energy F can be minimized by solving a set of PDEs denominated the Cahn-Hilliard [119] and Allen-Cahn [120] equation:

$$\frac{\partial \eta_p}{\partial t} + L_{pq} \frac{\partial F}{\partial \eta_q} = 0, \qquad (2.4)$$

where  $L_{pq}$  and  $M_{ij}$  are kinetic parameters related to the GB mobility.

### SPPs simulations

The particles are introduced in the model as an additional phase field variable, with a new energy term in the total free energy term of the system [44]. This new energy term is built so that the additional energy generated by a particle placed on a GB is equal to the intersection area multiplied by the surface tension.

The pinning effect of the particles is based on the fact that when a particle and grain boundary interact, the total amount of grain boundary in the system is reduced. Therefore, new GB area is created when the GB passes the particle.

Several 2D [44, 112, 121] and 3D [114, 113] PF simulations of the Smith-Zener pinning, with complex SPP shape [117], particle coherency [115, 121] and evolution [122] of SPPs have been considered.

The effect of particle shape had been studied in [111, 112] where spheroid particles were studied with different aspect ratios and sizes.

More advanced simulations have been done, where the particle evolution is taking into consideration, this had open the possibility of simulate some of the different particle mechanisms that takes place when particles evolve such as: coarsening [123, 124, 125], Ostwald ripening [126, 127, 128, 129, 130], dissolution [131, 132, 133, 134], precipitation [135, 136], splitting [137] in 2D and 3D.

The splitting particle mechanism has been investigated and reproduced by a PF approach for a coherent particle under applied uniaxial stress in [137]. Here, the splitting is attributed to the inhomogeneity of the diffusion potential inside the precipitate particle due to the elastic strain field. The authors observed that this mechanism is affected by the initial particle shape (see Figure 2.8). For instance, for spherical particles, there was no split, while square particles that present a high aspect ratio can split as presented in Figure 2.8. The supersaturation and the applied stress also influence the splitting process.



Figure 2.8: Evolution of an initially rectangular isolated particle compared with the observed experimental splitting [137].

The precipitation and growth of the  $\gamma'$  phase in the *DD*6 superalloy was studied in [135] (Figure 2.9). The PF model was coupled with an explicit nucleation algorithm to reproduce the precipitation of the  $\gamma'$  phase, which has a cuboidal shape and was considered under the influence of an elastic driving force. The growth of the particles can be divided into three stages: fast growth, coarsening, and quasi-static coarsening. The coarsening stage is consistent with the LSW theory.



Figure 2.9: Simulated microstructure evolution for different time steps of DD6 superalloy submitted to  $1120^{\circ}C$ . a. t = 0, b. t = 0.006, c. t = 0.012, d. t = 120, e. t = 0, f. t = 240 [min], [135].

Figure 2.9 illustrates the precipitation process for the DD6 material submitted to 4h of thermal treatment at  $1120^{\circ}C$ . As observed, the precipitation has almost been completed in the process from a. to b. Only a few particles were formed from b. to c. (rectangular areas). Then, precipitate evolution continues until it gradually slows down and stabilizes (see [135] for further details).

The coarsening of the CMSX-4 Ni-Al superalloy was investigated experimentally and simulated using a PF model incorporating elastic driving forces in [125]. The initial microstructure was extracted from SEM images (real morphology). Figure 2.10 shows the simulation results for different time steps. We observe that big particles grow faster and small particles dissolve and coalesce with the particles around. The particles shape is close to a square but with rounded corners and a low curvature to the sides.



Figure 2.10: Time evolution of the CMSX-4 material, comparison of the PF simulation results with the experimental samples [125].

The results of the PF simulation and the theoretical prediction of the coarsening model compared with the experimental data are in good agreement. Indeed, the coarsening rates calculated by the PF approach reproduce the coarsening law of the particles. Thus, the particles meet at their corners and coalesce.

3D PF simulations of the coarsening of  $\gamma'$  particles had been studied in [123]. The authors were able to simulate particle evolution comparable to experimental data. The simulated morphological changes agree with experimental observations regarding particle density, morphology, and spatial correlations.

### 2.1.5 Level-Set model

The LS method was first introduced in [138]. This method enables calculating the position of an interface  $\Gamma(t) = \partial G$  governed by a velocity field  $\overrightarrow{v}$ . The interface is represented as the interpolated zero-isovalue of a function  $\varphi(x, t)$  classically, defined as the Euclidean distance to the interface [139]:

$$\begin{cases} \varphi(x,t) = \pm d(x,\Gamma(t)), x \in \Omega\\ \Gamma(t) = \{x \in \Omega, \varphi(x,t) = 0\} \end{cases}$$
(2.5)

where  $d(x, \Gamma(t))$  is the Euclidean distance between a point x and the boundary  $\Gamma(t)$  of the considered object G and  $\Omega$  the calculation domain. When the interior of G is not the empty set, such as for grains in polycrystals, the sign convention assuming  $\varphi$  positive inside and negative outside is often adopted. Hence, the interface migration can be deduced by the resolution of a convection equation with  $N_G$  the number of grains:

$$\begin{cases} \frac{\partial \varphi_i}{\partial t} + \vec{v}_i \cdot \nabla \varphi_i = 0\\ \varphi_i(t=0,x) = \varphi_i^0 \quad \forall i \in [\![1, N_G]\!] \end{cases}$$
(2.6)

When the GG phenomenon is considered, the minimization of the system energy is driven by a mean curvature flow (minimization of the interface energy), so the grain boundaries velocity can be approximated by the following relationship:

$$\vec{v}_i = -M_i \gamma_i \kappa_i \vec{n}_i, \tag{2.7}$$

where  $M_i$  is the local grain boundary mobility,  $\gamma_i$  the local grain boundary energy,  $\kappa_i = \nabla \cdot \vec{n}_i$  the local mean curvature (i.e. the curvature in 2D and the trace of the curvature tensor in 3D), and  $\vec{n}_i = -\frac{\nabla \varphi_i}{\|\nabla \varphi_i\|}$  the outward unitary normal vector to the interface. When the material is considered isotropic,  $M_i = M$  is assumed only temperature-dependent (through an Arrhenius law) and  $\gamma_i = \gamma$  is considered as constant [35] (see Eq. 1.4).

A more complex expression of M could be considered with a heterogeneous description of  $\gamma$  (dependence to the misorientation angle) [30, 36, 37], anisotropic description of  $\gamma$  (dependence to the inclination of the grain boundaries) [39, 38], and more detailed description of M (dependence to the misorientation or solute aspect) [140]. Theoretically, each grain of a polycrystal must be represented by its own LS function. In order to reduce the computation time and memory storage, several non-neighboring grains in the initial microstructure (separated by a certain number of grains) can be grouped to form Global Level Set (GLS) functions thanks to Graph coloration techniques. However, the grains belonging to the same function can no longer be distinguished. Consequently, when two child grains of a GLS grow and meet each other, numerical coalescence occurs, i.e. both grains merge to form a single one. Thus different strategies are considered to avoid these numerical coalescence events. A re-coloration algorithm, such as the one proposed by Scholtes et al. [141, 142] can be considered at each time step. The number of GLS functions, N is then always drastically reduced compared to the number of grains  $N_G$ . This method is efficient in terms of memory and performance, allowing to use LS-FE simulations with a large number of grains even for 3D simulations [143]. In the following, the index i will denote the set of GLS used to describe the entire polycrystal and not the individual grain Id anymore.

Assuming that  $||\nabla \varphi_i|| = 1$ ,  $\forall i \in [\![1, N]\!]$ , i.e.  $\varphi_i$  remain distance functions all along the simulation, Eq.(2.6) can be reformulated as a set of diffusive equations meaning that the calculations of the mean curvatures and the normal vectors at grain interfaces are no longer required.

$$\begin{cases} \frac{\partial \varphi_i}{\partial t} - M \gamma \Delta \varphi_i = 0\\ \varphi_i(t=0,x) = \varphi_i^0 \quad \forall i \in \llbracket 1, N_G \rrbracket \end{cases}$$
(2.8)

where  $\varphi_i^0$  is the initial distance function of each GLS. A drawback of the LS formulation lies in the fact that, after the resolution of Eq.(2.8), the GLS are generally no longer distance functions  $||\nabla \varphi_i|| \neq 1$ . This is particularly problematic when a specific remeshing technique depending on the distance property is used at the interface. In addition, the diffusive formulation proposed in Eq.(2.8) requires a distance function at least in a thin layer around the interface in order to properly compute the mean curvature flow mechanism.

Finally, the conditioning of the transport problem when stored energy due to plastic deformation is considered also depends on the regularity of the LS function [144]. For these reasons, the GLS functions need to be reinitialized to restore their metric property at each time step. Numerous approaches exist to solve this reinitialization procedure in regular grids or unstructured FE meshes [145, 146, 147]. A direct, fast, and accurate approach usable in unstructured FE mesh proposed by Shakoor et al. was recently developed [148]. Discussions concerning the residual errors inherent to this approach are also discussed in [149].

The presence of non-physical vacuum regions at the multiple junctions, due to the front-capturing description of the grains and following the resolution of Eq.(2.8), is well known. The following classic procedure, proposed in [150], is

generally used to treat it:

$$\hat{\varphi}_i = \frac{1}{2} \left( \varphi_i - \max_{j \neq i} \varphi_j \right), \quad \forall i = 1 \dots N_{GLS},$$
(2.9)

where  $\hat{\varphi}_i$  is then used as the corrected GLS function.

### **SPPs** simulations

In existing LS formulations, the particles are represented as static holes in the domain. No assumption is then needed to simulate the pinning effect generated by the particles. Indeed, the SZP effect generated by the presence of particles can be naturally taken into account by imposing the relevant boundary conditions related to the SZP phenomenon at the interfaces between GBs and SPPs. More precisely, the influence of SPP on microstructure evolution can be taken into account by imposing a Neumann type limit condition on the GLS at the surface of the precipitates following the Young-Herring surface tension equilibrium:

$$\frac{\nabla\varphi}{||\nabla\varphi||} \cdot \vec{n} = \nabla\varphi \cdot \vec{n} = \sin(\alpha) = \frac{\gamma_p^2 - \gamma_p^1}{\gamma}, \qquad (2.10)$$

where, as illustrated in Figure 1.4,  $\vec{n}$  is the external unitary normal vector to the precipitate,  $\alpha$  is the angle established by the balance of surface tensions at contact positions between the SPP and the grain boundary. So, when the particle is assumed incoherent with the matrix (quite common for superalloys materials [9, 45]),  $\gamma_p^1 \simeq \gamma_p^2$ , which leads to  $\alpha \simeq 0$ . A null Neumann boundary condition is applied at the precipitate/grain boundary interface through the respect of Eq.(2.10).

To our knowledge, it does not exist simulations that consider particle evolution in the context of a LS model. However, several mechanisms have been reproduced, such as the abnormal grain growth of low stored energy grains for the *Inconel* 718 material in [151]. In this work, the material was submitted to particular thermal treatments. The driven force was provided by capillarity and residual stored energy to simulated post-dynamic recrystallization and GG considering the SPPs.

Figure 2.11 shows one of the obtained results; some grains grow more than others due to the stored energy differences across boundaries.

The possibility of simulated actual morphology of the particles was presented in [152], where the experimental particle shape was extracted from Electron backscatter diffraction (EBSD) maps or secondary electron (SE) images. Two years later, Scholtes et al. proposed in [141] several improvements to the LS method. That makes possible the 3D simulations of GG in the presence of spherical and real SSPs shapes as presented in Figure 2.12.

Although the actual morphology of the particles can be considered, a limitation of this method is the impossibility of reproducing the particle evolution.



Figure 2.11: Evolution of a microstructure (*Inconel* 718) with an initial energy distribution, and a static particle population (red points) with surface fraction  $f_{spp} = 2\%$  and annealing at 1258 K for different times. a. Initial microstructure. b. Microstructure at t = 3300s [151].



Figure 2.12: 3D FE mesh of a  $44 \cdot 44 \cdot 44 [\mu m^3]$  simulation domain containing 10,000 SPPs with radius  $r = 0.35 [\mu m]$ . The mesh is composed of 7 million of elements approximately. The color code refers to the mesh element quality. Precipitates are shown in white [153].

This is quite necessary if we aim to reproduce the material's behavior; since in the manufacturing process, the material is subjected to high temperatures where particle evolution occurs. The high numerical cost due to the fine mesh used to reproduce the shape of the particles is another weakness of this approach.

# 2.2 Summary and discussion

Model	Туре	Complexity	Space discretization	Interfaces	Computational performance	Large modeling of concomitant mechanisms
Monte Carlo	Stochastic	Low	Fixed	Sharp	High	Low
Cellular Automata	Stoch./Deter.	Low	Fixed/CAFE	Sharp	High	Low
Vertex	Deterministic	High	Only GB	Sharp	High en 2D	High
FE Front-Tracking	Deterministic	High	Volumic and remeshing capabilities	Sharp	High en 2D	High
Multi Phase-Field	Deterministic	Medium		Diffuse	Low	High
FE Level-Set	Deterministic	Medium		Sharp	Low	High

Table 2.1: Brief summary of the characteristics of the different full-field models.

Table 2.1 present roughly the characteristics of each usable numerical approach at the mesoscopic scale.

The Monte Carlo and Cellular Automata methods were the first framework used to simulate microstructural evolutions. They are easy to implement and very efficient but suffer also of important limitations. Typically a poor description of the curvature is directly linked to the grids considered to describe the microstructure. For MC approaches, the correlation between the MC step and the physical time can be difficult to exhibit when concomitant mechanisms occur [83].

Finally, the modeling of mechanisms involving large deformation of the polycrystals, like DRX can not be considered easily.

The Vertex models have a high computational performance in 2D; however, their 3D implementation involves numerous topological changes on the microstructure that are difficult to treat. Another weakness of this approach is the impossibility to reproduce intragranular phenomena. However, the recent TRM model [99] can overcome this limitation. To our knowledge, particle evolution was not considered in these front-tracking framework.

The PF model uses a diffuse description of the interfaces. The microstructural evolution is obtained by minimizing the free energy of the system. The particles are represented by adding a new space-dependence energy term in the total free energy system.

The LS model, where the interfaces are implicitly represented through a sharp description was introduced. Convective/diffuse equations can be used to describe the grain boundary network evolution.

A weakness of this approach is that the particle population is described as holes in the domain simulation and does not evolve. The principal advantage of the LS is the possibility of considering substantial deformations, which opens the possibility of reproducing more realistic thermomechanical treatments like the ones encounter in the metal forming industry. The LS approach is also particularly suitable for the modeling of numerous concomitant mechanisms in a unique numerical framework.

The PF and LS models can handle topological changes, making them very attractive to simulate the microstructural evolutions. Nevertheless, the major weakness of the PF and LS models based on a FE resolution remains their computational cost, which is high for simulations involving a large number of FE elements. An alternative to reduce the calculation time would be the implementation of an FFT [154, 155]; however, modeling large polycrystal deformation would not be possible.

These different elements explain why a FE-LS method was chosen in the following to develop a full-field framework enabling to consider evolutive second phase particles.

# Résumé en Français du Chapitre 2

Ce chapitre présente les différentes méthodes numériques à champ complet existantes dans la littérature pour simuler à l'échelle mésoscopique les phénomènes métallurgiques à considerer en mise à forme à chaud. La description des formalismes ainsi que l'implémentation des particules de seconde phase dans chacune des méthodes sont décrites. Une présentation des travaux les plus représentatifs a été proposée.

L'intérêt d'améliorer le formalisme LS existant à été décrit. C'est en effet à ce jour la formulation la plus efficace pour simuler de manière simultanée un grand nombre de mécanismes physiques, possiblement dans un contexte de grandes déformations en 2D et 3D. CHAPTER 2. NUMERICAL METHODS

# Chapter 3

# Simulation of the Smith-Zener Pinning phenomenon with an evolutive second phase particles population

As discussed in the first chapter, in numerous polycrystalline materials, grain size is controlled by second phase particles (SPPs) that hinder the grain boundaries (GBs) by pinning mechanisms. The Smith-Zener pinning (SZP) model describes the physical interaction between SPPs and GBs. Both of them can evolve when applying a heat treatment to the material. As industrial forging processes involve hot deformation steps near the solvus temperature, it is thus of prime importance to characterize the evolution of the SPPs due to their impact on the final microstructure, notably on the grain size.

As illustrated in the second chapter, the Level-Set (LS) method is classically used to describe the influence of SPPs on grain growth (GG) by considering the simulated particles as inert and represented by static holes in the used finite element (FE) mesh. Here, a new formalism to model the GG mechanism under the influence of the SZP phenomenon, able to take into account evolving particles, is proposed. It involves the representation of SPPs by a LS function and a particular numerical treatment around the grain interfaces encountering the SPPs, making possible the modeling of SPPs evolution without altering the undergoing pinning pressure. Validation and comparison of the new method regarding previous FE-LS formulation in 2D and 3D simulations and an application of a polycrystal GG under the influence of dissolving particles are described.

A version of this chapter was partially published in [21].

# **3.1** Introduction

The first page below offers a summary of the elements discussed in more detail in the two previous chapters.

The SZP describes the interaction between particles and GBs [12]. For materials like the  $\gamma/\gamma$  superalloys that present a matrix phase ( $\gamma$ ) and a particle phase ( $\gamma$ ), GG can be hindered by the  $\gamma$  phase, enabling to control the mean grain size of the microstructure during the forming processes of these materials. This is also presented in other materials like steels, for instance, where the particles facilitate the grain refinement [156, 157]. The described phenomenon was first discussed by Smith [12] following a Zener's idea [13]. Since these first developments to equate this phenomenon, many variants have been developed to dispel some of the initial hypotheses (see [14, 158, 43, 42]).

In the context of GG, the classical Smith-Zener model relates the average radius of grains  $\overline{R}$  of a given stable microstructure with the average radius  $\overline{r}_{spp}$  of precipitates and their volume fraction  $f_{spp}$ . This equation provides the stationary mean grain radius based on the balance of driving pressures.

Nowadays, it is well known that the original equation overestimates the limit grain size due to an overestimation of the capillarity driving pressure  $P_G$  or an underestimation of the pinning pressure  $P_Z$  which both rely on numerous hypotheses. These pressures depend on several factors like particle morphology, particle/matrix interaction (coherency), grain and particle size distributions...

Finally, the Smith-Zener equation can be generalized as presented in Eq. 1.11.

Due to the importance of predicting the properties of the materials, this phenomenon has mainly been studied with full-field numerical methods at the polycrystalline scale.

The two following paragraphs summarized briefly the state of the art fully detailed in the second chapter.

One can cite probabilistic techniques such as Monte Carlo (MC) [43, 72, 74, 73, 75, 159] and Cellular Automata (CA) [88, 93] methods where SPPs are described thanks to voxels/cells with a specific orientation. These sites are not allowed to be reoriented, so particles are assumed immobile, and the actual interactions between SPPs and GBs are roughly taken into account. For small SPPs (comparatively to the voxel size), some variants exist where an equivalent pinning pressure is introduced for voxels containing SPPs [160, 83]. Front-tracking models such as vertex methodologies [96], where the grains are linear segments that simulate boundaries, are also used. In these approaches, the SPPs are described as "pinning centers" where the pinning pressure must be overcame to liberate pinning GBs. Thus, while these approaches are practical and enable an accurate calculation of grain boundary curvature, they limit the description of the SPPs (shape, evolution, size) drastically.

Recently, the use of front-capturing methods like the phase-field (PF) [44, 111, 112, 113, 114, 115, 116, 117, 118] and level-set (LS) [152, 141, 150, 144, 35, 143, 149] have been developed to describe grain interfaces thanks to implicit math-

ematical functions. In the PF model, the particles are usually represented by adding a new space-dependent energy term in the total free energy system, and complex shape [117] or properties [115] of SPPs can also be considered. In the LS approach, they are described as holes in the domain where Neumann boundary conditions are imposed to the LS functions to respect the Young-Herring equilibrium in the contact positions between GBs and SPPs.

To our knowledge, the simulation of GG in the presence of SSPs based on the LS method has initially be proposed by Agnoli et al. [152], further studied and improved by Scholtes et al. [141] in a 3D context. In their model, it is possible to introduce a spherical SPP population and generate particles with a real morphology extracted from Electron backscatter diffraction (EBSD) maps or secondary electron (SE) images. However, this approach has some limitations mainly because the SPPs are described with static holes and refined FE meshes at their interfaces are required:

- the simulation of material deformations seems not straightforward when one wants to take into account the SPP behavior as the SPPs are not described in volume in the considered FE mesh,
- the previous remark could be crucial when DRX is investigated. Indeed, it is well-known that the SPPs interfaces are favorable sites for the appearance of new grains. Thus, a fine description of the mechanical fields at the SPPs interfaces is necessary but impossible, at yet, in the existing LS formulations,
- the simulation time can be drastically increased by the remeshing operations around the particles and the GBs, mainly in 3D,
- the evolution of SPPs, due to diffusive mechanisms, such as precipitation/dissolution, Ostwald ripening, agglomeration, spheroidization can not be considered (as SPPs are represented as static in the existing LS formulations).

For these reasons, a new LS approach to model GG mechanisms in the presence of meshed SPPs and able to reproduce evolving particles is proposed. In this perspective, the description of SPPs is made by a new LS function over the domain calculation without considering holes in the FE mesh. A numerical treatment around the grain interfaces surrounding the SPPs, that ensures a good description of the pinning pressure and makes it possible to migrate the SPPs interface has been developed. At the same time, the new formalism opens the possibility to simulate material deformations more naturally by taking into account the mechanical behavior of SPPs. Validation and comparisons of the proposed numerical framework comparatively to previous formulations [152, 141] followed by the GG simulations of a representative nickel base superalloy microstructure are presented.

# **3.2** Interface treatment

# 3.2.1 Level-set modeling of polycrystals

The full-field simulations presented in this work are based on the level set (LS) method in a P1 FE framework.

This method was presented in section 2.1.5 and to resume, the simulation of GG under the effect of capillary forces in a FE/LS context involves the following steps at each time step:

- Grain boundary migration is calculated by solving Eq. 2.8 for each active LS function. In order to limit the computational cost, a Graph coloration/recoloration technique is used [142] allowing to reduce the number of LS functions by grouping non-neighboring grains in common LS functions, called global Level-Set (GLS) functions. Each GLS contains numerous grains that are separated by a minimum distance, hence  $N_{GLS} \ll N_G$ [142].
- After the resolution of Eq. 2.8, voids or overlaps can have appeared at the GB and their junctions. In order to treat this issue and remove such non-physical regions, the methodology proposed in [150] and described by Eq. 2.9 is used for each GLS function.
- All active GLS functions are reinitialized  $(\|\nabla \varphi_i\| = 1)$  with the direct reinitialization method proposed in [148]. Indeed, one of the weaknesses of the LS formulation is to not naturally conserve the metric property of a LS function initially defined as a distance function. To ensure the validity of the diffusive form of Eq. 2.8 to describe curvature flow migration, distance functions are required at least near the interfaces.

Several approaches have been developed to treat the reinitialization procedure in regular grids, or unstructured FE meshes [147, 145, 146]. Here the parallel and direct reinitialization algorithm proposed by Shakoor et al. [148] was adopted as a fast and accurate method. Discussions concerning the residual error of this approach are detailed in [149].

- The negative GLS functions are removed from the system of equations.
- To avoid numerical coalescence, the re-coloring technique presented in [141, 142] is applied.
- Interface remeshing operations following the methodologies proposed in [152, 161] are performed if required.

# 3.2.2 Particle grain boundary interaction

In previous FE-LS studies [152, 141], where the particles are represented as static holes in the domain, no extra assumptions are needed to simulate the pinning

effect generated by the particles. The SZP effect can easily be accounted for by imposing Neumann-type boundary conditions on the interfaces between GB and SPP that meet the Young-Herring surface tension equilibrium.

As illustrated in Figure 1.4,  $\alpha$  is the angle established by the equilibrium of the surface tensions at the contact point between the SPP and GB. In our case the particle is assumed to be incoherent, meaning  $\gamma_p^1 \simeq \gamma_p^2$ , then  $\alpha \simeq 0$ , which can be obtained through a null Neumann boundary condition at the SPP (see Eq. 2.10).

In our new formalism, where the particles are represented by one new LS function,  $\varphi_{SPP}$ , the GLS fields describing the grains are initially modified with simple topological operations (following by a reinitialization step) to introduce the presence of the SPPs, without modifying the particle interface:

$$\forall i \in [\![1, N]\!] \quad \hat{\varphi}_i (t = 0, x) = \min \left(\varphi_i (t = 0, x), -\varphi_{SPP} (t = 0, x)\right). \tag{3.1}$$

A reinitialization procedure follows this operation as the resulting LS functions are not (when the intersection is not empty) a distance function, even if GLS and  $\varphi_{SPP}$  are. Of course, the function  $\varphi_{SPP}$  (t = 0, x) can be easily estimated as the distance function to the union of simple objects (as circular or spherical particles) but also obtained through the FE-immersion of an experimental map [152, 141].

See Figure 3.1, for an illustration of the resulting modification for two neighboring GLS.

### 3.2.3 SPP treatment

As already discussed (see Eq. 2.9), the appearance of voids or overlaps, especially at the multiple junctions after solving the convective-diffusive equations, was first treated by [150] and implement in several cases, in 2D and 3D, using the LS method [141, 35, 143].

In order to respect the Young-Herring equilibrium without hollowing out the SPPs, we propose, at each time step, to simply extend this treatment by taken into account  $\varphi_{SPP}$  in the procedure:

$$\hat{\varphi}_{i}(t,x) = \frac{1}{2} \left( \varphi_{i}(t,x) - \max\left(\max_{j \neq i} \left(\varphi_{j}(t,x)\right), \varphi_{SPP}(t,x)\right) \right), \quad \forall i = 1 \dots N, \quad \forall x \in \Omega$$

$$(3.2)$$

Thereafter, we will call this the Modified Multiple Junctions Treatment (MMJT). As illustrated in the following, in the zones without SPPs, MMJT is equivalent to the classical numerical treatment (Eq. 2.9) whereas when SPPs are present, it enables, by successive iterations, to impose the Young-Herring equilibrium for incoherent SPPs.



Figure 3.1: General procedure to modify the GLS functions, only the positive side of the GLS fields is represented by a given color: a) Initial GLS fields of two neighboring grains (red and blue respectively) and one particle (*yellow*), b) Intersections of grains and SPP fields, c) modification of the GLS, the particle LS  $\varphi_{SPP}$  do not change in the process.

In Figure 3.2, the GG evolution of one grain (green part denoted by  $\varphi_1$ ) in the presence of one spherical particle (yellow part denoted by  $\varphi_{spp}$ ) is represented to see the effect of the MMJT. Typical FE mesh size and time step are chosen following classical strategy in the context of FE-LS modeling of GG [162, 163]. When the diffusive equation (Eq. 2.8) is solved on  $\varphi_1$ , the grain overlaps the particle LS  $\varphi_{spp}$  (see Figure 3.2.b). Thus, it is necessary to apply the MMJT (see Figure 3.2.c).

However, it is clear that after applying Eq. 3.2 to  $\varphi_1$ , the SPPs/grain interface is not properly defined. In fact, the LS describing the grains have only moved



Figure 3.2: Grain growth evolution of one grain in the presence of one SPP represented as an LS and the effects presented on the interfaces. a) Initial LS field of one grain ( $\varphi_1$ ) and one particle ( $\varphi_{spp}$ ), b) Resolution of the diffusive equation, the grain shrinks due to the curvature flow but also overlaps the particle field, c) The modified multiple junctions treatment (MMJT) is applied one time to correct the overlap, d) The MMJT is applied 1, 2, 3 and 10 times to satisfactorily describe the initial particle/grain interface.

half of the necessary distance to correct the non-physical overlap. This problem can be corrected by the use of multiple consecutive calls of Eq. 3.2 by keeping  $\varphi_{spp}$  unchanged; these calls reduce each time by a factor 2 the overlapped region between the SPP and the grain interfaces. Thereafter, we will denote by  $\#_{mmjt}$ the number of calls of the MMJT. Now, the remaining question is how many times do we need to call Eq. 3.2 to reproduce the grain/particle interface correctly? Figure 3.3 illustrates the previous example with a precipitate of diameter  $(D_{spp} = 10 \ \mu m)$ . As the error is described by a geometric series when not multiples joints are present (only one interface particle/grain), it can be expressed as follows:

$$Error\% = \frac{D_{overlap_0}}{D_{spp}} \cdot \frac{1}{2^{\#_{mmjt}}} \cdot 100.$$
(3.3)

where  $D_{overlap_0}$  is the initial overlap of the particle after the resolution of the diffusive equation (Eq. 2.8). So, the  $\#_{mmjt}$  chosen to obtain a given error depends on the initial overlap  $D_{overlap_0}$ .



Figure 3.3: Internal grain diameter  $D_i$  as a function of the number of calls of Eq. 3.2 for a SPP with a diameter  $D_{spp} = 10 \ \mu m$ .

# **3.3** Numerical results

In this section, a comparative study will be presented for different test cases. GG mechanism, in the presence of spherical and irregular particles in 2D and 3D, is considered. These cases will be developed with our new formalism and compared with the former one proposed in [152, 141, 142]. Some parameters of the new methodology will be discussed, as well as the impact of the FE mesh used.

# 3.3.1 Spherical particle

The first case corresponds to a dimensionless simulation (with unitary reduced mobility  $M\gamma = 1$ ). The grain (initial surface  $S_0 = 0.5014$ ) and the particle (static radius  $r_{spp} = 0.1$ ) are both immersed on a 2D domain of  $1 \times 1$  as represented in Figure 3.4.



Figure 3.4: Initial state of a grain growth case in presence of a spherical particle.

Shrinking of the red grain and the correct interaction between the grain interface and the SPP, as a function of  $\#_{mmjt}$ , are investigated as illustrated in Figure 3.5. The use of the MMJT enables to respect progressively the correct Young-Herring equilibrium at the multiple junction between the SPP, the red grain, and the blue one. This equilibrium is responsible for the abrupt modification of the local mean GB curvature (curvature in 2D and sum of the principal curvatures in 3D), explaining the pinning aspect of the SPP in the mean curvature flow of the GB. Thus, the proposed methodology enables, as the former one [152], to take into account naturally the Smith-Zener effect (without the introduction of any approximated Smith-Zener driving pressure in the kinetics equation Eq.



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Figure 3.5: Comparison of the grain surface evolution for both methods at different times, to see how the red grain overpasses the particle and the influence of  $\#_{mmjt}$  for the case of a grain evolution in the presence of a spherical SPP with a mesh size (h = 0.0025).

2.8). As one interface is static (the SPP), the MMJT progressively enforces the orthogonality between the grain interface and the SPP (as it enforces naturally a  $120^{\circ}/120^{\circ}/120^{\circ}$  equilibrium for classical triple junctions between three evolving grains [164]). Physically, these remarks highlight that the proposed MMJT strategy is, at yet, adapted to describe only incoherent particles.

### Mesh convergence:

The mesh convergence study has been realized in both strategies, i.e. when the particles are simulated as a new distance function (Figure 3.6.a - New formalism) and when they are treated as holes in the domain (Figure 3.6.b - Former formalism [152, 141]) with an equivalent numerical strategy (exact solver for the diffusive equation and time step value). Isotropic unstructured (triangular elements) FE meshes with a homogeneous mesh size h are considered; the coarsest mesh corresponds to h = 0.025 and the others to a refinement by factors 2, 5,

and 10 this value. The finest with h = 0.0025 is considered, for each approach, as the reference case in terms of precision. The evolution of the red grain surface of this reference case is used as an indicator of the precision, and the table 3.1 summarizes the errors obtained for both methods with the different meshes comparatively to the reference case.



Figure 3.6: Mesh convergence study for the new and former formalism concerning the red grain. Surface evolution in presence of a spherical and static SPP. (see Figure 3.4).

As a result of this analysis, we observe a more significant influence of the mesh size in the former formalism that presented an error of 15.06% against 8.42% in the new formalism for the coarsest mesh. These results validate the accuracy of the new method in terms of spatial resolution.

# Impact of $\#_{mmjt}$ :

As established in section 3.2.3, to satisfactorily describe the grain/particle interface, it is necessary to apply several times (in the same increment) the MMJT to properly respect the orthogonal condition at the grain/particle boundary.

Mesh size	New formalism	Former formalism
h = 0.025	8.42	15.06
h = 0.0125	5.38	5.77
h = 0.005	0.48	1.01

Table 3.1: Surface error [%] comparatively to the reference case (h = 0.0025).

For a given mesh size, we applied the MMJT method 1, 2, 3, 4, and 10 times to establish how many iterations are needed to represent the grain/particle interaction well. Figure 3.7 illustrates the results obtained and the residual error by comparison with the case where  $\#_{mmjt} = 10$  (reference value for each mesh size).

We can deduce from Figure 3.7 that this parameter could induce some errors when the mesh size is coarse. Nevertheless, for reasonable mesh size, the convergence in terms of  $\#_{mmjt}$  is very fast.

### Comparison of the new and former formalism:

For the new formalism, the reference value in each case (different mesh size) is defined as the simulation obtained for a  $\#_{mmjt} = 10$ . These simulations were compared to the results obtained with the former method for the different mesh sizes. From Figure 3.8, we obtain, by considering the former method as the reference method, a maximal difference around 12% for the coarsest mesh. This difference decreases with the mesh size, reaching 3% for the finest mesh.

It must also be highlighted that the shape of the L2 difference curves (right side of the Figures 3.7 and 3.8) can be explained by slightly different times of unpinning between both methodologies leading sometimes to a fast increase of the cumulative L2 difference in time, but with very similar topology evolutions and obtained steady or quasi-steady states. This remark can also be applied to Figures 3.10, 3.11, 3.14 and 3.17.

This result demonstrates that the proposed methodology enables to predict the SPP/GB interaction correctly compared to the classical FE-LS treatment of Smith-Zener pinning [152, 141]. Figure 3.5 illustrates the evolution of the grain boundary at different stages of the simulation for the two models, the new one (right side) for which we can appreciate the influence of  $\#_{mmjt}$  and the former method (left). The kinetics of the grain is well described as well as the grain/particle interaction. However, the main difference between the two methods remains a slight difference between the moment when the grain overpasses the particle completely. This difference decreases with the mesh size.



Figure 3.7: Impact of  $\#_{mmjt}$  for different mesh sizes: a. h=0.025, b. h=0.0125, c. h=0.005, d. h=0.0025 and their respective L2 error compared with the reference value ( $\#_{mmjt} = 10$ ).



Figure 3.8: Comparison of the grain surface evolution for the two formalisms with different mesh sizes and the corresponding L2 difference by considering the former method as the reference.
### 3.3.2 The peanut-shaped particle case

This test makes it possible, by considering an irregular peanut-shaped particle as illustrated in Figure 3.9, to observe if the new method enables to well describe GG mechanism when the grain boundary evolves from a concave to a convex surface. Numerical parameters are identical to the previous test case. As the mesh convergence of our model was already tested, giving a good agreement with the former method, it will not be discussed again for this configuration. Nevertheless, the study of  $\#_{mmjt}$  for this concave/convex geometry remains interesting and is considered.

### Precision of the new formalism:

According to the findings for the spherical particle, the number of calls of Eq. 3.2 does not have a significant impact on the description of the grain surface evolution, and the produced error is reduced with the mesh size.

In Figure 3.10, one can see the grain surface evolution for different values of  $\#_{mmjt}$  and the error produced compared with the reference value ( $\#_{mmjt} = 10$ ) for a coarse mesh h = 0.025 (Figure 3.10.a) and for a fine mesh h = 0.0025 (Figure 3.10.b). In the first case, we obtain an error below 5% and the error is reduced to 3% for the second one. These results validate the hypothesis that the  $\#_{mmjt}$  does not affect significantly the GG kinetics.



Figure 3.9: Initial state of the peanut-shaped particle case.



Figure 3.10: Impact of  $\#_{mmjt}$  for different mesh size a. h=0.025, b. h=0.0025, and their respective L2 error for the peanut-shaped particle case.

### Comparison of the new and former formalism:

The comparison of the two models is made for a coarse and a fine mesh (see Figure 3.11). We obtain a maximal error of 7.86% for the coarse mesh and of 2.82% for the fine one by considering the former approach as the reference case. The grain boundary evolution of both methods is represented in Figure 3.12, for the finest mesh (h = 0.0025) case. We can observe the impact of  $\#_{mmjt}$  for the new method (left side) and the former one (right side). A very similar evolution of the grain interface around the peanut-shaped particle is obtained for the reference value of the new formalism compared with the former one.



Figure 3.11: Comparison of the grain surface evolution for the two formalisms for two different mesh sizes: a. the coarse mesh h = 0.025, b. the finest mesh h = 0.0025 and the corresponding  $L^2$  errors for each peanut-shaped particle case taking the former formalism as the reference.



Figure 3.12: Comparison of the grain surface evolution for the two formalisms at different times, to appreciate the influence of  $\#_{mmjt}$  for the proposed method for the peanut-shaped particle case for the finest mesh size (h = 0.0025).

### $3.3.3 \quad 2D/3D$ cloud of particles cases

A first 2D case is considered to test the kinetic behavior of our new method in the presence of several circular particles and to compare it again with the former method. The simulation domain corresponds to a dimensionless square of  $1 \times 1$ . We consider one grain interface (red dome) in the presence of several spherical particles of different sizes as illustrated in Figure 3.13. An unitary reduced mobility is considered, the initial grain surface is  $S_0 = 0.7413$  whereas the radius of particles varies from  $\bar{r}_{spp} = 0.02$  to  $\bar{r}_{spp} = 0.03$  and a isotropic mesh size h = 0.005is considered.

In Figure 3.13, one can observe the grain interface at different times for both approaches. The results are very similar. The comparison of the grain surface evolution is detailed in Figure 3.14. The difference between the models is more significant when the grain encounters most of the particles. The grain surface presents a maximal difference around 10% which begins to decrease when the grain approaches the equilibrium state (total pinning of the grain interface as

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illustrated in the last picture Figure 3.13.e). Once again, these results illustrate that, if the new and former methods can present a slight difference in terms of kinetics of grains interacting with SPPs, this difference remains scarce and does not influence the equilibrium angles between GB and SPP or the quasi-static equilibrium resulting from this interaction.



Figure 3.13: Comparison of the grain surface evolution in presence of multiples spherical particles for the two formalism at different times.



Figure 3.14: Comparison of the grain surface evolution for the two formalisms and the L2 difference obtained for the grain surface between the new formalism and the former one for the 2D multiparticles case.

The last case of this section was realized to confirm that the proposed new methodology is also totally usable in a 3D context. Thus, a similar 3D configuration (see Figure 3.15) of the previous test case is considered. In Figure 3.16, one can observe the comparative evolution for different times and view angles. Once again, from these images, we can appreciate the strong similarity between both simulations. The grain volume evolution is presented in Figure 3.17, as well as the L2 difference (below 2%).



Figure 3.15: Initial state of the 3D case, a grain in presence of a cloud of particles.



Figure 3.16: Comparison of the grain volume evolution for the two formalisms at different times and views (lateral and frontal) for the 3D multiparticles case.

One can conclude this part by the fact that the proposed new methodology is able, for static SPPs, to perform similar simulations than the one proposed in [152, 141, 142] in 2D and 3D cases. Thus, more complex configurations with an evolution of the SPPs can now be considered.



Figure 3.17: Comparison of the grain volume evolution for the new and former formalism with the L2 difference obtained for the grain volume in each formalism.

# **3.4** Particle dissolution

As a real thermomechanical route generally involves a considerable variation of temperature, modeling the SPPs evolution can be vital if one wants to predict quantitatively and qualitatively the microstructure evolution, notably the grain size distribution. Therefore, modeling evolving particles during GG is necessary to treat actual industrial processes and accurately predict microstructural evolution.

To our knowledge, in the context of the LS method, the SPPs have only been simulated as static objects [152, 141, 142] and represented as holes. In the following, one strategy to consider the evolution of the particles based on the new method previously described is proposed. Finally, this strategy will be tested by considering a realistic 2D GG case with evolving SPPs.

### 3.4.1 Evolving SPPs

Once the initial mesh is generated, the polycrystal can be created statistically or experimentally from an EBDS map. Then the grains fields  $\varphi_i$  are modified to introduce the new particles field  $\varphi_{SPP}$  (see Figure 3.1). A convective equation Eq. 2.6 is applied to  $\varphi_{SPP}$ : a velocity  $v_{spp}$  is computed thanks to prescribed data concerning the radius temporal evolution of the particles, then, a smoothed velocity field v is computed via a Laplacian (Eq. 3.4) with Dirichlet boundary conditions  $v_{spp}$  established at the particle interfaces. Finally, v is used to compute the velocity field  $\vec{v}$  oriented towards the center of each precipitate to be applied to  $\varphi_{SPP}$  (see Figure 3.18 for an example):

$$\begin{cases} \Delta v = 0\\ v = v_{spp} \quad at \quad \Gamma_{spp} \end{cases}$$
(3.4)

and

$$\vec{v} = v \cdot \vec{n} = -v \cdot \nabla \varphi_{SPP},\tag{3.5}$$



Figure 3.18: Smoothed precipitate interface velocity  $(\vec{v})$  for three circular precipitates (black circles).

with  $\vec{n}$  the unitary inside normal vector to the SPP,  $v_{spp}$  the velocity that we want to impose to the SPPs and  $\vec{v}$  the resulting velocity field that we really impose, through a convection equation to  $\varphi_{SPP}$ .

All this process is made at each increment of the simulation. The smoothed velocity is implemented to reduce/eliminate the instabilities produced by the solution of the local convective finite element problem and to reduce the concentration of the isovalues of the level set function around the particles. A reinitialization of  $\varphi_{SPP}$  is also required at each time step to ensure a good description of the particles' isovalues at each increment.

The solution of the diffusive Eq. 2.8 is now applied to the grains, followed by several calls of the MMJT given by the Eq. 3.2 to correct the overlaps and vacuums generated at the multiple junctions between grains and between grains and particles. Finally, the procedure ends with the recoloring technique and the GLS reinitialization.

# 3.4.2 Grain growth polycrystal case with random SPPs distribution

The initial microstructure is composed of around 50000 grains generated with a Laguerre-Voronoi tessellation [165]. The arithmetic mean grain radius  $\bar{R}$  is equal to 5  $\mu m$  and an initial polydisperse spherical particle population with a surface fraction  $f_{spp} = 6\%$  (around 19000 SPPs) is generated. All SPPs are assumed incoherent. Figure 3.19 illustrates the initial state of a domain of 200  $\mu m \ge 200$ 

 $\mu m$ . The values of M and  $\gamma$  are chosen as representative of the AD730 nickel base superalloy and come from the database of the DIGIMU© software [161]. The mobility M is then expressed as an Arrhenius law and depends on the temperature T, i.e.  $M = M_0 * e^{-Q/RT}$  where  $M_0$  is a constant  $M_0 = 2.981 \cdot 10^{14} \ [mm^4/Js], Q$ is the thermal apparent activation energy for mobility  $Q = 3.87 \cdot 10^5 \ [J/mol], R$ is the ideal gas constant  $R = 8.314 \ [J/(molK)], T$  is the absolute temperature,  $\gamma$ is assumed as constant  $(6 \cdot 10^{-7} \ [J/mm^2])$  and the  $\gamma'$  solvus temperature  $T_{\gamma'_{solvus}}$ is estimated at around 1110 [°C].





A thermal treatment of t = 3 [h] is considered. More precisely, an isothermal treatment at 990 [°C] for 45 [min] is followed by a linear increase of the temperature until 1120 [°C] in 90 [min] and the material is maintained at this temperature for others 45 [min] (Figure 3.22.a). The main interest of such thermal treatment is to clearly see the SPP evolution near the solvus temperature (for the reference case) and also to simulate different particles dissolution velocities that consequently produce different effective solvus temperatures (total disappearance of SPPs in full-field simulations) and to study the impact on such evolution on the mean grain size evolution.

An empiric particle velocity  $v_{spp}$  is defined here as  $v_{spp} = \beta \cdot (T - 990)$  where  $\beta [mm \cdot s^{-1} \cdot C^{-1}]$  is a constant that we made vary from  $10^{-9}$  to 0 to appreciate the complete particle dissolution at different rates (Figure 3.22.b). A such totally empiric expression enables here to illustrate the capabilities of our methodology for a large range of  $v_{spp}$  without the precise use of experimental data for SPPs disappearance velocity (it will be discussed in the next chapter).

The time step for the simulation was set to 5 [s], which is small enough to correctly describe the interactions between the GBs and the particle interfaces. The figures 3.20 and 3.21 illustrate the evolution of the microstructure at different stages of the simulation (2700, 5400, 8100 and 10800 [s]) for our reference case ( $\beta = 10^{-9} \ [mm \cdot s^{-1} \cdot \circ C^{-1}]$ ), showing how the particles disappear during the thermal treatment and the concomitant grain boundary network evolution (pinning followed by classical GG).

Figure 3.22.c shows the particle surface evolution as a function of time. For the reference case ( $\beta = 10^{-9} \ [mm \cdot s^{-1} \cdot C^{-1}]$ ), the  $f_{spp}$  is static for the first 45 [min] and then completely dissolved near the effective solvus temperature (~ 1120 [°C]). When the parameter  $\beta$  increases, the particles velocity also increases and the particles dissolve faster. Thus, the effective  $T_{\gamma'_{solvus}}$  decreases when the parameter  $\beta$  increases (see Figure 3.22.d).

Finally, Figure 3.22.e illustrates the arithmetic mean grain radius. For the reference case, at the beginning of the simulation, the grains are pinned by the particles, and there is not significant GG, but when the particles start to dissolve, the grains start to grow and finally, when no more particles remain, the grains can grow freely as for classical GG mechanism. When the constant  $\beta$  increases, the particles dissolve in less time, enabling the grains to grow freely earlier than in the reference case. When  $\beta = 0 \ [mm \cdot s^{-1} \cdot C^{-1}]$  (null SPP velocity), the fraction of the particles is fixed for all the simulation, and it can be appreciated how the particles block the GG, resulting in a large difference of the mean grain size at the end of the simulation (see Figure 3.22.e).

The mean grain size evolution as a function of  $\beta$  illustrates how it will be possible to play with a thermal treatment near the solvus temperature while keeping a reasonable impact on the GG kinetics.

Such an aspect has, of course, a strong industrial interest. A simple and imposed  $v_{spp}$  profile was proposed here as an illustration, but the methodology proposed in Eqs. 3.4 and 3.5 will be directly usable with more complex evolution laws or data directly coming from experimental observations.

The computation time for the 50000 initial grains took 67.3 [h] when performed on 24 processors, which is a promising result given the high number of grains and the possibility to reproduce the particle evolution.



Figure 3.20: Microstructure evolution at different times for a random SPPs distribution.



Figure 3.21: Microstructure evolution at different times for a random SPPs distribution.



Figure 3.22: Results of the 2D-50000 grains case with evolving particles.

## 3.4.3 Grain growth polycrystal case with heterogeneous SPPs distribution

### Homogeneous grains size distribution

In this case, we assume a heterogeneous bimodal particle distribution; the initial microstructure is similar to the previous polycrystal. A simulation domain of 200  $\mu m \ge 200 \ \mu m$  with an initial number of grains around 50000 is considered. The arithmetic mean grain radius  $\bar{R} = 5 \ \mu m$  and a spherical particles population with an initial surface fraction  $f_{spp} = 6\%$  divided in big SPPs with  $f_{spp_{big}} = 4\%$  of radius  $\bar{r}_{spp_{big}} = 2 \ \mu m$  and small SPPs with  $f_{spp_{small}} = 2\%$  of radius  $\bar{r}_{spp_{small}} = 1 \ \mu m$ , distributed in two bands in the domain as illustrated in Figure 3.24.a. This case is developed to illustrate the possibility of treated a heterogeneous particle population and to reproduce more accurately the real microstructure of the AD730 nickel base superalloy, which is classically composed of two SPPs populations (see Figure 3.23) and also to observe if a heterogeneous GG can take place.



Figure 3.23: Classical microstructure of the AD730 superalloy.

The material properties are the same as for the previous polycrystal case, and the material is submitted to the same thermal treatment (see Figure 3.22.a).

The particle velocity  $v_{spp}$  is also established with the same kinetic law (see Figure 3.22.b). The small particles will be dissolved before the large particles; thus, the bands region will present a classical GG mechanism sooner than the entire domain.

The Figures 3.25 and 3.26 illustrate the evolution of the microstructure at different stages of the simulation (2580, 4950, 6880 and 10800 [s]) for our reference case ( $\beta = 10^{-9} \ [mm \cdot s^{-1} \cdot C^{-1}]$ ), showing the particle and GG evolution during the thermal treatment.



Figure 3.24: Schematic and simulation initial microstructure for a heterogeneous SPPs dispersion.

For the initial isothermal treatment where the particles do not evolve, the grain size is smaller in the zones composed of small SPPs. This can be easily explained by a bigger resulting pinning pressure in this zone than in the zone with large SPPs (see Figure 3.25.a). When the temperature increases, the particles begin to dissolve, and the grains evolve, especially at the regions composed of small particles where the small grains of these regions begin to grow (see Figure 3.25.b). Once the small SPPs are entirely dissolved, the grains can grow freely, and as represented in Figure 3.25.c (white ellipses), some grains can grow more than others. Thus a heterogeneous grain evolution occurs, where some grains are

likely to grow more and more, leading potentially to abnormal grain growth.

The thermal treatment ends with a maintain of temperature at 1120 [°C] which is superior to the solvus temperature, so no particles remain and all the microstructure evolves following a pure GG mechanism.



Figure 3.25: Microstructure evolution at different times for a heterogeneous SPPs distribution. White ellipse illustrates a zones with smaller grains.

Figure 3.27 shows a comparison of the grain size distributions obtained for both cases (random and heterogeneous SPP) represented in histograms. The initial grain distribution for the two cases is very similar, as can be seen in the Figure 3.27 a. As the TT progresses, we can see in the Figure 3.27 b, c. that the population of small grains is more representative in the case of heterogeneous particle distribution because more precipitates can hinder the microstructure, then when the small particles dissolve, abnormal grain growth occurs, and larger grains in the heterogeneous case in 3.27 d. are observed.



d. t = 10800 s

Figure 3.26: Microstructure evolution at different times for a heterogeneous SPPs distribution. White ellipses illustrate zones with bigger grains.

Finally, the equilibrium state is reached in both cases, since all the SPPs were completely dissolved and the grain has grown freely.

This case presents many intragranular particles in the two bands composed of small particles. These intragranular particles do not obstruct the GB, and their presence does not affect the grain size evolution. Therefore, another test will be developed considering a bimodal grain size distribution, where the grains of the bands with small particles will present smaller grains than the other grains of the domain.



Figure 3.27: Histogram comparison (arithmetic mean grain size) of the simulations with random and heterogeneous SPPs distribution for the reference case  $(\beta = 10^{-9} \ [mm \cdot s^{-1} \cdot \circ C^{-1}])$  at different times.

### Heterogeneous (bimodal) grains size distribution

For this case, the same heterogeneous particle distribution as in the previous case was used with  $f_{spp} = 6\%$ ,  $f_{spp_{big}} = 4\%$  of radius  $\overline{r}_{spp_{big}} = 2 \ \mu m$  and small SPPs with  $f_{spp_{small}} = 2\%$  of radius  $\overline{r}_{spp_{small}} = 1 \ \mu m$  for a simulation domain of  $200\mu m \ge 200\mu m$  with 70000 initial grains, Figure 3.28 shows the initial state of the numerical microstructure.



 $Initial\ state$ 

Figure 3.28: Initial numerical microstructure for a heterogeneous SPPs dispersion with a bimodal grain size distribution.

The grains of the microstructure consists of a bimodal distribution, with smaller grains in the region of the bands. A fictitious HT was performed in order to generate the desired initial microstructure. Indeed, for the first 100s we apply a gradient for the temperature as a function of the distance (x-axis) where the temperature of the domain with large particles was considered higher than in the band regions. Therefore, the grains could grow more out these band regions due to the variation in temperature and the fact that the large particles are further apart than the small ones. After the pre-calculation that allowed us to introduce the bimodal microstructure, the simulation parameters were similar to those of the previous case as well as the thermal trajectory (see Figure 3.22.a), where the small particles dissolve before than the big ones.

It can be seen that the initial state presented in the Figure 3.28 does not contain a large number of intragranular particles as in the previous test case because the initial grain size of the band regions is smaller than in the rest of the domain; thus, most of the particles will play an essential role in hindering the GB before dissolution.

Figure 3.29 shows the evolution of the microstructure at different times of the simulation (2580, 4950, 6880 and 10800 [s]) for the reference case ( $\beta = 10^{-9}$ 

 $[mm \cdot s^{-1} \cdot^{\circ} C^{-1}]),$  we can see the evolution of particles and GG during heat treatment.



Figure 3.29: Microstructure evolution at different times for a heterogeneous SPPs distribution with a bimodal microstructure.

For the initial isothermal treatment where the particles do not evolve, the grain size remains smaller in the regions of small SPPs. As mentioned before, this can be explained by the higher pinning pressure exerted by the small SPPs (see Figure 3.29.a). As the temperature increases, the particles begin to dissolve and the grains evolve, but no great change is observed due to the strong pinning generated by the particles. (see Figure 3.29.b). The particles evolution continues and the small SPPs disappear, letting the grains to grow freely in these regions (see Figure 3.29.c). The evolution of the grains continues, especially in the regions of initial small SPPs, which are free of obstacles before the rest of the domain, and we can see some larger grains in these zones (see Figure 3.29.d).

Once all SPPs are entirely dissolved, the grains can grow freely and, as shown in Figure 3.29.d (white ellipses), some grains may grow larger than others. Thus, heterogeneous grain evolution begins to take place, where some grains are likely to grow larger and larger, potentially leading to abnormal grain growth.

# 3.5 Summary and discussion

A new methodology to describe second phase particles in FE-LS modeling of GG was proposed. This approach was validated by comparison with a pre-existing approach.

The use of a LS function to represent SPPs enables better integration of SPPs in the model when an evolving SPP population is considered. This new formalism opens new perspectives regarding full-field recrystallization simulations as opposed to SPPs described by holes in the FE mesh. SPPs mechanical behavior could be modeled, and it could be expected a non-negligible influence of such numerical capability on the prediction of nucleation events and recrystallization kinetics.

A first methodology has been proposed in order to take into account evolving particles in the context of FE-LS modeling of GG.

This new capability has been illustrated for a nickel base superalloy in the context of a thermal treatment near the  $\gamma'$  solvus temperature. This new method provides a FE-LS framework able to accurately consider the physical evolution of SPPs (due to dissolution/appearance of particles or Ostwald ripening) and is expected to predict the concomitant grain size distribution and topology evolutions quantitatively. Moreover, complex local heterogeneous SPPs velocities could also be investigated. Indeed, it is well known that precipitates' growth and/or dissolution can be accelerated by contact with evolving GBs. The proposed formalism could be adapted to discuss this phenomenon numerically.

The next chapter focuses on the performed experimental work during my PhD concerning the AD730 and N19 alloys in order to validate the proposed numerical strategy (see chapter 5 for crossed discussions).

# Résumé en Français du Chapitre 3

Dans ce chapitre, une approche FE-LS étendue a été développée pour prendre en compte l'évolution des particules de seconde phase.

Cette nouvelle approche à tout d'abord été comparée et validée à l'approche historique dans le cas des particules de seconde phase statiques representées par des trous dans le maillage d'élements finis.

Il a ensuite été illustré sa capacité à modéliser correctement les évolutions des particules tout en conservant une description correcte des interactions joints de grains/précipités. L'intérêt de cette nouvelle approche à ensuite été illustré sur de nombreuses microstructures virtuelles typiques de la morphologie des microstructures d'intérêts dans ce travail de thèse.

# Chapter 4

# Materials, experimental methods and results

This chapter presents first the  $\gamma/\gamma'$  nickel base superalloys studied in this thesis and the industrial fabrication process of each material since the production process conditions and the as-received microstructures. Then, the experimental plan is described with the respective aim of each set of heat treatments.

The sample preparation along with the different microscopy techniques used for particles quantification and grain size measurement are also presented. Finally the experimental results are presented and analyzed in view of identifying the grain boundary migration parameters (mobility and activation energy) and kinetics laws for precipitate size and fraction evolutions.

# 4.1 Materials

The evolution of the  $\gamma'$  phase precipitates has been studied in two  $\gamma/\gamma'$  nickel based superalloys, AD730 and N19. The first one is a cast-and-wrought material. For such heavily alloyed materials, the initial heterogeneities arising from the early stage of solidification usually can not be fully erased. The as-received AD730 billet microstructure indeed presents remnant heterogeneity, notably regarding the  $\gamma'$ phase topology. In contrast, the N19 alloy is made through a powder metallurgy route, that gives a more homogeneous microstructure.

# **4.1.1** *AD*730 **superalloy**

This material has recently been developed by Aubert & Duval [25]. Its chemical composition is presented in Table 1.1. This new alloy is aimed at being used for critical parts of aircraft engines submitted to very severe in-service conditions. It achieves a good compromise between its mechanical properties and cost (because it includes a relatively high amount of Fe).

## Industrial route for producing AD730 billets [166, 167]

The AD730 alloy is obtained by a triple fusion Vacuum Induction Melting (VIM)/ Electro-slag Remelting (ESR)/ Vacuum Arc Remelting (VAR) where the material is melt to create the first ingot and then purified (see Figure 4.1).



Figure 4.1: Processing steps from AD730 ingot to billet and then to turbine disk [166, 167].

Once the triple fusion is complete, some species are still highly segregated. Therefore, a supersolvus homogenization treatment is performed to allow chemical species to diffuse and thus homogenize the alloy. After homogenization, the conversion takes place to obtain a billet with a finer and homogeneous microstructure by a series of hot deformation and annealing steps, as follows:

• **Supersolvus forging** : this step is made to break down the solidification structures into fully recrystallized microstructure but with very coarse grains because of the high temperature and the absence of precipitates during forging.

- The supersolvus/subsolvus transition : Temperature is then decreased leading to the precipitation of  $\gamma'$  phase, which will help controlling grain sizes during subsequent steps.
- Subsolvus forging steps : the purpose is to refine the grain size and to obtain a fully recrystallized structure. The final stage is a radial forging (SMX forging) which deforms more the periphery of the ingot that the core.

Once the ingot has undergone all these operations, it is called a billet. Finally, the billet material undergoes different shaping steps such as forging and rolling in order to end up with the final part.

### As-received state

For this PhD thesis, the AD730 alloy has been delivered in the form of a bar with a diameter of 8 inches (204 mm) which has followed the previously described industrial fabrication process. Samples were cut out from this bar to investigate the  $\gamma'$  precipitates evolution. Figures 4.2 and 4.3 show the heterogeneous and complex microstructure of the as-received material, including two types of grains, the recrystallized and unrecrystallized ones. The recrystallized grains are created in the conversion process.

They have a compact morphology, low dislocation density and thus low stored energy. Large  $\gamma'$  precipitates mainly located at the GBs and with spheroidal morphology are associated with these grains, those large precipitates are referred to as primary precipitates. Much smaller secondary and tertiary precipitates are also present in the grains, but will not be considered as they will be dissolved in the studied heat treatment temperature ranges. The elongated unrecrystallized grains are inherited from the ingot conversion process and are characterized by higher dislocation densities (as revealed by orientation gradients) and thus higher stored energy. They also contain a high density of intragranular  $\gamma'$  precipitates that are smaller than the ones found at the GBs in the recrystallized regions. As presented in Vernier's PhD work [168], about 10% of the initial microstructure correspond to these unrecrystallized elongated grains.

The area fraction of primary  $\gamma'$  precipitates in the as-received AD730 material is 12.5% which is in agreement with other works [25]. Details of the phase quantification methods will be given in section 4.3.2. The arithmetic mean value of the grain size  $\overline{D}$  is about 9  $\mu m$  for the recrystallized grains and the unrecrystallized ones are several hundred of microns in length. In this PhD work, only the recrystallized grains will be considered with their respective precipitate population.



Figure 4.2: AD730 as-received state, EBSD map. The crystallographic orientation color code corresponds to the projection of a direction of the macroscopic frame into the crystal frame, black lines correspond to GB with a misorientation angle higher than 15° and ignoring twin boundaries.



Figure 4.3: In-Beam SE images showing  $\gamma'$  precipitate populations found in the as-received state of the AD730 alloy.

### 4.1.2 N19 superalloy

### Industrial route for producing N19 disks [24]

Powder metallurgy is an elaboration process (illustrated in Figure 4.4) that limits the problems of segregation of the alloying elements. As a result, more homogeneous microstructures can be obtained compared to those obtained with the cast-and-wrought route.

The first step of this process is Vacuum Induction Melting (VIM) process, where the different elements are melted. The powder is then obtained from the ingot by spraying the liquid metal under argon flow. This method allows a rapid solidification of the metal, forming small particles of similar composition, significantly reducing segregation elements [169].



Figure 4.4: Typical steps of a powder metallurgy route as used for the N19 superalloy [170].

The powder obtained after argon atomization is then sieved to separate ceramic particles that may have originated from the previous elaboration steps. The sieved powder is placed in steel containers for sintering and densification by hot isostatic pressing (HIP). The subsolvus spinning process then follows this step to end up with a bar.

The N19 alloy used in this work was produced and developed by Safran and the ONERA from the fusion of alloy elements through argon atomization and then placing in containers. Then, HIP, spinning and forging was performed by Safran Aircraft Engines to obtain the final material that was delivered for this PhD work.

### As-received state

Safran provided four parallelepipedic bars extracted from as forged disk of the N19 alloy, of about 195 mm diameter and 26 mm thick.

As expected, the microstructure of the initial N19 material is more homogeneous than that of the AD730 bar (Figure 4.5 and 4.6). The primary  $\gamma'$  precipitates form big clusters, some of them having similar size compared to the  $\gamma$ matrix arithmetic mean grain size  $\overline{D}$  of about 3  $\mu m$ .

The initial primary  $\gamma'$  fraction is 19%. Details of the phase quantification method will be given in section 4.3.2.



Figure 4.5: N19 as-received state, EBSD/EDS map. Color code corresponds to the crystal orientation, black lines correspond to grain boundaries with misorientation angle higher than 15° and ignoring twin boundaries. The orientation color coding is applied to the  $\gamma$  phase matrix. Primary  $\gamma'$  precipitates are colored in dark blue.



Figure 4.6: In-Beam SE images showing the  $\gamma'$  phase topology in the as received state of the N19 material.

### 4.1.3 Thermo-Calc simulations

Thermo-Calc software [171] was used to estimate the  $\gamma'$  phase volume fraction of each superalloy as a function of temperature by using the TCNI9 database and the compositions given in Table 1.1. Figure 4.7 presents the volume fraction of  $\gamma'$  phase as a function of temperature for the two alloys. The  $\gamma'$  phase volume fraction estimated at room temperature was calculated as 37% for AD730 and 43% for N19, which are in agreement with the values reported in the literature [25, 172].



Figure 4.7:  $\gamma'$  phase volume fraction vs temperature curves for the AD730 and N19 superalloys (obtained from Thermo-Calc calculations).

According to the curves of Figure 4.7, the full  $\gamma'$  phase dissolution temperatures  $(T_{solvus})$  estimated by *Thermo-Calc* correspond to 1108°C and 1138°C for AD730 and N19, respectively.

# 4.2 Experimental plan

Heat treatments were performed at different times and temperatures for the two materials (summarized in Table 4.1), followed by cooling at  $100^{\circ}C/\text{min}$  or water quenching. The evolution of primary  $\gamma$  precipitates was considered to be negligible during cooling at  $100^{\circ}C/\text{min}$ , as confirmed by comparison with the primary  $\gamma$  evolution obtained after water quenching. In table 4.1 the HT and sample analysis corresponding to the green crosses where realized during this PhD work, the purple one where the data provided by Aubert & Duval and the orange ones by Safran (SEM images were provided and analyzed in the context of this PhD work).

They have been divided into two groups: supersolvus and subsolvus experiments aiming at following grain growth in absence or presence of the  $\gamma'$  precipitates, respectively. All HT made at Cemef have been done in a Carbolite furnace - RWF 12/13 able for a maximum temperature of 1200°C. All the samples have been quenched in water immediately at the end of the HT. A thermocouple was attached to the sample to determine its actual temperature, which is slightly different from the furnace temperature due to temperature gradients inside the chamber. As already highlighted in Table 4.1, additional data from Safran and Aubert & Duval have been collected.

The supersolvus thermal treatments were made at different temperatures for several holding times for both materials in order to identify the grain boundary motion parameters such as the mobility (M) and activation energy  $(Q_m)$ . In the supersolvus domain, where no precipitate remains, the microstructure is expected to freely undergo grain growth.

For the AD730 material, supersolvus heat treatments were made for 10, 15, 20, 30, 60 and 240 min. For the N19 material additional shorter supersolvus HT were required (1, 2, 3 min), because of the higher solvus temperature and thus faster GG. The initial state for each isothermal supersolvus holding was determined as follows. The furnace was heated to the targeted temperature and then the sample was introduced. However, as the furnace opens, the temperature decreases. Once the sample reached the targeted temperature, the holding time begins and at the end of the HT the sample was taken out of the furnace and water quenched. Time "0" of the isothermal holding was this moment at which the sample gets to the targeted temperature in all following experiments.

The subsolvus HT were carried out to describe and study the precipitate dissolution and its effect on grain size evolution. Grain size and  $\gamma'$  precipitate size and area fraction were measured in all samples heat treated at Cemef. Only  $\gamma'$  precipitates size and area fraction were available in the data provided by Safran

AD730 heat treatments								
Domoin	Temperature	Holding time [min]						
Domain	[°C]	10	15	20	30	60	240	
	1060	Х		Х		Х	Х	
Subsolvus	1070	Х		Х		ХХ	XX	
	1080	Х		Х		Х	X	
	1090	Х		Х		Х	X	
	1100		X		Х	XX	XX	
Solvus	1110	Х		Х		X	X	
	1120				Х			
	1125				Х			
Supersolvus	1140	Х	Х		Х	Х		
	1150	Х		Х	Х			
	1160	Х	Х	Х	ХХ	Х		
	· · ·	a.						

and Aubert & Duval. All available data are summarized in Appendix A in Tables A.1 and A.2.

N19 heat treatments												
Demein	Temperature	Holding time [min]										
Domain	[°C]	1	2	3	10	20	30	60	120	240		
Subsolvus	1110				X		X		X	X		
	1120				X		X			X		
	1130				Х	Х	Х	Х		X		
	1140				Х	Х		X		X		
Solvus	1150	Х	X		X	X	X	x	X	X		
Supersolvus	1160	Х	Х		X	Х			ХХ	X		
	1180	Х		X	X	X			X	X		
$\overline{b}$ .												

Table 4.1: Heat treatments done for a. AD730 and b. N19 superalloys.

# 4.3 Microstructure characterization techniques

Scanning electron microscopy (SEM) was used to characterize microstructures and measure the precipitate area fraction, their size, morphology, the grain size (with EBSD) and size distribution for both precipitates and the grains.

The microstructural analysis was performed with two microscopes:

• A Tescan FERA 3 FEG-SEM equipped with a Oxford C-Nano EBSD detector. Secondary electron (SE) and In-Beam SE images were acquired, providing a contrast between the matrix and the precipitates suitable for

 $\gamma'$  phase quantification. Backscattered electron (BSE) images and electron backscatter diffraction (EBSD) maps were acquired also, to obtain the crystallographic orientation information and in turn grain sizes.

• A ZEISS Supra40 Field Emission Gun Scanning Electron Microscope (FEG-SEM) equipped with a Bruker combined EBSD-EDS system. The same analyses can be done with this microscope as with the former one but in addition, the coupling of EBSD with EDS allows to differentiate  $\gamma$  and  $\gamma'$  phases thanks to their differences in chemical composition.

### 4.3.1 Sample preparation

Samples were prepared in order to obtain high-contrast images between the  $\gamma$  matrix and the  $\gamma$  precipitates. Allowing the segmentation of the  $\gamma$  matrix and the  $\gamma'$  precipitates and a surface quality suitable for EBSD.

The preparation protocol begins with a mechanical polishing down to 1  $\mu m$  grit. Then, there were two options. First, samples are further polished using a colloidal silica solution (OPS by Struers) on a vibrating polishing machine. This protocol is adequate for EBSD analyses, however, it slightly hollows out the precipitates, and the phase contrast is relatively poor (Tables 4.2 a,b.).

The second protocol is electrolytic polishing in a solution of 10% perchloric acid and 90% methanol at 10°C, under 35 V for 5-8 s. It provides more relief between the phases and in-Beam SE images with a strong phase contrast (Table 4.2 c.). This second protocol allows also EBSD to be performed. This sample preparation procedure was applied to all the samples prepared at Cemef. The sample preparation methods have been used for both the AD730 and N19 alloys.

The sample preparation method used by the industrial partners was a mechanical polishing down to 0.25  $\mu m$  grid and etching (with citric acid or  $HCl/CH_3COOH$ ) (Table 4.2 d.).

### 4.3.2 Precipitate quantification and image processing

In-Beam SE images were acquired with an acceleration voltage of 3 kV, at 3-5 mm working distance and with 60  $\mu m$  diaphragm size. The images provided by the industrial partners were analyzed by optical microscopy. Five to ten images of 100  $\mu m \times 75 \mu m$  (for AD730 and N19) were taken for each sample for the sake of statistical representativity. 290-350 precipitates/image were found in AD730 microstructure after annealing at temperatures below 1060°C. The higher the annealing temperature, the lower the number of precipitates found, reaching 180-230 precipitates/image at 1090°C. The same behavior was observed for N19 alloy images, where the number of precipitates decreases as the temperature increases.

Image J software [173] was used for image binarization and quantitative analysis of the  $\gamma'$  phase area fraction, size distribution and morphology. Phase segmen-

Test	Polishing protocol and microscopic images					
a	SE image after1.Mechanical polishing down to 1 μm.2.Vibration polishing for 2h (colloidal silica solution).SEM conditions: HV: 15 kV WD: 15 mm	- The precipitates are slightly hallows. - Not suitable to make the SPP segmentation due to the poor $\gamma/\gamma'$ contrast. - Suitable for EBSD				
b	BSE image after1.Mechanical polishing down to 1 μm.2.Vibration polishing for 2h (colloidal silica solution).SEM conditions: HV: 15 kV WD: 11 mm	<ul> <li>Chemical contrast is visible.</li> <li>Not suitable to make the phase segmentation.</li> </ul>				
c	In-Beam SE image after1.Mechanical polishing down to 1 μm.2.Electrolytic polishing, solution of 10% perchlorid acid and 90% methanol at 10°C under 35V-6s.SEM conditions: HV: 15 kV WD: 10 mm	<ul> <li>Slight particle relief.</li> <li>Quality suitable for EBSD.</li> <li>Optimal conditions for the particle segmentation.</li> </ul>				
d	Optical image after1. Mechanical polishing down to 0.25 μm.2. Chemical Eetching with citric acid or (HCI/CH3COOH\).	- No EBSD data. - Optimal conditions for the particle segmentation.				

Table 4.2: SEM images obtained with the respective protocol conditions and images features.

tation leads to a binary image where, the precipitates can be analyzed, excluding the ones at the edges as shown in Figure 4.8.



 $\overline{D}_{spp}[\mu m]$ c.  $\gamma'$  precipitate size distribution

3

Figure 4.8: Precipitate quantification after phase segmentation with ImageJ. a. In-Beam SE images. b. After segmentation. c.  $\gamma'$  precipitate size distribution.

### 4.3.3 Grain size analysis by EBSD

0 + 0 = 0

The EBSD technique consists in acquiring the diffraction patterns of the backscattered electrons known as Kikuchi patterns and then analyzing them to determine the orientation of the crystal [174].

The EBSD maps have been acquired with an acceleration voltage of 20 kV, 15-16 mm (Supra40), or 22-24 mm (FERA 3) working distance and 120  $\mu m$  diaphragm size. They were analyzed and post-processed with MTEX [175, 176, 177], an open MATLAB toolbox. A GB was defined when the disorientation between two neighboring points exceeded 10°. Twin boundaries (60° < 111 >) have been ignored in the grain detection procedure. Twins are thus considering to be intragranular defects.

A typical example of an EBSD map is shown on Figure 4.2, where the color code corresponds to the projection of an axis of the macroscopic frame into the crystal frame.
The step size of the maps was adapted, as the grains became larger and larger when increasing the annealing temperature. For instance, for the 4*h* HT at 1070°*C*, an EBSD map of size 1130  $\mu m \times 840 \ \mu m$  with a step size of 1.13  $\mu m$  was acquired, giving a total number of 5120 grains. For the 4*h* HT at 1100°*C*, two EBSD maps were taken to reach a good statistical representativity of the sample; the size of the maps was 2180  $\mu m \times 1700 \ \mu m$  with a step size of 4  $\mu m$  and they included a total number of 2272 grains.

#### Phases identification in a EBSD map

The  $\gamma'$  phase is indistinguishable from the  $\gamma$  matrix [178] because of their crystallographic structure being to close to each other.

Therefore, in the case of AD730, a size criterion has been applied to separate the precipitate from the matrix. The particles with a size  $0.3 \,\mu m \leq D_{spp} \leq 4 \,\mu m$ were considered as  $\gamma'$  primary precipitates as shown in Figure 4.9. But of course, the smallest  $\gamma$  grains are also considered as  $\gamma'$  precipitates which leads to  $\gamma'$  phase fraction overestimation.

For the N19 material, it was not possible to distinguish the primary  $\gamma'$  precipitates from the grains ( $\gamma$  phase) with the same strategy because the primary  $\gamma'$  precipitates or precipitate clusters have a size similar to that of the grains. Therefore, EBSD analysis was coupled with energy dispersive X-ray spectrometry (EDS) [179] to distinguish the two phases based on their chemical compositions. However, one drawback of this technique is that the spatial resolution of the EDS is limited to about  $0.5 \,\mu m$  under the current acquisition setup. For these EBSD/EDS analyzes, two maps of size 120  $\mu m \times 90 \,\mu m$  with a step = 0.08  $\mu m$  were acquired, giving a total number of around 1600 grains for the samples annealed at 1110°C and 1200 grains at 1130°C.

Figure 4.10 shows an example of an EBSD/EDS map for the N19 as-received state. Both phases are indexed, and can be separated as grains and precipitates. However, because of the lack of spatial resolution in these combined EBSD/EDS analyses the smaller precipitate are not considered (Figure 4.10 c.), and to be more precise in the characterization of precipitates, the precipitate populations have been quantified from In-Beam SE images (see section 4.3.2) as done for the AD730 alloy.

One of the advantages of EDS-EBSD coupling is that both analyses are performed simultaneously, limiting the offset or distortion between crystallographic and chemical data. Nevertheless, as described before, one drawback of this technique is the spatial resolution of the EDS. This spatial resolution depends on the composition of the material and the energy of the incident electron beam [180, 181]. Also, this coupling technique requires a relatively long time of acquisition and stability of the beam to not disturb the acquisition.

After separating  $\gamma'$  precipitates from  $\gamma$  matrix grains by either of the two quoted methods, grain size distributions and average values have been obtained from the EBSD.



Figure 4.9: EBSD map of the as-received state of AD730. All pixels are indexed as being  $\gamma$  phase and then a size threshold is used to differentiate the precipitates and the grains.



Figure 4.10: Comparison of the  $\gamma'$  phase distribution. a. EBSD/EDS coupling of the N19 material. Green phase correspond to the  $\gamma$  matrix and the red phase to the  $\gamma'$  precipitates  $f_{spp} = 12\%$ . b. After segmentation of the In-Beam images  $f_{spp} = 12.4\%$ . c. Comparison of the  $\gamma'$  precipitate size distribution.

## 4.4 Experimental grain size and precipitate evolution kinetics

### 4.4.1 Area fraction of primary $\gamma'$ precipitates after annealing for 4 hours

Figure 4.11 shows micrographs after 4 h isothermal HT carried out at different temperatures for each alloy. Note that the  $\gamma'$  precipitates of each alloy have different size distributions and morphologies, being directly related to the previous thermomechanical treatments.

However, a common point is the dissolution process itself, which is the reduction of the volume fraction of primary  $\gamma'$  precipitates with increasing temperature. In addition, as visible from the crystallographic contrast of some images, the pri-



 $\succ \gamma Matrix \rightarrow Primary \gamma' precipitates$ 

Figure 4.11: SEM SE micrographs obtained using the first preparation protocol after 4h heat treatment for AD730 at a.  $1060^{\circ}C$ , b.  $1090^{\circ}C$ , c.  $1110^{\circ}C$  and optical images obtained using the second preparation protocol for N19 at d.  $1110^{\circ}C$ , e.  $1130^{\circ}C$  and f.  $1140^{\circ}C$  (N19). Bright particles are the primary  $\gamma$ '-phase precipitates undissolved during annealing.

mary  $\gamma'$  dissolution leads to an increase in the grain size (Figure 4.11 b,c, for instance), which will be better described in the following by using EBSD maps.

Figure 4.12 shows the evolution of  $\gamma'$  phase fraction as a function of T. The primary  $\gamma'$  fraction significantly evolves (dissolution mechanism) when the temperature is close to the solvus temperature  $(50^{\circ}C - T_{solvus} \leq T \leq T_{solvus})$ , and remains constant when the temperature is far from the solvus point, as illustrated in Figure 4.12 a.c. The presence of secondary and tertiary  $\gamma'$  precipitates was not considered in the measurement of the  $\gamma'$  phase fraction. However, they are reminded on these graphs with labels  $\gamma'_{II}$  and  $\gamma'_{III}$ , respectively. An experimental or effective  $T_{solvus}$  could be determined as the temperature at which the area fraction of primary  $\gamma'$  is equal to zero after 4h isothermal holding. Figure 4.12 also exhibits a comparison between the *Thermo-Calc* curves and the experimental data. Note that thermochemical simulation values does not match exactly with the experimental  $T_{solvus}$  obtained for the N19 alloy. For temperatures close to  $T_{solvus}$ , the Thermo-Calc curve and the experimental points nevertheless follow the same evolution and shape. A shift of the *Thermo-Calc* curve was therefore done to find a better fit with the experimental points, an offset of  $12^{\circ}C$  were applied to the N19 alloy (Figure 4.12 b, dashed line.)

For the AD730 alloy (Figure 4.12 a), there is a much better fit between experimental points and the *Thermo-Calc* results. From these results, it was thus defined that the effective  $T_{solvus}$  values for 4h annealing corresponds to  $1108^{\circ}C$ 

and  $1150^{\circ}C$  for the AD730 and N19 superalloys, respectively.

The  $f_{spp}(T)$  curves can be described by a polynomial fit as a function of  $T - T_{solvus}$  difference, as presented in Figure 4.13.

The dissolution kinetics is faster, i.e., the slope of the curve is steeper, for the N19 alloy compared to the AD730 alloy. This is physically relevant as the annealing temperatures and  $T_{solvus}$  are higher for the N19 alloy. The equations, i.e. phenomenological models, that describe the data plotted in Figure 4.13 are Eq. 4.1 for AD730 alloy and Eq. 4.2 for N19:

$$f_{spp_{eq}AD730}(T) = -0.00084(T - T_{solvus})^2 - 0.3(T - T_{solvus}), \qquad (4.1)$$

$$f_{spp_{eg}N19}(T) = -0.001(T - T_{solvus})^2 - 0.35(T - T_{solvus}), \qquad (4.2)$$

where  $f_{spp_{eq}}$  is the equilibrium volume fraction considered to be reached after 4 hours and actually corresponds to the measured primary  $\gamma'$  precipitate fraction since secondary and tertiary precipitates are dissolved within the 50°C range below the solvus point. The shape of the  $f_{spp}(T)$  curves, as well as the values of the obtained second order coefficients, illustrate that linear approximations, although less precise, would also be acceptable.

# 4.4.2 Area fraction of primary $\gamma'$ precipitates as a function of time and temperature

The main purpose of this part is to establish a model to describe the area fraction of the primary  $\gamma'$  phase as a function of time and temperature within the 50°C window below  $T_{solvus}$ . Through experimental data and Thermo-Calc, two equations that reproduce the fraction of primary  $\gamma'$  as a function of temperature were obtained in section 4.4.1 (Eqs. 4.1 and 4.2). A JMAK model was then used to describe the diffusion-controlled dissolution of spherical precipitates. This relation has already been used in several works to represent the dissolution of  $\gamma'$ phase in polycrystalline nickel base superalloys [68, 45, 182], as follows:

$$f_{spp}(t,T) = f_{eq}(T) + (f_0 - f_{eq}(T)) \cdot exp\left(\frac{-t}{t_1}\right).$$
(4.3)

The area fraction of precipitates at thermodynamic equilibrium is represented by  $f_{eq}$ ,  $f_0$  corresponds to the primary  $\gamma'$  area fraction at t = 0s (as-received material), and  $t_1$  is a time-dependence constant.

Assuming that Eq. 4.3 is valid to describe the  $\gamma'$  phase dissolution of the two studied alloys, and that Eq. 4.1 and 4.2 are valid over the range  $50 - T_{solvus} \leq T \leq T_{solvus}$  to provide the equilibrium area fraction of (primary)  $\gamma'$  precipitates  $(f_{spp_{eq}alloy} = f_{eq})$ , the following expression is obtained:

$$f_{spp}(t,T) = f_{spp_{eq}alloy}(T) + (f_0 - f_{spp_{eq}alloy}(T)) \cdot exp\left(\frac{-t}{t_1}\right).$$
(4.4)



Figure 4.12:  $\gamma'$  area fraction after 4h annealing treatment as a function of temperature for alloys a. AD730 and b. N19.



Figure 4.13: Area fraction of primary  $\gamma'$  precipitates after 4h isothermal treatment and the respective curves obtained trough a phenomenological fits established on the shape of the *Thermo-Calc* curves (described in equations 4.1 and 4.2). Fit at the raw *Thermo-Calc* results for *AD*730; fit after a +12°*C* shift for the N19 alloy.

This time dependant model was validated through several heat treatments ranging from 10 min to 4 hours at different temperatures for each alloy. Some of the SE images taken for different HT are presented in Figures 4.14 and 4.15 for the AD730 and N19, respectively. The initial primary precipitate area fraction  $f_0$  and the identified value of the  $t_1$  constant were respectively 12.5% and 1300s for the AD730 and 18.5% and 250s for the N19. It is worth mentioning that the constant  $t_1$ , once identified for the best fit of one curve (one temperature), is in fact quite precise for all temperatures in the analyzed range, contrary to the work presented in [45] where the parameter  $t_1$  needed to be identified for each temperature.

Figure 4.16 shows a good agreement between the experimental results and Eq. 4.4 model predictions. The evolution of the  $\gamma'$  phase area fraction could then be described in the considered range of time and temperature.



Figure 4.14: Primary  $\gamma'$  precipitate dissolution of the AD730 alloy for two selected temperatures (1060°C and 1100°C) and three holding times at each temperature.



Figure 4.15: Primary  $\gamma'$  precipitate dissolution of the N19 alloy for two selected temperatures (1110°C and 1130°C) and three holding times at each temperature.



Figure 4.16: Comparison between the phenomenological model Eq. 4.4 (dashed lines) and experimental results (points) of the primary  $\gamma'$  phase fraction at different times and temperatures for a. AD730 and b. N19 superalloys.

## 4.4.3 Primary $\gamma'$ precipitate size evolution

Figure 4.17 illustrates the evolution of the mean size of primary precipitates  $(\overline{D}_{spp})$  for the two alloys. At short annealing times, the average precipitate size decreases, as could be expected from the dissolution process. However, after 1h of heat treatment, a slight increase in the mean size of the precipitates can be seen. This behavior suggests the presence of the Ostwald ripening phenomenon, for which large precipitates grow and the small ones shrink and disappear, while keeping the particle fraction constant.



Figure 4.17: Mean primary  $\gamma'$  precipitate size in the a. AD730 and b. N19 superalloys.

The driving force of this mechanism is the minimization of interfacial energy through the Gibbs Thomson effect. A composition gradient in the matrix between precipitates of different curvature radii induces a diffusion flux from the small precipitates towards the large ones leading to the shrinkage and disappearance of small precipitates and growth of the larges ones. Thus eventually leads to a decrease of the free energy associated with the overall matrix-precipitate interface area.

## 4.4.4 Precipitate dissolution rate derived from the JMAK model

The precipitate area fraction as a function of time and temperature has already been established using a JMAK model given by Eq. 4.4 where the precipitate area fraction is described as a function of the initial and the equilibrium fractions.

The precipitate dissolution rate  $v_{spp}$  (defined here as the rate at which the precipitate interface moves) can then be described using the following relation:

$$v_{spp}(t,T) = \frac{S}{2\pi\overline{r}_{spp}(t,T)N_{spp}(t,T)}\frac{df_{eq}(T)}{dt}$$

$$= -\frac{S}{2\pi\overline{r}_{spp}(t,T)N_{spp}(t,T)}\frac{f_1}{t_1} \cdot exp\left(-\frac{t}{t_1}\right),$$
(4.5)

with S the total considered area,  $\bar{r}_{spp}(t,T)$  the arithmetic mean precipitate radius and  $N_{spp}(t,T)$  the remaining number of precipitates. This velocity expression will be introduced in the full-field simulations presented in chapter 5 to reproduce the precipitate interface velocity observed experimentally.

A phenomenological model can also be used to represent the precipitation and Ostwald ripening mechanisms as presented in appendices B and C, respectively.

#### 4.4.5 Grain size evolution at subsolvus temperatures

The smaller the primary  $\gamma'$  phase fraction in the microstructure, the larger the average grain size. This result has already been evidenced numerous times in different alloys [183, 9, 184, 185, 186, 14] and can be explained by the SZP phenomenon.

Figure 4.18 a illustrates the evolution of the experimental arithmetic mean grain size for the AD730 alloy. The grain size does not change significantly when the annealing is performed at a temperature far from  $T_{solvus}$ , as the grains boundaries are blocked by the rather high  $\gamma'$  phase fraction present in the initial material (12.5%). However, for higher temperatures (> 1080°C), where the dissolution of the precipitates becomes significant, the grains may grow more and more, especially when the microstructure is free from precipitates (at  $T_{solvus} = 1110°C$ ).

For the N19 alloy (Figure 4.18 b), a similar behavior is observed, with only slight grain growth at temperatures far below  $T_{solvus}$ . However, for temperatures close to  $T_{solvus}$ , a very significant evolution is noted in the first minutes, but after 1*h* heat-treatment, the average grain size tends to keep the same value. The N19 superalloy behavior is thus different from that of the AD730 for temperatures

close to the solvus domain. It will be necessary to understand why the mean grain size of this PM material behaves differently and then consider this effect in the simulations to compare the experimental data with the numerical results.



Figure 4.18: Mean grain size after subsolvus heat-treatments for a. AD730 for b. N19 superalloys.

### 4.4.6 Supersolvus experimental data used for the identification of GB migration parameters

The procedure used to obtain values of the GB migration parameters that are consistent with the experimental data for each material is presented below. Supersolvus data are then used so that the grain growth is mostly controlled by the GB properties, without any interference with the SPPs.

The main parameter controlling the migration of a GB submitted to a driving force is then its mobility. The temperature dependence of the GB mobility can be approximated through an Arrhenius law as presented in Eq. 1.4.

Here the mobility is considered to be the same for all GB, independently of their crystallographic properties. The values of M,  $M_0$  and  $Q_m$  can be obtained from the experimental GG data when there are no precipitates present in the material to hinder GB motion, i.e., at the supersolvus domain.

The procedure used to calculate  $M_0$  and  $Q_m$  (Eq. 1.4) can be summarized by the following steps [187]:

• Supersolvus heat treatments were performed at different temperatures for several holding times for the AD730, up to  $50^{\circ}C$  above the  $T_{solvus}$  and for the N19 material, up to  $30^{\circ}C$  above  $T_{solvus}$  as presented in Figure 4.19. It can be noticed again, here in the supersolvus domain, that the grain size of the N19 alloy stagnates at about 20  $\mu m$  whereas the grain size of the AD730 alloy keeps increasing with time and temperature.



Figure 4.19: Grains mean size after supersolvus heat treatments for a. AD730 and for b. N19 superalloys.

This stagnation could be due to the presence of Prior Particle Boundary (PPB) that block the grains. Such kind of additional precipitate population is common in PM materials and they have been largely studied experimentally [188, 189, 190]. The PPB form at the surface of the initial powder grains and they usually consist of oxides, carbides, or oxy-carbides. It is commonly accepted that PPB particles result from segregations or surface contamination. A slight dissolution [191] and coarsening [192, 193] of these precipitates have been observed in materials submitted to high temperatures. These precipitates will be further studied in the next Chapter.

• A first approximation of the reduced mobility  $(M\gamma_{BT})$  is obtained for each temperature by the best fit of the experimental data with a Burke & Turnbull law (BT) [194]:

$$\overline{R}^2 - \overline{R}_0^2 = \frac{1}{2} (M\gamma)_{BT} t.$$
(4.6)

with  $\gamma$  the grain boundary energy set here and in the following to 0.6  $J/m^2$ , which corresponds to GB energy in pure nickel at 1000°C [195].

• As the reduced mobility  $(M\gamma)$  is actually model-dependent [187], these values will have to be further refined based on full-field simulations as described in sections 5.3.1 and 5.3.2.

## 4.5 Summary and discussion

In this chapter, we presented the material fabrication process of the two studied superalloys. The AD730 which are produced by the cast and wrought route which leads to a material with relatively heterogeneous microstructure.

On the other hand, the N19 material is produced by powder metallurgy, a route which gives a more homogeneous and fine final microstructure with a higher content of precipitate population. The initial particle population has similar size as the initial grains in the N19 alloy. The N19 material has a higher  $\gamma' T_{solvus}$ , which is a great advantage because the material can be used at higher temperatures without degrading the material properties.

The studied materials were presented as well as the experimental methods used to get data regarding the evolution of precipitates and matrix grains during annealing at different temperatures and times. The experimental plan was detailed for each material as well as the aim of each set of experiments. A phenomenological model was also established for describing the evolution of precipitate area fraction and size as a function of time and temperature by combining *Thermo-Calc* simulations and a JMAK model [45, 182].

The identification of the precipitates and grains characteristics is essential and has been done with particular attention to run the full-field simulations and compare the numerical results with the experimental data to validate the simulations (see the next chapter).

This study was also extended for the René 65 material in [22] in the context of the PhD thesis project of I. Soares Janeiro. The obtained curve to reproduce the experimental behavior of the AD730 can be used to describe René 65 behavior, for temperatures in the 50°C range below the  $T_{solvus}$ . This shows that the overall area fraction evolution is similar for both cast-and-wrought materials at the equilibrium and transition state due to similar chemical composition and solvus temperatures.

## Résumé en Français du Chapitre 4

Dans ce chapitre, nous avons présenté le processus de fabrication des deux superalliages étudiés. L'alliage AD730 est élaboré par la voie dite « coulé-forgé » qui mène à des hétérogénéités de microstructure résiduelles dans les billettes du fait des très fortes teneurs en éléments d'alliages. Ces hétérogénéités se traduisent par la présence de grains recristallisés relativement fins et équiaxes (majoritaires) et de longs grains non recristallisés, avec un état de précipitation de la phase  $\gamma'$ différent dans les deux types de grains. Dans ce travaux de thèse, nous avons travaillé sur la partie recristallisée équiaxe exlusivement. D'autre part, le matériau N19 est produit par métallurgie des poudres. Ce procédé de fabrication donne une microstructure finale plus homogène et plus fine. La fraction de phase  $\gamma'$  dans l'alliage N19 est plus élevée que dans l'alliage AD730 du fait de leurs compositions chimiques. La taille des précipités  $\gamma'$  est également plus élevée et leur morphologie est moins régulière dans l'alliage N19, en raison notamment de l'agglomération des précipités primaires. Enfin l'alliage N19 présente une température de solvus de la phase  $\gamma'$  supérieure (valeur effectives constatées après 4h de traitement thermique : 1050°C pour l'alliage N19 et 1108°C pour l'alliage AD730).

Dans ce chapitre, le plan d'expériences réalisé au cours de la thèse (traitements thermiques) est également présenté ainsi que les méthodes d'analyse microstructurale quantitative employées. Les résultats sont analysés afin d'établir des lois décrivant la cinétique d'évolution de la phase  $\gamma'$  primaire et la cinétique de croissance de grains dans chaque alliage. Ainsi, un modèle phénoménologique a été établi pour décrire l'évolution de la fraction de phase  $\gamma'$  en fonction de la température et du temps de traitement, en s'appuyant sur une description phénoménologique des résultats de simulations ThermoCalc et le modèle de JMAK Une loi d'évolution de la taille des précipités (ou vitesse de migration de leur interface) a ensuite été déduite du modèle précédent. Cette dernière sera employée pour faire évoluer les précipités dans les simulations en champ complet présentées dans le chapitre 5.

La cinétique de croissance de grains est décrite à la fois en présence de précipités en cours de dissolution (dans la plage de 50°C sous la température de solvus) et dans le domaine supersolvus. Une première estimation de la valeur des paramètres régissant la cinétique de migration des joints de grains a été déduite d'un modèle simple de croissance de grains (modèle en champ moyen de Burke & Turnbull). Ces valeurs devront être affinées dans le chapitre 5 car elles sont modèles-dépendantes. Pour l'alliage N19, la croissance de grains stagne à une valeur de taille des grains de l'ordre de 20  $\mu m$ , même dans le domaine supersolvus. Ce phénomène connu en métallurgie des poudres est attribué à la présence de très fines particules initialement présentes à la surface des grains de poudre. Leur présence et leur effet devra être pris en compte dans les simulations en champ complet du chapitre 5. Enfin, le travail présenté dans ce chapitre a également été réalisé pour l'alliage coulé-forgé René 65, dans le cadre de la thèse d'Ilusca Soares Janeiro (en cours, Chaire TOPAZE). Les trois alliages, AD730, René65 et N19, sont inclus dans un article qui a récemment été publié [22]. Les deux alliages coulés-forgés (qui ont par ailleurs des compositions chimiques relativement proches) se comportent de manière très similaire, les évolutions de la phase  $\gamma'$  pouvant être décrites avec la même loi.

## Chapter 5

## Full-field simulations of grain growth along with second phase precipitate dissolution

The dissolution kinetics of the primary  $\gamma'$  precipitates during subsolvus solution treatments were investigated for two nickel base superalloys (*AD*730 and *N*19) in previous chapter 4. A temperature and time dependency equation was established to describe the evolution of primary  $\gamma'$  precipitates of each material using experimental data, *Thermo-Calc* simulation results and a Johnson-Mehl-Avrami-Kolmogorov (JMAK) model.

Here, the dissolution kinetics of precipitates is simulated using the Level-Set (LS) method (presented in chapter 3) and the former phenomenological model established in chapter 4. These simulations aim to predict grain size evolution in the transient and stable states while primary  $\gamma'$  precipitates are dissolving. Furthermore, it is illustrated how a population of Prior Particle Boundaries (PPB) particles can be considered in the numerical framework in order to be predictive for the grain size evolution in the powder metallurgy N19 superalloy.

The contents of this chapter have been published in [22].

### 5.1 Introduction

The primary  $\gamma'$  precipitates control the grain size by hindering grain boundary (GB) motion through the Smith-Zener pinning (SZP) mechanism [12, 13, 14, 15]. In order to properly control the grain size during a specific subsolvus solution treatment, the evolution of these primary precipitates as a function of time and temperature must be taken into account.

The dissolution of primary  $\gamma'$  precipitates in nickel base superalloys has mainly been studied experimentally [69, 70, 10, 11, 45, 182] but not so much in full-field numerical works [21, 131, 134] at the polycrystalline scale.

Such simulations may help clarifying, predicting, and controlling microstructure evolution when precipitate dissolution occurs during heat treatments (HT), at subsolvus and near-solvus temperatures. In this PhD work, only the effect of primary  $\gamma'$  precipitates has been considered, as secondary and tertiary ones are dissolved during the considered subsolvus solution treatments.

The full-field models are of great interest as they can reproduce microstructure evolutions and heterogeneities. Several methods have been developed to reproduce the SZP mechanism at the polycrystalline scale, considering the presence of SPP in the microstructure as presented and detailed in chapter 2.

All these full-field approaches can integrate SPP into their formulation. A spherical shape to describe them is commonly adopted, although the influence of more complex shapes has also been investigated [81, 117, 152]. In some cases, real precipitate morphology, obtained from Electron BackScatter Diffraction (EBSD) maps [174, 196, 196], secondary electron (SE) images [152, 141] or backscattered electron (BSE) images [152], has been used. However, all these works consider the precipitates as static objects. This reasoning is valid at low temperature (relatively to  $T_{solvus}$ ) where precipitates do not evolve (or very slowly) or at high temperature where precipitates dissolve very fast, in which case SZP does not need to be modeled at all. However, at a temperature just below  $T_{solvus}$ , precipitate dissolution and grain growth can be concomitant. Moreover, the Ostwald ripening mechanism can also take place at long annealing times.

To our knowledge, very few numerical works consider the precipitate evolution, using the PF method [115, 122] or the LS approach [21]. This work adopts the LS approach presented in [21], where a special treatment at the precipitate/GB interface is made to model incoherent precipitates (i.e., assuming the precipitate/GB contact angle to be  $90^{\circ}$ ).

The results of the LS full-field simulations are presented and compared with the experimental data presented in chapter 4 for validation.

## 5.2 Full-field model description

The full-field modeling method was presented in section 2.1.5 and 3.2. Here it is used to reproduce the full-field simulations of the GG process under the influence of the primary  $\gamma'$  precipitate dissolution, the later being described thanks to the calibrated phenomenological model presented in Chapter 4.

#### **Description of SPP**

The multiple junctions treatment based on [150] has been applied both for 2D and 3D simulations of GG in single phase materials or with static SPP using the LS approach [141, 162, 35, 143]. A Modified Multiple Junctions Treatment (MMJT) has recently been proposed in [21] and presented in section 3.2.3 to consider the precipitates as a LS function in FE-LS GG modeling. This new description of the particles opens the possibility of making SPP evolve. The MMJT is described by Eq. 3.2.

Without SPP, the MMJT is equivalent to the numerical treatment described by Eq. 2.9. When SPP are present, it enables by successive iterations to impose the Young-Herring equilibrium for incoherent SPP (see [21] for further details).

#### SPP interface velocity

To allow the precipitate interfaces to move, a convective equation is considered for  $\varphi_{SPP}$  as described in section 3.4.1. A SPP interface velocity,  $v_{spp}$ , is introduced at the SPP interfaces in order to reproduce the precipitate dissolution given by the phenomenological model previously developed in section 4.4.4. More precisely, a continuous velocity field, v is applied through the Laplacian equation Eq. 3.4 considering Dirichlet boundary conditions [197] at the precipitate interfaces. In the end, this smoothed velocity field v is implemented to calculate the velocity  $\vec{v}$  oriented towards the center of each precipitate (see Figure 3.18).

## 5.3 full-field simulations for the refinement of GB migration parameters and of GG

Before performing GG simulations, the parameters controlling the GB migration of the considered material must be identified for each alloy to provide the best agreement between the numerical results and the experimental data.

#### 5.3.1 AD730 superalloy

#### a. Parameter identification and validation

Section 4.4.6 describes the procedure used to calculate a first estimate of  $M_0$  and  $Q_m$  which needs to be further refined as detailed below.

Supersolvus heat treatments were performed between  $1120^{\circ}C$  and  $1160^{\circ}C$  for several holding times (see Figure 4.19 a). The first approximation of  $M\gamma_{BT}$  obtained through Eq. 4.6 was then used to run the full-field simulations, where a large number of grains must be considered in order to be as representative as possible. Consequently a highly performing Lagrangian model called ToRealMotion (TRM) [99] was preferred to the LS methodology in this identification to improve the "Number of grains/calculation time" ratio. In the case of the AD730 alloy, 180000 initial grains were simulated, with an arithmetic mean diameter  $\overline{D} = 9.2$  $\mu m$  and a domain size of 4.3 mm x 4.3 mm. Then, a  $L^2$  error minimization was made between the numerical results and experimental solvus data to obtain the fitted  $M\gamma$  value. Finally, the  $Q_m$  and  $M_0$  parameters were refined using the new  $M\gamma$  values and the Arrhenius law, leading to  $M_0 = 2.9 \cdot 10^{25} m^4/J \cdot s$ ,  $Q_m = 9.8 \cdot 10^5 J/mol$ .

TRM supersolvus GG simulations were again performed with the identified GB mobility parameters, then compared with the experimental data of the AD730 material. Figure 5.1 presents the results of supersolvus GG simulations at supersolvus temperatures for the AD730 material.



Figure 5.1: GG experimental data and simulations for different supersolvus heat treatments, to calibrate and validate the GB migration parameters  $M_0$  and  $Q_m$  values for the AD730 superalloy.

The results are in good agreement with the experimental points. It can therefore be considered, that the approach used to identify the GB properties gave a good approximation of  $M_0$  and  $Q_m$  values at the supersolvus domain and they will be extrapolated to the subsolvus domain. Once the material parameters have been validated, we can proceed with the full-field GG simulations taking into account the identified parameters in this section and the  $\gamma'$  phase dissolution described in section 4.4.2.

#### b. GG with primary $\gamma'$ precipitate dissolution

A pre-simulation was made in order to generate an initial numerical microstructure similar to the one observed experimentally. Where the SPP are mainly located at the GB. Thus, a short heat treatment of  $400s \approx 6 \min$  at  $1070^{\circ}C$  was simulated starting from an initial smaller mean grain size than the one measured experimentally in order to get the boundaries stopped at the SPP.

The resulting initial state (after 400s) presented in Figure 5.2 consists of about 13000 grains initially generated with a LS Laguerre-Voronoi tessellation [165]. The arithmetic mean grain radius is then equal to  $\overline{D} = 9.4 \ \mu m$ . An initial polydisperse and spherical precipitate population was generated by using the experimental  $\gamma'$  precipitate size distribution, an initial area fraction  $f_{spp} = 12.5\%$  and a mean size  $\overline{D}_{spp} = 2.1 \ \mu m$  (about 27000 SPP were generated). All SPP are considered to be incoherent.



Figure 5.2: Initial numerical microstructure of the AD730 superalloy.

The size of the domain is  $1 mm \ge 1 mm$ , and a time step of 5s was chosen to correctly describe the precipitate/GB interaction.

The simulation parameters are  $M_0 = 2.9 \cdot 10^{25} \ m^4/J \cdot s$ ,  $Q_m = 9.8 \cdot 10^5 \ J/mol$  as calculated in section 5.3.1.a,  $\gamma = 0.6 \ J/m^2$  [195], and  $T_{solvus} = 1108^{\circ}C$ , as determined in section 4.4.1.

Different isothermal heat treatments of 4 hours were considered from  $T = 1060^{\circ}C$  (i.e.,  $T_{solvus} - 50^{\circ}C$ ) up to the solvus temperature. The precipitate interface velocity  $v_{spp}$  is calculated using Eq. 4.5.

The evolution of the primary  $\gamma'$  precipitate area fraction for the AD730 alloy is given by Eqs. 4.1 and 4.4. The Figure 5.3 presents the results obtained from the LS simulations. Regarding the precipitate area fraction evolution (Figure 5.3 a), the full-field simulations reproduce, as expected, the phenomenological model. Most importantly, these results agree with the experimental grain size evolution, even if the mobility was extrapolated from the supersolvus domain to the subsolvus domain.

Figure 5.3 b illustrates the arithmetic mean precipitate size evolution. For the numerical treatment at 1060°C where the primary  $\gamma'$  phase fraction does not change, neither does the size of the precipitates. For temperatures between 1070°C and 1090°C, the size of the precipitate decreases during the first minutes due to the dissolution mechanism, but then the average size increases as the smallest precipitates disappear and eventually reach a stable value. Finally, for the temperatures 1100°C and 1110°C, the dissolution is more significant and the mean size of the precipitates decreases until the  $\gamma'$  phase fraction stops evolving, but for  $T_{solvus} = 1108°C$ , the precipitates were mostly dissolved after 1*h* of annealing.

As discussed in the experimental section 4.4.3, the Ostwald ripening mechanism is likely to occur after 1h when the  $\gamma'$  phase fraction remains constant. However, this phenomenon is not taken into account in the mean-field model used to describe the evolution of the precipitates thus nor in the full-field simulations. Nonetheless, the Ostwald ripening can also be considered with a phenomenological approach as detailed in Appendix C. The evolution of the numbers of precipitates and grains along the simulation is shown in Figure 5.3 c,d respectively.

Finally, Figure 5.3 e illustrates the arithmetic mean grain size evolution for all tests. When the heat treatment is far from the  $T_{solvus}$  (i.e.,  $1060^{\circ}C$ ,  $1070^{\circ}C$ ) the SPP completely blocks the grains boundaries. However, for temperatures between  $1080 - 1100^{\circ}C$  where the precipitates dissolve, the grains can grow until they find their new equilibrium state, which is reached when precipitate dissolution ends (if the Ostwald ripening mechanism were taken into account, the grains would be expected to keep evolving slowly, as the increase of mean precipitate radius at constant fraction reduces the pinning pressure and releases the GB). For the HT at the solvus temperature ( $1110^{\circ}C$ ), the precipitates dissolve rapidly and the grains can grow freely once no precipitate remains. These tests show that the used numerical framework can consider the evolution of SPP and predict the grain size not only at long annealing times when the SPP have reached their equilibrium fraction but also in the transient regime. These results validate the possibility of reproducing the precipitate dissolution with a phenomenological model used in LS full-field simulations to predict the material behavior with a good compromise between accuracy and simplicity.



Figure 5.3: Simulation results for the AD730 superalloy for different temperatures from  $T_{solvus}$  to 50° below it.

#### 5.3.2 N19 superalloy

#### a. Parameter identification and validation

Since the N19 is a PM superalloy, the identification procedure was slightly different. This type of alloy has indeed a specific GG evolution. At supersolvus temperatures, the mean grain size evolves up to a maximum size (about 20  $\mu$ m) and then remains constant. Thus, the identification process was only carried out during the first 20 minutes of heat treatments, where GG was observed to occur. The supersolvus treatments were made at 1160°C and 1180°C, once the first approximation of  $M\gamma_{BT}$  was obtained through Eq. 4.6. The first sets of TRM simulations were run with the precalculated parameters, 40000 initial grains were considered, with arithmetic mean diameter  $\overline{D} = 3 \ \mu m$  and a domain size of 0.5  $mm \ge 0.5 \ mm$ . Then, a  $L^2$  error minimization was made using the numerical results and the experimental supersolvus data to obtain a fitted  $M\gamma$  value. Finally, the Arrhenius law was used to calculate the parameter values of the N19 material, which are  $M_0 = 0.51 \ m^4/J \cdot s$  and  $Q_m = 3.3 \cdot 10^5 \ J/mol$ .

Figure 5.4 presents the experimental data and the supersolvus simulations (dashed lines) obtained with the identified properties. These simulations only match the experimental points at the HT onset. These results show a lack in the material description that we need to consider to reproduce and predict the real microstructure evolution.



Figure 5.4: GG simulations at supersolvus temperatures to calibrate and validate the identified the GB migration parameters  $M_0$  and  $Q_m$  values for the N19 superalloy.

The reason for the grain size stagnation is likely to be that Prior Particle Boundary (PPB) particles block the grains boundaries as often observed in PM materials. Then, to correctly predict GG, it is necessary to consider this additional type of particle population in the simulations.

#### b. Simulation of Prior Particles Boundary (PPB) particles

Some numerical works have been developed to account for the presence of PPB, for instance, using a particle packing in [198] or a FE framework in [199].

Here the PPB particles will be treated as a population of oxides that will not evolve. Thus, they will be considered as static particles located at the interfaces of the initial powder grains. Therefore, to generate the PPB network, a Laguerre-Voronoi tessellation was used considering the experimental size distribution (in 3D) of the initial powders (mean  $D_{powder} = 25 \ \mu m$ ), followed by the generation of the PPB particles at the boundaries of the tessellation in 2D (see Figure 5.5). The PPB particles are considered as a new distance function in our LS approach. Once the PPB particles were generated, the actual initial polycrystal was statistically generated (several grains/crystals per powder grain), and finally, the GG simulation was performed.

The validation of the identified properties for the N19 material was made once again with the new considered topology with  $f_{PPB} = 2\%$  and a homogeneous PPB particle size  $D_{PPB} = 1 \ \mu m$ . Although these values are large compared to the actual characteristics of the PPB [191, 192], they enable, in Smith-Zener meaning (i.e. by respecting the ratio  $f_{PPB}/D_{PPB}$  while increasing by a factor of about 10 the respective values of  $D_{PPB}$  and  $f_{PPB}$ ) to respect a representative pinning pressure. In terms of numerical cost of the meshing adaptation and subsequent simulations, it would not be realistic to consider a large number of nanoparticles.

The results are presented in Figure 5.4 (solid lines). The mean grain size prediction is in very good agreement with the experimental data, where the PPB particles effectively block the grain boundaries. The predicted final mean grain size (about 20  $\mu$ m) corresponds approximately to the mean size of the initial powders as a result of the strong pinning pressure exerted by the small PPB particles.

Once the material parameter values have been established, the full-field GG simulations can be performed, taking into account the identified GB migration parameters, the  $\gamma'$  precipitate dissolution described in section 4.4.2, and by considering or not PPB particles.

#### c. GG with primary $\gamma'$ precipitate dissolution

A pre-simulation was also made as for the AD730, but for a HT of 100s at 1110°C. The initial state (after 100s) consists of around 33000 grains initially generated with a LS Laguerre-Voronoi tessellation [165]. The arithmetic mean grain diameter  $\overline{D} = 3 \ \mu m$  and an initial spherical primary  $\gamma'$  precipitate population with a area fraction  $f_{spp} = 18.5\%$  and a mean size  $\overline{D}_{spp} = 1.3 \ \mu m$  (about





Figure 5.5: Successive zooms illustrating the initial statistical microstructure used to describe the N19 superalloy with the black PPB particles.

32000 incoherent SPP) are considered.

The size of the domain is 0.5 mm x 0.5 mm (see Figure 5.6 but without the red particles), and the time step was 5s. The simulation parameters were  $M_0 = 0.51 m^4/J \cdot s$ ,  $Q_m = 3.3 \cdot 10^5 J/mol$  as calculated in section 5.3.2.a,  $\gamma = 0.6 J/m^2$  [195] and  $T_{solvus} = 1150^{\circ}C$  as determined in section 4.1.3.

Simulations considering or not the PPB particles, in addition to the evolving  $\gamma'$  precipitates, were carried out to understand how they affect the GG behavior.

#### • Simulations without considering the PPB particles

Different isothermal heat treatments (4h) around  $T_{solvus}$  were simulated. The primary  $\gamma'$  precipitate interface velocity was calculated according to Eq. 4.5.



Figure 5.6: Initial state of the microstructure used in the simulations for the N19 material with  $\gamma'$  precipitates (black) and PPB particles (red).

The primary  $\gamma'$  phase area fraction evolution for the N19 was obtained by Eqs. 4.2 and 4.4.

Figure 5.7a shows that the  $\gamma'$  phase area fraction evolution matches the phenomenological model and the experimental data. The mean primary  $\gamma'$  precipitate size evolution is presented in Figure 5.7b where there is a slight decrease at the beginning of the simulation. Then, as the small precipitates continue to dissolve, the mean grain size tends to increase. Figure 5.7c,d illustrate the number evolution of the  $\gamma'$  precipitates and grains, respectively. The GB kinetics is so fast that it makes the smallest grains disappear during the first minutes of the simulations.

Finally, Figure 5.7e shows the arithmetic mean grain size evolution for all simulations. As discussed before, the mean grain size evolves during the  $\gamma'$  precipitate dissolution. However, the grain size reaches a stable value once the dissolution stops. As expected, a grain size larger than the one observed experimentally is reached because *PPB* particles were not considered in these simulations. Indeed, for the HT at 1150°C, the predicted grain size continues to increase contrary to the experimental points that stagnate at about 20  $\mu m$ .

#### • Simulations considering the PPB particles

The PPB particles were generated at the Laguerre-Voronoi tessellation boundaries based on the experimental granulometry of the initial powder of this material (see section 3.2). The initial state is the same used previously but with an additional static particle population ( $f_{PPB} = 2\%$  of size  $\overline{D}_{PPB} = 1 \ \mu m$ , red particles on Figure 5.6 ).

Figure 5.8 illustrates the microstructure evolution for some HT at different times and temperatures along the simulation and the results are summarized in Figure 5.9.

The  $\gamma'$  area fraction evolution (Figure 5.9a) is similar to the one of (Figure 5.7a) because the primary  $\gamma'$  interface velocity is the same. Considering the static PPB particles, the predicted arithmetic mean grain size is in much better agreement with the experimental data as illustrated in Figure 5.9c. The LS simulations describe very well the limiting grain size independently of the temperature.

Figure 5.10 illustrates the microstructural evolution for both the considered cases (with or without PPB particles) at  $1150^{\circ}C$ , and allows us to visualize how the grains continue to grow when no  $\gamma'$  precipitate remains and PPB particles are not taken into account. On the contrary, when static PPB particles are simulated, the grains boundaries are blocked by those particles, limiting the mean grain size slightly below the initial powder size. These results can be better observed in Figure 5.11 where the grain size histogram evolution are compared with or without PPB particles.



Figure 5.7: Simulation results for the N19 superalloy without considering the PPB particles.



Figure 5.8: GG simulations for the N19 material with both evolving primary  $\gamma'$  precipitates (black) and static *PPB* particles (red) at subsolvus temperatures for different heat treatment and times (200s, 500s, and 14400s). Zoomed area of the 0.5 mm x 0.5 mm simulation domain.



Figure 5.9: Simulation results for the N19 superalloy taking into account both the evolving primary  $\gamma'$  phase and the static PPB particles.



Figure 5.10: Comparison of the simulated microstructure evolution of the N19 superalloy considering or not the PPB particles (red) at  $1150^{\circ}C$ . Zoomed area of the 0.5 mm x 0.5 mm simulation domain.



Figure 5.11: Comparison of the grain size distribution evolution of the N19 superalloy during the simulation without and considering the PPBs.

## 5.4 Summary and discussion

A new approach using a mean-field model to describe the dissolution of second phase particles was introduced in a finite element framework using the LS model to simulate grain growth in  $\gamma/\gamma'$  nickel base superalloys as primary  $\gamma'$  precipitates dissolve at temperatures close to  $T_{solvus}$ .

The LS simulation results respect the imposed phenomenological equation describing the precipitate dissolution rate, and thus, there is a good agreement with the experimental data concerning the second phase precipitate evolution. These results confirm the possibility of considering a simple phenomenological model to describe the precipitate evolution to get good accuracy in terms of the predicted precipitate and grain sizes, including both the transient regime at short times and the steady-state at longer annealing times.

Moreover, the possibility of considering the Prior Particles Boundaries (PPB) particles has been introduced in the simulations of the powder metallurgy N19 alloy. The numerical results are consistent with the observations, where these PPB particles limit the mean grain size to about the mean size of the initial alloy powder at any temperature.

## Résumé en Français du Chapitre 5

Ce chapitre présente les résultats de simulation en champ complet de la croissance de grains dans les deux alliages étudiés alors que les précipités  $\gamma'$  primaires se dissolvent progressivement, dans la plage de température de 50°C sous le solvus. Ces simulations sont réalisées par la méthode Level-Set introduite dans le chapitre 3, avec une vitesse de dissolution des précipités décrite par le modèle phénoménologique établi dans le chapitre 4. Les particules de seconde phase sont ici décrites par une fonction Level-Set spécifique supplémentaire, ce qui permet de les faire évoluer au cours du temps.

Les paramètres contrôlant la cinétique de migration des joints de grains, dont une première valeur avait été estimée par le modèle de Burk & Turnbull dans le chapitre 4, ont été affinés en réalisant des simulations de croissance de grains en champ complet par la méthode TRM (qui permet de traiter de très grands nombres de grains dans des temps raisonnables). Ces valeurs de paramètres affinées ont ensuite été utilisées dans les calculs de croissance de grains avec dissolution concomitante des précipités, par la méthode Level-Set. Les résultats obtenus reproduisent très bien les évolutions de tailles de grains observées expérimentalement pour l'alliage AD730, non seulement en régime stationnaire après recuits de longue durée, mais également dans le régime transitoire où les précipités sont en train de se dissoudre progressivement.

Pour l'alliage N19, dont la taille de grains est limitée par la présence de particules aux interfaces des grains de poudres originaux (dites particules PPB), les simulations ne prenant pas en compte la présence de telles particules mènent bien entendu à des tailles de grains excessives lors de long recuits.

Elles ont donc été décrites avec une population supplémentaire de particules dont les attributs de taille et de fraction surfacique ont été choisis de manière à respecter une pression d'ancrage de Smith-Zener équivalente, toute en ne pénalisant pas trop les temps de calcul. Ces particules supplémentaires sont statiques à la différence des précipités  $\gamma'$  qui évoluent, selon le modèle phénoménologique calibré au chapitre 4 pour l'alliage N19. Les résultats de simulation reproduisent très bien les résultats expérimentaux en termes de taille de grains, dans le régime transitoire de dissolution de la phase  $\gamma'$  comme en régime stabilisé où la taille de grains stagne du fait des particules PPB. Ces simulations en champ complet respectent également évidemment les cinétiques expérimentales d'évolution des précipités  $\gamma'$  imposées par le modèle phénoménologique. CHAPTER 5. SIMULATIONS WITH PRECIPITATE DISSOLUTION
# Chapter 6 Conclusions and outlooks

The main objectives of this work was to be able, at the mesoscopic scale, to simulate the second phase particles evolution and to study the dissolution of the primary  $\gamma'$  particles in the AD730 et N19 nickel base superalloys experimentally in order to reproduce them numerically and thus correctly simulate the grain growth of these materials.

When the superalloys are subjected to heat treatments, the particles phase tends to evolve, and their evolution plays an essential role in the microstructure behavior. For this reason, it is vital to reproduce their kinetics in the numerical framework to correctly predict the material's behavior in terms of the size and distribution of grains and particles.

In chapter 1, the different mechanisms responsible for the particle evolution were described. It was detailed that even when the fraction of the particles remains constant, the particles can present evolution in their morphology and distribution. Until now, none of these mechanisms could be represented numerically in the LS full-field framework because the particles were represented as static holes in the domain.

However, it is possible to represent some of these mechanisms with other numerical methods, such as Monte Carlo, Cellular Automata, or Phase Field, as presented in chapter 2. For this PhD work, it was decided to concentrate mainly our developments on the LS method; even if it is not the fastest method, this method is the most versatile because it can treat most of the physical phenomena occurring during material manufacturing process, even in the complex context of large deformations. Therefore, a new methodology capable of simulating the evolution of incoherent primary particles  $\gamma'$  was proposed and developed in a LS context.

This new methodology consists in representing the particles as a new distance function different from that of the grains, and in modifying the multiple junctions treatment in such a way that it takes into account the particle interface to eliminate the voids or overlapping that have appeared at the GB and their junctions after the resolution of the convective/diffusive equations. Some tests were carried out to guarantee the orthogonality contact of the particle/GB interface and the possibility of reproducing different particle shapes in the new formalism. The results were compared with the former method (SPP represented as holes), obtaining an excellent equivalence between them. Additionally, in the new formalism, it was possible to reproduce the dissolution of the particles thanks to a convective equation applied to the particle distance function. The velocity is then smoothed with a Laplacian equation considering Dirichlet limit conditions at the particle interfaces. Finally, the previously established velocity is used to obtain the velocity field oriented towards the interior of each particle (dissolution) that must be applied to the distance function of the particles.

Once it was established how to consider the evolution of the particles in the LS-FE methodology, the first tests with polycrystals were carried out to study the impact of the dissolution rate.

The particle evolution in a polycrystals case was developed for the first time with the LS model. The results clearly showed the strong influence of the precipitates evolution on the Smith-Zener pressure, obtaining a significantly smaller mean grain size when the evolution of the particles is not considered than when the evolution of these precipitates is taken into account in the simulations. It was also observed that the distribution of the particles (bimodal and heterogeneous distribution) could trigger abnormal grain growth in the microstructure due to the earlier disappearance of particles in certain areas.

After developing the numerical part, an identification process was carried out to find the grain boundaries motion parameters, thus obtaining the values of activation energy  $(Q_m)$  and the mobility prefactor  $M_0$  for both materials of interest.

Then, different heat treatments were carried out to study the dissolution of the primary particles in the studied  $\gamma/\gamma'$  superalloys (AD730 and N19). Isothermal treatments were carried out for each material at different times to quantify the transitory and equilibrium state dissolution. The Thermo-Calc software was used to establish the  $\gamma'$  phase evolution of each superalloy as a function of the temperature at the equilibrium state (4h) to obtain the solvus temperature of each material. The Thermo-Calc curves were compared with the experiments and it was observed that the experimental points described the same behavior as the Thermo-Calc curves. However, it was decided to offset the curve until the experimental points in the case of the N19 alloy, thus, obtaining an effective dissolution temperature for each material of 1108°C and 1150°C for the AD730 and N19 superalloys, respectively.

Once the evolution of the particles was established as a function of temperature, a JMAK phenomenological model was used to introduce the time dependence and thus describe the transient state of the particles dissolution. This simple model allowed us to obtain an equation capable of describing the particle evolution as a function of time and temperature, giving a good agreement concerning the experimental data.

The velocity of the particle interfaces was extracted from the phenomenological model, and was introduced in the full-field simulations, giving consistent results compared with the microscopic observations. This demonstrates that a simple mean-field model can be used to describe the evolution of the particles and then the dissolution kinetics can be integrated into the LS simulations to describe and predict microstructural evolutions correctly. The great advantage of this procedure is that phenomenological models can be used to describe easily different mechanisms such as dissolution, Ostwald ripening, or even precipitation easily and then be integrated into the full-field description as detailed in appendices B and C.

It was also possible to study, analyze and simulate the behavior of PPB particles that are especially present in powder metallurgy superalloys such as the N19 material. It was found that the existing particles at the interfaces of the initial powder particles usually consist of oxides, carbides, or oxy-carbides and they result from segregations or surface contamination. These PPB particles block the grain growth around the size of the initial powder particles, even at the supersolvus domain. So, it is necessary to consider them in the numerical formalism to reproduce the experimentally observed microstructural evolution for these materials.

Although we have provided a precise framework for modeling grain growth in the presence of an evolving primary  $\gamma'$  phase, which allowed us to represent more accurately the microstructure of the nickel base superalloys and open the possibility of reproducing some of the mechanisms that occur during this particle evolution, the work of integrating and simulating different phenomena considering the particle kinetics is far from finished. The methodology must be extended to investigate the ReX, DRX and PDRX phenomena "in the presence of second phase particles. In the case of DRX, a particular behaviour law could be considered to affect different properties to the matrix and to the particles (e.g. hardness) to describe correctly the material forming.

Another important aspect would be to consider a more physical description of the dissolution/precipitation of the second phase particles through the FE modeling of the diffusive solid/solid phase transformation mechanisms. This is the main perspective of the PhD of N. Chandrappa [200].

Another aspect concern the modeling of 3D polycrystals with evolving particles in order to observe the effect of pinning in three-dimensional microstructures. The first tests with particles from a 3D microscopy study were carried out comparing the impact of the experimental morphology of the particles with their statistical representation using spherical particles. The first results are described below in section 6.1.

Finally, as previously detailed, although the LS method is the most adequate to represent the different microstructural phenomena, it is not the fastest numerical method. Indeed, the calculations can take several days and even weeks if 3D simulations are considered, despite that the method was already largely optimized by Scholtes et al. [142]. That is why an extension of the ToRealMotion (TRM) has begun to be developed in order to take into account static and dynamic SPP. The methodology for introducing the particles in this numerical method and comparing specific tests with the LS method is presented below in section 6.2.

## 6.1 3D simulations with real particles morphology first tests

We have received a stack of images used by M. Charpagne in her 3D microstructural study [201] dedicated to the formation of annealing twins during GG. Here we will use the precipitate population data to perform a 3D particle reconstruction to simulate the GG mechanism in the presence of a 3D real morphology particle population with a statistically generated microstructure (grains). Moreover, considering or not the dissolution of the particles and the impact of their shape, a population of spherical particles will be generated that respect the distribution of those observed experimentally to validate whether the shape of the particles may have some influence in 3D simulations.

The data corresponds to the *René* 65 material [201] after 1*h* of heat treatment at 1065°. A TriBeam microscope which consists of a Dual-beam Focused Ion Beam-Scanning Electron Microscope (FIB-SEM) coupled with a Femtosecond laser [202, 203] was used for the data acquisition. 325 BSE images of size 160  $\mu m \times 140 \ \mu m$  were taken, with a pixel size 0.16 in X and Y direction and a Z spacing between slices of 0.5  $\mu m$ .

### 6.1.1 Post-processing and reconstruction

Due to the interaction of the laser, the image quality can decreased, and it can became difficult to perform a correct segmentation of the primary  $\gamma'$  precipitates from the matrix (see Figure 6.1). Therefore, a post-processing of the image was required to correctly describe the precipitates in order to generate the 3D reconstruction of the  $\gamma'$  phase. For this purpose, the ImageJ software [173] was used through a script to improve the image quality as follows:

- the image to be processed is imported,
- a band-pass filter is used to reduce repeated streaks (noise),
- a Gaussian blur filter to blur the image is applied, followed by a minimum filter to dilate the particles,
- the image format is changed to 8 bits, and its contrast is improved,

• images are binarized using a gray level threshold.



Figure 6.1: Post-processing segmentation path for one image of the SPP dataset. a. Raw image, b. Bandpass filter, c. Gaussian blur filter plus minimum filter, d. enhanced contrast filter and e. final segmented image [204].

This procedure is repeated for the entire image stack to make 3D reconstruction possible.

The reconstruction process was carried out in Dream3D software [205]. At first, the data were imported, assigning the thickness of the slices  $(0.5 \ \mu m)$ . Then, an alignment between the slices was performed, followed by the segmentation of particles with a size threshold to erase the smallest particles. As the binarization was performed for 2D images, we can find small entities that are not real precipitates (the selected threshold value was 10 voxels to be considered as a particle). Finally, the reconstructed primary  $\gamma'$  phase is exported with all the characteristics that describe the precipitates, such as equivalent diameter, sphericity, or the number of voxels per particle. The Figure 6.2 shows the final 3D reconstruction of the primary  $\gamma'$  phase with a domain volume  $146 \times 91 \times 164 \ \mu m^3$ , a minimum particle size ( $D_{spp} = 0.33 \ \mu m$ ) and a volume fraction  $f_{spp} = 10\%$ . The post-processing procedure was carried out by F. Jaime [204] as part of his PhD work.

### 6.1.2 3D GG simulations

The reconstructed total domain was divided into 8 subdomains of  $74 \times 91 \times 42$   $\mu m^3$  in order to limit the computational cost of the simulation, but also to avoid visualization difficulties inherent to the large size of the output files.

The eight simulations were averaged to obtain the evolution of the particle fraction and the mean grains size of the entire volume.



Figure 6.2: Final 3D reconstruction of the SPP dataset performed in Dream3D software.

The influence of the particle morphology was studied, immersing the reconstruction of the particles and comparing the results with a population of spherical particles located randomly and generated with the same distribution as the experimental ones. The effect that the particle dissolution has on the 3D grain growth was also analyzed, attributing a constant dissolution speed to the particles and comparing them with the case where the particles do not evolve. A static heterogeneous isotropic mesh was used in order to avoid the numerical cost of 3D remeshing operations. Thus, a fine mesh size  $(h = 5.4 \times 10^{-4} \text{ mm})$  was considered around and inside the particle and a coarser mesh  $(h = 2.8 \times 10^{-3} \text{ mm})$  away from the interfaces.

In the Figure 6.3 it is possible to compare the reconstruction of a particle and its respective numerical representation. It is observed that the pixelized edges of the reconstruction are smoothed in the numerical representation, which makes the particles appear more natural.



Figure 6.3: Comparison of 3D reconstruction and the numerical immersed precipitates.

The obtained results are presented in the Figure 6.4. The evolution of the volume fraction of the particles is similar for both cases (spherical shape and real SPPs). The only difference is that the case of the immersed particles suffers a loss in the SPP volume at the beginning of the simulation due to reinitialization and the initial irregular particle shape. Figure 6.4.b. illustrates the evolution of the mean grain size and it can be seen that the morphology of the particles does not have a great influence on the predicted mean grain size. These results are in

agreement with the 2D study carried out by Agnoli et al. [152]. However, this little influence of the morphology could also be correlated to the semi-spherical shape of the considered SPP. More complex discussions could probably arise for more complex shapes of the precipitates. Figures 6.4.c. and d. illustrates the number of particles and grains throughout the simulation, respectively.



Figure 6.4: 3D Results.

## 6.2 ToRealMotion (TRM) particles integration

In [99, 100], a method was introduced for the modeling of Grain Growth (GG) in a front-tracking context. The method consists of the movement of interfaces using a Lagrangian scheme that updates the positions of the nodes of the mesh defining the GB, while a remeshing procedure treats topological changes and maintains the mesh quality. This Front-tracking method has now been extended to the study of Recrystallization (ReX) [101] and anisotropic grain boundary properties [103, 31]. In this section, a methodology for applying the TRM model to SPP/GB interactions will be presented.

### 6.2.1 Particles of ideal shape and initial discretization

This 2D model uses circles to track the particles, i.e. during our simulations, on top of the data structure of the TRM model presented in [99], a list of circles is added (a circle *i* defined by a position and radius  $x_i, y_i, r_i$ ). These circles are tracked until their radius  $r_i = 0$  or until the end of the simulation.



Figure 6.5: Example of the immersion of circular particles (cyan) in a meshed domain (orange). a. Initial mesh, b. mesh after the remeshing defining the boundaries of particles with an explicit mesh.

Of course, being a front-tracking model, it is necessary to explicitly define the SPP/GB interfaces using a body-fitted mesh, for which, similarly to [101], a remeshing procedure has been constructed: initially, all segments crossed by the boundaries of a circle, are cut using the *edge-split* operator (see Fig. 6.5). Then, using a *surface reconstruction* procedure, [99], the elements contained within the circular domain are extracted from their original surface<sup>1</sup> and are attributed to a new surface representing the discrete particle.

### 6.2.2 Kinetics at SPP/GB interfaces

The movement of multiple junctions is critical during the Smith-Zener pinning phenomenon as they account for most of the SPP/GB interactions. In this model, we have made use of a free-movement/projection approach (see Fig. 6.6): during a time step, nodes belonging to the SPP/GBs interfaces are allowed to move freely

<sup>&</sup>lt;sup>1</sup>In the TRM context, we define a *surface* as a set of elements and nodes. Surfaces typically represent a *grain*, but in this scope, they represent grains and particles.

according to their velocity  $\vec{v_i}$ . This velocity is computed using the methodologies presented in [99, 101, 103, 31], then a projection of these nodes is made to the interfaces defined by the perfect circles. The projection is made to the nearest interface point (in this case, in the radial direction).



Figure 6.6: Example of the cycle of the movement of a multiple junction (MJ) in the boundary of a particle (cyan). The initial position of the MJ is displayed in red by semi-transparent shading in all frames. a. Initial state and computation of the dynamics of the interfaces, b. displacement of the interfaces using the time-step and velocity, c. projection of the MJ of the meshed particle to the circle defining the particle domain, d. and e. new cycle, similar to b. and c.

In this text, the velocity is computed considering isotropic GG, which translates to multiple junctions with a Herring's equilibrium state at  $120^{\circ}-120^{\circ}-120^{\circ}$ . After the projection of SPP/GB MJs, if the circle is sufficiently big, the equilibrium state should be around  $90^{\circ}-180^{\circ}-90^{\circ}$ . Also, it is assumed that the time step is sufficiently low to provide smooth evolutions.

Figure 6.6 shows the typical behavior of a multiple junction (MJ) at a SPP/GB when an adjacent GB is curved. Note that after one cycle of move-project, the MJ has moved from its original position. This is because the immediate adjacent edge of the GB makes an angle more obtuse on the green than on the orange grain, and thus, it makes the MJ move to a more symmetrical position. If this process is repeated several times, a movement of the MJ becomes apparent, as if it were gliding on the particle surface.

Finally, it is also possible to do evolve the particles by changing the radius  $r_i$  of a circle defining the particle. Figure 6.7 illustrates how this is done in the TRM model during a time step: first, the velocity computation and free movement of nodes are done (Figs. 6.7 a. and b.) then the circle defining the particle is shrunk (which is equivalent to reduce the value of  $r_i$ , Fig. 6.7 c.), and finally, the projection procedure takes place (Fig. 6.7 d.).

### 6.2.3 Comparisons between the LS and TRM model

#### a. GG simulations with monodisperse SPP

The first set of simulations consist of four domains of 0.4 mm x 0.4 mm with different Laguerre-Voronoi tessellations [165] with around 1800 initial grain for each domain. The arithmetic mean grain size is equal to  $\overline{D} = 14 \ \mu m$  and an initial monodisperse and spherical precipitate population with an initial surface fraction  $f_{spp} = 5\%$  and an arithmetic mean size  $\overline{D}_{spp} = 2 \ \mu m$  (around 768 SPP were generated), all SPP are considered to be incoherent.

The GG migration parameters were calculated in section 5.3:  $M_0 = 2.9 \cdot 10^{25}$  $m^4/J \cdot s$ ,  $Q_m = 9.8 \cdot 10^5 J/mol$ ,  $\gamma = 0.6 J/m^2$ , which corresponds to the properties of the AD730 superalloy.

An isothermal heat treatment of 3 hours (10800s) were considered at  $T = 990^{\circ}C$ . The precipitate interface velocity were set to  $v_{spp} = 10^{-5} mm/s$ .

Figure 6.8 presents the comparison of the predicted grain size evolution between the TRM and LS model. First, the behavior obtained by the TRM method considering or not the presence and evolution of the SPP is illustrated in Figure 6.8.a. The result are coherent and well reproduce the Smith-Zener pinning. The results show an excellent agreement between the predictions of the two models.



Figure 6.7: Example of the life cycle of the evolution of a particle. a. Computation of the dynamics of the interfaces, b. displacement of the interfaces using the timestep and velocity, c. transformation of the circle defining the particle, here the circle has been shrunk, d. projection of the interfaces of the meshed particle to the circle defining the particle domain.

#### b. GG cases from [21]

For this test, the same case presented in [21] and in section 3.4.2 were reproduce considering static particles. The initial microstructure consists of around 50000 grains generated with a Laguerre-Voronoi tessellation [165] for a domain size of  $2mm \times 2mm$ . The arithmetic mean grain diameter  $\bar{D} = 9.5 \ \mu m$  and an initial monodisperse spherical particle population ( $\bar{r}_{spp} = 2 \ \mu m$ ) with a surface fraction  $f_{spp} = 6\%$  (around 20000 SPPs) were considered. Here we consider the same thermal path [990°C, 1120°C] and grain boundary properties.

The predict results are quite similar for both models. However, the mean grain size predicted by the LS method is slight bigger, then the number on grains



Figure 6.8: Comparison between the TRM and LS models concerning a. the mean grain size evolution b. GG when no particles are considered in the microstructure c. GG with a fixed SPP population d. GG under the effect of particles dissolution. Results for each case are averaged from the results of the four different initial tessellations, with the range of results indicated by the semi-transparent shading of the same color as the corresponding curve.

at the end of the simulation is inferior than the one obtained with the TRM model.

Most important the simulations performed with the TRM model were more than two times faster than one obtain with the LS method. Nevertheless, further analysis must be done considering the precipitate evolution.

### c. Domain size influence

The domain size influence was made to study the convergence of the model, thus optimizing the set of simulations.

The smallest domain contains 500 grains and 130 SPP, and the biggest one 50000 grains and 11200 SPP. Figure 6.10.a. presents the different domains with





Figure 6.9: Comparisons between the LS and TRM model of a polycrystal case with 50000 initial grains. a. Numerical heat treatment. b. Precipitate particle fraction. c. Mean grain size. d. Number of grains. e. Simulation time. f. Number of increments.

the respective grains and particles for each case. Figures 6.10.b and c. illustrate the mean grain size evolution and the L2 error considering the most significant case as the reference case.

Small domains show small errors (around 3 %) compared to the largest domain. This means that it could be preferable in terms of computation resources to run several small test cases as was made in section 6.2.3.a. than one big case presented in section 6.2.3.b. However, these results may be different using dynamic SPP and will need to be discussed further.



Figure 6.10: Domain size evolution: a. different tested cases with the respective number of grains and particles. b. mean grain size evolution and c. the obtained L2 error.

The proposed methodology is therefore promising but still needs to be extended to 3D like the rest of the TRM approach.

## CHAPTER 6. CONCLUSION AND OUTLOOKS

Appendices

# Appendix A Grain and primary $\gamma'$ precipitate data

Tables A.1 and A.2 present all the data concerning the precipitates and the grains for all the HT done for the AD730 and N19 respectively as well as the data obtained from the images provided by the industrial partners Safran and Aubert&Duval.

AD730 heat treatments											
Domain	Temperature [*C]	Holding time [min]	Analyzed images	Precipitates							
				Fspp [%]	Dspp [µm]	Analyzed SPP	D [µm]	Dmax [µm]	Standard deviation [µm]	Analyzed grains	Data
As-received		0	10	12.5	2.0	3293	9.4	59.5	9.1	1980	Cemef
	1060	10	10	10.5	1.9	3091	10.3	43.3	6.0	1081	Cemef
		20	10	11.5	2.1	2725	10.3	94.2	8.3	909	Cemef
		60	10	12.3	1.6	5128					Safran
		240	5	12.4	1.8	1800					Safran
	1070	10	10	10.6	2.1	2473	10.5	132.1	7.7	1120	Cemef
		20	10	9.9	1.9	3040	11.8	116.0	16.4	1036	Cemef
Subsolvus		60	10	10.2	1.4	5464	13.2	93.0	9.8	2066	Cemef
		240	10	10.7	1.6	4490	12.5	95.9	8.1	4119	Cemef
		10	10	10.0	1.9	3049	11.3	194.2	16.4	2161	Cemef
	1080	20	10	9.5	1.9	3089	13.5	126.6	18.3	1621	Cemef
		60	10	8.0	1.4	4430					Safran
		240	10	7.3	1.5	3587					Safran
	1090	10	10	8.5	2.0	2261	14.1	198.8	19.6	3021	Cemet
		20	10	8.3	1.8	2959	16.7	171.9	21.2	2387	Cernet
		60	9	5.5	1.3	2974					Safran
		240	10	5.5	1.5	2/19					Safran
	1100	15	4	8.4	2.0	41/	13.1	52.7	8.8	547	Cemer
		30	4	6.1	2.1	494	13.7	59.9	10.3	638	Cemer
		50	4	2.8	1.2	595	16.9	173.0	12.9	1115	Cemer
		240	10	2.3	1.6	1701	43.9	267.4	46.1	2272	Lemer
<u>8</u>	1110	10	10	5.3	1.9	1/01	27.3	169.8	29.5	1050	Cemer
- No		20	10	4.0	1.7	1408	32.0	200.1	23.2	2070	Cefferen
Š		240	10	0.7	1.2	043	100	218	29	2078	Sarran
		240	0	1.50	1.3	20	022	020	50	202	Camef
Supersolvus	1120	50	10	1.00	12	20	140	495	00.0	1005	Cerner
		240	10	1.00	1.2	12	292	1261	165	475	Safran
	1125	240					97.7	342	33.8	75	Cemef
	1125	00					235	1831	132	982	Safran
	1130	240					387	1144	204	348	Safran
	1140	10					100.0		201	0.0	A&D
		15					95.0	271.7	54.6	112	Cemef
		30					112.70	328.1	70.6	76	Cemef
		60					210.5	711.8	145.1	57	Cemef
	1150	10					146.3			0.	A&D
		20					201.5				A&D
		30					238.7				A&D
		60					375	1128	193	445	Safran
	1160	15					161.8	507.0	108.3	98	Cemef
		30					231.3	769.4	162.7	73	Cemef
		60					302.2	1122.5	213.3	226	Cemef

Table A.1: Heat treatments done for the AD730 superalloy.

N19 heat treatments											
Domain	Temperature [*C]	Holding time [min]	Analyzed images	Precipitates							
				Fspp [%]	Dspp [ µm ]	Analyzed SPP	D [µm]	Dmax [µm]	Standard deviation [µm]	Analyzed grains	Data
As-received		0	5	19	1.2	3725	3.0	12.4	1.2	746	Cemef
	1110	10	10	12.8	1.1	7417	3.4	9.3	1.6	535	Cemef
		30	10	12.0	1.1	6788	3.5	9.2	1.6	704	Cemef
		120	3	9.5	1.2	1619	3.8	5.9	1.9	675	Cemef
		240	5	12.0	1.3	2066	5.0				Safran
		10	10	12.0	1.1	6219	3.7	9.8	1.7	674	Cemef
	1120	30	10	9.0	1.1	5041	3.9	11.1	1.8	594	Cemef
Nus		240	5	10.0	1.4	1746	6.0				Safran
Subsol	1130	10	10	6.1	1.1	3476	5.2	32.4	3.9	642	Cemef
		20	10	5.4	1.2	2746	6.1	39.2	4.8	1531	Cemef
		60	6	6.5	1.3	1731	6.1	30.7	3.8	2055	Cemef
		240	5	5.4	1.5	836	10.0				Safran
	1140	10	10	3.2	1.2	1753	11.2	44.6	7.9	514	Cemef
		20	10	2.7	1.3	1285	11.5	51.8	7.7	530	Cemef
		60	5	2.4	1.3	564	12.2	55.3	8.2	2120	Cemef
		240	5	2.2	1.4	347	15.2				Safran
	1150	1					3.2	11.1	1.2	1637	Cemef
		2					8.1	54.7	1.0	1780	Cemef
		10	10	1.5	1.2	836	15.0	53.9	9.2	1493	Cemef
NUS		20	5	0.8	1.2	168	16.6	56.7	9.5	1270	Cemef
Sol		30					16.7	16.1	10.5	2153	Cemef
		60					17.5	60.1	10.0	2225	Cemef
		120					20				Safran
		240					22				Safran
Supersolvus	1160	1					3.8	13.0	1.6	2287	Cemef
		2					14.6	52.8	9.5	1222	Cemef
		10					18.4	54.0	10.4	1039	Cemef
		20					18.7	64.3	10.2	1022	Cemef
		120					19.9	64.1	10.2	927	Cemef
		240					22				Safran
	1180	1					11.1	51.3	9.6	1713	Cemef
		3					17.6	58.5	10.1	1125	Cemef
		10					20.9	59.3	10.5	844	Cemef
		20					19.8	65.1	9.5	958	Cemef
		120					22				Safran
		240					22				Safran

Table A.2: Heat treatments done for the N19 superalloy.

# Appendix B

# Precipitation in our LS framework

The nucleation model developed by Sholtes et al. [142] was adapted to insert new small particles.

Indeed, even if only particles dissolution was considered in the manuscript, all the developments were realized in order to also consider precipitation in near future. This appendix explains the methodology and provides a first illustration.

The particle distances function  $\varphi_{spp}$  are modified in order to take into account the new inserted particles. These new small particles referred to as «nuclei» can be inserted based on a phenomenological model by inserting a given number of particles at a given time or based on the experimental data, and they can be generated in a predefined position or fulfilling a criterion (energy, particle concentration ...).

This method does not describe the «particle nucleation» in a thermodynamic sense but allows to consider the precipitation by a phenomenological approach.

The initial state presented in Figure B.1 a. consists of about 600 grains generated with a LS Laguerre-Voronoi tessellation [165] for a domain size of 0.3  $mm \ge 0.3 mm$ .

The arithmetic mean grain radius is equal to  $\overline{D} = 12 \ \mu m$  and an initial polydisperse and spherical precipitate population with an initial area fraction  $f_{spp} = 3.6\%$  and a mean size  $\overline{D}_{spp} = 1.5 \ \mu m$  (about 2500 SPP were generated) were considered. All SPP are considered to be incoherent.

A heat treatment of  $1090^{\circ}C$  for 60 min was simulated, with a time step of 5s. The simulation parameters were the one obtained for the AD730 in section 5.3.1:  $M_0 = 2.9 \cdot 10^{25} \ m^4/J \cdot s$ ,  $Q_m = 9.8 \cdot 10^5 \ J/mol$ ,  $\gamma = 0.6 \ J/m^2$  [195].

Two stages can be observed in Figure B.1 c,d. The particle precipitation (nucleation), where the new small particles appear, and the particle growth, where the existing particles can grow following a phenomenological law.



Figure B.1: Numerical representation of the particle precipitation. a. Initial state b. Particle surface evolution c. Final state d. Number of particles.

# Appendix C

# Additional analyses confirming the occurrence of Ostwald ripening

## C.1 Experimental data validation

Isothermal treatments were carried out for different holding times for the AD730 superalloy as described in chapter 4. The dissolution of the particles takes place at the beginning of each TT but after one hour the  $\gamma'$  phase surface fraction reaches a stable value. However, as can be seen in Figures 4.17 and 5.3 b. the average precipitate size slightly increases from one hour to four hours of TT. This is likely to be due to Ostwald ripening, as confirmed by the thermal analyses of the precipitates size distribution below.

The particle size distribution was analyzed for all the temperatures and two HT durations (1h and 4h). The left side of the Figure C.1 shows the histograms directly obtained with the 2D experimental data. The population of small particles is reduced from 1h to 4h while the large particles get bigger and bigger and as a result, the histograms get wider. This behavior is consistent with the Ostwald ripening mechanism described in section 1.3.1.

The right side of the Figure C.1 show the histograms of the particle size distribution coming after Saltykov conversion from 2D to 3D data [206].

Figure C.2 shows experimental images after the different HT for 1h (left images) and 4h (right images). After 1h of heat treatment the particles are small and spherical, but after 4h they become bigger and some of them are agglomerated [207]. Most of them keep a spherical shape, which validates the usability of the Saltykov method.



Figure C.1: Comparison of the particle size distribution of the AD730 material after 1h and 4h of TT at different temperatures a.  $1060^{\circ}C$ , b.  $1070^{\circ}C$  and c.  $1080^{\circ}C$ . Histograms on the left side are the 2D experimental data obtained from SE images, and histograms on the right are obtained after a Saltykov 2D to 3D conversion.



Figure C.1: Comparison of the particle size distribution of the AD730 material after 1h and 4h of TT at different temperatures d.  $1090^{\circ}C$ , e.  $1100^{\circ}C$  and f.  $1110^{\circ}C$ . Histograms on the left side are the 2D experimental data obtained from SE images, and histograms on the right are obtained after a Saltykov 2D to 3D conversion.

# $AD730 \ material$



Figure C.2: SE images of the AD730 alloy after 1h and 4h heat treatments at different temperatures: a.  $1060^{\circ}C$ , b.  $1080^{\circ}C$  and c.  $1100^{\circ}C$ .

## C.2 Mean-field models for Ostwald ripening and LS simulation

Several theories reproduce the Ostwald ripening mechanism (see section 1.4.1), but many of them are not valid for high values of particles fraction. Moreover, our future objective is to study this mechanism in the AD730 superalloy, which has an important fraction of  $\gamma'$  phase. Therefore, we need a model capable of taking high precipitate fractions into account.

One of the first model concerning the Ostwald ripening was proposed in [64]. This model provides analytical equations for the coarsening mechanism. However, the weak point of this method is that it can only consider a maximum particle surface fraction of 8.5% in 2D and a maximum volume fraction of 6% in 3D; these limits are too low to reproduce the initial fraction of particles of our material.

The method developed by Voorhees and Glicksman (VG theory) [61, 62] is based on a computer algorithm that solves the multiparticle diffusion problem (MDP), where spherical particles interact with each other through its diffusion field. So the equation of the diffusion field within the matrix must be solved through the solution of a large linear system of equations.

This procedure can be quite complex with a large number of particles.

Instead of looking for detailed solutions to the MDP, the same authors also studied the average behavior of a group of dispersed particles in a matrix. Thus, a mean-field statistical model for Ostwald ripening was presented in [208].

For this new approach, the interaction environment is defined by a mean potential  $\alpha = \rho_{cr}^{-1}$ , where  $\rho_{cr} = r_{spp_{cr}}/r_{spp_{Av}}$  is the ratio of the radius of the critical particle, i.e. the particle for which  $dr_{spp}/dt = 0$  (neither grows nor shrinks), to that of the average particle. The mean potential  $\rho$  operates at a distance  $\rho_0$  from the center of the typical particle of size class  $\rho = r_{spp}/r_{spp_{Av}}$ .

They introduced the constant  $B(\rho)$  whose magnitude mesure the growth  $(B(\rho) > 0)$  or shrink  $(B(\rho) < 0)$  of the particle phase and it can be expressed as follows:

$$B(\rho) = \frac{(\alpha \rho - 1)\rho_0}{\rho_0 - \rho} \tag{C.1}$$

The constant  $B(\rho)$  are inherently related to the particle's coarsening rate by the following expression:

$$\frac{dr_{spp}}{dt} = \frac{B(\rho)}{r_{spp}^2}.$$
(C.2)

From Eq. C.1, we can deduce that the growth or shrinkage rate of the particles depends on its size relatively to the average. In this way, the particles for which  $r_{spp} < r_{sppAv}$  shrink, while particles for which  $r_{spp} > r_{sppAv}$  grow. Then, the expression for  $B(\rho)$ , can be modified to consider the particle fraction in the equation:

$$B(\rho) = \begin{cases} (\rho/\rho_{cr} - 1)(1 - f_{spp}^{1/3})^{-1}, & \rho > \rho_{cr} \\ (\rho/\rho_{cr} - 1)(1 - f_{spp}^{1/3}\rho/\rho_{cr})^{-1}, & \rho < \rho_{cr} \end{cases}$$
(C.3)

## C.3 Numerical simulations in LS context

A coarsening test at  $1060^{\circ}C$  was reproduced, taking the experimental data obtained after 1 h as the initial state and a duration of 3h at the same temperature, to compare the simulation results with the experimental data obtained after 4h. The previous described model was used to evolve the SPPs.

The initial state presented in Figure C.3 a. consists of about 1000 grains generated with a LS Laguerre-Voronoi tessellation [165] (arithmetic mean grain radius equal to  $\overline{D} = 10 \ \mu m$ ) and an initial polydisperse and spherical precipitate population was generated by using the experimental  $\gamma'$  precipitate size distribution with an initial area fraction  $f_{spp} = 12\%$  and an arithmetic mean size  $\overline{D}_{spp} = 2.1 \ \mu m$  (about 4300 SPP were generated). All SPP are considered to be incoherent.

The size of the domain is 0.3 mm x 0.3 mm, and a time step of 5s was considered to describe the precipitate/GB interaction correctly. The simulation parameters were the one obtained in section 5.3.1:  $M_0 = 2.9 \cdot 10^{25} \ m^4/J \cdot s$ ,  $Q_m = 9.8 \cdot 10^5 \ J/mol$ ,  $\gamma = 0.6 \ J/m^2$  [195].

The particle radius distributions obtained after 3h of numerical annealing describe well the experimental data as presented in Figure C.3.



Figure C.3: Numerical and experimental comparison of the particle size distribution of the AD730 material for 1h = initial state and 4h=after 3h of heat treatment.

APPENDIX C. OSTWALD RIPENING IN OUR LS FRAMEWORK

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## RÉSUMÉ

Dans la plupart des superalliages polycristallins à base nickel, la taille des grains est contrôlée par les particules de seconde phase. Le modèle de Smith-Zener décrit cette interaction physique. Les processus de forgeage industriels impliquent des étapes de déformation à chaud parfois proches de la température de dissolution des particules. Par conséquent, il est essentiel de comprendre et de prévoir l'évolution de ces particules afin de contrôler correctement les distributions de taille de grains obtenues lors de ces traitements thermomécaniques.

Deux superalliages base nickel ont été étudiés (AD730 et N19) avec une série de traitements isothermes et d'analyses microstructurales détaillées basées sur les techniques SEM et EBSD. Plus précisément, les précipités  $\gamma'$  (fraction, tailles et morphologies) ainsi que l'évolution de la taille des grains ont été analysées. Un modèle de type Johnson-Mehl-Avrami-Kolmogorov (JMAK) a été établi et validé pour décrire l'évolution temporelle des précipités de chaque matériau en fonction de la température à l'aide des données expérimentales acquises et du logiciel *Thermo-Calc*. Des simulations numériques ont ensuite été effectuées dans le cadre d'une formulation à champ complet pour simuler les phénomènes de croissance de grain.

Plus précisément, une approche Level-set (LS) de description des joints de grains dans un contexte éléments finis (EF) avec une population de particules de seconde phase évoluant selon les cinétiques précédemment établies, a été développée. L'approche numérique a été largement validée grâce à de nombreux tests académiques mais aussi surtout par son caractère prédictif vis-à-vis des données expérimentales capitalisées durant la thèse.

## MOTS CLÉS

Croissance de grains, ancrage de Smith-Zener, methode Level-Set, simulations à champ complet, superalliages, dissolution des particules.

## ABSTRACT

In most polycrystalline nickel base superalloys, the grain size is controlled by second phase particles which pin the grain boundaries. The Smith-Zener model describes this physical interaction. Industrial forging processes can involve hot deformation steps near the solvus temperature, where second phase particle dissolution occurs. Therefore, it is essential to understand and predict their evolution to properly control the grain size obtained after a specific subsolvus solution treatment and, in turn, the related material properties.

Two nickel base superalloys were studied (AD730, and N19) through a series of isothermal treatments and detailed microstructural analyses based on SEM and EBSD techniques. More precisely, the primary  $\gamma'$  precipitates (fraction, sizes, morphologies) as well as the grain size evolution were analyzed. A temperature-time codependency equation, through a Johnson-Mehl-Avrami- Kolmogorov (JMAK) model, was established to describe the evolution of precipitates of each material using experimental data and the *Thermo-Calc* software. Numerical simulations could then be performed using a full-field modeling framework for simulating grain growth (GG) phenomenon.

In fact, a Level-Set (LS) method, within a finite element (FE) context, was improved in order to deal with second phase particle evolving accordingly to the previously established kinetic models. This new numerical framework was largely validated thanks to academic test cases but most importantly by its predictive nature concerning the experimental results capitalized during the PhD.

## KEYWORDS

Grain growth, Smith-Zener pinning, Level-Set, full-field simulations, superalloys, particle dissolution kinetics.