SIMULATION STUDY OF DIRECTIONAL COARSENING (RAFTING) OF $\gamma$ IN SINGLE CRYSTAL Ni-Al

Dissertation

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ABSTRACT

Dislocation propagation in and work hardening of $\gamma$ channels and directional coarsening (rafting) of $\gamma'$ precipitates are the major microscopic processes taking place during high temperature deformation of single crystal Ni-base superalloys. Understanding of those processes is crucial for developing improved models of creep and fatigue of turbine blades in aircraft engines. Recent investigations of rafting in superalloys demonstrate clearly the importance of elastic modulus difference between the $\gamma$ and $\gamma'$ phases and dislocation-level activities in the $\gamma$-channels in determining the kinetic pathway of the processes. The elastic modulus difference can lead to the non-uniform distribution of stresses through the interaction with the lattice misfit and external load. While work hardening in the $\gamma$ channels has a direct effect on differentiation of the stress state in the vertical and horizontal channels and on $\gamma/\gamma'$ interface coherency and energy, and hence influences the diffusive flow and morphological changes of the $\gamma/\gamma'$ microstructure. In turn, changes in particle shape and coherency of the interface alter the local stress state and thereby the Peach-Koehler force on dislocations. Although existing models treating these processes separately can offer a qualitative explanation about the direction of rafting for typical superalloys, a complete quantitative understanding of rafting phenomena requires these processes to be treated simultaneously in a common framework because of their intimate coupling.
The objective of this thesis is to develop an integrated computational approach in simulating simultaneous evolution of both $\gamma/\gamma'$ microstructure and dislocations in an elastically anisotropic and inhomogeneous system by using a single, consistent phase field methodology. In particular, the phase field dislocation model is used to simulate the initial dislocation $\gamma$ channel filling process and calculate stress distribution associated with complex three-dimensional (3D) dislocation configurations in the $\gamma$-channels. The relative contributions from elastic modulus inhomogeneity and $\gamma$-channel plasticity are quantified by the dislocation-level simulations through the analysis of the spatial variation of solute atoms' chemical potential, which show that $\gamma$-channel plasticity plays the dominant role in controlling the rafting process (rafting type and kinetics). Micrometer-scale simulations are carried out that takes into account plastic deformation in $\gamma$-channels described by local channel dislocation densities from individual active slip systems. The rafting kinetics and the corresponding creep deformation are characterized at different values of applied stress, lattice misfit and precipitate volume fraction. The simulation predictions agree well with experimental observations and the models developed can be utilized in design of new superalloys and optimization of existing ones.
This is dedicated to my parents and my wife
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CHAPTER 1

INTRODUCTION

Ni-base single crystal superalloys are widely used as turbine engine blades. During their high temperature (around 1100°C) and low stress (about 120MPa ~ 150MPa) service, Ni-base single crystal superalloys’ initial microstructure which features cuboidal shaped $\gamma$ phase embedded in $\gamma$ matrix is altered through a directional coarsening process. During this process, $\gamma'$ precipitates coarsen and coalesce into “rafts” either perpendicular or parallel to the loading axis. And that is where the name “rafting” comes from. As a critical issue concerning the integrity of the turbine engine, the rafting phenomenon has been widely studied both experimentally and theoretically.

The modeling efforts of the rafting mechanism can be categorized roughly into two approaches: ‘elastic’ and ‘elastic-plastic’. For ‘elastic’ models, the influence from $\gamma$-channel dislocations is ignored and the cause of rafting is attributed to the interactions among external applied stress, lattice misfit and elastic modulus difference between the $\gamma$ and $\gamma'$ phases. While for ‘elastic-plastic’ models, matrix plasticity is considered crucial in determining the rafting direction and rafting kinetics. The details of those models will be briefly reviewed in this chapter. Although both approaches can be applied to qualitatively explain the direction of
rafting for typical superalloys, one of the major discrepancies between the elastic and elastic-plastic models is their prediction of the relationship between the modulus mismatch and the rafting directionality. For example, for a given sign of lattice misfit and direction of applied uniaxial load, the elastic model predicts that the rafting direction depends on the sign of the modulus difference (e.g., which phase is softer), while the elastic-plastic model predicts that the sign of the modulus difference should not affect the rafting direction. Experimental efforts have attempted to resolve this discrepancy, but no definite conclusion could be reached [3]. So, in order to develop a comprehensive model which can perform quantitative analysis and solve the discrepancy once for all, it is important to explore the experimental findings to better understand the physics of the rafting phenomenon and analyze the pros and cons of different rafting models. In this chapter, experimental works and modeling efforts on channel dislocation structure and rafting process in the past 30 years are briefly summarized.

1.1 Microstructure of single crystal Ni-base superalloys

Single crystal Ni-base superalloys are well known for their high strength under high temperature (>1000°C), which can be attributed to their unique microstructures. These so-called blade alloys are single crystalline materials with high volume fraction (up to 70%) of cuboidal γ' particles separated by narrow γ channels. The edge size of γ' cube is about 0.4 um to 0.6um, and the γ channel is about 0.1 um wide (Fig 1.1 [4]). The chemical composition of real alloys involves with many elements, but Al usually has the second highest molar fraction in superalloys next
CMSX-4 Single Crystal

Figure 1.1: Microstructure of coherent $\gamma/\gamma'$ single crystal blade alloy. [4]

only to Ni and many modelling work are based on Ni-Al binary system. A part of the Ni-Al binary phase diagram is shown in Fig 1.2 [5].

The $\gamma'$ phase has $L1_2$ ordered structure with Ni atoms occupying all the face center positions. And $\gamma$ phase is fcc solid solution. These two phases are initially coherent, but as an external stress is applied, dislocation multiplication will break the coherency of the $\gamma/\gamma'$ interfaces. Because the lattice constants of the two phases are usually not the same for single crystal superalloys, there is internal misfit stress in the material in order to maintain the initial coherency across the interface. The misfit here is used to address the lattice constant difference between $\gamma$ and $\gamma'$ phase. $\delta = 2(a_{\gamma'} - a_{\gamma})/(a_{\gamma'} + a_{\gamma})$. (Some authors like to define it as $\delta = (a_{\gamma'} - a_{\gamma})/a_{\gamma}$) So, positive misfit means $\gamma'$ phase has larger lattice constant than $\gamma$ phase. Most commercial Ni-base superalloys exhibit negative lattice misfit under high temperature. Also,
for most Ni-base superalloys, lattice constant will decrease as temperature goes up [6]. Fig 1.3 [7] shows the coherent lattice in a $\gamma/\gamma'$ system. In order to maintain coherency, the original cubic lattice is distorted. So the lattice constants of both phases used in the definition of the lattice misfit is for the unconstrained state. And now, the accurate measurement of the misfit is possible even at elevated temperature with the development of experiment techniques. [8, 9, 7, 10]

Most commercial Ni-base superalloys have negative lattice misfit and high volume fraction of $\gamma'$ phase (about 70%). And the volume fraction of $\gamma'$ will slightly decrease under high temperature tension creep test (depend on temperature and working stress) [11].
1.2 Dislocations in single crystal Ni-base superalloys

The applied stress is placed along [001] direction which corresponds to the centrifugal force direction. Dislocation segments are deposited on the $\gamma/\gamma'$ interface after the alloy is put into service, because under small stress ($< 150\text{MPa}$), dislocations are not able to cut into the $\gamma'$ phase to create APBs. So dislocations can only move in the $\gamma$ channels. In fcc material, there are 12 slip systems. The slip planes are $\{111\}$ and the Burgus vector should be $\frac{1}{2}(110)$ type. Because the slip planes are of $\{111\}$ type and the $\gamma'$ cubes are aligned along the $\langle 001 \rangle$ directions, dislocations will be deposited on $\{001\}$ interfaces with $\langle 110 \rangle$ line directions because dislocations’ further glide is impeded by the $\gamma'$ precipitates.

T.M. Pollock et al [13] found for negative misfit single crystal alloy under tension ($825^\circ\text{C}, 450\text{ MPa}$), dislocations preferentially glide in horizontal $\gamma$ channels (channel perpendicular to the stress axis) first. And for negative misfit alloy under small stress($50\text{MPa}$) and high temperature($1050^\circ\text{C}$), dislocations were generally absent from the thin matrix layers between the merging precipitates (parallel
to the applied tensile stress), but were abundant in the layers of matrix above and below the plate at the \( \gamma/\gamma' \) interfaces normal to the applied stress. (Schematic drawing of dislocation \( \gamma \) channel filling can be seen in Fig. 1.4 [12]). Similar phenomena have been verified by others [14, 15], as shown in Fig 1.5 [15].

C. Carry and J.L. Strudel proposed that interfacial dislocations might move towards the edges of the \( \gamma' \) particles by a combined glide-climb process [16]. U. Tetzlaff and H. Mughrabi further suggested as dislocations in horizontal channels reached the edge of \( \gamma' \) particle, they can glide across the vertical channel and build a dislocation arrangement at the vertical \( \gamma/\gamma' \) interface provided the local stresses are large enough. [17]
Figure 1.5: $\gamma$ channel dislocations for $T=1123K$ and $\varepsilon = 0.14\%$. (a) Horizontal (001) matrix channel: rectangular net-work of 60 degree interfacial dislocations. (b) Vertical (100) matrix channel: short dislocation loops with screw interfacial segments. [15]
C. Carry and J. L. Strudel also found during the secondary creep, strain is mainly the result of climb of dense edge dislocation networks in the $\gamma / \gamma'$ interfaces: the dislocation density ($10^9/cm^2$) is kept constant. And according to their explanation, at medium stress level, dislocations movement can only proceed by climbing in the $\gamma/\gamma'$ interface until they reach the edge of a cube, glide shortly and meet another cube face where they resume climbing. Effective creep parameters are typical of a diffusion controlled process: edge dislocations climbing in $\gamma/\gamma'$ interfaces. But such explanation will require channel dislocations’ slip plane to be $\{011\}$ and line direction $\langle 100 \rangle$. According to them, for dislocations on the interfaces normal to the stress axis $[001]$, edge component of the trailing dislocation tends to compensate the lattice misfit and locally lower the Al and Ti content of the matrix. While the exactly opposite thing happens on interfaces parallel to the stress axis. The whole picture brought up by them is like this: dislocation first penetrate into $\gamma$ channel by glide of leading screw segments leaving dislocation dipoles with edge components on $\gamma/\gamma'$ interfaces. As dislocation segments pile up, back stresses build up and inhibit the further glide of bowing out dislocations. After that, climbing of the deposited dislocations will be the rate controlling and strain producing mechanism for the secondary creep. [18]

M. Probst-Hein et al used DDM (discrete dislocation models) to estimate the influence of $\gamma'$ volume fraction and misfit on dislocation density in different $\gamma$ channels. (As shown in Fig 1.6 [19]) And found misfit will attract a dislocation loop from one microscopic crystallographic slip system into one type of $\gamma$ channel while make it more difficult for the same dislocation loop to enter other types of $\gamma$ channels. [19, 20]
Figure 1.6: Channel dislocation configuration used in DDM model. [19]

Figure 1.7: Simulation of dislocation channel filling using phase field dislocation model. [21]
Some preliminary simulation work of dislocation \( \gamma \) channel filling using phase field dislocation model has already yielded promising results (refer to Fig 1.7 [21]), which can explain the channel preference of dislocations due to lattice misfit and external load direction (page 159 of [21]).

1.3 Dislocation network

The formation of dislocation network in \( \gamma \) channel is also a widely discussed topic, which is believed to be critical to the creep behavior of Ni-base blade alloy. And also, the formation of a complex dislocation network is taking place simultaneously with the rafting process. So, a clear understanding of the dislocation network may be helpful for exploring the rafting mechanism.

D.F. Lahrman et al used over-age heat treatment to produce incoherent \( \gamma' \) under 1079°C. From TEM analysis under different diffraction condition, a dislocation network configuration was rationalized. From their observation and analysis, dislocation segments in the network are all of pure edge \( 1/2 \langle 110 \rangle \) type. [22]

In TEM observation, A. Singh et al found the rearrangement of dislocation networks by cross-slip and climb around \( \gamma' \) precipitates. And they also confirmed the predominantly edge character of those \( a/2 \langle 110 \rangle \) type dislocations around \( \gamma' \) phase. Several scenarios of dislocation nucleation around a spherical precipitate were proposed. And their calculation shows the lattice misfit in those superalloys is too small to spontaneously nucleate misfit dislocations around precipitates due to coherency stress. While dislocation nucleation was seen near carbides and then form Orowan loops that penetrate into \( \gamma \) channels. They attribute this to the large
lattice misfit between carbides and γ matrix. In their experiments, they also observed some roughly rectangular loops of predominantly edge character. These kinds of rectangular loops are not reported in other papers. [23]

R.C. Reed et al [24] also proposed a so-called octahedral and squares dislocation networks from TEM observation which is similar to the one suggested by D.F. Lahrman et al [22] as shown in Fig 1.8 [22]. It is said the network is formed by six different dislocations. And all dislocation segments in the network are of edge type. In their experiments of CMSX-4 superalloy, rafting occurs quickly and completes within 10h of creep test under 1150°C and 100MPa. And a creep hardening effect is observed at that temperature. The decrease of creep rate is due to the formation of rafted structure which eliminates the vertical γ channel and prevents the further glide/climb of \( \{111\}\langle110\rangle \) creep dislocations. The major difference between
R.C. Reed and D.F. Lahrman [22]’s explanation of network is that the formation of dislocations in the former case is due to [001] creep load, and the latter case is due to over-age heat treatment. But they both came up with the similar configuration of mostly edge type dislocation networks. If we start from available fcc slip systems and only consider dislocation slip on \{111\} planes and the local dislocation reaction at the intersections, the resultant dislocation network may give us a configuration similar to the one proposed in these two papers (with same geometry and same Burgers vector) but rotated by 45 degree with [001] axis. The rotation of $1/2 \langle 110 \rangle$ dislocations may better relief the misfit stress and lower system’s energy but need the assistance of climb to rearrange on (001) $\gamma/\gamma'$ interface. And one thing needs to mention is that the dislocation network in over-aged sample may be different from the one formed under applied stress.

Also, in C. Carry and J.L.Strudel [16]’s 250MPa creep test, dislocations at the onset of secondary creep are also said to be almost pure edge type.
T.M. Pollock et al systematically studied the creep of CMSX-3 alloy under inter-
mediate temperature (850°C) and relatively high applied stress (552MPa) in 1992. [13] Useful information was provided in this review about the forming of dislocation networks. The creep at moderate temperature was divided into several stages. First, an incubation period features the initial dislocation multiplication in the ini-
tial dislocation free superalloy. Then, followed by the primary creep when three dimensional nodal network of dislocation begins to form. Next is the steady state creep stage, a quasi-stationary structure is achieved. In the tertiary stage, $\gamma'$ precipitates begin to be sheared by dislocations and finally lead to failure. During incubation period, long and strait dislocations begin to bow out into $\gamma$ channel. The sources of dislocations are still far apart. As dislocations mostly go into horizontal channels (negative misfit alloy under tension), a lot of cross slips were observed using TEM which showed a 90-degree turn for penetrating dislocation loops (Fig 1.9 [13]). The dislocation segments deposited on the horizontal channel interface are 60 degree mixed type. When entering primary creep stage, dislocations from different sources begin to meet and inter-penetrate into a single channel, leading to reactions between different dislocations. And as dislocation density increases, some dislocations went into the vertical channels. Once the steady state creep in reached, all $\gamma$ channels are filled with dislocations (under 552MPa stress, which should be considered comparatively large) and multiple slip systems have been activated. At this time, the three-dimensional network is complete and the disloca-
tion density maintains stable with spacing of dislocations in the range of 50-120nm (60-80nm is most commonly observed). The networks lie in $\{111\}$ plane and are made up of 3 sets of dislocations with three coplanar $a/2\langle011\rangle$ type Burgers vector.
Burgers vector of \( a/2[110] \) and \( a/2[\bar{1}10] \) are less frequently observed because they have no resolved shear stress under [001] tension. But dislocation reaction can produce those types of dislocations. And \( a/2[110] \) and \( a/2[\bar{1}10] \) dislocations are often edge in orientation. Also, as cross slip is often observed, the sharp 90-degree turn of the dislocation loop will be rounded off very rapidly and the dislocation segments at the corner will align along \( \langle 001 \rangle \) directions. This rounding out requires a combination of core diffusion and volume-diffusion-controlled-climb of those dislocations. This climb continues to straighten out initial 60-degree mixed dislocations with sense vector \( \langle 110 \rangle \) to cube direction. Thus, dislocation line length is reduced. This may explain the [001] line direction dislocations observed in other experiments, shown in Fig 1.10 [25]. With same loading time, alloy with higher misfit will first develop dislocation network align along \( \langle 100 \rangle \) direction. (Based on this observation of this rounding off phenomenon, we can further use P-K force analysis to carefully check the cross slip process, and reach the conclusion that the rounding out process will always produce pure edge dislocations.) Using TEM observation and the relation between climb velocity and diffusion speed, the effective diffusivity at 825°C was estimated to be \( 3.1 \times 10^{16} cm^2/s \). In-situ annealing reveals a gradual and quasi-viscous coarsening of dislocation network without jerky collapse. The authors calculated the Orowan bowing out stress in the channel and the misfit stress which explains the preference of horizontal channel for dislocations in negative misfit alloy under tension. Solid solution resistance and dislocation resistance on the moving dislocations are also calculated.

Dislocation networks developed from local reactions between creep dislocations is discussed in reference [26]. Different geometries of network formation
Figure 1.10: TEM observations of dislocation networks. (a) misfit -0.16%; (b) misfit -0.33% [25]
resulted from local reactions between creep dislocations gliding on stressed octahedral slip systems are presented (see Fig 1.11 [13]). Reorientation and rearrangement of interfacial dislocations to maximally accommodate misfit are considered. And from experiment, it is found, dislocations do not tend to form Orowan loops around individual $\gamma'$ particles, but extend long distances through $\gamma$ channel. Dislocation reactions are calculated and dislocation type and line direction are also scrutinized. An interesting finding from experiment is that the $a/2[110]$ and $a/2[1\bar{1}0]$ dislocations, which have no resolved stress for slip in the specimen (with load along [001]), tend to be relatively straight and in their mismatch orientation ($\langle 100\rangle$ line direction). This result is consistent with the prediction made by dislocation reaction on $\gamma/\gamma'$ interface.
R.R. Keller et al analyzed the $\gamma/\gamma'$ interface dislocation networks after (001) tensile creep by TEM and determined the extra half plane of dislocation’s edge component. They found dislocations lying on precipitate faces parallel to the creep stress will aggravate the coherency strains while dislocations on faces perpendicular to the creep stress relax the coherency strains (negative misfit alloy). From their TEM experiment of (001) samples, dislocation networks were found on both type of interfaces, parallel and perpendicular to the stress axis. Networks consisted of three types of dislocations with non-coplanar $1/2\{110\}$ type Burgers vector. Hexagonal arrangements were found. Interfacial dislocation line vectors were found to lie between 70 and 90 to Burgers vector, implying these were mostly edge type.[27]

1.4 Experimental results of directional rafting

From T.M. Pollock and A.S. Argon’s classic paper [28], the relation of rafting direction with the sign of misfit and applied load direction is concluded as follows: negative misfit alloys stressed in tension or positive misfit alloys stressed in compression will develop rafts transverse to the direction of the externally applied stress, which is referred to as N(normal) type, while positive misfit alloys stressed in tension and negative misfit alloys stressed in compression will develop rafts parallel to the direction of externally applied stress, which is called P(parallel) type. It is stated in the paper: upon loading at high temperatures, Ni-base single crystals often exhibit a normal primary creep response, with initially rapid creep rates which decay to a minimum after a relatively brief transient. But an incubation period prior to the onset of primary creep was found in some alloys.
Figure 1.12: SEM image of rafting for alloys with different loading direction and sign of misfit (R1: positive misfit. R3: negative misfit.) (a) Microstructure resulted from the thermal exposure alone. The third face shows a fourier transform of the corresponding image. (b) Stress-annealed for 8 h at 130MPa compressive (R1) and tensile stress (R3) along [001]. (c) Stress-annealed for 8h at 130MPa tensile (R1) and compressive stress (R3) along [001] [3]
<table>
<thead>
<tr>
<th>Experiment year</th>
<th>Material</th>
<th>Misfit in reference</th>
<th>Test temperature</th>
<th>Test stress</th>
<th>Misfit at temperature</th>
<th>Raft type (ten)</th>
<th>Raft type (comp)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1971</td>
<td>Udiment 700</td>
<td>0.02% (RT)</td>
<td>950°C</td>
<td>147MPa</td>
<td>−0.3%</td>
<td>N</td>
<td>P</td>
</tr>
<tr>
<td>1979</td>
<td>Ni-15Al</td>
<td>0.56% (750°C)</td>
<td>750°C</td>
<td>147MPa</td>
<td>0.56%</td>
<td>P</td>
<td>N</td>
</tr>
<tr>
<td>1979</td>
<td>Alloy01</td>
<td>−0.4%</td>
<td>850°C</td>
<td>190-300MPa</td>
<td>0.38%</td>
<td>P</td>
<td></td>
</tr>
<tr>
<td>1980</td>
<td>Alloy 143</td>
<td>−0.78% (RT)</td>
<td>1038°C</td>
<td>207MPa</td>
<td>−0.82%</td>
<td>N</td>
<td></td>
</tr>
<tr>
<td>1983</td>
<td>Nasair 100</td>
<td>−0.36% (RT)</td>
<td>1000°C</td>
<td>148MPa</td>
<td>−0.45%</td>
<td>N</td>
<td></td>
</tr>
<tr>
<td>1983</td>
<td>Nasair 100</td>
<td>−0.36% (RT)</td>
<td>1000°C</td>
<td>strain rate</td>
<td>8.4 x 10^-7/s</td>
<td>−0.45%</td>
<td>P</td>
</tr>
<tr>
<td>1983</td>
<td>CMSX2</td>
<td>0.14% (RT)</td>
<td>1050°C</td>
<td>120MPa</td>
<td>−0.33%</td>
<td>N</td>
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</tr>
<tr>
<td>1984</td>
<td>Ta alloy</td>
<td>−0.23% (1050°C)</td>
<td>1050°C</td>
<td>140MPa</td>
<td>−0.23%</td>
<td>N</td>
<td></td>
</tr>
<tr>
<td>1983 1984 1985</td>
<td>Alloy 143</td>
<td>−0.8% (RT)</td>
<td>927-1038°C</td>
<td>143-234MPa</td>
<td>−0.23%</td>
<td>N</td>
<td></td>
</tr>
<tr>
<td>1984</td>
<td>Alloy C</td>
<td>−0.23% (RT)</td>
<td>1000°C</td>
<td>148MPa</td>
<td>−0.31%</td>
<td>N</td>
<td></td>
</tr>
<tr>
<td>1985</td>
<td>Alloy E</td>
<td>Nil (RT)</td>
<td>1000°C</td>
<td>148MPa</td>
<td>−0.22%</td>
<td>N</td>
<td></td>
</tr>
<tr>
<td>1987</td>
<td>Alloy H</td>
<td>−0.17% (1000°C)</td>
<td>1000°C</td>
<td>128MPa</td>
<td>−0.17%</td>
<td>N</td>
<td></td>
</tr>
<tr>
<td>1989</td>
<td>Nasair 100</td>
<td>−0.4% (760°C)</td>
<td>760°C</td>
<td>600-690MPa</td>
<td>−0.4%</td>
<td>N</td>
<td></td>
</tr>
<tr>
<td>1989</td>
<td>SRR99 100</td>
<td>−0.21% (980°C)</td>
<td>980°C</td>
<td>170MPa</td>
<td>−0.21%</td>
<td>N</td>
<td></td>
</tr>
<tr>
<td>1992</td>
<td>CMSX3 100</td>
<td>−0.38% (1050°C)</td>
<td>1050-1060°C</td>
<td>50-138MPa</td>
<td>−0.38%</td>
<td>N</td>
<td></td>
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<tr>
<td>1992</td>
<td>PMA</td>
<td>0.77% (RT)</td>
<td>1050°C</td>
<td>150MPa</td>
<td>0.5%</td>
<td>P</td>
<td></td>
</tr>
</tbody>
</table>

Table 1.1: Summary of experimental rafting behavior [2]
Also, from electron microscopy analysis, they found the incubation period represents the time required to percolate dislocations through the cross-section of the initially dislocation-starved material[13]. But in that paper, experiments are done at intermediate temperature. As temperature increases to higher than 900°C, the incubation period is immeasurably short. Also, they mentioned large scale coalescence of the precipitates was observed only in regions where significant densities of dislocations were present locally. All those experiment results emphasis the importance of dislocations to rafting. And rafting will complete before steady state creep is achieved.

A 3D reconstruction of the rafted structure can provide a clear view about how the rafts look like [3](Fig 1.12). And a detailed table is shown in Table 1.1, in which rafting direction, loading condition and lattice misfit corrected with thermo expansion of various experiments done before were summarized [2]. And the rafting process usually completes around 20 hours under typical experiment condition.

1.5 Modeling of rafting in Ni-base superalloys

There are many ways to classify the models developed to explain the rafting process. The method can be analytical solutions or simulations using finite element, Monte Carlo and phase field. The approach can be energy perturbation (kinetics) or total energy minimization (finite energy levels for different morphologies). The model can be pure elastic, or elastic-plastic. The energy change due to rafting can be attributed mostly to interfacial energy or bulk chemical energy. Focus may be placed on interface motion by elastic driving force or from diffusion concern. So it’s hard to place them strictly into a certain catalog. Here, we just list
several models selected from literature, and try to compare the characters of each model.

1.5.1 FEM

A lot of analysis was based on 2D finite element method[28][25]. Von-Mises equivalent stress is calculated, which shows clearly the bias caused by lattice misfit and applied stress direction. But the simple yield analysis can’t be used direct to explain the rafting. As mentioned by T.M. Pollock[28], in order to get rafting, the creep strain should be as large as 12% to achieve rafting just from \( \gamma \) channel flow. Actually, rafts develop with creep strains as small as 0.5-0.8%. While the Von-Mises stress analysis is useful to determine the dislocation density in different \( \gamma \) channels.

Even 3D FEM calculation was performed using real elastic modulus measured from experiments by M. Osawa et al [29]. They calculated the enthalpy change of rafting for 3 different negative misfit alloy, considering coherency strain, interfacial energy and the external stress. The result of their partially relaxed model turned out to be that 2 alloys will have rafting normal to the tensile axis while the other will raft parallel to the tensile direction which is against the experiment result. They explained that by mentioning the dislocations distribution in the material which will prefer \( \gamma \) channel normal to tensile direction instead of parallel channel, and that prevented the relaxation parallel to the tensile axis. The method they used here is based on strain-controlled boundary condition, that is, applying fixed total strain. Actually, as rafting happens under external stress, stress-controlled
boundary condition will better describe the process, and the total mechanical en-
ergy should be the sum of total strain energy and the potential energy of the exter-
nal load (the external work).

As a comparison with their FEM stress state analysis, T.M. Pollock and A.S. Argon [13] used a so-called “approximate analytical model” to do the estimation. This three-dimensional plasticity problem of compression of thin disks between rigid platens is first described by Schroeder and Webster [30]. After modification, this model is used to estimate the critical applied stress of tensile creep resistance. The basic idea is: a rigid plate in the matrix can increase the load carrying ability by a factor before slippage between plate and matrix happens. And this factor is related to the bulk tensile resistance and plate geometry.

1.5.2 Monte Carlo

Based on linearly elastic material behavior and fully coherent interfaces. Monte Carlo simulation is often performed based on the FEM analysis. Many models rely on the elastic strain energy and an isotropic interfacial energy term [31, 32, 33, 34, 35]. One example of the simulation result can be found in Fig 1.13 [35].

J.L.Valles and D.J.Arrell [36] focused on $\gamma/\gamma'$ interface and the energy reduc-
tion due to the relaxation of lattice misfit and applied stress by the formation of interface dislocations which will be specified latter.

1.5.3 Phase Field

D.Y. Li and L.Q.Chen used Eshelby’s treatment of coherent inclusion to study the rafting problem[37]. They took into account the different degenerate order-
ing states for $\gamma'$ phase and used 2D Phase Field method to simulate the rafting
process caused by binary diffusion. Two phase field variables were used: composition $c$ and long range order parameter $\eta$. Anisotropy and the modulus mismatch between the precipitates and matrix (ratio of $c_{11} - c_{12}$ between precipitate and matrix =1.21) were also considered using the method developed by Khachaturyan et al[38]. As shown in Fig 1.14 [37], the result is similar to other analytical solutions using pure elastic approach, that is, the sign of modulus mismatch, sign of lattice misfit and direction of applied stress will dictate the rafting direction.

J. Wang et al also used 2D phase field method to predict the rafting direction under applied stress. They used Cluster Variation Method to determine the gradient coefficient to match the interfacial energy[39]. Basically, they got the same result as D.Y.Li, L.Q.Chen.[37]
1.5.4 Elastic model

A. Pineau used Eshelby’s theory of inclusions and inhomogeneities to predict the rafting under uniaxial applied stress[40]. Pure elastic approach and isotropic approximation were applied to calculate the total system elastic energy, which came up from three parts: inhomogeneity effect (from modulus mismatch), inclusion effect (from lattice misfit) and interaction energy. Prediction of the stable shape (simplified as plate, needle or spheroid) was based on total elastic energy, and his famous rafting prediction map was shown in Fig 1.15 [40], which relates the rafting direction with the applied stress, lattice misfit and modulus mismatch.

In the paper, comparison was made with experiment done by Tien et al to justify the theoretical analysis[41]. Actually, from S.Socrate et al’s analysis[2], the real lattice misfit should be corrected for higher temperature, which will reverse the sign of misfit and result in wrong prediction. Anyhow, this paper illustrated some pioneer work on rafting prediction of Ni-base superalloys using pure elastic analysis.
Figure 1.15: A. Pineau’s rafting prediction map, which lead to the lowest total elastic energy for sphere (S), plates perpendicular to the stress axis (P) and needles aligned with the stress axis (N). $E_p/E_m$ is the ratio of the Young’s modulus of the matrix and that of the precipitates, $\sigma_A$ is the applied stress and $\delta$ the misfit between the precipitate and the matrix. [40]
T. Miyazaki et al added cubic anisotropy into the analysis. They considered an isolated ellipsoidal particle of variable aspect ratio and predict the rafting direction by calculation of energy versus aspect ratio[42].

F.R.N. Nabarro et al also used Eshelby’s energy-momentum tensor to get analytical expression for rafting driving force[43]. Anisotropy of the material and 3D case are considered. They found the driving force of rafting is proportional to the product of the tension, the lattice misfit and the modulus misfit. But when the plastic strain exceeds $m \delta$, the sign of $\sigma \delta$ will dominant the rafting direction and the elastic constant mismatch between inclusion and matrix is no longer important, which is consistent with S. Socrate’s 2D analysis[2]. ($m$ is the modulus mismatch; $\delta$ is the lattice misfit; $\sigma$ is the applied stress). They also emphasized the importance of the existence of dislocations during creep because under high temperature and low stresses, the distortion strain of each $\gamma'$ particle during rafting is on the order of 15%, while the total creep strain of the sample as a whole is only about 0.02%, moreover, the deformation of each particle under typical conditions of rafting has the opposite sense to the bulk deformation of the sample (negative misfit alloy under tension forms rafting structure perpendicular to the tensile axis). In the paper, the authors stated that from pure elastic approach there is no driving force for rafting under external stress if matrix and precipitate have the same elastic constants. They also mentioned rafting is driven by the difference between the work done by the loading machine and the increase in internal energy, which may raise question about the approaches used by some other researchers, such as J.L. Valles’s interfacial energy reduction method. [36]
I. Schimidt and D. Gross got the closed-form results for the prediction of rafting direction using the assumption of elastic material behavior in matrix and precipitate [44]. They considered cubic symmetry and the interaction between particles and brought up the load-equivalent eigenstrain to predict the rafting. They basically get the similar result as A. Pineau [40] and F.R.N. Nabarro [43] with the emphasis that the rafting direction is related to the ratio of $c_{11} - c_{12}$ between precipitate and matrix from pure elastic approach. ($c_{11}$ and $c_{12}$ are stiffness constants)

The Phase Field simulation done by D.Y. Li, L.Q. Chen [37] is also based on a pure elastic model. In their calculation, precipitates are considered about 20% stiffer than the matrix (stiffness constant data of precipitate and matrix were obtained for NiAl system at 1023k).

### 1.5.5 Elastic-Plastic model

Although from pure elastic analysis, many interesting results can be achieved, the importance of the existence of dislocations can’t be ignored. Actually, many people found the plastic flow in $\gamma$ channels due to dislocations is the key to explain the rafting phenomena when pure elastic approach fails to provide correct prediction. Also, dislocations multiplication in $\gamma$ phase can be used to explain the critical plastic stain (about 0.1%) [45] and incubation period observed during rafting [16, 46, 47, 48, 49, 50]. (The incubation period is often observed at intermediate temperature (700-900°C) which is characterized by a very low and steady creep rate ($10^{-6} \dot{\varepsilon} < \dot{\varepsilon} < 10^{-5} \dot{\varepsilon}$))

A dislocation based criterion for raft morphology predicting during high temperature creep is proposed [51]. Dislocations will first fill the matrix channels where
they induce a relaxation of the coherency stresses. According to that, those precipitate faces which exhibit dislocations at the beginning of creep will expand by directional coarsening. A diffusion mechanism based on the effect of creep dislocations on the local $\gamma$ phase chemical composition is suggested to account for the observed relationship between dislocation activity and directional coarsening of precipitates. The chemical gradients induce crossed fluxes of $\gamma$ and $\gamma'$ elements which eventually lead to the directional coarsening of $\gamma'$ precipitates. In this paper, J. Buffiere et al emphasized that the edge component of mixed type dislocation will relieve or enhance the coherency stresses. And the force exerted by the coherency stresses on the edge dislocation in $\gamma$ channel is on the same order of magnitude as the force due to the external stress (provided the misfit in superalloy is about $10^{-3}$). The authors carefully examined the dislocation Burgers vector and sense vector under compression and tension followed by extra-half plane position analysis. The authors also proposed that rafting maybe involves with the effect of dislocations’ influence on the $\gamma$ phase local composition. They also mentioned that the presence of interfacial dislocations provides vacancies sinks and sources for diffusion and also provides faster diffusion paths.

High temperature bending creep experiment was done by M. Ignat et al to examine the relationship between rafting and stress state[52]. The relation between dislocation edge component and relaxing of the misfit is carefully analyzed. According to their experiments, square platelets developed transverse to the stress axis in the tensile region of the sample, while elongated platelets parallel to the stress axis were observed in the compressive region of the sample. Dislocation density was determined from TEM image analysis. It was reported, the density is
$15 \times 10^4 cm^{-1}$ for the networks of the compressive region and $25 \times 10^4 cm^{-1}$ for the network in the tensile region. And the dislocation network in compressive region is on four faces parallel to stress axis while the network in tensile region is on two faces perpendicular to the stress. According to the author, there are two competing mechanisms controlling the coalescence of the precipitates: the transport of matter driven by non-relaxed misfit and the interaction of dislocations relaxing the misfit and rearranging on the interfaces of the coalesced structure. For N type rafting, the transport of matter is activated on the four faces parallel to the stress direction, and for P type rafting, it is activated only on the two opposite faces perpendicular to the stress direction. A so called critical stress is also mentioned that only when tension exceeds 300MPa or compression exceeds 100MPa can significant morphological change develop. And this critical stress is related to the generation of dislocations. Superalloy CMSX2 used by the authors is supposed to have a negative misfit under high temperature. Although that is not specified in the paper, the rafting result is consistent with the behavior of a negative misfit alloy. The explanation for the difference of critical stress magnitude and dislocation network density is like this: for compression, dislocations will go into vertical channels first and for tension, dislocation will go into horizontal channel first. And the ratio between vertical channel and horizontal channel is two to one. So dislocation is easier to multiply under compression while higher tensile stress is needed to stack dislocation into horizontal channel. And the higher external stress and less available interface in horizontal channel may result in higher dislocation density.

S. Socrate and D.M. Parks worked under the framework of the finite element method (plane strain 2D model) to locally evaluate the generalized force acting on
a material interface which is work-conjugate with the normal displacement of the interface itself[2]. They found using pure elastic analysis will get the same rafting prediction result as A. Pineau did for the same alloy, although S. Socrate et al’s approach is more general, considering the initial cuboidal shape of the $\gamma'$ precipitates. But the predictions are not always true comparing with the experimental results[53][42]. Obviously, from their paper, if the equivalent Young’s modulus (still use isotropic approximation) of precipitate is lower than that of the matrix, the pure elastic analysis will give the exact opposite prediction. Also, in their paper, summary of experimental rafting behavior [41, 42, 18, 54, 55, 56, 57, 58, 59, 60, 61, 62, 63, 13] was made and the corresponding lattice misfit was corrected according to experiment temperature (Refer to Table 1.1). Only by assuming that $\gamma'$ particles are elastically stiffer than the matrix for all those alloys (which is not necessarily true), can their model correctly predict the rafting direction by the sign of misfit and loading direction.

So they modified their model to consider the plastic creep flow of the matrix, which indicates the influence of dislocations in the matrix can’t be ignored. And from their analysis, the absolute value of elastic interface driving forces are generally increased by one order of magnitude comparing with pure elastic analysis, and sometimes will invert the sign of the force providing prediction in agreement with experiments (for $|\text{matrix plastic strain}| > |\text{modulus mismatch} \times \text{misfit}|$, ie, $|\epsilon_p| > |m \times \delta|$, $\sigma\delta$ becomes the dominant term). They attribute the failure of pure elastic approach by the omission of the stress state alteration due to the localized creep process which relieving the $\gamma/\gamma'$ misfit. But the stress analysis of creep transient process is not performed in detail.
T. Ichitsubo et al predict the rafting by comparing the total mechanical energy under different rafting direction configuration. Their result is the rafting phenomena can’t be explained within the coherent elastic regime[64]. From their elastic approach, only transverse rafting can be achieved no matter the direction of load and sign of misfit. The difference between this result and other elastic analysis (Nabarro) was attributed to the consideration of the potential energy of the external force using a macroscopic viewpoint. And according to the authors, the effect of interfacial dislocations from creep on eigenstrain must be considered to correctly predict the rafting direction combining the sign of misfit and external stress direction. They also mentioned the possibility of rafting without external stress with critical amount of pre-induced plastic strain. Actually, T. Ichitsubo et al’s approach is flawed because they used an average elastic constant in their calculation with a finite energy level minimization approach.

N. Matan et al proposed that, there is a critical plastic strain (about 0.1%), exceed this value, even if annealing the sample without applied stress, rafting will still continue[45]. They explained it as the critical value to break the coherency of γ and γ′ phases interface. So they divided the rafting into a so called “elastic” regime and a “plastic” regime. That actually emphasized the importance of the role of dislocations. From their experimental observation, small strain sample has lower dislocation density and dislocations are confined in their slip planes with no more than 3 or 4 dislocations on any one of the γ/γ′ interfaces. While large strain sample has higher dislocation density and dislocation networks are formed at γ/γ′ interface. Actually, we can easily explain the observed critical plastic strain
as follow: Small strain produces little amount of dislocations, which are more mo-
ble (most remain on their slip planes), and less possible of being pinned by other
dislocations because only a few slip systems are locally activated. So, there will
be less dislocation interception. After that small applied load is removed, dislo-
cations may slip away from $\gamma/\gamma'$ interface, and dislocation dipoles may annihilate
with each other, which will make it harder to achieve further rafting during an-
nealing. But for larger strain case, dislocations from many slip systems multiply
and intercept. They become less mobile because of the pinning effect. So, even af-
ter the removal of external load, dislocations network can still exist on the interface
stably. As the existence of dislocations is very critical, it’s not hard to imagining
further rafting during annealing without applied load. The value of critical plastic
strain was estimated by calculation of the dislocation density needed to relieve the
lattice misfit in horizontal channel.

T. Ohashi et al formulated the dislocation density tensor using the concept of
geometrically necessary dislocations according to the plastic strain distribution in
$\gamma$ channels[12, 65]. Then they calculated the stress field created by array of pure
edge dislocations on $\gamma/\gamma'$ interface by approximation followed by the calculation of
strain energy density in horizontal and vertical $\gamma$ channel respectively considering
contributions from applied stress, lattice misfit and dislocations. The result shows
that the strain energy density difference between horizontal and vertical channel
is about $2mJ/m^3$, large enough to cause diffusion which will result in rafting. Their
attempt to link the plastic deformation with geometrically necessary dislocation
distribution is more appropriate than analyzing rafting just by observing the Von-
Mises stress distribution in $\gamma$ channels, because rafting can’t be explained merely
by γ channel plastic flow. But, their way to treat interface dislocations is lack of scrutiny. Simply considering pure edge dislocations aligned along [001] crystallographic direction from dislocation reaction is not very appropriate.

M. Veron et al used a 3-dimensional simulation with $15 \times 15 \times 15$ precipitates array to describe the morphology and kinetics of the rafting process[35]. Anisotropic misfit relaxation by dislocations generated during creep is considered as the driving force for rafting. In their model, diffusion flux due to chemical potential gradient and plastic flow are simultaneously computed. Local stresses are obtained by superposition of the macroscopic applied stress and the misfit stresses which depend on the distribution of dislocations. Modulus mismatch didn’t come into this model because only the bias between different types of channels is considered. According to the authors, when relaxation of misfit stressed by plasticity is allowed, the most loaded channels will first undergo plasticity and dislocations will lie first on the interfaces of the channels perpendicular to the stress axis. Actually, experiments show that it’s true only for negative misfit alloy under tension or positive misfit alloy under compression. As a matter of fact, the preference of channels by dislocations depends not only on loading direction but also on the sign of misfit. In their simulation of a -0.3% misfit system, the coherency stresses in horizontal channels are set to zero considering the misfit relieving interfacial dislocations, and the local stresses are calculated. The inhomogeneity of the elastic energy density is said to drive diffusive flux between different types of channel. The diffusion coefficient used is on the order of $10^{-14}m^2/s$, and the applied stress was set to be 200MPa. The result showed an N type rafting for tension and P type rafting for compression. And an increase of coarsening rate was observed for higher applied stress. In
their simulation, the kinetics during compression appeared four times slower than under tension. They finally presented a morphology map indicating the relation between rafting direction, misfit and applied stress. This map has some problems in its positive misfit region which can not be verified by experiments. That is because they put too much emphasis on plastic flow, and for positive misfit region, the plastic flow is supposed to work against diffusional flow. Actually, it was confirmed by many creep experiments that rafting completes when only 0.2% creep strain is obtained [66]. So, plastic flow may only be a secondary effect with respect to rafting comparing to diffusional flux.

The onset of measurable rafting was observed by O.Paris et al to coincide with the beginning of the formation of misfit strain-relieving dislocation network, implying a plasticity-driven mechanism[66]. In their experiment, after 4h under load, rafts were clearly discernible and a plastic strain of 0.2% was accumulated. And the slip dislocations are lying preferentially in the horizontal interfaces. The kinetics of raft formation was found to be non-linear with a three-stage behavior. The first (moderate increase in aspect ratio) and second stage (rapid increase) are believed to be due to the redistribution of γ' forming matter (such as Al,Ti,Ta) on the length scale of individual precipitates by diffusional mass flow. (The diffusion length scale of the precipitates is about 100nm and the interdiffusion coefficients in the range of $10^{-16}$ to $10^{-15} m^2/s$ at T=1253K, and the typical diffusion time $t \sim d^2/D$ is estimated to be of the order of minutes.) During the third stage, rafting slows down due to the diminishing driving force. The presence of dislocation at the interfaces alters the gradient in solute chemical potential compare to the coherent state, which is critical to the rafting process. And from experiments, fully coherent
interface is hard to maintain under high temperature long time annealing even for applied stress as low as 40MPa. That may be attributed to uneven distribution of channel width and thermally assisted dislocation motion.

Experiments were done by M.Fahrmann et al[3]. To examine the relation between rafting direction with elastic modulus mismatch under high temperature. For Elastic model, the reverse of the sign of modulus mismatch will change the direction of rafting (rafting direction depend on $[\sigma(E_{\gamma} - E_{\gamma})/\delta(E_{\gamma})^2]$ ) , while for elastic-plastic model, this parameter is deemed as insignificant (only depend on $[\sigma/\delta E_{\gamma}]$ ). Unfortunately, although the small differential in the elastic constants of the two phases is determined in their experiments, the result itself can’t be used to discriminate these two models. That is because for both of their tested alloy, positive values were obtained and the rafting direction can be correctly predicted by both models. Due to the technical difficulty of manipulating modulus mismatch at high temperature in alloy designing, the dispute between elastic model and elastic-plastic model still goes on.

1.5.6 Interfacial energy minimization

An interesting model was developed by D.J.Arrell et al[67]. They calculated the interfacial energy density on each cubic plane of the precipitate, and predict the rafted shape by comparing the calculated energy density. But they didn’t consider the work done by loading machine during the creep and rafting process. The interfacial energy they calculated is from the interface dislocations. In their model, assumptions are made that interface dislocations just relive the lattice misfit between the $\gamma/\gamma'$ phase, and only edge dislocations which can relive lattice misfit are
counted. The line direction and actual dislocation type (slip system) are not treated explicitly. The result is interesting. They showed the predicted rafting result for a negative misfit alloy. And the modulus mismatch is considered relates to temperature. From their prediction, the sign of modulus mismatch will have a crossover at about 1000k, which gives inversed result if only considers a uniaxial load. They explained the reason why only rafting for $E_{\gamma} > E_{\gamma}$ case is observed as rafting is realistic only under high temperature. This model can deal with multiaxial stress. But their assumption is too simple to be true. From experiments, interface dislocation networks are observed which may have spacing ranging from 20nm to several hundred nanometers depending on lattice misfit and applied load. So it’s not appropriate to assume that the interface dislocations exactly relieve the lattice misfit. Calculation shows [45], for an alloy with absolute lattice misfit value even as large as -0.3%, only several dislocations can fully relax the misfit which gives a dislocation spacing on the scale of 100nm, while in experiments, denser dislocations structure may appear depending on the magnitude of the external load. Anyway, it is still an interesting attempt to incorporate both lattice misfit and modulus mismatch based on the understanding of interfacial dislocation network.

J.L. Valles and D.J. Arrell also used Monte Carlo simulation to reproduce the rafting under different stress state and temperature. But their model was still based on the interfacial energy anisotropy caused by different density of interfacial dislocations, which, according to them, “will completely relief the stress caused by lattice misfit and applied load”. [36]
1.5.7 Bulk chemical energy minimization

F.R.N. Nabarro once brought up a very simple model to calculate the thermodynamic pressure for rafting [68]. First, he admitted in elastic regime, if the elastic constants of $\gamma$ and $\gamma'$ are equal, there will be no tendency of rafting, but when plastic flow occurs, the difference in elastic constants becomes negligible. In his model, applied stress will increase only the free energy of $\gamma$ phase. He calculated the relation between enthalpy change and $\gamma/\gamma'$ interface displacement, and came to the conclusion that the initial chemical thermodynamic pressure on the interface is about 20 times the thermodynamic pressure arising from purely elastic effects. But he also estimated that the migration distance of interface under applied stress caused by bulk free energy is negligible. Which means the initial large thermodynamic pressure cause by bulk free energy can’t compete with the thermodynamic pressure from elastic effects which remains constant as interface migrates. So it’s hard to picture the result from this model, which basically rules out the possibility of rafting. The problem in his derivation is that only the equilibrium between one $\gamma$ channel and $\gamma'$ phase is considered. Actually, the free energy difference between different types of channel will continue the diffusion flux and move the interface forward. Also in his paper, he mentioned that only from the view of plasticity, the shift direction of $\gamma$ phase bulk free energy will be different for channel perpendicular to the stress and the channel parallel to the stress under a certain applied stress. Also, if a linear relation between local lattice misfit and solute concentration is assumed, the whole derivation will not be valid any more.
1.6 Potential of phase field model

As the $\gamma'$ rafting is ultimately a coarsening problem, the diffusion of solute atoms under the influence of non-uniform distribution of stresses in the system is the main cause leading to the symmetry breaking of the microstructure. So the focus of the study should start with the analysis of the local stress state. As illustrated in Fig. 1.16, the contributions of local stress come from external load, the back stresses caused by $\gamma$ channel dislocations, the misfit stress from the two-phase mixture, and the interaction between external load and modulus mismatch. Fortunately, phase field methods are able to capture all those factors contributing to the rafting process and offer quantitative data for in depth analysis.

With the recent development of phase field dislocation model [69, 70, 71] and microelasticity for elastically and structurally inhomogeneous systems [72], phase field method can analyze rafting contributions from both channel plasticity and
modulus inhomogeneity. Also, as phase field simulation of binary Ni-Al and ternary Ni-Al-Cr systems have already been linked to thermodynamic and mobility databases [73, 74], quantitative calculation of the rafting kinetics under different experimental conditions (such as temperature, composition) becomes possible. Moreover, special techniques have been developed in phase field for Ni-base superalloy studies including the treatment of anti-phase boundary, particle size distribution and different heat treatment processes [75, 76, 77], which makes the model capable of dealing with more complex and realistic situations.

In the following chapter, a brief introduction of the phase field method and its recent development will be presented which establishes the methodology foundation of the quantitative simulation of the rafting process.
CHAPTER 2

PHASE FIELF METHOD

This chapter provides a review of the basic formulation of the phase field method, Khachaturyan and Shatalov’s microelasticity theory and the recent development of new techniques such as Kim-Kim-Suzuki (KKS) model, microelasticity for modulus inhomogeneous systems and phase field dislocation model.

2.1 Basic formulation of phase field method

Phase field method was widely used to solve generalized diffusion problems based on Cahn-Hilliard equation for conserved field such as chemical concentration and Allen-Cahn equation for non-conserved field, such as crystal orientation, long-range order, crystal structure, and in plastic deformation case: the displacement field. These two equations can be regarded as metallurgical derivatives of the theories of Onsager and Ginzburg-Landau which represent a class of phenomenological continuum field approaches capable of describing continuous and quasi-continuous (the structural and/or chemical field variables are defined as continuous spatial functions which change smoothly rather than sharply across internal interfaces) phase separation phenomena in coherent and non-coherent systems. [78] Varieties of transformations in metal, ceramics and polymers, such as
spinodal decomposition, competitive partial ripening, non-isostructural precipitation and growth phenomena, grain growth, solidification and dendrite formation can be addressed in terms of corresponding chemical and structural phase field variables. [78, 79, 80, 81, 82, 83, 84, 85, 86, 87, 88, 89, 90, 91, 92, 93, 94, 95, 96]

The form of Cahn-Hilliard Equation is:
\[
\frac{\partial c(x,t)}{\partial t} = -\nabla \cdot \left[ -M_c \nabla \frac{\delta F}{\delta c(x,t)} \right]
\]
(2.1)
where \(c(x,t)\) is the concentration field variable, which is defined as the solvent volume fraction, \(x\) is the spatial coordinate, \(t\) is the time, \(F\) is the free energy function and \(M_c\) is the kinetic coefficient of diffusion (chemical mobility).

The form of Allen-Cahn equation (sometimes referred to as Time-dependent Ginzburg Landau equation) is:
\[
\frac{\partial \eta(x,t)}{\partial t} = -L \frac{\delta F}{\delta \eta(x,t)}
\]
(2.2)
where \(\eta(x,t)\) is the structure field variable and \(L\) is the kinetic coefficient.

The local chemical potential \(\mu_i(x)\) is given by the common derivative:
\[
\frac{\delta F}{\delta \xi_i(x,t)} = \mu_i(x)
\]
(2.3)
where \(\xi_i(x,t)\) is the generalized field form of the original functions \(c(x,t)\) and \(\eta(x,t)\).

The generalized total free energy in a compositional and structural inhomogeneous binary solution can be expressed through its energy density as a functional of that concentration field and its gradients in the form:
\[
F = \int_V \left( f(c(x,t), \eta(x,t)) + \kappa_c (\nabla c(x,t))^2 + \kappa_{c-\eta} (\nabla c(x,t)) \cdot (\nabla \eta(x,t)) + \kappa_{\eta} (\nabla \eta(x,t))^2 \right) dV
\]
(2.4)
where $V$ is the volume of the system and $f(c(x,t), \eta(x,t))$ is the bulk free energy density. The other terms in Eq. 2.4 are gradient energy describing the additional free energy due to chemical or structural inhomogeneity such as interfacial energy and grain boundary energy. $\kappa_c$, $\kappa_{c-\eta}$ and $\kappa_\eta$ are gradient coefficients.

## 2.2 Incorporation of Kim-Kim-Suzuki model

The main drawback of the “traditional” phase field formulation illustrated in the previous section is that in order to avoid numerical pinning during the computation process, the gradient energy penalty must be significant enough to maintain the diffusiveness of the phase variable across different phases (or grains). Due to the chemical energy contribution to the interfacial energy, there’s a certain limit in the interface thickness, that is, the limit of phase field calculation grid size. As a rule of thumb, the typical value of interfacial energy ($10 mJ/m^2$ to $500 mJ/m^2$) usually leads to relatively small lengthscale (i.e. simulation grid size limited to nanometer or sub nanometer scale) [97]. Attempts have been made to extend the lengthscale limit [76, 98, 99], while the most popular method is called Kim-Kim-Suzuki (KKS) model which involves with special treatment at the interface.

The idea of treating the free energy density in the interface region as a mixture of both phases with different compositions but constant ratio was first brought up by I. Steinbach et al [100]. Thus, the limit of the interface thickness can be relaxed, which makes possible large lengthscale phase field simulations with correct interfacial energy.

Here, an example of the KKS model treatment of a polycrystalline Ni-Al binary system with $(n - q)$ different $\gamma$ grain orientations coexisting with $q$ different
orientaions of \( \gamma \) precipitates can be helpful for explaining the formulation of this model.

Totally there are \( n \) phases (or grain orientations) and \( n - 1 \) independent order parameters in the system: \( \sum_{i=1}^{n} \eta_i = 1 \) (i.e. \( \eta_n = 1 - \sum_{i=1}^{n-1} \eta_i \)). A monotonously changing function \( P(\sum_{i=1}^{n-q} \eta_i) \) from \( P(0) = 0 \) to \( P(1) = 1 \) is used to interpolate the mixture of phases at interface. A commonly used form of this function is:

\[
P^{n-q}(\sum_{i=1}^{n-q} \eta_i) = (\sum_{i=1}^{n-q} \eta_i)^3 \left[ 10 - 15(\sum_{i=1}^{n-q} \eta_i) + 6(\sum_{i=1}^{n-q} \eta_i)^2 \right] \tag{2.5}
\]

As this form guarantees the first and second order derivatives with respect to \( \sum_{i=q+1}^{n} \eta_i \) is 0 at both ends (\( \sum_{i=q+1}^{n} \eta_i = 0 \) and \( \sum_{i=q+1}^{n} \eta_i = 1 \)), it provides a smooth transition across interface. And due to its symmetric form, we have:

\[
P\left( \sum_{i=q+1}^{n} \eta_i \right) = 1 - P\left( \sum_{i=1}^{n-q} \eta_i \right) \tag{2.6}
\]

The bulk chemical free energy density: \( f \) for this multiphase polycrystal is expressed as follow:

\[
f(c, \eta_{i=1..n-1}) = P\left( \sum_{i=1}^{q} \eta_i \right) f^{\gamma}(c_{\gamma}) + \left[ 1 - P\left( \sum_{i=1}^{q} \eta_i \right) \right] f^{\gamma'}(c_{\gamma'}) + \omega \sum_{i \neq j}^{n} \eta_i \eta_j \tag{2.7}
\]

where \( f^{\gamma}(c_{\gamma}) \) is the free energy function for \( \gamma \) phase with respect to its composition \( c_{\gamma} \) and \( f^{\gamma'}(c_{\gamma'}) \) is the free energy function for \( \gamma' \) phase with respect to its composition \( c_{\gamma'} \). \( \omega \) is the parameter related to the interfacial energy.

And the solute molar fraction: \( c \) is defined as:

\[
c = P\left( \sum_{i=1}^{n-q} \eta_i \right) c_{\gamma} + \left[ 1 - P\left( \sum_{i=1}^{n-q} \eta_i \right) \right] c_{\gamma'} \tag{2.8}
\]

The chemical potentials of both phases \( f^{\gamma}_{cl}(c_{\gamma}) \) and \( f^{\gamma'}_{cl}(c_{\gamma'}) \) need to be locally equal:

\[
f^{\gamma}_{cl}(c_{\gamma}) = \frac{df^{\gamma}(c_{\gamma})}{dc_{\gamma}} = f^{\gamma'}_{cl}(c_{\gamma'}) = \frac{df^{\gamma'}(c_{\gamma'})}{dc_{\gamma'}} \tag{2.9}
\]
For simplicity, the derivatives of the chemical potential are expressed as:

\[
\frac{d^2 f^\ell(c_\gamma)}{dc_i^2} = f_{cc}^\ell(c_\gamma)
\]

(2.10)

\[
\frac{d^2 f^\ell(c_\gamma)}{dc_i^2} = f_{cc}^\ell(c_\gamma)
\]

(2.11)

Regard \( c_\gamma \) and \( c_\gamma \) as functions of \( c \) and \( \eta_1, \eta_2, \ldots, \eta_{n-1} \). As \( c \) and \( \eta_i \) are independent variables, \( \frac{dc}{d\eta_i} = 0 \). Also, as \( f_{cl}^\ell(c_\gamma) \) is a function of \( c \) and \( \eta_i \):

\[
\frac{\partial f_{cl}^\ell(c_\gamma)}{\partial \eta_i} = \frac{\partial f_{cl}^\ell(c_\gamma)}{\partial c_i}. \tag{2.12}
\]

Thus, the derivatives of compositions of different phases with respect to one of the order parameter \( \eta_i \) corresponding to a \( \gamma \) grain orientation \( (1 \leq i \leq n - q) \) are:

\[
\frac{\partial c_\gamma}{\partial \eta_i} = -P'(\sum_{i=1}^{n-q} \eta_i)c_\gamma f_{cc}^\ell(c_\gamma) + P'(\sum_{i=1}^{n-q} \eta_i) c_\gamma f_{cc}^\ell(c_\gamma) \left[ 1 - P'(\sum_{i=1}^{n-q} \eta_i) f_{cc}^\ell(c_\gamma) \right]
\]

(2.13)

Where \( P'(\sum_{i=1}^{n-q} \eta_i) = \frac{\partial P(\sum_{i=1}^{n-q} \eta_i)}{\partial \eta_i} \).

The partial derivative of free energy density with respect to \( \eta_i \) is:

\[
\frac{\partial f}{\partial \eta_i} = P'\left( \sum_{i=1}^{n-q} \eta_i \right) f^\ell(c_\gamma) + P\left( \sum_{i=1}^{n-q} \eta_i \right) f_{cl}^\ell(c_\gamma) \frac{\partial c_\gamma}{\partial \eta_i} - P'\left( \sum_{i=1}^{n-q} \eta_i \right) f_{cl}^\ell(c_\gamma) + \omega \left[ |\eta_i| - |\eta_i| \right]
\]

(2.14)

If the curvatures of the free energy of both phases are the same (i.e. \( f_{cl}^\ell(c_\gamma) = f_{cc}^\ell(c_\gamma) \)), Eq. 2.14 can be greatly simplified as:

\[
\frac{\partial f}{\partial \eta_i} = P'\left( \sum_{i=1}^{n-q} \eta_i \right) f^\ell(c_\gamma) - c_\gamma f_{cl}^\ell(c_\gamma) - f^\ell(c_\gamma) + c_\gamma f_{cl}^\ell(c_\gamma) + \omega \left[ |\eta_i| - |\eta_i| \right]
\]

(2.15)

And the governing equation of order parameters is:

\[
\frac{\partial \eta_i}{\partial t} = M_\eta \left[ \kappa_\eta \nabla^2 \eta_i - \frac{\partial f}{\partial \eta_i} \right]
\]

(2.16)
For γ phase, the evolution of η_i (1 ≤ i ≤ q) can be obtained from:

\[ \frac{\partial \eta_i}{\partial t} = M_\eta \left[ \kappa_\eta \nabla^2 \eta_i - P'(\sum_{i=1}^{n-q} \eta_i) \left[ f_i^\tau(c_\gamma) - c_\gamma f_{c_\gamma}^\tau(c_\gamma) - f_i^\tau(c_{\gamma'}) + c_{\gamma'} f_{c_{\gamma'}}^\tau(c_{\gamma'}) \right] - \omega \left[ |\eta_i| - |\eta_i| \right] \right] \]  
\[
(2.17)
\]

Where \( M_\eta \) is the kinetic coefficient of the order parameter and \( \kappa_\eta \) is the gradient coefficient. The interfacial energy (or grain boundary energy) is determined by \( \kappa_\eta \) and \( \omega \).

Similarly, governing equation for order parameters \( \eta_i \) of the γ phase (q < i < n):

\[ \frac{\partial \eta_i}{\partial t} = M_\eta \left[ \kappa_\eta \nabla^2 \eta_i + P'(\sum_{i=1}^{n-q} \eta_i) \left[ f_i^\tau(c_\gamma) - c_\gamma f_{c_\gamma}^\tau(c_\gamma) - f_i^\tau(c_{\gamma'}) + c_{\gamma'} f_{c_{\gamma'}}^\tau(c_{\gamma'}) \right] - \omega \left[ |\eta_i| - |\eta_i| \right] \right] \]
\[
(2.18)
\]

And:

\[ \frac{\partial \eta_n}{\partial t} = 1 - \sum_{i=1}^{n-1} \frac{\partial \eta_i}{\partial t} \]
\[
(2.19)
\]

As \( f_i^\tau(c_\gamma) = f_{c_\gamma}^\tau(c_{\gamma'}) \), get derivative of both sides with respect to c: \( f_{c_\gamma}^\tau(c_{\gamma'}) \frac{\partial c_{\gamma'}}{\partial c} = f_{c_{\gamma'}}^\tau(c_{\gamma'}) \frac{\partial c_{\gamma'}}{\partial c} \). Also, get derivative of c on both sides of Eq. 2.8: 1 = \( P(\sum_{i=1}^{n-q} \eta_i) \frac{\partial c_\gamma'}{\partial c} + \left[ 1 - P(\sum_{i=1}^{n-q} \eta_i) \frac{\partial c_{\gamma'}}{\partial c} \right] \), which leads to:

\[ \frac{\partial c_\gamma}{\partial c} = \frac{f_{c_\gamma}^\tau(c_{\gamma'})}{P(\sum_{i=1}^{n-q} \eta_i) f_{c_\gamma}^\tau(c_{\gamma'}) + \left[ 1 - P(\sum_{i=1}^{n-q} \eta_i) \right] f_{c_{\gamma'}}^\tau(c_{\gamma'})} \]
\[
(2.20)
\]

\[ \frac{\partial c_{\gamma'}}{\partial c} = \frac{f_{c_{\gamma'}}^\tau(c_{\gamma'})}{P(\sum_{i=1}^{n-q} \eta_i) f_{c_{\gamma'}}^\tau(c_{\gamma'}) + \left[ 1 - P(\sum_{i=1}^{n-q} \eta_i) \right] f_{c_{\gamma'}}^\tau(c_{\gamma'})} \]
\[
(2.21)
\]

And the derivative of Eq. 2.7 is:

\[ \frac{\partial f}{\partial c} = P(\sum_{i=1}^{n-1} \eta_i) f_{c_\gamma}^\tau(c_{\gamma'}) \frac{\partial c_\gamma'}{\partial c} + \left[ 1 - P(\sum_{i=1}^{n-1} \eta_i) \right] f_{c_{\gamma'}}^\tau(c_{\gamma'}) \frac{\partial c_{\gamma'}}{\partial c} \]
\[
(2.22)
\]

If \( f_{c_\gamma}^\tau(c_{\gamma'}) = f_{c_{\gamma'}}^\tau(c_{\gamma'}) \), Eq. 2.22 can be simplified as:

\[ \frac{\partial f}{\partial c} = f_{c_\gamma}^\tau(c_{\gamma'}) = f_{c_{\gamma'}}^\tau(c_{\gamma'}) \]
\[
(2.23)
\]
And the diffusion equation is:

\[
\frac{1}{V_m} \frac{\partial c}{\partial t} = \nabla \cdot M_c \nabla \left( \frac{\partial f}{\partial c} \right) = \nabla \cdot M_c \nabla f^{\gamma}(c_\gamma)
\]  

(2.24)

2.3 Phase field microelasticity theory for homogeneous modulus system

2.3.1 Elastic energy calculation

Incorporating elastic energy into the free energy functional can be realized following the work of J.D. Eshelby [101] and A.G. Khachaturyan [87].

There are three basic assumptions of Khachaturyan and Shatalov’s microelasticity theory: 1) The parent and transformed phase particles have the same elastic moduli (the homogeneous moduli approximation). 2) The average length dimension of an inclusion and the average distance between the nearest inclusions are small compared to the typical length dimensions of the crystal. 3) The boundary condition of the multiphase mixture is stress-free. (this assumption is later extended to both stress-controlled and strain-controlled boundary conditions)

The inclusion phases are assumed to be coherent with the matrix. The so-called Eshelby’s circle is used to calculate the related elastic energy due to the phase transformation: step 1: cut the untransformed inclusion out of the matrix; step 2: transform the inclusion under stress-free condition, step 3: apply surface traction to deform the transformed inclusion back to its untransformed shape, step 4: put the particle back into its original hole in the matrix, step 5: “weld” the particle to the matrix, step 6: let the particle relax until reach elastic equilibrium. And the energy corresponds to each step is calculated accordingly.
In order to do so, a shape function is introduced (considering there are \(v\) different types of inclusions):

\[
\tilde{\theta}_p(r) = \begin{cases} 
1 & \text{in the volume unit } V_p, \\
0 & \text{outside the volume unit } V_p.
\end{cases} \tag{2.25}
\]

As it is assumed that the interface is coherent, the energy used to cut out the inclusion in step 1 cancels with the energy released by weld the inclusion back in step 5. While the transformation under stress-free condition in step 2 will not cause energy change. So the elastic strain energy change during the whole phase transformation process occurs during step 3 and step 6.

For step 3, surface transition is applied to the transformed inclusion and restore its original shape, so the total strain to accomplish that is: \(\varepsilon_{ij} = -\varepsilon_{ij}^*(p)\), which is the negative of the stress-free transformation strain: \(\varepsilon_{ij}^*(p)\). Using Hooks law, the internal stress induced is: \(\sigma_{ij} = C_{ijkl}[-\varepsilon_{ij}^*(p)]\), and the mechanical energy change for \(p\)-th inclusion is \(\Delta E_{self}^*(p) = \frac{1}{2}C_{ijkl}\varepsilon_{ij}^*(p)\varepsilon_{kl}^*(p)\). Considering all kinds of particles, the energy related to step 3 is:

\[
\Delta E_3 = \frac{1}{2} \sum_{p=1}^{V} V_p C_{ijkl} \varepsilon_{ij}^*(p)\varepsilon_{kl}^*(p) \tag{2.26}
\]

And as Hook’s law is used, linear elasticity is assumed.

For the relaxation in step 6, Taylor expands the local stress at the 0 total strain to the first order term:

\[
\sigma_{ij}(r) = \left[\sigma_{ij}(r)\right]_{\varepsilon_{ij}(r)=0} + \left[\frac{\partial \sigma_{ij}(r)}{\partial \varepsilon_{kl}(r)}\right]_{\varepsilon_{ij}(r)=0} \varepsilon_{kl}(r) + O(\varepsilon_{ij}(r)) \tag{2.27}
\]

The relaxation energy density can be expressed as the work done during relaxation for unit volume material:

\[
f_{relax}(r) = \int_0^{\varepsilon_{ij}(r)} \sigma_{ij}(r) d\varepsilon_{ij}(r) = -\sigma_{ij}^*(r)\varepsilon_{ij}(r) + \frac{1}{2}C_{ijkl}\varepsilon_{kl}(r)\varepsilon_{ij}(r) \tag{2.28}
\]
Where \( \sigma_{ij}^*(r) = C_{ijkl}^* \epsilon_{kl}^*(r) \) is the local stress-free transformation strain stress. And the shape function is used for the integration of relaxation energy density:

\[
\Delta E_{relax} = \int_V \left[ - \sum_{p=1}^V \sigma_{ij}^*(p) \tilde{\theta}_p(r) \epsilon_{ij}(r) + \frac{1}{2} C_{ijkl} \epsilon_{kl}(r) \epsilon_{ij}(r) \right] dV \tag{2.29}
\]

where \( \sigma_{ij}^*(p) = C_{ijkl} \epsilon_{kl}^*(p) \) is the stress-free transformation strain stress corresponding to the \( p \) type inclusions.

Based on the afore-mentioned assumption 2, the elastic strain \( \epsilon_{ij}(r) \) can be divided into homogeneous part \( \bar{\epsilon}_{ij} \) and heterogeneous part \( \delta \epsilon_{ij}(r) \):

\[
\epsilon_{ij}(r) = \bar{\epsilon}_{ij} + \delta \epsilon_{ij}(r) \tag{2.30}
\]

And the relaxation energy can also be divided into homogeneous part and heterogeneous part \( \Delta E_{relax} = E_{relax}^{homo} + E_{relax}^{heter} \):

\[
E_{relax}^{homo} = - \sum_{p=1}^V V_p \sigma_{ij}^*(p) \bar{\epsilon}_{ij} + \frac{V}{2} C_{ijkl} \bar{\epsilon}_{ij} \bar{\epsilon}_{kl} \tag{2.31}
\]

\[
E_{relax}^{heter} = \int_V \left[ - \sum_{p=1}^V \sigma_{ij}^*(p) \Delta \tilde{\theta}_p(r) \delta \epsilon_{ij}(r) + \frac{1}{2} C_{ijkl} \delta \epsilon_{kl}(r) \delta \epsilon_{ij}(r) \right] dV \tag{2.32}
\]

Where \( \Delta \tilde{\theta}_p(r) = \tilde{\theta}_p(r) - V_p/V \).

The mechanical equilibrium requires the variation of homogeneous strain and local displacement \( u(r) \) to reduce the relaxation energy to minimum:

\[
\frac{\partial \Delta E_{relax}}{\partial \bar{\epsilon}_{ij}} = 0 \tag{2.33}
\]

\[
\frac{\partial \Delta E_{relax}}{\partial u_i(r)} = 0 \tag{2.34}
\]

Note that \( \delta \epsilon_{ij} = \frac{1}{2} \left( \frac{\partial u_i}{\partial r_j} + \frac{\partial u_j}{\partial r_i} \right) \).

Calculated from Eq.2.33 and Eq.2.31, the homogeneous relaxation energy can be written as:

\[
E_{relax}^{homo} = -\frac{V}{2} \sum_{p=1}^V \sum_{q=1}^V C_{ijkl} \epsilon_{ij}^*(p) \epsilon_{kl}(q) \frac{V_p V_q}{V V} \tag{2.35}
\]
Calculated from Eq. 2.34 and Eq. 2.32:

\[ C_{ijkl} \frac{\partial^2 u_k}{\partial r_i \partial r_j} = \sum_{p=1}^{\nu} \sigma^*_i j(p) \frac{\partial \Delta \tilde{\theta}_p(r)}{\partial r_j} \]  

(2.36)

Using Fourier transform to change partial differentiation in real space into multiplication in k-space. And because of the orthogonality of the Fourier series, the coefficients have to be equal term by term on both sides of Eq. 2.36:

\[ C_{ijkl} k_j k_v l_i(k) = -i \sum_{p=1}^{\nu} \sigma^*_i j(p) k_j \Delta \theta_p(k) \]  

(2.37)

Where:

\[ v(k) = \int_V dV \cdot u(r) e^{-ikr} \]  

(2.38)

And:

\[ \Delta \theta_p(k) = \int_V dV \cdot \Delta \tilde{\theta}_p(r) e^{-ikr} \]  

(2.39)

The solution of Eq. 2.37 is:

\[ v_i(k) = -i \sum_{p=1}^{\nu} G_{ij}(k) \sigma^*_j k p k \Delta \theta_p(k) \]  

(2.40)

where \( G_{mi}(k) C_{ijkl} k_k l_i = \delta_{mj} \).

Rewrite the heterogeneous relaxation energy in k-space:

\[ E_{relax}^{\text{heter}} = \int \frac{d^3 k}{(2\pi)^3} \left[ -i \sum_{p=1}^{\nu} \sigma^*_i j(p) \Delta \theta_p(k) k_j v_i^*(k) + \frac{1}{2} C_{ijkl} k_j l_i v_i(k) v_i^*(k) \right] \]  

(2.41)

Using Eq. 2.38 and Eq. 2.35, and combining with Eq. 2.26:

\[ E_{elast} = \frac{1}{2} \sum_p V_p C_{ijkl} \varepsilon_{ij}^*(p) \varepsilon_{kl}^*(q) - \frac{V}{2} \sum_{p,q} C_{ijkl} \varepsilon_{ij}^*(p) \varepsilon_{kl}^*(q) \frac{V_p}{V} \frac{V_q}{V} \]

\[ -\frac{1}{2} \sum_{p,q} \int \frac{d^3 k}{(2\pi)^3} [k_i \sigma_{ij}(p) G_{jk}(k) \sigma_{kl}(q) k_l] \Delta \theta_p(k) \Delta \theta_q^*(k) \]  

(2.42)
Using Parseval theorem:

\[
\int \frac{d^3 k}{(2\pi)^3} \Delta \theta_p(k) \Delta \theta_q^*(k) = \int_V \Delta \tilde{\theta}_p(r) \Delta \tilde{\theta}_q^*(r) dV = \delta_{pq} V p - \frac{V p V q}{V} \tag{2.43}
\]

Also, there is:

\[
\Delta \theta_p(k) = \int dV \left[ \tilde{\theta}_p(r) - \frac{V_p}{V} \right] e^{-i kr} = \begin{cases} \theta_p(k) & \text{if } k \neq 0, \\ 0 & \text{otherwise.} \end{cases} \tag{2.44}
\]

Using Eq. 2.43 and Eq. 2.44 to rewrite Eq. 2.42:

\[
E_{elast} = \frac{1}{2} \sum_{p,q} \int \frac{d^3 k}{(2\pi)^3} \left[ C_{ijkl} \varepsilon_{ij}^*(p) \varepsilon_{kl}^*(q) - (n_i \sigma_{ij}^*(p) \Omega_{jk}(n) \sigma_{kl}^*(q) n_l) \right] \theta_p(k) \theta_q^*(k) \tag{2.45}
\]

In which the integration excludes a volume \((2\pi)^3/V\) at \(k = 0\) for the stress free boundary condition. \(n = k/k\) and \(\Omega_{ij}^{-1}(n) = C_{ijkl} n_k n_l\).

Generalize Eq. 2.45 to the stress-controlled boundary condition, that is, the boundary is under external stress: \(\sigma_{ij}^{ex}\):

\[
E_{elast} = \frac{1}{2} \sum_{p,q} \int \frac{d^3 k}{(2\pi)^3} B_{pq}(n) \theta_p(k) \theta_q^*(k) - \sigma_{ij}^{ex} \int_p \varepsilon_{ij}^*(p) \tilde{\theta}_p(r) d^3 r
\]

and:

\[
B_{pq}(n) = \begin{cases} n = 0 & B_{pq}(n) = 0, \\ n \neq 0 & B_{pq}(n) = \left[ C_{ijkl} \varepsilon_{ij}^*(p) \varepsilon_{kl}^*(q) - (n_i \sigma_{ij}^*(p) \Omega_{jk}(n) \sigma_{kl}^*(q) n_l) \right]. \end{cases} \tag{2.46}
\]

Moreover, for the clamped boundary condition (that is, there’s no macroscopic strain):

\[
E_{elast} = \frac{1}{2} \sum_{p,q} \int \frac{d^3 k}{(2\pi)^3} B_{pq}(n) \theta_p(k) \theta_q^*(k) \tag{2.47}
\]

Where:

\[
B_{pq}(n) = \begin{cases} n = 0 & B_{pq}(n) = C_{ijkl} \varepsilon_{ij}^*(p) \varepsilon_{kl}^*(q), \\ n \neq 0 & B_{pq}(n) = \left[ C_{ijkl} \varepsilon_{ij}^*(p) \varepsilon_{kl}^*(q) - (n_i \sigma_{ij}^*(p) \Omega_{jk}(n) \sigma_{kl}^*(q) n_l) \right]. \end{cases} \tag{2.48}
\]
2.3.2 Local elastic stress calculation

Imaging that one introduces a stress detector described by a Dirac delta function at the position \( r = r_0 \) by treating it as a \( q \) type inclusion. The shape function corresponds to the detector is: \( Q(r) = \delta(r - r_0) \). And in order to avoid confusion, all other real inclusions in the system are referred to as \( p \) type.

The interaction energy between the real inclusions and the detector is defined as [102, 103]:

\[
E_{pq}^{int} = - \int_V \varepsilon_{ij}^{q0}(r) \sigma_{ij}(r) dV = \int_V e_{pq}^{q}(r) dV
\]

(2.49)

Where the local interaction energy density: \( e_{pq}^{q}(r) = - \int_V dV \varepsilon_{ij}^{q0}(r) \sigma_{ij}(r) \). Note that the interaction energy defined here is just the interaction between the stress detector (described by a delta function) and all other inclusions. The pairwise interactions between inclusions and interactions between volume elements within a finite precipitate are not considered.

The local stress free transformation strain of the detector is defined as:

\[
\varepsilon_{ij}^{q0}(r) = Q(r) \delta_{im} \delta_{jn}
\]

(2.50)

\( \delta_{ij} \) is the Kronecker delta. \( m,n \) correspond to the stress component of interest: \( \sigma_{mn} \).

Thus

\[
E_{pq}^{int} = - \int_V Q(r) \delta_{mn} \delta_{ij} \sigma_{ij}(r) dV = - \int_V \delta(r - r_0) \sigma_{mn}(r) dV = -\sigma_{mn}(r_0)
\]

(2.51)

Also, the interaction energy can be written in \( k \)-space:

\[
E_{pq}^{int} = \int \tilde{e}_{pq}^{q}(k) \frac{d^3k}{(2\pi)^3}
\]

(2.52)

So:

\[
\sigma_{mn}(r_0) = \int \tilde{e}_{pq}^{q}(k) \frac{d^3k}{(2\pi)^3}
\]

(2.53)
Also,
\[
\sigma_{mn}(r_0) = \int \tilde{\sigma}_{mn}(k)e^{ikr_0} \frac{d^3k}{(2\pi)^3} \tag{2.54}
\]

From Eq. 2.53 and Eq. 2.54, there is:
\[
\tilde{\sigma}_{mn}(k)e^{ikr_0} = -\tilde{\epsilon}_{pq}^{pq}(k) \tag{2.55}
\]

From Khachaturyan and Shatalov’s microelasticity theory described in the previous section, the interaction between inclusions and the detector can also be written as [87, 102, 103]:
\[
E_{pq}^{\text{int}} = \int \sum_p B_{pq}(n)\theta_p(k)\tilde{Q}^*(k)\frac{d^3k}{(2\pi)^3} \tag{2.56}
\]
where \(\theta_p(k)\) is the shape function of the real inclusion (type p) expressed in k-space. And \(B_{pq}(n) = [C_{ijkl}\varepsilon_{ij}^*(p)\delta_{km}\delta_{ln} - (n_j\sigma_{ij}^*(p)\Omega_{jk}(n)(C_{klm}\delta_{sm}\delta_{nt})n_l)]\). \(\varepsilon_{ij}^*(p)\) is the stress free transformation strain of the p-th inclusion and \(\sigma_{ij}^*(p) = C_{ijkl}\varepsilon_{kl}^*(p)\). \((B_{pq}(0)\) depends on boundary condition.\)

So,
\[
\tilde{\epsilon}_{pq}^{pq}(k) = \sum_p B_{pq}(n)\theta_p(k)\tilde{Q}^*(k) \tag{2.57}
\]

Because there is:
\[
\tilde{Q}^*(k) = \tilde{Q}(-k) = \int Q(r)e^{ikr}dr^3 = \int \delta(r - r_0)e^{ikr}dr^3 = e^{ikr_0} \tag{2.58}
\]
So:
\[
\tilde{\epsilon}_{pq}^{pq}(k) = \sum_p B_{pq}(n)\theta_p(k)e^{ikr_0} \tag{2.59}
\]
Thus
\[
\sigma_{mn}(k) = -\sum_p B_{pq}(n)\theta_p(k) \tag{2.60}
\]
In real space, the elastic stress can be obtained using the following equation:

\[
\sigma_{mn}(r) = -\int \sum_p B_{pq}(n)\theta_p(k)e^{i\mathbf{kr}}\frac{d^3k}{(2\pi)^3} \tag{2.61}
\]

Or, from Eq. 2.37, the displacement field in k-space: \(\mathbf{v}(k)\) can be obtained. And then, use the definition of the strain to get the local total strain. Finally, subtracting the local transformation strain from the total strain leads to the elastic strain:

\[
\varepsilon_{ij}(r) = \bar{\varepsilon}_{ij} + \frac{1}{2}\left\{n_j\Omega_{kl}(n) + n_i\Omega_{kj}(n)\right\}\bar{\sigma}_{kl}(k) - \varepsilon^*_i(r) \tag{2.62}
\]

where \(\varepsilon^*_i(r) = \sum_{p=1}^P \varepsilon^0_{ij}(p)\theta_p(r)\) and \(\sigma^*_i(r) = C_{ijkl}\varepsilon_{kl}(r)\). Thus local stress is: \(\sigma_{ij}(r) = C_{ijkl}\varepsilon_{kl}(r)\). (Homogeneous strain \(\bar{\varepsilon}_{ij} = \frac{1}{V} \int_{V'} \varepsilon^0_{ij}(r)d^3r + S_{ijkl}\sigma^*_{kl}\) for stress-controlled boundary condition.)

### 2.4 Microelasticity for inhomogeneous modulus systems

Based on J.D. Eshelby’s [101] classic work for ellipsoidal inclusion in elastically isotropic media, Y.U. Wang et al formulated the microelasticity for elastically and structurally inhomogeneous solid [72, 104].

First of all, the stiffness tensor \(C_{ijkl}\) is defined as a function of spatial position \(r\):

\[
C_{ijkl}(r) = C_{ijkl}^0 - \Delta C_{ijkl}(r) \tag{2.63}
\]

Where \(C_{ijkl}^0\) is the stiffness of the reference phase, and \(\Delta C_{ijkl}(r)\) is the spatial variation of the modulus.

A so called virtual strain \(\varepsilon^0_{ij}(r)\) is created to satisfy the following equation:

\[
C_{ijkl}^0 \left[\varepsilon_{kl}(r) - \varepsilon^0_{kl}(r)\right] = \left[C_{ijkl}^0 - \Delta C_{ijkl}(r)\right] \left[\varepsilon_{kl}(r) - \varepsilon^*_k(r)\right] \tag{2.64}
\]

Where \(\varepsilon_{ij}(r)\) is the total strain at position \(r\) and \(\varepsilon^*_i(r)\) is the real stress free transformation strain from the inclusions.
The strain energy of the equivalent elastically homogeneous system (which has homogeneous modulus, but the stress free transformation strain of the inclusion is modified so the elastic stress and total strain field match with the original inhomogeneous modulus system) is:

\[ E^{\text{homo}} = \frac{1}{2} \int_V C_{ijkl}^0 \left[ \varepsilon_{ij}(r) - \varepsilon_{ij}^0(r) \right] \left[ \varepsilon_{kl}(r) - \varepsilon_{kl}^0(r) \right] d^3 r \tag{2.65} \]

Note: the equivalent homogeneous system can only provide the local elastic equilibrium with the same stress distribution and local total strain in the system comparing to the elastically and structurally inhomogeneous system. While the total elastic strain energy is different.

The strain energy of the original elastically and structurally inhomogeneous system is:

\[ E^{\text{inhomo}} = \frac{1}{2} \int_V \left[ C_{ijkl}^0 - \Delta C_{ijkl}(r) \right] \left[ \varepsilon_{ij}(r) - \varepsilon_{ij}^0(r) \right] \left[ \varepsilon_{kl}(r) - \varepsilon_{kl}^0(r) \right] d^3 r \tag{2.66} \]

And the difference between \( E^{\text{homo}} \) and \( E^{\text{inhomo}} \) is:

\[ \Delta E = E^{\text{inhomo}} - E^{\text{homo}} \]

\[ = \frac{1}{2} \int_V \left[ C_{ijkl}^0 \Delta S_{mn pq}(r) C_{pq kl}^0 - C_{ijkl}^0 \right] \left[ \varepsilon_{ij}(r) - \varepsilon_{ij}^0(r) \right] \left[ \varepsilon_{kl}(r) - \varepsilon_{kl}^0(r) \right] d^3 r \tag{2.67} \]

Where \( \Delta S_{ijkl}(r) = \Delta C_{ijkl}^{-1}(r) \)

Put the elastic energy formula derived in the previous section (Eq. 2.45) into Eq. 2.67, for strain-controlled boundary condition:

\[ E^{\text{inhomo}} = \frac{1}{2} \int_V \left[ C_{ijkl}^0 \Delta S_{mn pq}(r) C_{pq kl}^0 - C_{ijkl}^0 \right] \left[ \varepsilon_{ij}(r) - \varepsilon_{ij}^0(r) \right] \left[ \varepsilon_{kl}(r) - \varepsilon_{kl}^0(r) \right] d^3 r \]

\[ + \frac{1}{2} \int \frac{d^3 k}{(2\pi)^3} \left[ C_{ijkl}^0 \tilde{\varepsilon}_{ij}(k) \tilde{\varepsilon}_{kl}(k) - n_j \tilde{\sigma}_{ij}(k) \Omega_{jk}(n) \tilde{\sigma}_{kl}(k) n_i \right] \]
\[-\tilde{\varepsilon}_{ij} \int_{V} C_{ijkl}\tilde{\varepsilon}_{kl}(\mathbf{r})d^3r + \frac{V}{2} C_{ijkl}\tilde{\varepsilon}_{ij}\tilde{\varepsilon}_{kl} \]

(2.68)

Where \( \tilde{\varepsilon}_{ij}(\mathbf{k}) = \int_{V} \varepsilon_{ij}(\mathbf{r})e^{-ikr}d^3r \), and \( \tilde{\sigma}_{ij}(\mathbf{k}) = C_{ijkl}\tilde{\varepsilon}_{ik}(\mathbf{k}) \).

For stress-controlled boundary condition:

\[
E^{inhomo} = \frac{1}{2} \int_{V} \left[ C_{ijkl}\Delta S_{mnop}(\mathbf{r})C_{pqkl} - C_{ijkl}(\mathbf{r}) \right] \left[ \varepsilon_{ij}(\mathbf{r}) - \varepsilon_{ij}^0(\mathbf{r}) \right] \left[ \varepsilon_{kl}(\mathbf{r}) - \varepsilon_{kl}^0(\mathbf{r}) \right] d^3r \\
+ \frac{1}{2} \int \frac{d^3k}{(2\pi)^3} \left[ C_{ijkl}\tilde{\varepsilon}_{ij}(\mathbf{k})\tilde{\varepsilon}_{kl}(\mathbf{k}) - n_i\tilde{\sigma}_{ij}(\mathbf{k})\Omega_{jk}(\mathbf{n})\tilde{\sigma}_{kl}(\mathbf{k})n_l \right] \\
- \frac{1}{2V} C_{ijkl} \int_{V} \varepsilon_{ij}^0(\mathbf{r})d^3r \int \varepsilon_{kl}(\mathbf{r})d^3r' - \sigma_{ij}^t \int_{V} \varepsilon_{ij}^0(\mathbf{r})dV - \frac{V}{2} \tilde{\sigma}_{ijkl}\sigma_{kl}^t \sigma_{kl}^t 
\]

(2.69)

The integration in k-space excludes a volume of \((2\pi)^3/V\) around the point \( \mathbf{k} = 0 \).

As a functional of the virtual strain, the inhomogeneous elastic energy reaches minimum when there is:

\[
\frac{\delta E^{inhomo}}{\delta \varepsilon_{ij}^0(\mathbf{r})} = 0 
\]

(2.70)

The time-dependent Ginzburg-Landau type equation is used for obtaining the energy minimizer:

\[
\frac{\partial \varepsilon_{ij}^0(\mathbf{r}, \tau)}{\partial \tau} = -L_{ijkl} \frac{\delta E^{inhomo}}{\delta \varepsilon_{kl}^0(\mathbf{r}, \tau)} 
\]

(2.71)

Where \( L_{ijkl} \) is the kinetic coefficient which needs to be positive. A convenient choice is \( L_{ijkl} = LS_{ijkl}^0 \) [105].

The explicit form is:

\[
\frac{\partial \varepsilon_{ij}^0(\mathbf{r}, \tau)}{\partial \tau} = -LS_{ijkl}\Delta C_{klmn}(\mathbf{r})\left\{ \frac{1}{2} \int \frac{d^3k}{(2\pi)^3} [n_m\Omega_{no}(\mathbf{n}) + n_n\Omega_{mn}(\mathbf{n})]\tilde{\sigma}_{op}(\mathbf{k})n_p e^{ikr} \\
- \Delta S_{mnop}(\mathbf{r})C_{opqr}^0[\varepsilon_{qr}(\mathbf{r}) - \varepsilon_{qr}^*(\mathbf{r})] - \varepsilon_{mn}^*(\mathbf{r}) + \varepsilon_{mn}^0 + S_{mnop}^0\sigma_{op}^t \right\} 
\]

(2.72)

where \( \varepsilon_{ij}^0 = (1/V) \int_{V} \varepsilon_{ij}^0(\mathbf{r})d^3r \).

Thus, the virtual strain of the equivalent elastically homogeneous system is obtained, which provides the same stress and total strain distribution in the system comparing to the original elastically inhomogeneous system.
The total strain in the system is:

\[ \varepsilon_{ij}(\mathbf{r}) = \bar{\varepsilon}_{ij} + \frac{1}{2} \int \frac{d^3k}{(2\pi)^3} \left[ n_i \Omega_j k(n) + n_j \Omega_i k(n) \right] \bar{\sigma}_{kl}(k) n_l e^{ikr} \]  

(2.73)

Where \( \bar{\varepsilon}_{ij} = \varepsilon_{ij}^0 + \tilde{\sigma}_{ijkl}^0 \sigma_{kl}^{\text{ex}} \).

And the stress is:

\[ \sigma_{ij}(\mathbf{r}) = C_{ijkl}^0 \left[ \varepsilon_{kl}(\mathbf{r}) - \varepsilon_{kl}^0(\mathbf{r}) \right] \]

\[ = C_{ijkl}^0 \left[ \left( \frac{1}{2} \int \frac{d^3k}{(2\pi)^3} \left[ n_i \Omega_{lm} k(n) + n_l \Omega_{km} k(n) \right] \sigma_{mn}(k) n_n e^{ikr} \right) + \varepsilon_{kl}^0 - \varepsilon_{kl}^0(\mathbf{r}) \right] + \sigma_{ij}^{\text{ex}} \]  

(2.74)

### 2.5 Phase field dislocation model

Phase field dislocation model was first developed by Y.U. Wang et al in 2001 [69]. A dislocation loop in the slip plane is described as a coherent misfitting platelet inclusion [106]. Inside the plate is the sheared region and the boundary between sheared and unsheared region on the slip plane is defined as the dislocation line. The thickness of the plate is the interplanar spacing \( d \). As shown in Fig 2.1 [21], the region inside the loop is sheared by a Burgers vector \( b \). The stress-free strain is expressed as \( \varepsilon_{ij}^0 = \frac{b n_i + b n_j}{2d} \). \( n \) is the slip plane normal.

A field variable \( \eta \) is used to describe a certain type of dislocation (defined by the slip plane and Burgers vector). Sheared region is marked as 1, unsheared is 0. The energy formulation for dislocation evolution is made up of 4 terms:

\[ E = E^{\text{crystal}} + E^{\text{gradient}} + E^{\text{elastic}} + W^{\text{ext}} \]

The first term is crystal energy, which is from the energy increase of atoms deviating from their equilibrium positions. The second term is gradient energy. This term can be addressed as the correction of non-linear elastic energy and make sure dislocation profile is diffuse along its slip plane in order to observe the kinetics of dislocations. The third term is the Elastic
energy. The last term is the external work done on the system because of a stress-controlled boundary condition. More sophisticated and accurate dislocation models are developed based on this which will be introduced in detail in the following chapter. Dislocation interaction is considered by modifying the formulation of gradient and crystal energy term and the introduction of γ surface can assure more accurate crystal energy calculation [70, 71, 107, 108].
CHAPTER 3

CONTRIBUTION TO RAFTING FROM CHANNEL DISLOCATIONS

3.1 Phase field model of γ rafting

Rafting of Ni-base blade alloys is a process dominated by simultaneous preferential dissolution/deposition of γ through diffusive mass flux in the γ-channels [28]. In the phase field model described below, the evolution of γ/γ’ microstructure proceeds via solute redistribution subject to chemical potential gradients, which includes contributions from the existing γ/γ’ microstructure, channel dislocations and external load, while the evolution of dislocations is governed by their elastic interactions with the γ/γ’ microstructure under external load.

A general form of the total free energy of a system that consists of both precipitate microstructure and phase field description of dislocations will be formulated on the basis of gradient thermodynamics [79] and phase field microelasticity [83, 84, 87]:

\[ F = F[\{\phi\}, \{\eta\}] \]
\[
\begin{align*}
&= \int dx \left[ f(\{\phi\}) + f^d(\{\eta\}) + \sum_{p, p'=1}^N \frac{\alpha_{pp'}}{2} \nabla \phi_p \nabla \phi_{p'} + \sum_{q, q'=1}^M \frac{\beta_{qq'}}{2} \nabla \eta_q \nabla \eta_{q'} \right] \\
&+ \frac{1}{2} \int \frac{dg}{(2\pi)^3} \left[ C_{ijkl} \tilde{\varepsilon}_{ij}^T(g) \tilde{\varepsilon}_{kl}^T(g) - n_i \tilde{\sigma}_{ij}(g) \Omega_{jk}(n) \tilde{\sigma}_{kj}^T(g) n_j \right] \\
&- \int dx \left[ \sigma_{ij}^{appl}(x) \varepsilon_{ij}(x) \right]
\end{align*}
\]  

(3.1)

which characterizes the precipitate microstructure in terms of a set of \( N \) fields, \( \{\phi_p(x) : p = 1, \ldots, N\} \), and the dislocations in terms of a set of \( M \) fields, \( \{\eta_q(x) : q = 1, \ldots, M\} \). Since \( \{\phi\} \) and \( \{\eta\} \) are themselves functions of spatial position \( x \), the total free energy, \( F \), in Eq.3.1 is a functional. The first integral gives the "local" energy densities of the two-phase mixture, \( f \), and of the dislocations, \( f^d \), and the contributions from short-range spatial correlations due to the gradients of \( \{\phi\} \) and \( \{\eta\} \). \( \alpha_{pp'} \) and \( \beta_{qq'} \) are the gradient-energy coefficient matrices. The second integral corresponds to the elastic energy associated with a coherent two-phase microstructure and dislocations, where \( C_{ijkl} \) is the elastic stiffness tensor, \( \tilde{\varepsilon}_{ij}^T(g) \) is the Fourier transform of the stress-free strain tensor, \( \tilde{\sigma}_{ij}(g) = C_{ijkl} \tilde{\varepsilon}_{kl}^T(g) \), and \( \Omega_{jk}(n) \) is the Green function tensor inverse to \( \Omega_{jk}^T(n) = C_{ijkl} n_i n_j \). The superscript asterisk indicates the complex conjugate. The last integral is the mechanical work under an external load, \( \sigma_{ij}^{appl}(x) \). We may assume that the stress-free strain, \( \varepsilon_{ij}^T \), is contributed by both dislocations and precipitates in a linear combination:

\[
\varepsilon_{ij}^T(x) = \sum_{p=1}^N \varepsilon_{ij}^{T0,\phi_p}(x) \phi_p(x) + \sum_{q=1}^M \varepsilon_{ij}^{T0,\eta_q}(x) \eta_q(x)
\]  

(3.2)

where \( \varepsilon_{ij}^{T0,\phi_p} \) and \( \varepsilon_{ij}^{T0,\eta_q} \) are constant strain tensors given respectively by \( \varepsilon_{ij}^{T0,\phi_p} \equiv (\partial \varepsilon_{ij}^T/\partial \phi_p)_{\phi_p=0} \) and \( \varepsilon_{ij}^{T0,\eta_q} \equiv (\partial \varepsilon_{ij}^T/\partial \eta_q)_{\eta_q=0} \).
The driving forces for the evolution of precipitate microstructure and dislocations are given by the variation of $F$ with respect to the corresponding fields:

\[
\frac{\delta F}{\delta \phi_p} = \frac{\partial f}{\partial \phi_p} - \alpha_p \nabla^2 \phi_p(x) + \left\{ C_{ijkl} \tilde{\epsilon}_{ij}^T(g) \epsilon_{kl}^{T0,\phi_p} - n_i \tilde{\sigma}_{ij}^T(g) \Omega_{jk}(n) \sigma_{kl}^{T0,\phi_p} n_l \right\}_x - \sigma_{ij}^{appl}(x) \epsilon_{ij}^{T0,\phi_p}
\] (3.3)

\[
\frac{\delta F}{\delta \eta_q} = \frac{\partial f^d}{\partial \eta_q} - \beta_q \nabla^2 \eta_q(x) + \left\{ C_{ijkl} \tilde{\epsilon}_{ij}^T(g) \epsilon_{kl}^{T0,\eta_q} - n_i \tilde{\sigma}_{ij}^T(g) \Omega_{jk}(n) \sigma_{kl}^{T0,\eta_q} n_l \right\}_x - \sigma_{ij}^{appl}(x) \epsilon_{ij}^{T0,\eta_q}
\] (3.4)

In the above, $\{ \ldots \}_x$ denotes an inverse Fourier transform, $\sigma_{ij}^{T0,\phi_p} \equiv C_{ijkl} \epsilon_{kl}^{T0,\phi_p}$ and $\sigma_{ij}^{T0,\eta_q} \equiv C_{ijkl} \epsilon_{kl}^{T0,\eta_q}$.

The time evolutions of $\{ \phi \}$ and $\{ \eta \}$ then follow the Onsager type kinetics mentioned in Chapter 2, which may be written in a compact form as

\[
\frac{\partial \{ \phi, \eta \}}{\partial t} = \hat{M} \frac{\delta F}{\delta \{ \phi, \eta \}}
\] (3.5)

where $\hat{M}$ is an operator that relates the driving force on a field proportionally to its rate of change. In the simplest forms, $\hat{M} = M \nabla^2$ for a conserved field and $\hat{M} = -L$ for a non-conserved field, with $M$ and $L$ being scalars.

In what follows we will give the detailed forms of Eqs.3.1 through 3.5 for the present study of rafting in Ni-base alloys.

### 3.1.1 Chemical free energy for $\gamma/\gamma'$ microstructure

For a binary Ni-Al alloy without considering the effect of anti-phase domains, the composition of Al is all one needs to characterize the $\gamma/\gamma'$ microstructure, i.e.,
\{\phi\} \rightarrow c(x) \text{ (mole fraction)}. The local chemical free energy density for the two-phase mixture may be approximated by a double-well shaped function:

\[ f \equiv f^d(c) = f_0 V_m^{-1} (c - c_{\gamma}^c)^2 (c - c_{\gamma'}^c)^2 \]  

(3.6)

where \( f_0 = 1.058 \times 10^8 J/mol \), \( V_m = 1 \times 10^{-5} m^3/mol \), \( c_{\gamma}^c = 0.16 \) and \( c_{\gamma'}^c = 0.229 \). Although more sophisticated free energy models [75, 109] can be applied to incorporate the effect of anti-phase domains during rafting, it is not the focus of the present work. The above parameters give the equilibrium compositions of the \( \gamma \) and \( \gamma' \) phases, respectively, \( c_{\gamma}^c = 0.16 \) and \( c_{\gamma'}^c = 0.229 \), and a free energy hump between the two phases, \( \Delta f = 149.9 J/mol \), (Fig. 3.1), which corresponds to a binary Ni-Al alloy at approximately 1300K, as compared to the free energy model given by Ansara et. al. [109].

The gradient-energy coefficient in Eq.3.1 is chosen as \( \alpha^* = 0.2 \), which yields an interfacial energy of 223\text{mJ/m}^2 and an interfacial width of 33.3nm for the \( \gamma/\gamma' \)
interface. The high value of the interfacial energy is required to maintain a diffuse interface at length scales of experimental relevance. Since particle shape during coarsening is governed by the ratio of elastic energy over interfacial energy rather than their individual values, rescaling the elastic energy contribution from lattice misfit through the relation \( \frac{(Ce^2)^3}{\gamma^2} \)\(^{[75]} \) to maintain a desired particle shape for the \( \gamma' \) particles reduces the effective interfacial energy down to 50.2\( mJ/m^2 \), which is a reasonable value for Ni-base superalloys.

### 3.1.2 Free energy for dislocations

As already mentioned in Chapter 2, for dislocations the conventional local free energy can be replaced by a sinusoidal function that mimics a periodic potential energy in a crystal with respect to a continuous shear displacement between two adjacent atomic planes\(^{[69]} \). Furthermore, this local energy can be directly related to the generalized stacking fault energy\(^{[70]} \). A simple form that takes into account all 12 primary slip systems in an f.c.c. crystal may be given as\(^{[70]} \)

\[
f^d = \frac{\mu(b/d)^2}{2\pi^2} \left[ 4 - \cos\pi (\eta_{[110]}^{[011]} - \eta_{[111]}^{[011]} ) \cos\pi (\eta_{[111]}^{[101]} - \eta_{[111]}^{[110]} ) \right. \\
\times \cos\pi (\eta_{[111]}^{[010]} - \eta_{[111]}^{[010]} ) - \cos\pi (\eta_{[111]}^{[010]} - \eta_{[111]}^{[101]} ) \\
\times \cos\pi (\eta_{[111]}^{[010]} - \eta_{[111]}^{[010]} ) \cos\pi (\eta_{[111]}^{[110]} - \eta_{[111]}^{[110]} ) \\
- \cos\pi (\eta_{[111]}^{[011]} - \eta_{[111]}^{[011]} ) \cos\pi (\eta_{[111]}^{[101]} - \eta_{[111]}^{[101]} ) \\
\times \cos\pi (\eta_{[111]}^{[110]} - \eta_{[111]}^{[110]} ) \cos\pi (\eta_{[111]}^{[111]} - \eta_{[111]}^{[111]} ) \\
\left] \right)
\]

(3.7)
where $\mu$ is the shear modulus of the $\{111\}\langle110\rangle$ slip system: $\mu_{\{111\}\langle110\rangle}$, $b$ and $d$ are respectively the magnitude of $1/2\langle110\rangle$ Burgers vector and that of the inter-planar distance of $\{111\}$ slip planes. In Eq. 3.7 we have specifically labeled the disregistry field, $\eta$, with the slip plane and slip direction respectively as the sub- and super-scripts. For dislocations, the gradient term $\sum_{q,q'=1}^{M} (\beta_{qq'} / 2) \nabla \eta_q \nabla \eta_{q'}$ in Eq. 3.1 may be replaced by \[ \frac{\beta}{2} \sum_{q,q'=1}^{M} \left[ (n(q) \times \nabla \eta_q) \cdot (n(q') \times \nabla \eta_{q'}) \right] \] (3.8)

which automatically takes into account the pair-wise interactions among different slip systems according to Frank’s rule. In the above equation, $n(q)$ denotes the slip plane normal associated with the field $\eta_q$.

3.1.3 Stress-free strain

If we assume that the lattice parameter of the binary Ni-Al alloy is a linear function of composition (i.e., Vegard’s law), then the stress-free transformation strain field can be described by the following equation

$$\varepsilon_{ij}^{T,c}(x) = \frac{2(a_{\gamma'} - a_{\gamma})}{(a_{\gamma'} + a_{\gamma})} \cdot \frac{1}{c_{\gamma'}^{c} - c_{\gamma}^{c}} \left[ c(x) - c_{\gamma}^{c} \right] \delta_{ij} \equiv \varepsilon_{ij}^{T,c}(x) \delta_{ij}$$

(3.9)

where $a_{\gamma'}$ and $a_{\gamma}$ are the equilibrium lattice parameters of the $\gamma'$ and $\gamma$ phase, respectively, and $\delta_{ij}$ is the Kronecker delta. The eigenstrain field of a dislocation can be expressed as a dyadic product of the slip plane normal, $n(q)$, and the Burgers vector, $b_0(q)$, associated with $\eta_q$ [69], i.e.,

$$\varepsilon_{ij}^{T,\eta_q}(x) = \frac{n(q) \otimes b_0(q) + b_0(q) \otimes n(q)}{2d} \eta_{q}(x)$$

(3.10)
Substitute Eqs. 3.9 and 3.10 into Eq. 3.2, we have

$$
\varepsilon_{ij}^T(x) = \frac{2(a_{\gamma'} - a_{\gamma})}{(a_{\gamma'} + a_{\gamma})} \cdot \frac{1}{c_{\gamma'} - c_{\gamma}} \left[ c(x) - c_{\gamma} \right] \delta_{ij} + \sum_{q=1}^{M} \frac{n(q) \otimes b_0(q) + b_0(q) \otimes n(q)}{2d} \eta_q(x)
$$

(3.11)

Throughout this chapter, $\delta = 2(a_{\gamma'} - a_{\gamma})/(a_{\gamma'} + a_{\gamma})$ is chosen as $\pm 0.3\%$ for the cases of positive and negative lattice misfit, respectively.

3.1.4 Simultaneous evolution of precipitates and dislocations

The time-evolution of a $\gamma/\gamma'$ microstructure in a binary Ni-Al alloy is characterized by Eq. 3.5, written in terms of a concentration field:

$$
\frac{1}{V_m^2} \frac{\partial c(x,t)}{\partial t} = M_c \nabla^2 \frac{\delta F}{\delta c}
$$

(3.12)

where $V_m$ is the molar volume, $M_c$ is the chemical mobility that is assumed constant throughout the simulations for simplicity. $M_c = 1 \times 10^{-17} J^{-1} \text{mol}^2 \text{m}^{-1} \text{s}^{-1}$ corresponds to an interdiffusivity of $D_{NiAl} = 1 \times 10^{-16} \text{m}^2 \text{s}^{-1}$ in Ni-Al at 1253K [66]. A semi-implicit spectra method [110] is used in the numerical solution of Eq. 3.12.

Dislocations considered in the present study are all gliding dislocations. Their motion is governed by Eq. 3.5 for the non-conserved disregistry fields:

$$
\frac{\partial \eta_q(x,t)}{\partial t} = -L_\eta \frac{\delta F}{\delta \eta_q}
$$

(3.13)

where $L_\eta$ characterizes dislocation mobility and is assumed constant for all dislocations. In practice, relaxation of dislocation configurations is much faster than that of precipitate microstructures which requires long-range diffusion. Therefore the coupling of the two dynamic evolution processes is insensitive to the value of
as long as it is sufficiently large to yield a much faster kinetics as compared to solute diffusion.

3.2 Validation of phase field dislocation method

Since the stress field associated with channel dislocations directly influences $\gamma'$ rafting, the accuracy of stress calculation in the phase field dislocation model is critical for an accurate prediction of the rafting kinetics. To validate the phase field model of dislocations, critical stresses for two simple dislocation configurations in an elastically isotropic solid are calculated and the results are compared with analytical solutions. The first configuration is an infinitely long straight screw dislocation dipole and the second one is a circular dislocation loop. The critical stresses for these two configurations are related closely to those for dislocations filling in the $\gamma$-channels. In the phase field calculations, the dipole distance and the loop radius are varied and the critical stresses are determined. In both cases the applied stress is parallel to the slip plane and along the Burgers vector direction.

Two different methods are used to calculate the critical stress, the first one (referred to hereafter as Method-1) is to start with a sub-critical value of the applied stress and gradually increase the stress magnitude until the state of the dipole or the loop switches from shrinkage to expansion. The increment of the stress is reduced successively until the final critical stress is found within a pre-determined accuracy. The second approach (referred to hereafter as Method-2) starts with a given dislocation configuration and a small perturbation is applied (e.g. the dipole distance or loop radius is varied by one pixel on the discrete numerical grid). The critical stress is then approximated by $\Delta E^{el}/\Delta(\int dx \mathbf{e}_{ij}^{s} \eta_{q}(x))$, where $\Delta E^{el}$
and \( \Delta(\int d\mathbf{x} \varepsilon_{ij}^{T,\eta_i}(\mathbf{x})) \) are the differences in total elastic energy and total stress-free strain between the two states, respectively. Because of the finite (in contrast to an infinitesimal) variation in the dislocation configuration considered in the phase field calculations, Method-2 yields an “averaged” stress around the true critical stress. The error decreases with the ratio of \( \Delta(\int d\mathbf{x} \varepsilon_{ij}^{T,\eta_i}(\mathbf{x})) / (\int d\mathbf{x} \varepsilon_{ij}^{T,\eta_i}(\mathbf{x})) \). An advantage of Method-2 over Method-1 is that it can provide an estimate of the critical stress even when a large phase field grid size is used, where dislocations becomes pinned numerically on the discrete grid and Method-1 fails accordingly. It is, however, limited to simple dislocation geometries such as straight dipoles and circular loops in isotropic media. For more complicated 3D dislocation configurations in elastically anisotropic media such as \( \gamma \)-channel filling dislocations, Method-1 is used to determine the critical stresses.

3.2.1 Critical stress of a straight dislocation dipole

The stress component \( \tau_{yz} \) of a straight screw dislocation located at the origin \( \text{Fig. 3.2 in an elastically isotropic media is plotted as a function of distance, H. The open circles are obtained from the phase field calculations with an un-relaxed (sharp) core and the solid line is from the analytical solution for a Volterra type dislocation: } \tau_{yz} = \mu b / (2\pi H) \). The results agree well with each other. Fig. 3.3 shows the critical shear stress as a function of the separation, H, between two dislocations in a dipole. The dotted line is from analytical solution and the discrete symbol with error-bar is obtained from phase field calculations. Since in the phase field calculations the smallest perturbation cannot be smaller than a grid size, the error-bars in Fig. 3.3 correspond to results obtained by reducing or increasing the separation,
Figure 3.2: Comparison between phase field calculation (circle) and analytical solution (line) of $\tau_{yz}$ as a function of distance for a straight screw dislocation.

$H$, by one grid point. Note that when the distance between the two dislocations in the dipole becomes smaller, the error-bar becomes larger because the ratio of one grid size over the separation distance, $H$, becomes bigger. However, as long as the simulation grid size is fine with respect to the separation distance, the phase field calculations agree well the analytical solutions.

Figure 3.4 shows the shear stress $\tau_{xz}$ distribution near a screw dislocation at the center. It is worth noting that due to the intrinsic property of the phase field dislocation model, dislocation is always described as the boundary between sheared and un-sheared region, and that’s the reason why there is a corresponding screw dislocation at the corner of the simulation cell which has an inverted sense vector direction and forms a screw dislocation dipole with the center one. Because the periodic boundary condition used in calculation, the configuration can be thought as periodic repeating screw dislocation dipoles in 2D. And by superimposing the
Figure 3.3: Critical stress of a dipole of two infinitely long straight dislocations obtained by the phase field model (with error bars) and by analytical solution (-). (The error-bars correspond to results obtained by reducing or increasing the separation, H, by one grid point in phase field)
analytical solution of shear stress for a single screw dislocation, the result is plotted on top of the numerical simulation result by using phase field dislocation model. From the figure, it can be seen that the stress distribution calculated from phase field dislocation method (using the stress calculation method for micro-elasticity theory explained in Chapter 2) agrees well with the result from analytical solution. The small undulation of the phase field result is due to the discreteness of the grid based calculation.

### 3.2.2 Critical stress of a dislocation loop

For a Volterra type dislocation, in order to avoid singularity in the elastic energy calculation, a core cutoff $\rho$ is introduced in the analytical expression of the critical shear stress [111]:

$$\tau_c \cdot r = \frac{\mu b}{4\pi} \times \left[ \ln\left(\frac{4r}{\rho}\right) - 1 \right]$$

(3.14)

where $r$ is the dislocation loop radius and $\tau_c$ is the critical shear stress. A linear relation between $\tau_c \cdot r$ and $\ln(r)$ is expected with a constant slope of $\mu b / (4\pi)$. As shown in Fig. 3.5, the crosshairs are the values obtained by Method-2, while the line is fitted to the crosshairs with the slope fixed to $\mu b / (4\pi)$. The results show that the slope from the phase field calculations agrees well with the analytical solution. From the analytical solution, the intercept of the line is $\frac{\mu b}{4\pi} \times \left[ \ln(4r/\rho) - 1 \right]$, which relates to the core cutoff $\rho$ for Volterra type dislocation. In Fig. 3.5, a cutoff radius $\rho = 0.15b$ is found when the slope of the fitting line is fixed to $\mu b / (4\pi)$. Calculations based on the Peierls dislocations by the microscopic phase field dislocation model indicate a similar result [112].
Figure 3.4: Stress component $\tau_{xz}$ near screw dislocations (smooth curves are obtained from analytical solution and wavy curves are from phase field calculation)
3.2.3 Critical channel dislocation density

Analytical solutions for critical stress are available only for a few simple cases. For more complicated dislocation configurations, discrete dislocation models (DDM) have been used widely in stress distribution and elastic energy calculations. For example, Probst-Hein et. al. [19] have applied DDM to calculate the critical number of dislocations in γ-channels under an applied shear stress. Straight dislocations from one slip system in one type of γ-channels in an elastically isotropic crystal were considered to simplify the calculations. The misfit stress on a cross-section of the γ/γ′ microstructure was first calculated using a finite element method (FEM) and the result was used as input to the DDM model with uniform property along the dimension normal to the cross-section.
To make a direct comparison with this DDM calculation, a similar approach is employed in the phase field calculations. The misfit stress from a 3D $\gamma/\gamma'$ microstructure is first calculated and the stress field on a 2D cross-section along $\gamma'$ cube face diagonal (i.e., $\bar{1}10$ plane) is used in a 2D phase field dislocation model (equivalent to infinitely long straight dislocations normal to the cross-section). Figure 3.6 (a) shows the von-Mises stress distribution for a $-0.3\%$ misfit alloy on the cross-section. The critical number of dislocation dipoles in a $\gamma$-channel is calculated using Method-2. Perturbation is made only to the dislocation dipole in the middle of the $\gamma$-channel. The critical resolved stress is then calculated with respect to the channel dislocation density and lattice misfit. Instead of fixing the applied shear stress (85MPa) and counting the critical number of channel dislocation loops (CNCDL)[19], different number of dislocation dipoles are introduced in the $\gamma$-channel and the critical shear stresses required to stabilize the dislocation dipole in the middle are calculated. The dislocation configuration used in the calculation is shown schematically in Fig. 3.6 (b). The volume fraction of $\gamma'$ is $53.3\%$. According to Probst-Hein et. al. [19], the volume fraction of $\gamma'$ (from $50\%$ to $70\%$) has little effect on the CNCDL. The length of $\gamma'$ edge plus $\gamma$-channel width is $1\mu m$. The dislocations all belong to the same $1/2[101](\bar{1}11)$ slip system. As shown in the Fig. 3.6 (c), for $+0.05\%$ lattice misfit, a critical shear stress of 85MPa corresponds to a CNCDL of 12 (by linear interpolation). This result agrees well with the DDM calculation result which is about 11. Extrapolation of the critical shear stress for the cases of $-0.02\%$ and $-0.3\%$ lattice misfit up to 85MPa yields respectively $\sim 18$ and $\sim 44$ dislocation dipoles in the phase field calculations while the DDM calculations predicted $\sim 20$ and $\sim 50$ dislocation dipoles. The discrepancy between the two
could be caused by the linear extrapolation in the phase field calculations. Such an extrapolation is necessary because the density of channel dislocations that can be considered in the phase field calculations is limited by the smallest mesh size [71, 21], for example, the inter-dislocation distance has to be significantly larger than the mesh size in order to obtain a reasonable accuracy.

3.3 Simulation results of rafting using phase field dislocation method

3.3.1 Initial $\gamma/\gamma'$ microstructure

In the initial microstructure, the $\gamma'$ cube edge length is set to be 400nm and the $\gamma$ channel length is 93.3nm. The volume fraction of the $\gamma'$ phase is 53.3%. The elastic constants are assumed to be the same for both phases, e.g., $C_{11} = 231\, \text{GPa}$, $C_{12} = 149\, \text{GPa}$, and $C_{44} = 117\, \text{GPa}$ [113]. The Burgers vector is $b = 0.25\, \text{nm}$. Simulations are carried out with a 3D system of 128 $\times$ 104 $\times$ 180 grid points and periodic boundary conditions are applied along all three dimensions. Figure 3.7 show the fully relaxed microstructure with $\pm 0.3\%$ lattice misfit that is used as the initial input for dislocation channel filling simulations. Hereafter the $\gamma$-channels parallel to [001] orientation (the loading axis) will be referred to as parallel channels (there are two parallel channels: (100) and (010) channels), and the $\gamma$-channel normal to [001] orientation will be referred to as normal channels ((001) channels).

3.3.2 Channel dislocation configuration

As observed from experiments [13, 28, 15], dislocations in the matrix phase of Ni-base superalloys are mostly $1/2\langle 110 \rangle$ type. Considering all 12 slip systems in an f.c.c single crystal, only 8 of them will have non-zero Schmidt factor under [001]
Figure 3.6: (a) von-Mises stress distribution in a γ/γ microstructure of −0.3% lattice misfit (isotropic elasticity with shear modulus of 74.8GPa, the stress is normalized by 1.3839GPa); (b) Dislocation configuration used in calculation: dislocation dipoles in one γ channel; (c) Critical number of dislocation loops in the γ channel predicted by the phase field model for three different values of lattice misfit under 85MPa applied shear stress.
Figure 3.7: Initial $\gamma/\gamma'$ microstructure (with $\pm 0.3\%$ misfit). The edge length of the $\gamma'$ particles is 400nm and the $\gamma$ channel width is 93.3nm.

Compression or tension. For simplicity, only two of them are included in the simulations, i.e., $(1/2[011](\bar{1}\bar{1}1) \text{ and } 1/2[011](\bar{1}\bar{1}1))$. All the other remaining activated slip systems can be represented by these two through symmetry operations along the four-fold [001] axis and therefore will have similar effects. As the present rafting study focuses on the symmetry breaking of the initial cubic shape of the precipitates, this simplification will not influence the type of rafting. In addition, as dislocations start to fill in the initially dislocation-free channels, it is reasonable to assume that only one or two slip systems may dominate locally due to the scattered dislocation sources.

With the relaxed $\gamma/\gamma'$ microstructure as input, dislocations from the two slip systems are deposited on the $\gamma/\gamma'$ interface alternately on multiple (\bar{1}\bar{1}1) slip planes. Because cutting of $\gamma'$ particles is considered a rare event before rafting completes,
dislocations are confined in the $\gamma$-channels and completely wrap around $\gamma'$ particles. Subsequent relaxation of the system leads to equilibrium dislocation configurations. Because of the complicated geometry of the channel dislocations, Method-1 described in Section 3 is used to determine the equilibrium channel dislocation configurations. For the case with $-0.3\%$ lattice misfit under 152MPa [001] tension, different channel dislocation densities (with dislocation spacing along the $\gamma/\gamma'$ interface varying from 33nm to 98nm) are used as the initial configurations. Figure 3.8 (a) shows the dislocations channel distribution for spacing=33nm case. If we look more closely onto those individual slipped planes, on some $\bar{1}11$ planes, dislocations are no longer able to penetrate the normal channel (as shown on Fig. 3.8 (b) and (c)). As the initial dislocation spacing is an input for the simulation, the configuration (initial sheared plane spacing value) for the ensuing rafting simulation is chosen using the following criterion: The smallest value of sheared plane spacing is chosen provided with dislocations on each slipped plane are able to thoroughly penetrate through the normal channel ($\gamma$ channel normal to the applied tension direction) which is favored by the loading condition and lattice misfit.

The simulation results of different dislocation spacing show that as the interfacial dislocation spacing becomes smaller than 65nm, some of the dislocation dipoles in the normal channel start to collapse. This stable spacing is close to the ones observed in experiments [63, 27, 114, 51], i.e., 52nm under 170MPa tensile stress and 32nm under 200MPa tensile stress. Though by allowing dislocation cross-slip and rearrangement may lead to even higher dislocation density compare to the model prediction presented here, the following simulations in this chapter
Figure 3.8: (a) Equilibrium dislocation structure in a γ/γ' microstructure of −0.3% lattice misfit under 152MPa [001] tensile stress with sheared plane spacing=33nm; (b) One of the 1/2[101] dislocations on a (111) plane which fails to penetrate the normal channel; (c) One of the 1/2[011] dislocations on a (111) plane which fails to penetrate the normal channel. (Dislocations are the boundaries between the slipped and un-slipped regions)
will always choose interface dislocation spacing=65nm as the starting configuration. The equilibrium 3D dislocation configuration obtained is shown in Fig. 3.9 (a). Figure 3.9 (b) and (c) indicate that the dislocations stay in the normal channel and one of the parallel channels that is parallel to the Burgers vector direction. Figure 3.10 show the equilibrium dislocation configuration with 91.4MPa [001] tension. Compare Fig. 3.10 (b) with Fig. 3.9 (c), dislocation has been driven out of the parallel channel and a part of the normal channel. In these figures and the ones that will follow, dislocations are the boundaries between the slipped and un-slipped regions.

Figure 3.11(a) shows the equilibrium dislocation configuration obtained for the case with +0.3% misfit and 152MPa tension along [001]. The results indicate that dislocations from both slip systems (1/2[101](\overline{1}11) and 1/2[011](\overline{1}11)) prefer parallel channels in the case of positive misfit and under tension. More details can be seen on a \(\overline{1}11\) slip plane that contains 1/2[101] dislocations (Fig. 3.11(b)). The line direction of the dislocation is along [011], forming a 60 degree angle with the Burgers vector. While on another \(\overline{1}11\) plane with a 1/2[011] dislocation (Fig. 3.11(c)), the dislocation line direction is along [101], forming also a 60 degree angle with the Burgers vector. Increasing the applied stress to 274MPa squeezes the dislocations into the another parallel channel with pure screw segments deposited along the \(\gamma/\gamma\) interface (see Fig. 3.12 (a) and (b) for a \(\overline{1}11\) plane with 1/2[101] and 1/2[011] dislocations).

Following the analysis given by J. Buffiere and M. Ignat [51], the extra half plane position of the edge component of a dislocation is analyzed for different slip systems in different \(\gamma\)-channels under tension/compression and the results are
Figure 3.9: (a) Equilibrium dislocation structure in a $\gamma/\gamma'$ microstructure of $-0.3\%$ lattice misfit under 152MPa [001] tensile stress with sheared plane spacing=65nm; (b) Equilibrium structure of the $1/2[101]$ dislocations on $\overline{111}$; (c) Equilibrium structure of the $1/2[011]$ dislocations on $\overline{111}$. (Dislocations are the boundaries between the slipped and un-slipped regions)
Figure 3.10: Equilibrium dislocation structure on (¯111) for a negative lattice misfit of $-0.3\%$ under 91.4MPa [001] tensile stress. (a) $1/2[101]$ dislocations and (b) $1/2[011]$ dislocations. Dislocations are the boundaries between the slipped and un-slipped regions.

listed in Table 3.1. In the case of positive lattice misfit, the $\gamma$-channels in an initial dislocation-free $\gamma/\gamma'$ microstructure are under tension. When dislocations begin to fill in the channels under the applied stress, the tensile stress in the channels can be partly relieved by the insertion of extra half planes associated with the edge components of the dislocations into the $\gamma$-channels. In this case, the misfit stress provides an additional driving force for the dislocations to enter the $\gamma$-channels. If the extra half planes of the dislocations are in the $\gamma'$ phase, however, the misfit stress will resist entry of the dislocations into the channel. In the case of negative lattice misfit, the $\gamma$-channels are under compression and dislocations with their extra half plane in the $\gamma'$ phase will be favored by the misfit stress. The elastic interactions between the misfit stress and the stress fields of dislocations based on such
Figure 3.11: (a) Equilibrium dislocation structure in a $\gamma/\gamma'$ microstructure of $+0.3\%$ lattice misfit under 152MPa [001] tensile stress; (b) Equilibrium structure of the $1/2[101]$ dislocations on ($\bar{1}\bar{1}1$); (c) Equilibrium structure of the $1/2[011]$ dislocations on ($\bar{1}\bar{1}1$). (Dislocations are the boundaries between the slipped and unslipped regions)
Figure 3.12: Equilibrium dislocation structure on (\overline{1}11) for a positive lattice misfit of +0.3% under 274MPa [001] tensile stress. (a) 1/2[101] dislocations and (b) 1/2[011] dislocations. Dislocations are the boundaries between the slipped and un-slipped regions.

An analysis are listed in Table 3.2. Combining with Table 3.1, dislocation channel preference can be easily determined. For example, for a system with a positive lattice misfit under tension or negative misfit under compression, dislocations prefer channels parallel to the loading direction, while for positive lattice misfit under compression or negative misfit under tension, dislocations prefer channels normal to the loading direction. The phase field simulation results agree with these conclusions.

3.3.3 \(\gamma'\) rafting

The simulated equilibrium dislocation configurations and the initial \(\gamma/\gamma'\) microstructure under a given applied stress and lattice misfit serve as the inputs for
<table>
<thead>
<tr>
<th>Slip plane</th>
<th>Burgers vector</th>
<th>(100)Channel (parallel)</th>
<th>(010)Channel (parallel)</th>
<th>(001)Channel (normal)</th>
</tr>
</thead>
<tbody>
<tr>
<td>[001]Compression</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(111)</td>
<td>1/2[İ01]</td>
<td>γ' phase</td>
<td>Pure screw</td>
<td>γ channel</td>
</tr>
<tr>
<td></td>
<td>1/2[İ10]</td>
<td>*</td>
<td>*</td>
<td>*</td>
</tr>
<tr>
<td></td>
<td>1/2[0İ1]</td>
<td>Pure screw</td>
<td>γ' phase</td>
<td>γ channel</td>
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<td>γ' phase</td>
<td>Pure screw</td>
<td>γ channel</td>
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<td>1/2[İİ0]</td>
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<tr>
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<td>1/2[0İ1]</td>
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<td>γ' phase</td>
<td>γ channel</td>
</tr>
<tr>
<td>[001]Tension</td>
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<td></td>
</tr>
<tr>
<td>(11İ)</td>
<td>1/2[İ01]</td>
<td>γ channel</td>
<td>Pure screw</td>
<td>γ' phase</td>
</tr>
<tr>
<td></td>
<td>1/2[İİ0]</td>
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<tr>
<td></td>
<td>1/2[0İ1]</td>
<td>Pure screw</td>
<td>γ channel</td>
<td>γ' phase</td>
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<td>1/2[İ01]</td>
<td>γ channel</td>
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<td>γ channel</td>
<td>γ' phase</td>
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<tr>
<td>(İİ1)</td>
<td>1/2[İ01]</td>
<td>γ channel</td>
<td>Pure screw</td>
<td>γ' phase</td>
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<td></td>
<td>1/2[İİ0]</td>
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<td>*</td>
</tr>
<tr>
<td></td>
<td>1/2[0İ1]</td>
<td>Pure screw</td>
<td>γ channel</td>
<td>γ' phase</td>
</tr>
</tbody>
</table>

Table 3.1: Extra half plane position of dislocation edge component for different slip systems in different γ-channels under tension/compression.

<table>
<thead>
<tr>
<th>Misfit</th>
<th>Extra half plane in γ channel</th>
<th>Extra half plane in γ' phase</th>
</tr>
</thead>
<tbody>
<tr>
<td>+</td>
<td>Attractive</td>
<td>Repulsive</td>
</tr>
<tr>
<td>-</td>
<td>Repulsive</td>
<td>Attractive</td>
</tr>
</tbody>
</table>

Table 3.2: Interactions between stress fields from lattice misfit and from dislocations.
the rafting simulations. In these simulations, the $\gamma/\gamma'$ microstructure evolves (following Eq. 3.12) under the stress fields from both lattice misfit and channel dislocations while the dislocation configurations evolve instantaneously (following Eq. 3.13) as the $\gamma/\gamma'$ microstructure changes. The simulated microstructural evolution in a system having $-0.3\%$ lattice misfit and under 152MPa [001] tension is shown in Figure 3.13. The channels parallel to the applied load direction decrease in width and eventually close up. Correspondingly, the initial $\gamma'$ particles of a rounded cubic shape evolve into (001) plates (known as N-type rafting). Given the interdiffusivity employed in the simulations, i.e., $\tilde{D}_{NiAl} = 1 \times 10^{-16} m^2 s^{-1}$, the completely rafted structure is obtained after 14.7 hours, which is reasonably consistent with experimental observations [2, 28, 66, 56, 55, 115]. But whether such diffusivity reflects the effective diffusivity of commercial alloys needs to be established. In a system of $+0.3\%$ lattice misfit under 152MPa [001] tension, the $\gamma$ channels normal to the applied load direction decrease in width and $\gamma'$ rods parallel to the loading direction ([001] rods) develop after 7.2 hours (for $\tilde{D}_{NiAl} = 1 \times 10^{-16} m^2 s^{-1}$), resulting in a P-type rafting (Fig. 3.14). Simulations with $+0.3\%$ misfit under 152MPa [001] compression result in an N-type rafting and simulations with $-0.3\%$ misfit under [001] compression yields a P-type rafting.

A summary of all the simulation results is given in Table 3.3, from which one can see that the rafting type is determined by the product of the sign of lattice misfit and the loading direction (positive for tension and negative for compression). A P-type rafting will develop if the product is positive and an N-type rafting will develop if the product is negative.
<table>
<thead>
<tr>
<th>[001] axis</th>
<th>Misfit channel</th>
<th>Parallel channel</th>
<th>Normal channel</th>
<th>Rafting type</th>
<th>Final morphology</th>
</tr>
</thead>
<tbody>
<tr>
<td>Compression</td>
<td>Positive</td>
<td>Narrowing</td>
<td>Widening</td>
<td>N</td>
<td>(001)plate</td>
</tr>
<tr>
<td>Compression</td>
<td>Negative</td>
<td>Widening</td>
<td>Narrowing</td>
<td>P</td>
<td>[001]rod</td>
</tr>
<tr>
<td>Tension</td>
<td>Positive</td>
<td>Widening</td>
<td>Narrowing</td>
<td>P</td>
<td>[001]rod</td>
</tr>
<tr>
<td>Tension</td>
<td>Negative</td>
<td>Narrowing</td>
<td>Widening</td>
<td>N</td>
<td>(001)plate</td>
</tr>
</tbody>
</table>

Table 3.3: Rafting type w.r.t. the sign of misfit and loading direction.

The simulation results also indicate that P-type rafting completes faster than an N-type rafting under the same magnitude of lattice misfit and applied stress. This is because solutes from two types of parallel channels (i.e., (100) and (010)) jointly fill in the only normal channels (i.e., (001)) for the P-type rafting, while solutes from only the normal type of channels fill in both types of parallel channels for the N-type rafting. Therefore, the solute flux is effectively smaller for the N-type rafting than for the P-type rafting.

3.3.4 Spatial variation of chemical potential in \( \gamma \)-channels

In order to analyze the simulation results, the chemical potential variation in the \( \gamma \)-channels are calculated from the local stress according to \( \mu(x) = -\sigma_{ij}(x)\varepsilon_{ij}^T(x) = -\text{Trace}\{\sigma_{ij}(x)\}\varepsilon_{ij}^T(x) \), where \( \sigma_{ij} \) is the local stress and \( \varepsilon_{ij}^T \) is the dilatational misfit strain between the solute and solvent atoms given by Eq.3.2. In an elastically homogeneous system, the local stress includes contributions from the applied stress, lattice misfit and channel dislocations. Since the applied stress and the trace of the misfit strain are identical in different channels, only the stress field of the channel dislocations causes the chemical potential difference, and hence rafting will not occur in the absence of channel dislocations. However, the external load and lattice
Figure 3.13: Formation of N type raft in an alloy of −0.3% lattice misfit under 152MPa tensile stress along [001]. (a) t=3.6 hours. The arrows indicate the loading direction. (b) t= 7.2 hours; (c) t= 14.7 hours. Slipped regions rather than dislocations are shown in the figures by the shaded areas in the γ-channels.
misfit play an indirect role by biasing the equilibrium configurations of dislocations in each type of channels (see Table 3.1 and Table 3.2).

Figure 3.15 shows the chemical potential contours on the \((\overline{1}1\overline{1})\) planes calculated at \(t=2.4\) hour (with \(\dot{D}_{\text{NiAl}} = 1 \times 10^{-16} m^2 s^{-1}\)). The \(\gamma/\gamma'\) interface on the cross-section is outlined by the thick solid lines. Two different \((\overline{1}1\overline{1})\) cross-sections through the structure are presented in Fig. 3.15 (a) and (b) for a system with \(-0.3\%\) misfit under 152MPa tension. It is shown that the chemical potential in the normal channel is higher than that in the parallel channels. Therefore solute atoms will leave the normal channel and enter the parallel channels, leading to an N-type rafting. The results for a system with \(+0.3\%\) misfit under 152MPa tension are presented in Fig. 3.15 (c) and (d). In this case, the chemical potential in the parallel channels is higher than that in the normal channel and the solute atoms will leave the parallel
channels and enter the normal channel, leading to a P-type rafting. The value of chemical potential difference between the different channels is about $30 \sim 50 J/mol$.

### 3.4 Summary

A general phase field model is formulated to treat the simultaneous evolution of different types of defects in crystalline solids. The model is applied to study, for the first time within the same framework, work hardening in $\gamma$ channels and rafting of $\gamma'$ precipitates. The simulation results show clearly that rafting of $\gamma'$ precipitates is closely related to the type and distribution of dislocations at the $\gamma/\gamma'$ interfaces.
The sign of lattice misfit and the direction of the applied load determine the dislocation configurations in the $\gamma$-channels and the rafting directions. It is found that dislocations prefer normal channels (normal to the direction of applied load) for a positive misfit alloy under compression, and a negative misfit alloy under tension, while parallel channels were preferred by dislocations in a positive misfit alloy under tension and a negative misfit alloy under compression.

The kinetics of rafting is determined by a stress-induced diffusion process. Chemical potential calculations under different conditions verifies that the interaction energy between solute atoms and the local stress $\Delta\mu = -\delta(\sigma_1 + \sigma_2 + \sigma_3)$ can be used as an indication of the chemical potential difference to predict the diffusional flux and rafting direction. The chemical potential difference between different channels is about $30 \sim 50J/mol$ at 1300K under 152MPa tension or compression with $\pm 0.3\%$ lattice misfit. The simulation results show that the rafting process will complete within 14 hours if the interdiffusivity is $\tilde{D}_{NiAl} = 1 \times 10^{-16}m^2/s$ under typical creep conditions for single crystal Ni-base blade alloys, which is reasonably consistent with experimental observations. The P-type rafting develops faster than the N-type rafting given the same magnitude of lattice misfit and applied load.
CHAPTER 4

CONTRIBUTION TO RAFTING FROM MODULUS INHOMOGENEITY

Using an integrated phase field model of dislocation dynamics [69, 70, 116, 107, 71] and γ/γ′ microstructural evolution [117, 75, 77, 118], Chapter 3 has shown that the existence of γ channel dislocations under the influence of applied stress and misfit stress leads to rafting without the consideration of modulus inhomogeneity [119, 120]. Not only do the rafting directions predicted by the simulations agree with experimental observations, but the time required to reach a completely rafted state compares reasonably well with experimental results [28, 2, 66, 56, 55, 115].

In this chapter, the phase field microelasticity model for elastically inhomogeneous systems developed recently [72, 104] is first applied to determine quantitatively the spatial variations of chemical potential of solute atoms in γ channels due to lattice misfit, modulus difference and applied stress, without considering channel dislocations. The results are then compared to those due to channel dislocations obtained under the homogeneous modulus assumption [120]. Finally, the interplay between the contributions from channel plasticity and from elastic inhomogeneity is examined within the same model using a simplified approach for the channel plasticity. The main objective is to provide a quantitative comparison
between the two contributions under experimental conditions in terms of material parameters, temperature and external load.

In the following section, we will first introduce the phase field model used for this study and then focus on challenges associated with solving diffusion equations for elastically inhomogeneous systems. A scaling technique is introduced in order to simulate rafting phenomena taking place at experimentally relevant length and time scales. An effective channel plastic strain is introduced in Section 3, which leads to a simplified model taking into account the coupling between contributions from both the channel plasticity and the modulus inhomogeneity. In Section 4, simulation results on temporal evolution of the $\gamma/\gamma'$ microstructure and the corresponding chemical potential gradient at different stages of the rafting process in the different cases considered are presented. The kinetic features of rafting caused by modulus inhomogeneity, channel plasticity and a combination of the two are discussed in Section 5.

4.1 Phase-Field model of rafting in elastically inhomogeneous system

The total free energy of a system having an arbitrary coherent two-phase microstructure is formulated on the basis of the gradient thermodynamics [79] and phase field microelasticity theory for elastically inhomogeneous system [72, 104]:

$$F = \int dx \left\{ f(c(x)) + \frac{\alpha c}{2} [\nabla c(x)]^2 \right\} + \mathcal{E}_{\text{el}}^{\text{ext}} \left\{ \epsilon_{ij}^p(c(x)), \sigma_{ij}^{\text{ext}}, C_{ijkl}(c(x)) \right\} \quad (4.1)$$

The first integral is the chemical free energy that includes two contributions: the bulk chemical free energy density $f(c(x))$ as a function of local concentration $c(x)$, where $x$ is spatial coordinate, and the gradient energy density that accounts
for the spatial non-uniformity in concentration, with \( \alpha_c \) being the gradient energy coefficient. The second term is the elastic energy of the system, which depends on external applied stress \( \sigma_{ij}^{ex} \), the stress-free transformation strain (SFTS), \( \varepsilon_{ij}^{T}(c(x)) \), and the anisotropic elastic constants, \( C_{ijkl}(c(x)) \), which are different for different phases.

Similar to Eq. 3.9, the lattice parameter of a binary Ni-Al alloy is assumed to be a linear function of solute concentration (i.e., following the Vegard’s law). And the SFTS tensor \( \varepsilon_{ij}^{T}(c(x)) \) can be described as:

\[
\varepsilon_{ij}^{T}(c(x)) = \delta \frac{c(x) - c_\gamma^e}{c_\gamma^e - c_\gamma'^e} \delta_{ij}
\]

(4.2)

Where \( c_\gamma^e \) and \( c_\gamma'^e \) are the equilibrium compositions of the \( \gamma \) and \( \gamma' \) phase, respectively, \( \delta \) is the lattice misfit defined as \( \delta = 2(a_\gamma - a_\gamma')/(a_\gamma + a_\gamma') \), and \( a_\gamma \) and \( a_\gamma' \) are the equilibrium lattice parameters of the \( \gamma \) and \( \gamma' \) phase, respectively. \( \delta_{ij} \) is Kronecker delta.

The driving force for the evolution of precipitate microstructure is given by the variational derivative of the total free energy, \( F \), with respect to solute concentration, i.e.,

\[
\frac{\delta F}{\delta c(x)} = \frac{\partial f(c(x))}{\partial c(x)} - \alpha_c \nabla^2 c(x) + \frac{\delta E_{el}}{\delta c(x)} \left\{ \varepsilon_{ij}^{T}(c(x)), \sigma_{ij}^{ex}, C_{ijkl}(c(x)) \right\}
\]

(4.3)

where the last term on the right-hand side accounts for the elasticity effect from modulus mismatch that leads to \( \gamma' \) rafting under applied stress.

The time evolution of the concentration field (and hence the precipitate microstructure) is governed by the Cahn-Hilliard equation: Eq. 3.12 in Chapter 3 with same chemical mobility: \( M_c = 1 \times 10^{-17} J^{-1}mol^2m^{-1}s^{-1} \).

\[
\frac{1}{V_m} \frac{\partial c(x)}{\partial t} = \nabla \cdot \left\{ M_c \nabla \left[ \frac{\delta F}{\delta c(x)} \right] \right\}
\]

(4.4)
Semi-implicit spectral method is used to solve the equation [110].

### 4.1.1 Approximation of local chemical free energy

In order to do a fair comparison with the channel plasticity contribution from Chapter 3, the chemical free energy in this chapter uses the exact same formula and numerical value as the previous chapter:

\[
f \equiv f^d(c) = f_0 V_m^{-1} (c - c_e^\gamma)^2 (c - c_e^{\gamma'})^2
\]

where \( f_0 = 1.058 \times 10^8 \text{J/mol} \), \( V_m = 1 \times 10^{-5} \text{m}^3/\text{mol} \), \( c_e^\gamma = 0.16 \) and \( c_e^{\gamma'} = 0.229 \).

The gradient-energy coefficient \( \alpha_c \) in Eq. 4.1 is chosen as \( \alpha_c = 1.04 \times 10^{-6} \text{J/m} \), which yields a grid size of 6.66nm, an interfacial energy of \( \sigma = 223 \text{mJ/m}^2 \) and an interfacial width of 33.3nm for the \( \gamma'/\gamma' \) interface. The high value of the interfacial energy is required to maintain a diffuse interface at length scales of experimental relevance in the conventional phase field approach employed here [98, 99]. Such a high interfacial energy will, however, lead to a nearly spherical particle shape rather than a cuboidal shape for the typical lattice misfit and particle size found in Ni-base single crystal superalloys. In this case, the narrow \( \gamma \) channels separating arrays of \( \gamma' \) particles cannot be maintained when the \( \gamma' \) volume fraction is high. Since particle shape during coarsening is governed by the ratio of elastic energy over interfacial energy [121], i.e., \( (C\varepsilon^2R^3)/(\sigma R^2) = C\varepsilon^2 R/\sigma \), where \( R \) is the particle size, rather than by their individual values, for a given \( \sigma \) one can either increase the particle size \( R \) (and correspondingly the system size) or increase the elastic energy contribution to maintain a cube-like shape (because elastic energy scales with particle volume while interfacial energy scales with particle surface area). Because the former is limited by computational resources, the latter approach [75]
is adopted in this study. The implementation of this scaling scheme in an elastically inhomogeneous system turns out to be more complicated than that in an elastically homogeneous system [75] and a new method will be introduced later in Section 2.3. By this new method the interfacial energy has been scaled down effectively to about $50 mJ/m^2$, which is a reasonable value for Ni-base superalloys.

4.1.2 Microelasticity of elastically inhomogeneous system

To calculate the elastic energy of an arbitrary coherent mixture in an elastically anisotropic and inhomogeneous system, the iterative method developed recently by Y.U. Wang et. al [72, 104] is used. By introducing an effective strain, $\varepsilon^0_{ij}(x)$ (which is a function of position), and an arbitrary non-zero reference modulus, $C^0_{ijkl}$, the stress and total strain distribution in an elastically inhomogeneous system were obtained [72, 104] by solving the elasticity problem in an equivalent elastically homogeneous system. $\varepsilon^0_{ij}(x)$ is a variational solution that minimizes the total elastic energy. In practice it can be solved numerically in the time-dependent Ginzburg-Landau type equation [72]:

$$\frac{\partial \varepsilon^0_{ij}(x, \tau)}{\partial \tau} = -L_{ijkl} \frac{\delta E^{el}}{\delta \varepsilon^0_{kl}(x, \tau)}$$

(4.6)

Where

$$E^{el} = \frac{1}{2} \int_V \left[ C^0_{ijmn} \Delta \delta_{mpq}(c(x)) \cdot C^0_{pqkl} - C^0_{ijkl} \right] \times \left( \varepsilon^0_{ij}(x) - \varepsilon^T_{ij}(c(x)) \right) \cdot \left( \varepsilon^0_{kl}(x) - \varepsilon^T_{kl}(c(x)) \right) dx$$

$$+ \frac{1}{2} \int_V C^0_{ijkl} \varepsilon^0_{ij}(x) \varepsilon^0_{kl}(x) dx - \bar{\varepsilon}_{ij} \int_V C^0_{ijkl} \varepsilon^0_{kl}(x) dx$$

$$+ \frac{V}{2} N_i \delta^0_{ij}(k) \Omega_{jk}(n) \bar{\varepsilon}^0_{kl}(k) n_l$$

(4.7)
$L_{ijkl}$ is the kinetic coefficient tensor (a convenient choice will be $L_{ijkl} = L(C^0_{ijkl})^{-1}$ [105]) and the parameter $\tau$ describes the elastic relaxation process (similar to time in other kinetic equations). In Eq. 4.7, $\Delta S_{ijkl}(c(x)) = \left[ C^0_{ijkl} - C_{ijkl}(c(x)) \right]^{-1}$, $C_{ijkl}$ is the elastic modulus tensor, $\varepsilon^T_{ij}$ is the SFTS tensor, both of them are functions of local concentration $c(x)$ and thus functions of spatial coordinate, $x$. $\bar{\varepsilon}_{ij}$ is an averaged strain given by $\bar{\varepsilon}_{ij} = \frac{1}{V} \int_V \varepsilon_{ij}(x) dx$, $n_i$ is the $i$-th component ($i=1,2,3$) of a unit vector in the reciprocal space, $V$ is the system volume, $\tilde{\sigma}_{ij}^0(k) = C_{ijkl}^0 \tilde{\varepsilon}_{kl}^0(k)$, $\tilde{\varepsilon}_{ij}^0(k)$ is the Fourier transform of $\varepsilon_{ij}^0(x)$, $\Omega_{ij}^{-1}(n) = C_{ijkl}^0 n_k n_l$, and the superscript asterisk indicates the complex conjugate. The last integral term in Eq. 4.7 excludes a volume of $(2\pi)^3/V$ around the point at $k = 0$.

An interpolation function, $\eta(x)$, is used to describe the modulus change across the interface:

$$C_{ijkl}(x) = C^0_{ijkl} + \eta(x) \left( C^S_{ijkl} - C^0_{ijkl} \right) = [1 - \eta(x)] C^0_{ijkl} + \eta(x) C^S_{ijkl} \quad (4.8)$$

where $C^0_{ijkl}$ and $C^S_{ijkl}$ are two reference stiffness tensor. Since in our model the solute concentration is the only field that defines different phases, $\eta(x)$ is formulated as a function of $c(x)$:

$$\eta(x) = \frac{1}{4} \left\{ \tanh \left[ \frac{c(x) - (c_0^\gamma + c_0^\epsilon)/2}{\theta (c_0^\gamma - c_0^\epsilon)} \right] + 1 \right\} + 0.5 \quad (4.9)$$

where $\theta$ is a constant. Note that the reference modulus $C^0_{ijkl}$ is neither the modulus of the matrix phase $C^\gamma_{ijkl}$ nor the modulus of the precipitate phase $C^\epsilon_{ijkl}$. Instead, it is $C^0_{ijkl} = 2C^\epsilon_{ijkl} - C^\gamma_{ijkl}$ when $\theta \ll (c_0^\gamma - c_0^\epsilon)$. Inside the bulk of a precipitate, for example, $c(x) = c_0^\epsilon$ and, according to Eq. 4.9, $\eta(x) = 1$ and the modulus becomes $C_{ijkl}(x) = C^\epsilon_{ijkl} = C^S_{ijkl}$, while inside the bulk of the matrix phase, $c(x) = c_0^\gamma$, $\eta(x) = 0.5$, and the modulus becomes $C_{ijkl}(x) = C^\gamma_{ijkl} = (C^0_{ijkl} + C^S_{ijkl})/2$. By using Eq.
4.9 as the modulus interpolation function, we can avoid singularity when calculating \( \Delta S_{ijkl}(c(x)) = \left[ C^0_{ijkl} - C_{ijkl}(c(x)) \right]^{-1} \) for the total elastic energy in Eq. 4.7 because \( C_{ijkl}(c(x)) \) will never equal to \( C^0_{ijkl} \) in the system.

However, the form of this interpolation function contains one extra degree of freedom, \( \theta \), which alters the sharpness of the modulus transition from one phase to the other. Due to the property of the hyperbolic tangent function, different \( \theta \) values will lead to different \( \eta \) values inside the bulk phases even for the same composition. The dependence of the interpolation function (Eq. 4.9) on both composition \( c \) and \( \theta \) is illustrated in Fig. 4.1. Only when \( \theta \ll (c^c_{\gamma} - c^c_{\gamma'}) \), this modulus interpolation function becomes 0.5 and 1 for the equilibrium bulk concentrations. Based on the same one-dimensional (1D) concentration profile \( c(x) \) that represents a \( \gamma' \) particle in the center with \( \gamma \) channels on both sides (Fig. 4.2(a)), different \( \theta \) value is used to calculate \( \gamma' \) profiles. As shown in Fig. 4.2(b), the bulk value of \( \eta \) is close to 0.500 for the \( \gamma \) phase and close to 1.000 for the \( \gamma' \) phase for \( \theta = 0.1 \). When \( \theta = 0.5 \), the bulk value of \( \eta \) becomes 0.562 for the \( \gamma \) phase and 0.939 for the \( \gamma' \) phase. When \( \theta = 1.0 \), the bulk value of \( \eta \) changes to 0.637 for the \( \gamma \) phase and 0.864 for the \( \gamma' \) phase. In the latter two cases, the bulk value of \( \eta \) significantly deviates from 0.5 and 1. So, the value of \( C^S_{ijkl} \) in Eq. 4.8 no longer equals to the stiffness constants of the precipitate, \( C^\gamma_{ijkl} \). Instead, the parameter \( C^0_{ijkl} \) and \( C^S_{ijkl} \) are determined directly from the modulus for \( \gamma \) and \( \gamma' \) phase (\( C^\gamma_{ijkl} \) and \( C^\gamma_{ijkl} \)). That is, the value of \( \eta \) inside the bulk \( \gamma \) phase is \( \eta^\gamma = \frac{1}{4} \left[ tanh \left( -\frac{1}{2\theta} \right) + 1 \right] + 0.5 \) and the value inside the bulk \( \gamma' \) phase is \( \eta'^\gamma = \frac{1}{4} \left[ tanh \left( \frac{1}{2\theta} \right) + 1 \right] + 0.5 \). Different \( \theta \) values will result in different bulk \( \eta \) values and the two reference stiffness tensors can be determined from \( C^\gamma_{ijkl} \) and \( C^\gamma_{ijkl} \) by
Figure 4.1: The dependence of the interpolation function, $\eta(x)$, on concentration $c$ and parameter $\theta$. The interpolation function is introduced to describe elastic modulus variation across $\gamma/\gamma'$ interfaces. As shown in the plot, the parameter $\theta$ determines the sharpness of the modulus transition from one phase to the other.

the following equations:

$$C_{ijkl}^0 = \frac{\eta_i^\prime \cdot C_{ijkl}^\gamma - \eta_j^\prime \cdot C_{ijkl}^\gamma}{\eta_i^\gamma - \eta_j^\gamma}$$  \hspace{1cm} (4.10)

$$C_{ijkl}^\gamma = C_{ijkl}^\gamma \eta_i^\gamma + (1 - 1/\eta_i^\gamma) \frac{\eta_j^\prime \cdot C_{ijkl}^\gamma - \eta_j^\gamma \cdot C_{ijkl}^\gamma}{\eta_i^\gamma - \eta_j^\gamma}$$  \hspace{1cm} (4.11)

With the interpolation function, the elastic modulus $C_{ijkl}(x)$ equals exactly $C_{ijkl}^\gamma$ and $C_{ijkl}^\gamma$ in the respective bulk phase and varies continuously across the $\gamma/\gamma'$ interface. Tests show that if the modulus profile cross the interface is too sharp ($\theta = 0.1$),
Figure 4.2: Relations between (a) concentration field $c(x)$ and (b) Modulus interpolation $\eta(x)$ with different $\theta$ value along the center of the simulation cell.
it will be difficult for the interface to move due to numerical friction, especially in cases where the interface migration is driven mostly by modulus inhomogeneity. The use of larger $\theta$ value will, however, need larger simulation system to resolve microstructural details. In the following simulations, $\theta = 1.0$ is chosen to achieve a balance between smooth modulus interpolation in the boundary regions and computational efficiency.

The variation of the elastic energy with respect to concentration, 

$$\frac{\delta E_{el}}{\delta c(x)} \Bigg|_{\{c(x),\sigma_{ij}^{app},C_{ijkl}(c(x))\}}$$

in Eq.4.3 is calculated based on the assumption that elastic relaxation happens much faster than diffusion. That is, the time-dependent Ginzburg-Landau equation (Eq. 4.6) is used first to obtain a steady-state solution for the effective strain $\epsilon_{ij}^{0}(x)$ with a fixed concentration field. Then the effective strain is used as a constant to calculate the variation of the elastic energy with respect to concentration $c(x)$, as shown in the following equation

$$\frac{\delta E_{el}}{\delta c(x)} = \delta \left\{ \frac{1}{2} \int_{V} \left[ C_{ijmn}^{0} \Delta S_{mnpq}(c(x)) C_{pqkl}^{0} - C_{ijkl}^{0} \right] \cdot \left( \epsilon_{ij}^{0}(x) - \epsilon_{ij}^{T}(c(x)) \right) \cdot \left( \epsilon_{kl}^{0}(x) - \epsilon_{kl}^{T}(c(x)) \right) \right\}$$

(4.12)

where $\epsilon_{ij}^{T}(c(x))$ is given by Eq. 4.2 and $\Delta S_{mnpq}(c(x))$ is given by the following equation:

$$\Delta S_{ijkl}(c(x)) = \left[ C_{ijkl}^{0} - C_{ijkl}(c(x)) \right]^{-1}$$

$$= \left\{ \eta(x) \left[ C_{ijkl}^{0} - C_{ijkl}^{S} \right] \right\}^{-1} = \frac{4[C_{ijkl}^{0} - C_{ijkl}^{S}]^{-1}}{\tanh \left[ \frac{c(x) - (c_{f} + c_{l})/2}{\theta(c_{f} - c_{l})} \right] + 3}$$

(4.13)

Eq. 4.12 can be divided into two parts using the chain-rule

$$\frac{\delta E_{el}}{\delta c(x)} = \frac{1}{2} \left[ \frac{\partial C_{ijmn}^{0} \Delta S_{mnpq}(c(x)) C_{pqkl}^{0} - C_{ijkl}^{0}}{\partial c(x)} \right] \cdot \left( \epsilon_{ij}^{0}(x) - \epsilon_{ij}^{T}(c(x)) \right) \cdot \left( \epsilon_{kl}^{0}(x) - \epsilon_{kl}^{T}(c(x)) \right)$$

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\[ + \frac{1}{2} \left[ C_{jmn}^0 \Delta S_{mpq} (c(x)) C_{pqkl}^0 - C_{ijkl}^0 \right] \cdot \left[ \frac{\partial}{\partial c(x)} \left( \epsilon_{ij}^0(x) - \epsilon_{ij}^T(c(x)) \right) \right] \cdot \left( \epsilon_{kl}^0(x) - \epsilon_{kl}^T(c(x)) \right) \] (4.14)

The first term can be treated as a contribution from the modulus inhomogeneity while the second term from the transformation strain. If the morphological change is driven mainly by the transformation strain, then the evolution process is not sensitive to the choice of \( \theta \). If the modulus inhomogeneity is the dominant driving force, however, the choice of \( \theta \) must guarantee a smooth transition of the modulus across the interface to avoid numerical pinning.

In the simulations, Eq. 4.14 is substituted into Eq. 4.3 to calculate the driving force for diffusion and to simulate microstructural evolution through the Cahn-Hilliard equation (Eq. 4.4).

### 4.1.3 Scaling scheme in an elastically inhomogeneous system

As mentioned in Section 4.1.1, in order to maintain a cuboidal shape for the precipitates under the condition of enhanced interfacial energy, a scaling method needs to be implemented. For an elastically inhomogeneous system, the elastic relaxation process is obtained by iteratively solving Eq. 4.6, which can be further expanded as [72, 104, 105]:

\[
\frac{\partial \epsilon_{ij}^0(x, \tau)}{\partial \tau} = L_{ijst} \cdot \Delta C_{stk}^0 \left\{ \frac{1}{2} \int \left[ \frac{d\mathbf{k}}{(2\pi)^3} \left( n_k \Omega_{lmn}(\mathbf{n}) + n_k \Omega_{kmn}(\mathbf{n}) \sigma_{mn}^0(\mathbf{k}) n_n e^{\mathbf{k} \cdot \mathbf{x}} \right) \right] \right.
\]

\[
- \Delta S_{kmn}(c(x)) C_{mpq}^0 \left[ \epsilon_{pq}^0(x) - \epsilon_{pq}^T(c(x)) \right] \]

\[
- \epsilon_{kl}^T(c(x)) + \bar{\epsilon}_{kl}^0 + S_{klmn}^0 \sigma_{mn}^\text{ext} \right\} \] (4.15)

The solution of the effective strain \( \epsilon_{ij}^0(x) \) at \( \frac{\partial \epsilon_{ij}^0(x, \tau)}{\partial \tau} \approx 0 \) corresponds to the elastic equilibrium.
If the lattice mist is increased by a factor of $\phi$ (i.e., $\delta' = \phi \cdot \delta$) to maintain a cube-like shape, the stress-free transformation strain needs to be increased by $\phi$ times as well, e.g., $\varepsilon_{ijkl}^T(c(x)) = \phi \varepsilon_{ijkl}^T(c(x))$. If we also increase the external applied stress by $\phi$ times, i.e., $\sigma_{mn}^{ex} = \phi \cdot \sigma_{mn}^{ex}$, then the relaxation equation becomes:

$$
\frac{\partial \varepsilon_{ijkl}^0(x, \tau)}{\partial \tau} = L_{ijkl} \cdot \Delta C_{stkl}^0 \left\{ \frac{1}{2} \int \left[ \frac{d \mathbf{k}}{(2\pi)^3} \left[ n_k \Omega_{lm}(\mathbf{n}) + n_l \Omega_{km}(\mathbf{n}) \right] \tilde{\sigma}_{mn}^0(\mathbf{k}) n_n e^{i \mathbf{k} \cdot \mathbf{x}} \right]
- \Delta S_{klmn}(c(x)) C_{mpq}^0 \left[ \varepsilon_{pq}^0(\mathbf{x}) - \varepsilon_{pq}^T(c(x)) \right]
- \varepsilon_{kl}^T(c(x)) + \tilde{\sigma}_{kl}^0 + \sigma_{mn}^{ex} \right\} \tag{4.16}
$$

which becomes identical to Eq. 4.15 after multiplied by $\phi$ at both sides, e.g.,

$$
\frac{\partial [\phi \cdot \varepsilon_{ijkl}^0(x, \tau)]}{\partial \tau} = L_{ijkl} \cdot \Delta C_{stkl}^0 \left\{ \frac{1}{2} \int \left[ \frac{d \mathbf{k}}{(2\pi)^3} \left[ n_k \Omega_{lm}(\mathbf{n}) + n_l \Omega_{km}(\mathbf{n}) \right] \cdot [\phi \cdot \tilde{\sigma}_{mn}^0(\mathbf{k})] n_n e^{i \mathbf{k} \cdot \mathbf{x}} \right]
- \Delta S_{klmn}(c(x)) C_{mpq}^0 \left[ \phi \cdot \varepsilon_{pq}^0(\mathbf{x}) - \phi \cdot \varepsilon_{pq}^T(c(x)) \right]
- \phi \cdot \varepsilon_{kl}^T(c(x)) + \phi \cdot \tilde{\sigma}_{kl}^0 + \sigma_{mn}^{ex} \phi \cdot \sigma_{mn}^{ex} \right\} \tag{4.17}
$$

Note that $\sigma_{ij}^0(x) = C_{ijkl}^0 \varepsilon_{kl}^0(x) = C_{ijkl}^0 \cdot \phi \cdot \varepsilon_{kl}^0(x) = \phi \cdot \sigma_{ij}^0(x)$ and $\tilde{\sigma}_{ij}^0(\mathbf{k}) = \phi \cdot \tilde{\sigma}_{ij}^0(\mathbf{k})$.

Therefore starting from an identical microstructure (the same concentration field $c(x)$) and iteratively solving both Eq. 4.15 and Eq. 4.16 until they converge (i.e., $\frac{\partial \varepsilon_{ijkl}^0(x, \tau)}{\partial \tau} = 0, \frac{\partial \varepsilon_{ijkl}^0(x, \tau)}{\partial \tau} = 0$), we will obtain $\varepsilon_{ijkl}^0(x, \tau) = \phi \cdot \varepsilon_{ijkl}^0(x, \tau)$. This is an important result showing that for the same microstructure, if both the stress-free transformation strain (or the lattice misfit) and external applied stress are multiplied by a factor, $\phi$, the equivalent strain will be scaled up by the same factor in the whole system and, from Eq. 4.12, the elastic driving force for diffusion $\frac{\delta F_{el}}{\delta e(c(x))}$ will be scaled up by $\phi^2$ times. These relationships allow us to employ a larger lattice mismatch.
to maintain a cuboidal particle shape and then to scale the simulation results back to represent the system having the original lattice misfit by multiplying the chemical potential difference by $\phi^2$ and correspondingly multiplying the rafting rate (or divide the rafting time) by $\phi^2$.

### 4.2 Effective channel plastic deformation strain

Ideally, in order to study the coupling between channel plasticity and modulus inhomogeneity, a model needs to combine dynamics of individual channel dislocations and $\gamma/\gamma'$ microstructure evolution in an elastically inhomogeneous system. However, as the aforementioned elastically inhomogeneous solver uses an iterative method to determine the elastic equilibrium, such a model will involve three different time scales, i.e., the establishment of elastic equilibrium, the migration of channel dislocations and the diffusion of solute atoms. Thus, parametric study for the rafting kinetics is difficult because of the low computational throughput. Alternatively, a simplified method that characterizes channel plasticity via an effective plastic deformation strain can be implemented in order to achieve optimum efficiency and study the interaction between channel plasticity and modulus inhomogeneity at the experimentally relevant length and time scales.

In the phase field dislocation model, the sheared region enclosed by a dislocation loop is described as a plate with a thickness equal to the inter-planer spacing of the glide plan, $d$, and a plane normal, $\mathbf{n}$. The atoms inside the plate is sheared by a Burgers vector $\mathbf{b}$. The eigenstrain of the dislocation loop, or equivalently, the stress free transformation strain of the plate is given by $\varepsilon^T = [\mathbf{n} \otimes \mathbf{b} + \mathbf{b} \otimes \mathbf{n}]/(2d)[23]$. For channel dislocations in blade alloys, they shear the $\gamma$ phase in pairs and deposit
dislocation segments on the $\gamma/\gamma'$ interfaces as shown in Fig. 4.3(a). Although the deformation is actually localized in the atomic planes sheared by the individual dislocation dipoles, the total plastic strain of the channel can be described on average from a macroscopic view as indicated in Fig. 4.3(b). This is equivalent to a configuration shown in Figs. 4.3(c) and (d), which contains the same quantity of dislocation dipoles, but they are redistributed in the system with an equal spacing $D$ between neighboring sheared planes. Thus, by assuming that dislocations are evenly distributed in the $\gamma$ channels, an averaged plastic strain in the channel can be obtained for a given slip system: $\varepsilon^T = [\mathbf{n} \otimes \mathbf{b} + \mathbf{b} \otimes \mathbf{n}] / (2D)$.

Considering all active slip systems in the $\gamma$ channels under a given applied stress, the effective plastic strain becomes

$$
\varepsilon^{\text{dist},T}(\mathbf{x}) = \sum_{i=1..P} \frac{(\mathbf{n}_i \otimes \mathbf{b}_i + \mathbf{b}_i \otimes \mathbf{n}_i)}{2D_i(\mathbf{x})} 
$$

(4.18)

where $P$ is the total number of active slip systems and $D_i(\mathbf{x})$ is the average spacing between neighboring sheared planes of slip system $i$. Thus the plastic deformation in the system can be described through a new set of non-conserved fields, $D_i(\mathbf{x})$ ($i=1..P$), that characterize channel dislocation contents. Their spatial-temporal evolutions are governed by the time-dependent Ginzburg-Landau equations. For simplicity, we assume in this chapter that dislocations from a given slip system are evenly distributed in all channels. In this case $D_i(\mathbf{x})$ becomes a constant $D_i$. As channel dislocations are deposited on $\{001\}$ $\gamma/\gamma'$ interfaces along $\langle110\rangle$ directions, the distance between neighboring dislocation segments on the $\{001\}$ planes is $d_0' = \frac{\sqrt{2}}{2}D_i$ and the transformation strain can be related to $\gamma/\gamma'$ interface dislocation density as $\varepsilon_i^T = \frac{\sqrt{2}}{2d_0'} \frac{(\mathbf{n}_i \otimes \mathbf{b}_i + \mathbf{b}_i \otimes \mathbf{n}_i)}{2}$. Fig 4.4 shows the geometry setting of a dislocation loop wrapping around the $\gamma'$ particle by shearing $\gamma$ channels.
Figure 4.3: Schematics of effective channel plastic deformation strain (the non-uniform shear of materials by dislocations (a) leads to a macroscopic shape change/deformation strain (b). The same macroscopic shape change/deformation strain can also be produced with the uniform shear of the material (c) by an array of dislocations with equal spacing between their glide planes (d).)
Figure 4.4: Schematic of dislocations looping around $\gamma$ precipitate after shearing $\gamma$ channels
When an external stress is applied along the [001] direction, 8 of the 12 slip systems for an F.C.C. crystal will be activated (i.e., $n_1 = 1/\sqrt{3}(111), b_1 = 1/2[\bar{1}01]a_0; n_2 = 1/\sqrt{3}(111), b_2 = 1/2[0\bar{1}1]a_0; n_3 = 1/\sqrt{3}(\bar{1}11), b_3 = 1/2[101]a_0; n_4 = 1/\sqrt{3}(\bar{1}11), b_4 = 1/2[0\bar{1}1]a_0; n_5 = 1/\sqrt{3}(\bar{1}11), b_5 = 1/2[101]a_0; n_6 = 1/\sqrt{3}(\bar{1}11), b_6 = 1/2[011]a_0; n_7 = 1/\sqrt{3}(1\bar{1}1), b_7 = 1/2[011]a_0; n_8 = 1/\sqrt{3}(1\bar{1}1), b_8 = 1/2[\bar{1}01]a_0$; where $a_0 = 0.358nm$ is the lattice constant [7] for a typical Ni-base superalloy).

Table 4.1 shows the corresponding plastic strain with respect to the 8 slip systems, where $|b|$ is the modulus of the Burgers vector.

From the Schmidt factor analysis, the resolved shear stresses are the same for all the 8 activated slip systems. So, it’s reasonable to assume that the average channel dislocation density (or slip plane spacing) is the same, i.e., $d_i^{0=1\ldots8} = d_0$. And the total channel plastic strain can be obtained by summing over contributions from all the 8 slip systems:

$$\varepsilon^{\text{disl}} = \sum_{i=1\ldots8} \frac{\sqrt{6}}{2d_0} \frac{\left| n \otimes b + b \otimes n \right|}{2} = \frac{\sqrt{2}a_0}{d_0} \begin{bmatrix} -1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & 2 \end{bmatrix} \quad (4.19)$$

By analyzing the line directions of the interface dislocations from all 8 slip systems, the average distance between neighboring dislocation segments on \{001\} planes is $d_0/4$ (two perpendicular $\langle110\rangle$ orientation for each \{001\} plane, each hosts dislocations from 4 slip systems, which is demonstrated in Fig. 4.5). Thus this simplified model can take into account channel dislocation density obtained directly from experimental observations in rafting studies using the inhomogeneous modulus solver.
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<tr>
<td>1</td>
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<td>$\epsilon^{T1} = \frac{</td>
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<tr>
<td>2</td>
<td>(111); 1/2[011]</td>
<td>$\epsilon^{T2} = \frac{</td>
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<td>6</td>
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<td>8</td>
<td>(1-11); 1/2[-101]</td>
<td>$\epsilon^{T8} = \frac{</td>
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Table 4.1: Plastic strains for 8 activated F.C.C. slip systems under [001] tension.
4.3 Simulation Results

4.3.1 Input parameters and starting microstructure

The system size is $74l \times 74l \times 74l$, with the grid size, $l$, being 6.66nm. In the initial microstructure, the $\gamma'$ cube edge length is set to be 400nm and the $\gamma$ channel width is 93.3nm. The volume fraction of the $\gamma'$ phase is 53.3%. Periodic boundary condition is applied along all three dimensions to imitate a periodic array of identical $\gamma'$ precipitates aligned along the cubic orientations separated by narrow $\gamma$ channels.

From the theoretical analysis [43], the difference in the quantity $(C_{11} - C_{12})$ between the matrix and precipitate phases in an elastically anisotropic media will influence the direction of rafting due to elastic modulus difference. In Ni-base superalloys, the $\gamma'$ phase has larger $(C_{11} - C_{12})$ value than that of the $\gamma$ matrix at
elevated temperatures [22]. The elastic constants used in this study are chosen from the experimental data for one of the alloys studied in [3], i.e., Alloy R3 under 1273K. This alloy has the largest ratio of \((C_{11} - C_{12})\) between the \(\gamma\) and \(\gamma'\) phases, e.g., \(\frac{(C_{11}' - C_{11})}{(C_{11}' - C_{11})} = 85\%\), which means that \(\gamma'\) precipitates are about 15\% stiffer than the \(\gamma\) matrix (throughout this study, the term “hard” or “soft” is used to refer to a higher or lower value of \((C_{11} - C_{12})\)). The individual elastic constants for the \(\gamma\) phase are: \(C_{11}' = 163\), \(C_{12}' = 112\), and \(C_{44}' = 86\) GPa and for the \(\gamma'\) phase are: \(C_{11}' = 198\), \(C_{12}' = 138\), and \(C_{44}' = 97\) GPa. The lattice misfit, \(\delta = \frac{2(a_{\gamma} - a_{\gamma'})}{(a_{\gamma} + a_{\gamma'})}\), is chosen to be \(-0.3\%\), which is a typical value for blade alloys [40, 2]. All the material and processing parameters used in the simulations are listed in Table 4.2.

In the simulations, the initial \(\gamma'\) cube with sharp edges and corners is relaxed without external load to obtain the equilibrium shape assumed by the interplay between the coherency strain energy and the interfacial energy. The relaxation process is carried out until the maximum chemical potential nonuniformity in the entire system becomes less than 1 J/mol. The final equilibrium shape of the \(\gamma'\) particle obtained with a scaling factor \(\phi = 2.1\) for the lattice misfit (Section 4.1.3) is shown in Fig. 4.6, with continuous \(\gamma\) channels separating neighboring \(\gamma'\) particles. Fig. 4.6(a) is a 3D view, with white frame boxes and black dots in the centers of both types of \(\gamma\) channels indicating the sampling positions and regions for the chemical potential, while Fig. 4.6(b) shows the cross-section cutting through the center of the particle (the cross-section position is also indicated in (a)). On this cross-section, there are two types of \(\gamma\) channels that can be related to each other by the symmetry operation of the crystal. When external stress is applied along the vertical direction, this symmetry is broken and the two channels are referred to as
<table>
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<th>Temperature range</th>
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<td>Pre-rafting $\gamma$ channel width</td>
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</tr>
<tr>
<td>$C_{12}^{\gamma}$</td>
<td>112GPa</td>
</tr>
<tr>
<td>$C_{44}^{\gamma}$</td>
<td>86GPa</td>
</tr>
<tr>
<td>Elastic constants for $\gamma'$ phase</td>
<td></td>
</tr>
<tr>
<td>$C_{11}^{\gamma'}$</td>
<td>198GPa</td>
</tr>
<tr>
<td>$C_{12}^{\gamma'}$</td>
<td>138GPa</td>
</tr>
<tr>
<td>$C_{44}^{\gamma'}$</td>
<td>97GPa</td>
</tr>
<tr>
<td>Ni-Al interdiffusivity</td>
<td>$\bar{D}_{\text{NiAl}} = 1 \times 10^{-16} m^2 s^{-1}$</td>
</tr>
<tr>
<td>Interfacial energy after scaling</td>
<td>$\sigma = 50 mJ/m^2$</td>
</tr>
</tbody>
</table>

Table 4.2: Material and processing parameters used in simulation
Figure 4.6: The equilibrium shape of γ’ particles obtained under −0.3% lattice misfit without applied load. (a) 3D view; (b) (100) cross-section at the center of the precipitate as indicated in (a). Because of the periodic boundary condition used in the simulations, the computational cell shown in (a) represents a periodic array of identical γ’ precipitates aligned along the elastically soft directions (i.e., (100)) separated by narrow γ channels.

horizontal channel and vertical channels, respectively, according to their orientation with respect to the loading direction.

4.3.2 Chemical potential variation during rafting

A tensile stress of 152MPa is applied along the vertical direction of the system shown in Fig. 4.6(a). According to the scaling technique discussed in Section 4.1.3, the applied stress σ^ex is multiplied by a factor of φ = 2.1. Two validation simulation tests are carried out first with identical initial microstructure as the one shown in Fig. 4.6. In the first simulation the original un-scaled values of δ and σ^ex are
Figure 4.7: Contours of $\frac{\delta E_{el}}{\delta c(x)}$ on the central (100) cross-section of the computational cell obtained under lattice misfit $\delta = -0.3\%$ and applied stress $\sigma^{ex} = 152MPa$ (tension along [001] indicated by the arrows). The $\gamma'$ precipitates are 15% stiffer than the $\gamma$ matrix. (a) Test 1 with original $\delta$ and $\sigma^{ex}$. (b) Test 2 with scaled $\delta' = \phi \delta$ and $\sigma'^{ex} = \phi \sigma^{ex}$.

used while in the second simulation $\delta' = \phi \delta$ and $\sigma'^{ex} = \phi \sigma^{ex}$ are used. The driving force from the total elastic energy, i.e., $\frac{\delta E_{el}}{\delta c(x)}$, for these two cases are shown respectively in Fig. 4.7(a) and (b) for the middle cross-section of the system, with the arrows indicating the external load directions. The contours in the two plots verify that the scaling technique used scales up linearly $\frac{\delta E_{el}}{\delta c(x)}$ by a factor of $\phi^2 = 4.4$ everywhere (note the difference in scale in the two plots).

Figure 4.8 shows the iso-contours of chemical potential at the beginning of the simulation immediately after the external load is applied. The chemical potential in the horizontal channel is higher than that in the vertical channel. So diffusion of solute atoms along the chemical potential gradients from the horizontal channel

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Figure 4.8: Chemical potential contours on the central (100) cross-section of the computational cell obtained under −0.3% lattice misfit and 152MPa tension along [001] (indicated by the arrows). The \( \gamma' \) precipitates are 15% stiffer than the \( \gamma \) matrix.

to the vertical channel results in the dissolution of the \( \gamma' \) particle in the horizontal channel and growth in the vertical channel because the \( \gamma \) phase has a higher solute concentration as compared to the matrix phase. The corresponding morphological evolution of the \( \gamma' \) particle as a function of time is shown in Fig. 4.9, from which we can see clearly how the \( \gamma' \) particles transform gradually from the initial cuboids into plates or rafts normal to the applied stress direction. At time \( t = 61.6 \) h, the raft has already formed by joining its images across the periodic boundaries and eliminating all vertical \( \gamma \) channels, while the solute flow continues afterward to smooth out the \( \gamma/\gamma' \) interface.
Figure 4.9: Morphological evolution of γ' precipitates during rafting obtained under lattice misfit $\delta = -0.3\%$ and applied stress $\sigma^{\text{ext}} = 152\, MPa$ (tension along [001] indicated by the arrows in (a)). γ' precipitates are 15% stiffer than the γ matrix. (a) t=20.5 h, (b) t=41 h, (c) t=53.3 h and (d) t=61.6 h.
During the shape evolution, the chemical potential variation in the entire system is continuously monitored. The chemical potential contours on the central cross-section plane indicated in Fig. 4.6(a) is shown in Fig. 4.10, where (a) to (d) correspond to particle shapes at the moments shown in Fig. 4.9 (a) to (d). Note the difference of the scale at different times. In particular, the chemical potential difference between the horizontal and vertical channels is calculated as a function of time by using two methods. In the first method, the chemical potential difference on two numerical grid points, one located at the center of the horizontal channel and the other at the center of the vertical channel as indicated by the black dots in Fig. 4.6 (a), calculated by $\delta F/\delta c(x)$, are recorded, while in the second method the chemical potential difference between two averaged values calculated over 800 numerical grid points (20 by 20 by 2) in the central portion of the horizontal and vertical channels as indicated by the two white frame boxes shown in Fig. 4.6 (a) are recorded. The results are shown in Fig. 4.11. Note that the calculated chemical potential difference is scaled down (i.e., divided) by $\phi^2$ while the time axis is scaled up (i.e., multiplied) by $\phi^2$ according to the scaling scheme discussed in Section 4.1.3. The result obtained from the first method is shown by the solid line while the one obtained from the second method is shown by the dotted line. The two agree well with each other. Although the chemical potential difference fluctuates during the shape evolution, it stays positive, which means that the chemical potential in the horizontal channel is always higher than that in the vertical channel, so there is a continuous flow of solute atoms from the horizontal channels to the vertical channels, which drives the rafting process. The chemical potential difference decreases gradually from about $40 J/mol$ to $15 J/mol$ before the particles start to coalesce. The
coalescence occurs at the time of the spike in Fig. 4.11, due to the development of a transient negative curvature of the $\gamma'$ particle when it joins with its periodic-boundary images (indicated in the (001) cross-section plot of Fig. 4.12). These results show that for an alloy with $-0.3\%$ lattice misfit, precipitates being about $15\%$ stiffer than the matrix, an interdiffusivity of $10^{-16} m^2 s^{-1}$ ($\bar{D}_{\text{NiAl}} = 1 \times 10^{-16} m^2 s^{-1}$ in Ni-Al at $1253K$ [66]), under an applied tensile stress of $152MPa$, an N type rafting (normal to the applied stress direction) fully develops in about 50 hours.

For comparison, the chemical potential difference between the horizontal and vertical channels as a function of time is also calculated for a system with identical $\gamma/\gamma'$ microstructure, lattice misfit and applied stress, but with channel dislocations and under homogeneous modulus assumption (to be more specific, the elastic constants of the $\gamma$ phase are used for the calculation, i.e., $C_{11} = 163$, $C_{12} = 112$ and $C_{44} = 86GPa$), following the method developed in [120]. After initial relaxation under $152MPa$ applied tension, the dislocations are mainly concentrated in the horizontal channels with spacing about $65$ nm on the (001) $\gamma/\gamma'$ interface. The results of chemical potential differences in the horizontal and vertical channels are shown in Fig. 4.13. In the presence of channel dislocations, the two vertical channels are no longer symmetry related because the selectivity of channel dislocation type and filling process [120], the two lines in Fig.4.13 represent the chemical potential differences between the horizontal channel and each of the two vertical channels. The chemical potential difference also spikes before the rafting completes, due to the same reason as mentioned in the previous case.

One of the major differences found in this elastic-plastic model with homogeneous modulus assumption as compared to the elastic model with inhomogeneous
Figure 4.10: Rescaled iso-chemical potential contour on the central (100) cross-section plane lattice misfit $\delta = -0.3\%$ and applied stress $\sigma^{ex} = 152MPa$ (tension along [001]). $\gamma'$ precipitates are 15% stiffer than the $\gamma$ matrix. (a) t=20.5 h, (b) t=41 h, (c) t=53.3 h and (d) t=61.6 h.
Figure 4.11: Chemical potential difference between the horizontal and vertical channels obtained for an inhomogeneous modulus system under 152MPa tension along [001] without considering channel plasticity. The lattice misfit $\delta = -0.3\%$ and the $\gamma'$ precipitates are 15% stiffer than the $\gamma$ matrix.

Figure 4.12: The (001) central cross-section of $\gamma'$ particle at $t=53.3$ h showing high curvatures developed during particle coalescence, which lead to the spike in the chemical potential variation with time as shown in Fig. 4.11
Figure 4.13: Chemical potential difference between the horizontal and vertical channels obtained in a homogeneous modulus system with $-0.3\%$ lattice misfit under 152MPa tension along [001] and with the consideration of channel plasticity.
modulus is that for the latter case the chemical potential difference keeps increasing (from about $20 J/mol$ to $150 J/mol$) during the shape evolution process. The rafting completes in about 9 hours (indicated by the appearance of the spike in Fig. 4.13), that is much shorter than obtained for the elastically inhomogeneous system without channel plasticity. Therefore the plasticity contributes more to the rafting kinetics than does the $\gamma'/'\gamma'$ modulus difference.

Simulation results with different parameters (e.g., the sign of lattice misfit, the direction of applied stress and the sign of the modulus difference between the matrix and the precipitate) have confirmed the predictions from previous pure elastic models with respect to the directionality of rafting in Ni-base superalloys [41, 40, 42, 122, 31, 123, 43, 37, 65, 44]. That is, in general, the direction of rafting depends on the sign of the following products if no channel plasticity is taken into account: 

$$\frac{\sigma\varepsilon}{\delta \left[ (c_{11}' - c_{12}') - (c_{11} - c_{12}) \right]^2}.$$ 

For example: Fig. 4.14 shows the microstructural evolution for the same alloy (same elastic property and lattice misfit), but under 152 MPa compression along the vertical direction. The P-type rafting finishes after about 30 hours. The chemical potential difference between the horizontal and vertical channels is shown in Fig. 4.15 for this compression test. However, by comparing the magnitude of the chemical potential difference and rafting speed, it is clear that when the contribution from channel dislocations is considered, the rafting direction will be determined by the plasticity argument rather than by the pure elasticity argument. This has also been confirmed by simulations that consider simultaneously contributions from both effects, which are described in the following section.
Figure 4.14: Morphological evolution of $\gamma'$ precipitates during rafting obtained under lattice misfit $\delta = -0.3\%$ and applied stress $\sigma^{ex} = 152MPa$ (compression along [001] indicated by the arrows in (a)). $\gamma'$ precipitates are 15% stiffer than the $\gamma$ matrix. (a) t=12.3 h, (b) t=24.6 h, (c) t=36.9 h and (d) t=49.2 h.
Figure 4.15: Chemical potential difference between the horizontal and vertical channels obtained for an inhomogeneous modulus system under 152MPa compression along [001] without considering channel plasticity. The lattice misfit $\delta = -0.3\%$ and the $\gamma'$ precipitates are 15\% stiffer than the $\gamma$ matrix.

4.3.3 Rafting kinetics with combined contributions from channel plasticity and modulus inhomogeneity

Using the simplified model described in Section 4.2, we are able to observe the rafting process under the influence from both channel plasticity and modulus inhomogeneity. From previous phase field dislocation channel filling simulations [120] and experimental results [63, 27, 124, 25], we assume that the distance between neighboring dislocations on \{001\}$\gamma/\gamma'$ interfaces is 65nm under 152MPa applied tension along [001] direction (i.e., $d_0/4 = 65nm$). Thus the effective channel deformation strain is:

$$\varepsilon_{\text{disl,T}} = \begin{bmatrix} -0.195\% & 0 & 0 \\ 0 & -0.195\% & 0 \\ 0 & 0 & 0.389\% \end{bmatrix}$$
If we assume the same lattice misfit, i.e., $\delta = -0.3\%$, the total transformation strain becomes:

$$\varepsilon_\gamma^* - \varepsilon_{\text{disl}}^T = \begin{bmatrix} -0.105\% & 0 & 0 \\ 0 & -0.105\% & 0 \\ 0 & 0 & -0.689\% \end{bmatrix}$$

where the minus sign in front of $\varepsilon_{\text{disl}}^T$ is due to a fact that the lattice misfit strain is calculated using the precipitate phase as the reference while the effective plastic strain is calculated using the matrix phase as the reference. Two tests are performed first, both for system with $-0.3\%$ lattice misfit and under 152MPa tension along [001]. Test #1 uses the elastic constants listed in Table 4.2, with the precipitate phase being elastically “harder” than the matrix phase. In order to investigate the relative contribution from modulus mismatch, the elastic constants of the $\gamma$ and $\gamma'$ phases are switched in Test #2 (i.e. for $\gamma$ phase: $C_{11}^{\gamma} = 198, C_{12}^{\gamma} = 138$ and $C_{44}^{\gamma} = 97$GPa and for $\gamma'$ phase: $C_{11}^{\gamma'} = 163, C_{12}^{\gamma'} = 112$ and $C_{44}^{\gamma'} = 86$GPa), which creates a system with “soft” precipitates. The chemical potential difference between the horizontal and vertical channels as a function of time is plotted in Fig. 4.16 (a) for Test #1 (“hard” precipitate) and Fig. 4.16 (b) for Test #2 (“soft” precipitate). Both tests show that $\gamma'$ precipitates form plates normal to the applied tension direction (i.e., N-type rafting). Using the spike on the chemical potential difference plot to identify the completion of rafting, one obtains 3.4 hours from Test #1 and 3.9 hours from Test #2. The system with “hard” precipitates rafts faster, which is not surprising based on the simulation results presented above or the relative contributions from channel plasticity and from modulus mismatch. Even though the rafting type
is fully determined by contributions from channel plasticity, the difference in rafting completion time caused by modulus inhomogeneity is significant and cannot be ignored.

The first bump/dip on Fig. 4.16 (a) and (b) appear between 1 and 2 hours are due to the coalescence of $\gamma$ particle with its image along the initial cube edge which can be seen from Figs. 4.17 (a) and (b) for microstructures at 1.2 and 2.5 hours.

Further tests with combinations of different sign of misfit (positive misfit) and direction of applied stress (compression) all confirm that under the same [001] applied stress (tension or compression) and lattice misfit (positive or negative), the system with “hard” precipitates always completes the rafting process faster than does a system with “soft” precipitates, but the rafting type (P or N type) is solely determined by the applied stress and lattice misfit, the criterion following the elastic-plastic model.

Parametric study was performed with eight additional simulations with different parameters listed in Table 4.3. In all simulations the applied stress is 152MPa tension and the average dislocation spacing is either 50nm or 100nm on $\{001\}$ $\gamma/\gamma'$ interfaces. As the dislocation spacing depends not only on the applied stress magnitude but also on the microstructure (channel width and particle shape) and elastic property of the system, the choice of different dislocation spacing enables us to study the dependence of rafting kinetics on channel dislocation density. The definition of “hard” and “soft” phases is already specified in the previous simulation ($\gamma$ “harder”: use modulus data for test #1; $\gamma$ “harder”: use modulus data for test #2).
Figure 4.16: Chemical potential difference between the horizontal and vertical channels obtained for an inhomogeneous modulus system under 152MPa tension along [001] with the consideration of channel plasticity. The lattice misfit $\delta = -0.3\%$. (a) $\gamma$ phase is stiffer (rafting completes at 3.4 h as indicated by the spike); (b) $\gamma$ phase is stiffer (rafting completes at 3.9 h as indicated by the spike)
The chemical potential difference between the center of the horizontal channel (normal to the applied stress direction) and the center of the vertical channel (parallel to the applied stress direction) are plotted as a function of time in Fig 4.18. The sampling regions in both channels are indicated in Fig 4.6(a). The peaks or valleys in the plots associated with a transient period where two neighboring particles are coalescing that develops temporarily large negative $\gamma/\gamma'$ interface curvatures provide an easily identifiable feature to determine the time needed for the completion of rafting. The rafting completion time so determined is listed in Table 4.3. Note that if the neighboring particles are in anti-phase relationship which is ignored in this chapter, then coalescence shall not occur and the variation in chemical potential difference would behave differently. Corresponding study will be demonstrated in the next chapter. The results show that the complete rafting time
varies from 1.9 hour to 4.9 hours depending on the input parameters. Same as the analysis of test #1 and #2, a positive value of the chemical potential difference in Fig. 4.18 means that solute atoms have higher chemical potential in the horizontal channels, so the direction of the flux of solute atoms (aluminum) is from horizontal channel to the vertical channel, leading to an N type rafting, while a negative value means that solute atoms have higher chemical potential in the vertical channels, resulting in a P type rafting.

4.4 Discussion

The above simulations have quantified for the first time the variation of driving force for rafting and rafting completion time in a Ni-Al alloy under external load in three different cases: modulus inhomogeneity alone, channel plasticity alone, and the combination of the two. By analyzing the simulation results one can discriminate the relative contributions from each of these factors. For example, under the same conditions (starting microstructure, lattice misfit, interdiffusivity and applied stress), rafting takes about 50 hours to complete in the elastically inhomogeneous case without channel dislocations, about 9 hours in the elastically homogeneous case with channel dislocations and about 3.4 hours in the case of combined effect from both contributions.

It should be noted, however, that the result for the combined effect is obtained by implementing an effective plastic strain from channel dislocations in the elastically inhomogeneous model without actually considering individual dislocations. Even though a general expression of the effective plastic strain is formulated that
Figure 4.18: (a) to (h) Variations of the chemical potential difference between the centers and central regions of the vertical and horizontal channels with time under different test conditions (listed in Table 4.3)
<table>
<thead>
<tr>
<th>Test #</th>
<th>Hard phase</th>
<th>Lattice misfit</th>
<th>dislocation spacing</th>
<th>local inelastic strain for the precipitate phase $\frac{[\epsilon(x) - \epsilon_0]}{[\epsilon_{\gamma} - \epsilon_0]}$</th>
<th>rafting type</th>
<th>time to raft</th>
</tr>
</thead>
<tbody>
<tr>
<td>3</td>
<td>$\gamma$</td>
<td>$-0.3%$</td>
<td>50nm</td>
<td>$\begin{bmatrix} -0.05 &amp; 0 &amp; 0 \ 0 &amp; -0.05 &amp; 0 \ 0 &amp; 0 &amp; -0.8 \end{bmatrix}$</td>
<td>N</td>
<td>3.1h</td>
</tr>
<tr>
<td>4</td>
<td>$\gamma$</td>
<td>$-0.3%$</td>
<td>50nm</td>
<td>$\begin{bmatrix} -0.05 &amp; 0 &amp; 0 \ 0 &amp; -0.05 &amp; 0 \ 0 &amp; 0 &amp; -0.8 \end{bmatrix}$</td>
<td>N</td>
<td>3.5h</td>
</tr>
<tr>
<td>5</td>
<td>$\gamma$</td>
<td>$+0.3%$</td>
<td>50nm</td>
<td>$\begin{bmatrix} 0.55 &amp; 0 &amp; 0 \ 0 &amp; 0.55 &amp; 0 \ 0 &amp; 0 &amp; -0.2 \end{bmatrix}$</td>
<td>P</td>
<td>1.9h</td>
</tr>
<tr>
<td>6</td>
<td>$\gamma$</td>
<td>$+0.3%$</td>
<td>50nm</td>
<td>$\begin{bmatrix} 0.55 &amp; 0 &amp; 0 \ 0 &amp; 0.55 &amp; 0 \ 0 &amp; 0 &amp; -0.2 \end{bmatrix}$</td>
<td>P</td>
<td>2.1h</td>
</tr>
<tr>
<td>7</td>
<td>$\gamma$</td>
<td>$-0.3%$</td>
<td>100nm</td>
<td>$\begin{bmatrix} -0.175 &amp; 0 &amp; 0 \ 0 &amp; -0.175 &amp; 0 \ 0 &amp; 0 &amp; -0.55 \end{bmatrix}$</td>
<td>N</td>
<td>4.3h</td>
</tr>
<tr>
<td>8</td>
<td>$\gamma$</td>
<td>$+0.3%$</td>
<td>100nm</td>
<td>$\begin{bmatrix} -0.175 &amp; 0 &amp; 0 \ 0 &amp; -0.175 &amp; 0 \ 0 &amp; 0 &amp; -0.55 \end{bmatrix}$</td>
<td>N</td>
<td>4.9h</td>
</tr>
<tr>
<td>9</td>
<td>$\gamma$</td>
<td>$+0.3%$</td>
<td>100nm</td>
<td>$\begin{bmatrix} 0.425 &amp; 0 &amp; 0 \ 0 &amp; 0.425 &amp; 0 \ 0 &amp; 0 &amp; -0.05 \end{bmatrix}$</td>
<td>P</td>
<td>2.6h</td>
</tr>
<tr>
<td>10</td>
<td>$\gamma$</td>
<td>$+0.3%$</td>
<td>100nm</td>
<td>$\begin{bmatrix} 0.425 &amp; 0 &amp; 0 \ 0 &amp; 0.425 &amp; 0 \ 0 &amp; 0 &amp; -0.05 \end{bmatrix}$</td>
<td>P</td>
<td>3.0h</td>
</tr>
</tbody>
</table>

Table 4.3: Rafting type and completion time with respect to modulus mismatch; lattice misfit and channel dislocation density

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is able to account for variation in dislocation contents in different types of \( \gamma \) channels, the example presented in this chapter is under an additional assumption that the plastic deformation in all \( \gamma \) channels is the same. It is well known that in reality dislocations have channel preference during multiplication process under the influence of applied stress and coherency stress. For instance, under the typical service condition for commercial single crystal Ni-base superalloys (high temperature, low tensile stress, negative lattice misfit), \( \gamma \) channels normal to the external tension will undergo more plastic deformation as dislocations prefer the horizontal channels [15]. Also, dislocations propagating into the vertical channels are mostly pure screw type which does not interact with the solutes that are centers of dilatation in the system considered. Using the dislocation spacing obtained by experimental observation of \( \gamma/\gamma' \) interface dislocation structure in the horizontal channels to calculate the effective plastic strain is equivalent to an assumption that dislocations fill not only the horizontal channels (which is assisted by the misfit stress), but also the two vertical channels, one of which even repels dislocations by the misfit stress. Therefore, the equal plastic strain approximation over estimates the contribution from channel plasticity and this is the reason why the rafting completion time obtained in this case is shorter than that obtained when individual dislocations and their different distributions in different \( \gamma \) channels are considered. According to the extra half plane analysis [51, 120], for an alloy with negative misfit and under tension the edge components of dislocations in the horizontal channel and one of the vertical channels (if dislocations can enter this vertical channel) both promote the migration of solute atoms from the horizontal to the vertical channel, while the dislocations in the other vertical channel are pure screw type and do not
interact with the solute atoms. Thus it could be estimated that the contribution from channel plasticity to rafting in the effective plastic strain approximation has been exaggerated roughly by a factor of 2. If we simply multiply the rafting time by a factor of 2 to account for the exaggeration, then the rafting completion time for the case with both modulus inhomogeneity and channel plasticity would be 6.8 hours, which seems reasonable as compared to those obtained in the two separate cases. Calculations allowing different dislocation contents in different types of \( \gamma \) channels will be presented in the next chapter.

In addition to the rafting completion time, the variation in the chemical potential difference between the horizontal and vertical channels during rafting is also significantly different in the two separate cases. In the elastically inhomogeneous case without channel dislocations, after external stress is applied, the chemical potential difference drops immediately from about \( 60 J/mol \) to \( 40 J/mol \) after a very short period of time (which is almost indiscernible on Fig. 4.11). This sharp drop corresponds to an initial response by the system to the external stress through diffusion in the local area to re-establish the equilibrium concentration profile across the \( \gamma/\gamma' \) interface and the corresponding elastic relaxation. Then, the chemical potential difference decreases continuously from \( 40 J/mol \) to \( 15 J/mol \) as the horizontal channels widen up and the vertical channels close down. The driving force for rafting in this case is closely related to the shape of the precipitates because not only the misfit stress but also its coupling with the applied stress and modulus mismatch (which creates extra internal stress) are dependent of the precipitate shape [101]. As the aspect ratio of the precipitates increases, the precipitate shape becomes closer to the equilibrium shape and the driving force for rafting decreases.
For the homogeneous modulus case with channel dislocations, however, the typical chemical potential difference increases from $20J/mol$ to $150J/mol$ during the rafting process. The rafting driving force in this case is from solute-dislocation interactions through the dilatational back-stresses created by the channel dislocations. Using the analytical solution for Volterra type dislocations [111], the dilatational stress caused by a straight dislocation at spatial coordination $(x,y)$ (using dislocation line direction as $Z$ axis and slip plane normal as $Y$ axis, with the extra half plane pointing to $+Y$ direction) is

$$\sigma_{dila} = -A \times \left[ \frac{2x^2y^2 + 2y^3}{(x^2 + y^3)} \right],$$

where $A = \frac{(1-\nu)\mu b_{edge}}{2\pi(1-\nu)}$ is a positive constant, $\mu = C_{44} - (2C_{44} + C_{12} - C_{11})/3 = 45.67 GPa$ is the shear modulus of the slip system $\{111\} \langle 110 \rangle$, $b_{edge} = \sqrt{3}b/2 = \sqrt{6}a_0/4 = 0.155nm$ is the edge component of the Burgers vector of a 60 degree dislocation and $\nu = C_{12}/(C_{11} + C_{12}) = 0.407$ is the Poison ratio. The dislocations in the horizontal channel are illustrated in Fig. 4.19 on a $\overline{110}$ cross-section, where $W$ is the channel width measured along the $[001]$ direction, which is 93.3nm at the beginning and widens to about 230nm after the N-type rafting completes. The slip plane of those dislocation dipoles are $\{111\}$ plane and the distance between neighboring dislocations on the $(001)$ $\gamma/\gamma'$ interface is $S=65nm$. As the channel width $W$ gradually increases during $\gamma'$ rafting, the dilatational stress in the center of the channel (indicated in Fig. 4.19 with a solid circle) can be calculated by superimposing contributions from all individual channel dislocations. Fig. 4.20 shows how the total dilatational stress caused by the channel dislocations varies with the increase in channel width. When the channel width increases, the distance between the center of the channel and the channel dislocations also increases because the channel dislocations always stick to the $\gamma/\gamma'$ interface under the applied stress. Intuitively, it seems that as dislocations move...
farther apart, the stress contribution will be reduced, but as shown in Fig. 4.20, the stress contribution from the channel dislocations actually increases when the horizontal channel starts to open up. It reaches a maximum value at $W \sim 125\text{nm}$ before descending gradually. Such a variation can be readily understood if we evaluate the situation for a single dislocation dipole in the channel, as illustrated in Fig. 4.21. When $W$ increases, the solute-stress interaction or the dilatational stress at the fixed observation point (the distance from the point to the center of the dipole is $50\text{nm}$) first increases and then decreases as shown in Fig 4.22.

Another interesting feature is worth of mentioning. If one superimposes Fig. 4.11 and 4.13, it will have a crossover in the first several hours, which means that the elastic inhomogeneity could make a dominant contribution in terms of rafting driving force at the early stages over channel plasticity if one assumes that the two effects are simply additive. So, if the precipitate phase is “harder” than the
Figure 4.20: Dilatational stress at the center of the horizontal channel (shown by the solid circle in Fig 4.19) as a function of channel width.

Figure 4.21: Schematics of a single dislocation dipole in the $\gamma$ channel
matrix phase, then both effects (elastic inhomogeneity and channel plasticity) work together to form directional rafts whose type is determined by the sign of \( \frac{\sigma_{\text{ex}}^{\gamma}}{\delta(C_{11}^{\gamma} - C_{12}^{\gamma})} \).

If the precipitate phase is softer than the matrix phase, however, the two effects will compete against each other. Although the contribution from elastic inhomogeneity may dominate the rafting process at the beginning, the contribution from channel plasticity will quickly take over as the rafting process proceeds and the final rafting direction will still be determined by the sign of \( \frac{\sigma_{\text{ex}}^{\gamma}}{\delta(C_{11}^{\gamma} - C_{12}^{\gamma})} \). Even though positive modulus mismatch (i.e., the \( \gamma' \) phase is harder than the \( \gamma \) phase) is usually observed in experiments [3], one could design new alloys with a negative modulus mismatch at the aging temperature if it is desirable to slow down the rafting process.

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**Figure 4.22:** Variation of the dilatational stress at the observation point (Fig 4.21) as a function of channel width.
The morphological evolution during \( \gamma \) rafting in the two separate cases differs from that observed for the combine case using the effective plastic strain approximation. From Fig. 4.9 and Fig. 4.14, the channels width of the closing channels decreases uniformly until the \( \gamma \) precipitate coalesces with its neighbors (similar to the case presented in [120]), while in the latter case the precipitate first joins with its neighbors laterally at the original cube edge after a short period of time as shown in Fig. 4.17, and then forms a plate normal to the applied stress direction.

### 4.5 Summary

For the first time the relative contributions from modulus mismatch, channel plasticity and the combination of the two to \( \gamma \) rafting or directional coarsening in Ni-Al have been discriminated by computer simulation studies. Quantitative comparisons have been made for times to reach complete rafting, driving force variations and microstructural evolution during rafting among the three cases. It is found that channel plasticity dominants the rafting process. Under 152MPa uniaxial tension along [001] at 1250K, an alloy system having periodical array (aligned along the \{001\} directions) of cubic \( \gamma \) particles of uniform size of 400 nm, with \(-0.3\%\) lattice misfit and interdiffusivity of \( \tilde{D}_{\text{NiAl}} = 1 \times 10^{-16} \text{m}^2\text{s}^{-1} \), the rafting completion time is about 50 and 9 hours, respectively for considering contributions from (a) modulus inhomogeneity only (with \( (C_{11}^\gamma - C_{12}^\gamma)/(C_{11}^{\gamma'} - C_{12}^{\gamma'}) = 85\% \)) and (b) channel plasticity only (with dislocations mainly concentrated in the horizontal channels and having inter-dislocation spacing about 65 nm on the (001) \( \gamma/\gamma' \).
interface). When channel plasticity is considered, the rafting type (P or N) is determined solely by the direction of the applied stress and the sign of the lattice misfit (i.e., the sign of $\frac{\sigma}{\delta(C_{11} - C_{12})}$).

The simulations also show that if the precipitate phase has lower elastic stiffness than that of the matrix, the rafting process takes longer time to complete under the same applied stress, lattice misfit and dislocation density. It is also observed that the P type rafting completes faster than the N type rafting because in the former solute atoms diffuse from two vertical channels to one horizontal channel leading to a rod, while in the latter solute atoms diffuse from one horizontal channel to two vertical channels leading to a plate.

Even though modulus inhomogeneity does not change the rafting type even for systems with much “softer” precipitates ($(C_{11} - C_{12})/(C_{11} - C_{12}) = 85\%$), its contribution to rafting kinetics is significant and cannot be ignored for the level of modulus mismatch considered in this work. The phase field approach discussed in this paper is able to capture these features and facilitates parametric studies of rafting kinetics under different conditions. With the assistance of this model, the interplay between the two factors (elastic inhomogeneity and channel plasticity) can be characterized and utilized to offer opportunities for possible new design strategy to better control the rafting process. Also, the scaling method introduced for phase field simulations in systems with elastically inhomogeneous inclusions under external stress can improve significantly the computational efficiency by increasing the length scale.
CHAPTER 5

LARGE-SCALE THREE-DIMENSIONAL PHASE FIELD SIMULATION OF $\gamma'$-RAFTING AND CREEP DEFORMATION

In previous chapters, the relative contributions from $\gamma$ channel plasticity and from modulus inhomogeneity to $\gamma'$ rafting were discriminated quantitatively using defect level models. Formulated using Khachaturyan’s static concentration wave theory of atomic ordering [125, 87] and phase field microelasticity theories of coherent precipitates and dislocations [83, 84, 87, 75, 69, 126], these models consider simultaneous evolution of individual channel dislocations and $\gamma'/\gamma$ microstructures with multiple antiphase domains in elastically anisotropic and inhomogeneous systems [127, 128, 129]. These studies are, however, limited to a computational cell containing a single $\gamma'$ particle. Under the periodical boundary condition, this represents a perfect periodical array of $\gamma'$ particles. To account for effects of spacial variations in $\gamma'/\gamma$ microstructures on rafting kinetics, precipitate-matrix inversion and overall creep deformation, a new phase field approach at micrometer scale was developed in this chapter (referred to as the “Plastic deformation strain field method” throughout this chapter). A new set of local channel dislocation density fields was introduced to characterize plastic deformation in $\gamma$ channel by dislocations from individual active slip systems. To increase the length scale of
the phase field simulations, the Kim-Kim-Suzuki-Steinbach (KS) model [97, 100] was used to treat $\gamma/\gamma'$ interfaces.

In the following section, the detailed formulation of the new phase fields are presented, i.e., the plastic strain fields were first introduced, followed by phase field description of $\gamma'$ precipitates by concentration and long-range order (lro) parameter fields. These fields, coupled through their contributions to the stress-free transformation strain fields in Khachaturyan’s phase field microelasticity theory [83, 84, 87], describe simultaneously spatial-temporal evolution of both $\gamma/\gamma'$ microstructure and channel dislocations. In section 5.2, the model was first validated against the phase field dislocation model [127] and discrete dislocation model (DDM) [19] for dislocations filling in $\gamma$-channels, followed by simulations of initial $\gamma$-channel filling process by dislocations under external stress and the ensuing rafting process corresponding to different conditions. Rafting kinetics, evolution of the channel dislocation density, the overall creep deformation, and precipitate-matrix inversion are analyzed. The new method is made possible by the findings from the single-particle simulations that channel plasticity dominants the $\gamma'$ rafting type and kinetics over modulus inhomogeneity [128].

5.1 Plastic deformation strain field model formulation

5.1.1 Field description of plastic deformation

In the phase field dislocation model [69], a dislocation loop is described as a sheared platelet with thickness equals to the inter-planer spacing, $d$, of the glide planes, and the plane normal $n$. The region inside the plate is sheared by a Burgers vector $b$. Now let us extend this description to a spatial region that contains a
population of dislocations. The stress free transformation strain field of the region can be written as: 

\[ \varepsilon^p(x) = \sum_{i=1}^P \frac{(\mathbf{n}_i \otimes \mathbf{b}_i + \mathbf{b}_i \otimes \mathbf{n}_i)}{2D_i(x)}, \]

where \( P \) is the total number of active slip systems and \( D_i(x) \) is the average spacing between neighboring sheared planes of slip system \( i \). \( \mathbf{n}_i \) is the unit vector of the slip plane normal and \( \mathbf{b}_i \) is the Burgers vector for slip system \( i \).

Thus, we define our new phase fields as:

\[ \eta_i(x) = \frac{\mathbf{b}_i}{D_i(x)} \]  

which carries the physical meaning of the value of local plastic deformation strain produced by dislocations from a specific slip system, and the strain tensor can be written as:

\[ \varepsilon_{kl}^p(x) = \sum_{i=1}^P \frac{(\mathbf{n}_i \otimes \mathbf{b}_i + \mathbf{b}_i \otimes \mathbf{n}_i)}{2|\mathbf{b}_i|} \eta_i(x) = \sum_{i=1}^P \varepsilon_{kl}^p(i) \eta_i(x) \]  

The concept of the plastic strain field \( \eta_i(x) \) can be linked back to the shear of the \( \gamma \) channel by dislocations as indicated in Fig. 5.1. The solid lines represent the crystal plane already sheared by dislocations and the amount of plastic deformation are represented by different shades of gray. The darker of the shades, the larger of the dislocation density.

During early stages of the creep deformation in single crystal superalloys, dislocations are mostly confined in the \( \gamma \) matrix [28]. So for the current study, the deformation strain field is confined in the \( \gamma \) phase. As the \( \gamma \) matrix is an F.C.C. solid solution containing twelve \( 1/2\langle 110 \rangle \{ 111 \} \) slip systems, a phase field variable \( \eta_i(x) \) \((i = 1, 2, \ldots 12)\) is assigned to describe the extent of plastic deformation or the plastic strain caused by dislocations from each of the 12 slip systems. During deformation, dislocations will be deposited on the \( \gamma/\gamma' \) interfaces driven by misfit...
stress and applied stress. By considering the geometry of the $\gamma/\gamma'$ configuration shown in Fig 5.2, the distance between neighboring dislocations (from the same slip system) deposited on the $\gamma/\gamma'$ interface is

$$d_i^0 = \frac{\sqrt{6}}{2} D_i(x) = \frac{\sqrt{6}}{2} \frac{|b_i|}{\eta_i(x)} \quad (5.3)$$

For F.C.C, $D_i(x)$ is the distance between the neighboring $\{111\}$ glide planes that have been sheared by dislocations from $i$-th slip system. The schematic drawing of the plastic deformation in a $\gamma$ channel is illustrated in Fig. 5.2.

For single crystal Ni-Al, when considering an external stress applied along one of the cubic directions (an stress state experienced by single crystal turbine blades that work under centrifugal force), 8 of the 12 slip systems will have non-zero Schmidt factors and be activated. For a system under [001] tension, the activated slip systems are listed in Table. 5.1 together with their corresponding $\varepsilon^p(i)$. If [001]
Figure 5.2: The geometry of the $\gamma$ channel dislocations in Ni-Al single crystal superalloys

Compression is applied, a minus sign has to be added in front of the Burgers vector, because the slip direction (the relative displacement of the upper part of the crystal defined by the slip plane normal $\mathbf{n}$ with respect to the lower part of the crystal) is reversed for the same slip system. $\eta(x)$ always assumes non-negative values to guarantee the correct shear direction corresponds to the applied stress.

From Fig. 5.1, it is noteworthy that the value of $\eta(x)$ not only changes on the slip plane when crossing over a dislocation, but also varies normal to the sheared plane. Similar to the gradient energy formulation used in the phase field model of individual dislocations, the gradient of $\eta(x)$ has to be confined on the slip plane because across the slip plane the crystal is perfect and there should be no excess
<table>
<thead>
<tr>
<th>i=</th>
<th>Slip system</th>
<th>$\varepsilon^p(i)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>$n = \frac{1}{\sqrt{3}}(1\bar{1}1); b = \frac{1}{\sqrt{2}}[011]</td>
<td>b</td>
</tr>
<tr>
<td>2</td>
<td>$n = \frac{1}{\sqrt{3}}(\bar{1}11); b = \frac{1}{\sqrt{3}}[101]</td>
<td>b</td>
</tr>
<tr>
<td>3</td>
<td>$n = \frac{1}{\sqrt{3}}(\bar{1}11); b = \frac{1}{\sqrt{2}}[0\bar{1}1]</td>
<td>b</td>
</tr>
<tr>
<td>4</td>
<td>$n = \frac{1}{\sqrt{3}}(111); b = \frac{1}{\sqrt{2}}[\bar{1}01]</td>
<td>b</td>
</tr>
<tr>
<td>5</td>
<td>$n = \frac{1}{\sqrt{3}}(111); b = \frac{1}{\sqrt{2}}[0\bar{1}1]</td>
<td>b</td>
</tr>
<tr>
<td>6</td>
<td>$n = \frac{1}{\sqrt{3}}(\bar{1}11); b = \frac{1}{\sqrt{2}}[101]</td>
<td>b</td>
</tr>
<tr>
<td>7</td>
<td>$n = \frac{1}{\sqrt{3}}(\bar{1}11); b = \frac{1}{\sqrt{2}}[011]</td>
<td>b</td>
</tr>
<tr>
<td>8</td>
<td>$n = \frac{1}{\sqrt{3}}(1\bar{1}1); b = \frac{1}{\sqrt{2}}[\bar{1}01]</td>
<td>b</td>
</tr>
</tbody>
</table>

Table 5.1: Activated slip systems in $\gamma$ channels under [001] tension and their corresponding $\varepsilon^p(i)$. 
energy. Following [69, 70], we have.

\[ E_{\text{grad}} = \frac{\beta}{2} \sum_{i=1}^{8} [n_i \times \nabla \eta_i(x)] \cdot [n_i \times \nabla \eta_i(x)] dV \]  

(5.4)

where \( \beta \) is the gradient energy coefficient. Thus, the gradient energy of the deformation strain fields, \( E_{\text{grad}} \), can be solely attributed to the core energy of the dislocations. As the dislocations are confined in the \( \gamma \) channels, reactions among dislocations from the 8 active slip systems are energetically unfavorable and thus not considered in Eq. 5.4. The coefficient \( \beta \) is set to be \( 7.8 \times 10^{-4} J \cdot m^{-1} \) to guarantee a smooth transition of the deformation strain field profile across the sheared-unsheared boundaries on glide planes.

5.1.2 Phase field description of \( \gamma/\gamma' \) microstructure with multiply anti-phase domains for \( \gamma' \)

The \( L1_2 \) ordered structure of \( \gamma' \) precipitates has been described by using Khachaturyan’s static concentration wave theory [125] in phase field models where three long-range order parameters were introduced to characterize the four symmetry related antiphase domains. In order to increase the length scale of the phase field simulations in this study, however, we used four phenomenological long-range order parameters to describe the four types of antiphase domains [73], i.e., \( \phi_q(x) = 0 \) within the \( \gamma \) matrix and \( \phi_q(x) = 1 \) inside the \( q \)-th ordered \( \gamma' \) domain. The equilibrium composition of \( \gamma \) and \( \gamma' \) phases are fitted to Ansara’s free energy model [109] under 1300K, i.e. \( c^c_\gamma = 0.160 \text{at}\% \text{Al} \) and \( c^c_\gamma = 0.229 \text{at}\% \text{Al} \). The Kim-Kim-Suzuki-Steinbach (KS) model [97, 100, 118] was implemented and the simulation grid size is chosen as \( l_0 = 20 \text{nm} \). The chemical free energy of the system is described by two parabolic functions corresponding to the \( \gamma \) and \( \gamma' \) phases respectively (Fig. 144).
Figure 5.3: Bulk chemical free energy density of $\gamma$ and $\gamma'$ phases with respect to composition

\begin{align*}
  f'(c_{\gamma}(x)) &= f_0 V_m^{-1} [c_{\gamma}(x) - c_{\gamma}]^2 \\
  f'(c_{\gamma'}(x)) &= f_0 V_m^{-1} [c_{\gamma'}(x) - c_{\gamma'}]^2
\end{align*}

(5.5)

where $f_0 = 3.2175 \times 10^4 \text{J/mol}$, $V_m = 1 \times 10^{-5} \text{m}^3/\text{mol}$ is the molar volume (assumed constant). $c_{\gamma}(x)$ and $c_{\gamma'}(x)$ are the composition fields of $\gamma$ and $\gamma'$ phases required by the KS model, and the local composition is calculated as:

$$c(x) = [1 - h(\phi_q(x))] c_{\gamma}(x) + h(\phi_q(x)) c_{\gamma'}(x)$$

(5.6)

where $h(\phi_q(x))$ is an interpolation function [118]:

$$h(\phi_q(x)) = \sum_{q=1}^{4} \left[ \phi_q^3(x) \left( 6\phi_q^2(x) - 15\phi_q(x) + 10 \right) \right]$$

(5.7)

This function is used to distinguish the disordered and ordered phases, i.e., $h(\phi_q(x)) = 0$ corresponds to the $\gamma$ phase and $h(\phi_q(x)) = 1$ corresponds to the $\gamma'$ phase.

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The local equilibrium between the two phases requires: \( \frac{df(c_\gamma(x))}{dc_\gamma(x)} = \frac{df'(c_\gamma'(x))}{dc_\gamma'(x)} \) which leads to \( c_\gamma(x) = c_\gamma(x) + (c_\gamma' - c_\gamma) \). The local chemical free energy is given by:

\[
f(c(x), \phi_\gamma(x)) = [1 - h(\phi_\gamma(x))]f'(c_\gamma(x)) + h(\phi_\gamma(x))f''(c_\gamma'(x)) + \omega \cdot g(\phi_\gamma(x))
\] (5.8)

where \( \omega \) is a parameter related to the \( \gamma/\gamma' \) interfacial energy and interface width, which will be determined later together with the gradient energy coefficient, and:

\[
g(\phi_i(x)) = \sum_{i=1}^{4} \phi_i^2(x)(1 - \phi_i(x))^2 + \theta \sum_{i=1}^{4} \sum_{j \neq i} \phi_i^2(x) \cdot \phi_j^2(x)
\] (5.9)

The parameter \( \theta \) is related to the anti-phase domain boundary (APB) energy. By choosing \( \theta = 10 \), it guarantees that the APB energy is higher than twice of the \( \gamma/\gamma' \) interfacial energy, which prevents the coalescence of different \( \gamma' \) domains during the precipitation process after solution treatment.

5.1.3 Elastic energy formulation

According to Khachaturyan’s phase field microelasticity theory [83, 84, 87], the total elastic energy of a system of various types of mutually interacting stress-carrying defects under the stress-controlled boundary condition (system is subjected to uniform external stress: \( \sigma_{ij}^e \)) is given by:

\[
E^{el} = \frac{1}{2} \sum_{p,q} \int \frac{d^3k}{(2\pi)^3} B_{pq}(n)\theta_P(k)\theta^*_q(k) - \sigma_{ij}^{ex} \sum_p \varepsilon^{ex}_{ij}(p)\hat{\theta}_P(x)dV
\] (5.10)

where \( p, q \) stands for different types of defects, \( V \) is the volume of the system, \( n \) is a unit vector in k-space, \( \hat{\theta}_P(x) \) is the shape function describing the spatial distribution of the \( p \)-th type defects in real space while \( \theta_P(k) \) is its Fourier transform. \( B_{pq}(n) \) is defined as:

\[
B_{pq}(n) = \begin{cases} n = 0 & B_{pq}(n) = 0, \\ n \neq 0 & B_{pq}(n) = \left[ C_{ijkl}\varepsilon^{ex}_{ij}(p)\varepsilon^{ex}_{kl}(q) - \left(n_i\sigma^{ex}_{ij}(p)\Omega_{jk}(n)\sigma^{ex}_{kl}(q)n_j\right)\right]. \end{cases}
\] (5.11)
where $C_{ijkl}$ is the stiffness tensor, $\varepsilon_{ij}^*(p)$ is the stress free transformation strain of the $p$-th type defect, $\sigma_{ij}^*(p) = C_{ijkl} \varepsilon_{kl}^*(p)$ and $\Omega_{ij}^{-1}(n) = C_{klhj} \eta_k n_l$.

For the rafting simulation, we will introduce two types of stress-carrying defects: $\gamma$ precipitates and dislocations. The related stress free transformation strain due to lattice misfit between the $\gamma$ and $\gamma'$ phases is defined as a linear function of $h(\phi_q(x))$:

$$
\varepsilon_{ij}^0 = \frac{2(a_{\gamma} - a_{\gamma'})}{(a_{\gamma} + a_{\gamma'})} \delta_{ij} h(\phi_q(x))
$$

(5.12)

Where $a_{\gamma'}$ and $a_{\gamma}$ are the equilibrium lattice parameters of the $\gamma'$ and $\gamma$ phases, respectively. For dislocations, Their eigenstrains or transformation strains are already formulated in Eq. 5.2. The total elastic energy of the current system (under external stress $\sigma_{ij}^p$) is then expressed as:

$$
E^{el} = \frac{1}{2} \int \frac{d^3 k}{(2\pi)^3} \left[ (B_{hh}(n) - \langle B_{hh}(n) \rangle) \tilde{h}(k) \tilde{h}(k)^* + \sum_{u=1,..,p} (B_{hh_u}(n) - \langle B_{hh_u}(n) \rangle) \tilde{h}(k) \tilde{\eta}_u(k)^* 
\right. \\
+ \sum_{u,v=1,..,p} B_{uu_v}(n) \tilde{\eta}_u(k) \tilde{\eta}_v(k)^* 
\left. - \int_V \sigma_{ij}^p \sum_{u=1,..,p} \varepsilon_{ij}^p(u) \eta_u(x) dV \right]
$$

(5.13)

Where $\tilde{h}(k)$ is the Fourier transform of the function $h(\phi_q(x))$ (shape function for $\gamma'/\gamma'$ microstructure) and $\tilde{\eta}_u(k)$ is the Fourier transform of $\eta_u(x)$ (shape functions of the sheared regions). $B_{hh}(n) = C_{ijkl} \varepsilon_{ij}^0 \varepsilon_{kl}^0 - n_l \sigma_{ij}^0 \Omega_{jk}(n) \sigma_{kl}^0 n_i$; $B_{hh_u}(n) = C_{ijkl} \varepsilon_{ij}^0 \varepsilon_{kl}^p(u) - n_l \sigma_{ij}^0 \Omega_{jk}(n) \sigma_{kl}^p(u) n_i$. And $\langle B_{hh}(n) \rangle = \frac{1}{4\pi} \oint B_{hh}(n) dO_n$ ($dO_n$ is the solid angle element normal to the unit vector $n$); $\langle B_{hh_u}(n) \rangle = \frac{1}{4\pi} \oint B_{hh_u}(n) dO_n$. $B_{uu_v}(n) = C_{ijkl} \varepsilon_{ij}^p(u) \varepsilon_{kl}^p(v) - n_l \sigma_{ij}^p(u) \Omega_{jk}(n) \sigma_{kl}^p(v) n_i$, the integrant $\int$ excludes the point $k = 0$, which corresponds to the afore-mentioned stress-controlled boundary condition. As the equilibrium compositions of the $\gamma$ and $\gamma'$ phases are fitted to a coherent $\gamma'/\gamma'$ phase diagram, part of the elastic energy depending on the volume of the $\gamma'$ precipitates is included in the bulk chemical free energy [126]. In order to maintain the volume fraction
of γ phase given by the coherent phase diagram, only the configuration dependent part elastic energy contributed from lattice misfit is considered. That is why the averaged value of \( B_{hh}(n) \) and \( B_{n\eta}(n) \) are deducted from the elastic energy calculation [126]. From Eq. 5.2, \( \varepsilon_{ij}^p(u) \) are the plastic deformation strain related to the \( u \)-th slip system, and their corresponding values are listed in Table 5.1. Also, \( \sigma_{ij}^p(u) = C_{ijkl} \varepsilon_{kl}^p(u) \) and \( \sigma_{ij}^0 = C_{ijkl} \varepsilon_{kl}^0 \). The term: \( \int_V \sigma_{ij}^p \sum_{u=1}^p \varepsilon_{ij}^p(u) \eta_u(x) \) represents the work done by the external stress through plastic deformation. The macroscopic plastic strain can be obtained from this model as:

\[
\bar{\varepsilon}_{ij}^p = \frac{1}{V} \int \sum_{u=1}^p \varepsilon_{ij}^p(u) \eta_u(x) dV
\]  

(5.14)

As rafting is essentially a stress-induced shape coarsening process driven by the inhomogeneous distribution of stress, the microstructural evolution is a result of the competition between the elastic energy and the interfacial energy. Since γ/γ interfacial energy scales with area while elastic energy scales with volume, the interplay is a function of particle size. To represent correctly such an interplay, the KS model [97, 100] was implemented to carry out simulations at the experimentally relevant length scales.

5.1.4 Kinetics equations

The kinetic equations that govern the time evolution of concentration, order parameters and plastic strain fields read:

\[
\frac{\partial c(x)}{\partial t} = \nabla \cdot \left[ M_c \nabla \left( \frac{\delta F}{\delta c(x)} \right) \right]
\]  

(5.15)

\[
\frac{\partial \phi_q(x)}{\partial t} = -L_\phi \frac{\delta F}{\delta \phi_q(x)}
\]  

(5.16)

\[
\frac{\partial \eta_i(x)}{\partial t} = -L_\eta \frac{\delta F}{\delta \eta_i(x)}
\]  

(5.17)
where $F$ is the total free energy in the system, which is described as:

$$
F = \int_V dV \left[ f(c(x), \phi_q(x)) + \frac{\kappa_\phi}{2} \sum_{q=1}^{4} (\nabla \phi_q(x))^2 \right] + E^{\text{grad}} + E^{\text{el}}
$$

(5.18)

where $f(c(x), \phi_q(x))$ is the local chemical free energy defined in Eq. (5.8). $\kappa_\phi$ is the gradient energy coefficient of the long-range order parameters. By choosing $\kappa_\phi = 9.36 \times 10^{-15} J \cdot m^2 \cdot mol^{-1}$ and $\omega = 3.8996 \times 10^6 J \cdot m^{-3}$ in Eq. (5.8), the $\gamma'/\gamma$ interfacial energy becomes $\sigma = 0.01424 J \cdot m^{-2}$, which matches the experimental measured value [130]. $E^{\text{grad}}$ is defined in Eq. (5.4) and $E^{\text{el}}$ is given in Eq. (5.13).

The chemical mobility $M_c$ is assumed to be a constant: $M_c = 1.544 \times 10^{-16} J^{-1} mol^2 m^{-1} s^{-1}$, which corresponds to an interdiffusivity, $D_{NiAl} = 1.0 \times 10^{-16} m^2 s^{-1}$ in Ni-Al at 1253K [66], $L_\phi$ is the interface mobility and $L_\eta$ is the mobility of the plastic deformation field which reflects the dislocation mobility. $L_\phi = 5.79 \times 10^{-9} m^2 N^{-1} s^{-1}$ is chosen to guarantee diffusion controlled microstructure evolution. $L_\eta = 2.46 \times 10^{-6} m^2 N^{-1} s^{-1}$ is chosen to ensure that the plastic deformation of the $\gamma$-channel (i.e. dislocation glide) is much faster than the diffusion process.

**5.2 Validation of plastic deformation strain field method**

**5.2.1 Validation against Discrete Dislocation Model (DDM)**

The dislocation density (critical number of dislocation loops) in one of the $\gamma$-channels as a function of lattice misfit, $\gamma'$ volume fraction and external stress has been explored [19] using a discrete dislocation model (DDM). To validate the new phase field description of plastic deformation in $\gamma$-channels formulated above, simulations were carried out using exactly the same input parameters as those used in
the DDM model [19]. A $1\mu m \times 1\mu m \times 1\mu m$ computational cell with periodic boundary condition was created (Fig. 5.4 (a) shows the configurations of $\gamma/\gamma'$ microstructure and channel dislocations on the (10$\overline{1}$) cross-section of two unit cells). The lattice misfit is $-0.3\%$, and the $\gamma'$ volume fraction is $50\%$. Only one slip system was activated, i.e. $1/2[01\overline{1}](111)$, with $85MPa$ shear stress resolving on this slip system. Isotropic elasticity was used in this calculation with shear modulus $\mu = 74.8GPa$ (poison ratio: $\nu = 0.276$). A cube-shaped $\gamma'$ particle was placed at the center of the computational cell without relaxation. The equilibrium state of the plastic deformation strain from this slip system after dislocation channel filling is plotted along the diagonal of the $\gamma'$ cube ((10$\overline{1}$) cross-section containing two unit cells), as shown in Fig 5.4(b). The distance between neighboring dislocations in the $\gamma$-channel along the $\gamma/\gamma'$ interface can be calculated from the deformation strain similar to what has been explained earlier (Fig. 5.2). At the center of the (010) $\gamma$-channel, the distance between neighboring dislocations predicted by the phase field model is $d_0 = 22nm$, which agrees well with the one predicted by DDM [19], i.e., $d_0 = 23nm$.

5.2.2 Validation against phase field dislocation model

From previous calculations using the phase field description of individual dislocations ($\gamma'$ volume fraction is $53.3\%$ and $\gamma'$ particle edge length is about $400nm$) [127], when using two of the eight activated slip systems to simulate the dislocation channel filling process, i.e. $1/2[101](\overline{1}11)$ and $1/2[011](\overline{1}11)$, with equal distance between neighboring sheared planes along slip plane normal direction, the result showed that for spacing smaller than $65nm$, some dislocation dipoles are unable to
penetrate through the $\gamma$ channel favored by the loading condition and lattice misfit (for 152MPa tension along [001] and $-0.3\%$ lattice misfit, the favored channel was the horizontal channel). However, further increasing the dislocation density resulted with partially stabilized channel dislocation structure, that is, dislocations on some planes were not able to completely go through the channel leaving dipoles on the $\gamma/\gamma'$ interface (i.e., wrapping around $\gamma'$ particles), instead, they formed stable elongated dislocation loops in the channel, which is demonstrated in Fig. 5.5 (a) (the spacing between neighboring sheared plane equals 32nm, note that the simulation unit cell was rotated to facilitate the numerical calculation). The cross-section in the center of the horizontal channel is demonstrated in Fig. 5.5 (b) and (c) for the two slip systems respectively. The lines are the intersection of the dislocation sheared region (displacement field) and the cross-section plane. From the figure, it can be noticed that part of the displacement field was not able to penetrate
through the horizontal channel. The averaged dislocation density in the horizontal channel (considering both of the two slip systems used in the simulation) can be backed out for this partial channel filling configuration. In this case the averaged spacing of channel dislocations is 42nm (in the horizontal γ channel).

Using the plastic deformation strain fields approach introduced in the previous section, we were able to perform a calculation with similar model setting: a 25 × 25 × 25 3-D computational cell with periodic boundary condition (length scale \( l_0 = 20\text{nm} \)). The elastic stiffness constants are: \( C_{11} = 231\text{GPa}, C_{12} = 149\text{GPa}, C_{44} = 117\text{GPa} \) [113], which will be used throughout this chapter unless mentioned otherwise. The microstructure was pre-relaxed under the influence of misfit stress without loading. The volume fraction of the γ' phase is 51.2% and the edge length of the γ' particle is 400nm. Two plastic deformation strain fields were implemented corresponding to the two slip systems: \( 1/2[101](\bar{1}\bar{1}1) \) and \( 1/2[011](\bar{1}\bar{1}1) \). The equilibrium distribution of plastic deformation from the two slip systems under 152MPa tension along [001] is listed on table 5.2 with cross-sections going through the center of the γ' particle along three cubic directions. Compare to the result from individual dislocations simulation (Fig. 5.5 (a)), the channel preference and the dislocation (plastic deformation) distribution in the γ channels agreed well with each other. The calculated average dislocation spacing in the center of the horizontal channel is 36nm, slightly higher than that predicted from the individual dislocation model.
Figure 5.5: Dislocation channel filling simulated using phase field dislocation model (dislocation line is the boundary between sheared and un-sheared region). (a) Displacement fields and microstructure. (b) The cross-section of displacement field in the center of a horizontal channel corresponds to slip system: $1/2[101](-1-11)$. (c) The cross-section of displacement field in the center of a horizontal channel corresponds to slip system: $1/2[011](-111)$
Table 5.2: The distribution of plastic deformation from the two slip systems under 152MPa tension along [001], illustrated by cross-sections going through the center of the γ' particle along three cubic directions.

<table>
<thead>
<tr>
<th>Lattice misfit</th>
<th>(100) plane</th>
<th>(010) plane</th>
<th>(001) plane</th>
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<tbody>
<tr>
<td>−0.3%</td>
<td>[001]</td>
<td>[001]</td>
<td>[001]</td>
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<tr>
<td>152MPa [001]tension</td>
<td>[010]</td>
<td>[100]</td>
<td>[010]</td>
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<tr>
<td>Microstructure cross-section through the center of the γ' particle</td>
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<td><img src="image3.png" alt="Image" /></td>
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<td><img src="image8.png" alt="Image" /></td>
<td><img src="image9.png" alt="Image" /></td>
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<tr>
<td>n = ( \frac{1}{\sqrt{3}}(\bar{1}\bar{1}1) ), b = ( \frac{1}{\sqrt{2}}[101]</td>
<td></td>
<td>b</td>
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</tr>
<tr>
<td>n = ( \frac{1}{\sqrt{3}}(\bar{1}\bar{1}1) ), b = ( \frac{1}{\sqrt{2}}[011]</td>
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<tr>
<td>legend</td>
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<td><img src="image17.png" alt="Image" /></td>
<td><img src="image18.png" alt="Image" /></td>
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</table>
5.3 Simulation results using plastic deformation strain field method

5.3.1 Precipitation and growth after solution treatment

A $128 \times 128 \times 128$ computational cell was used with a grid size $l_0 = 20nm$, which represent a system of $(2.56 \mu m)^3$. The homogeneous modulus assumption was used because channel plasticity has been shown to dominate the rafting process in our previous studies [127, 128, 129]. As for most Ni-base single crystal superalloys, the elastic moduli of the $\gamma'$ precipitates are slightly higher than that of the $\gamma$ matrix under service temperatures [3], e.g., $(C_{11} - C_{12})_{\gamma'} - (C_{11} - C_{12})_{\gamma} = 0 \sim 15GPa$, it is expected that the actual rafting kinetics will be slightly faster than that predicted by using the homogeneous modulus assumption [128].

The simulations were started with a homogeneous $\gamma$ phase matrix having a composition of 0.195 at%Al, which is supersaturated at 1300K. Random fluctuation of composition and long-range order parameters [131] were introduced through the Langevin random force terms added in Eq. 5.15 and Eq. 5.16 to facilitate the nucleation of $\gamma'$ at 1300K. Figure 5.6 shows the microstructure obtained after 4.7 hours aging at 1300K. The volume fraction of $\gamma'$ phase is about 60%. The size of most $\gamma'$ particles ranges from 160 to 360nm.

5.3.2 Plastic deformation in $\gamma$-channels (corresponding to the initial dislocation channel filling process)

After external stress was applied along [001] direction, plastic deformation was produced in the $\gamma$-channels as the plastic strain fields $\eta_i(x)$ responded spontaneously to the local stress state change in the $\gamma$ channels. Since the plastic strain fields relax much faster than the evolution of the $\gamma/\gamma'$ microstructure, they soon reached the equilibrium state corresponding to the starting $\gamma/\gamma'$ microstructure.
Figure 5.6: Microstructure after 4.7 hours aging at 1300K (lattice misfit: $|\delta| = 0.3\%$; without external load)
The spatial distribution of these plastic strain fields in the \(\gamma\)-channels obtained for a negative \((-0.3\%)\) and positive \((0.3\%)\) lattice misfit under 152MPa tension are shown in Table 5.3 and Table 5.4, respectively.

When dislocations fill in the \(\gamma\)-channels under an applied stress, they lay down segments at the \(\gamma/\gamma'\) interfaces (assuming no shearing of \(\gamma'\)). Depending on the operating slip systems, some dislocation segments at the interfaces are pure screw type, while others are so called “60 degree” dislocations whose Burgers vector and sense vector form a 60 degree angle. According to the theoretical analyses [51, 127], it is expected that for an alloy with a negative lattice misfit under tension, the interaction between the misfit and applied stress fields will create a larger effective stress for dislocations in the \(\gamma\)-channels that are normal to the tensile axis (i.e., (001) channels or the horizontal channels), while the misfit stress in one of the vertical channels repels the dislocations. On the contrary, for a positive lattice misfit under tension, the dislocations will prefer one of the vertical channels that host 60 degree dislocations and avoid the horizontal channels. From the results shown in Table 5.3 (negative misfit under tension) and the 3D plot shown in Fig. 5.7, one can readily see that the simulation predictions agree well with these theoretical analyses.

For positive misfit under tension (Table 5.4), the plastic strain fields tend to concentrate in one of the vertical channels. As shown in Table 5.4 for the \(1/2[011]\) \((1\bar{1}1)\) slip system, the deformation strain is mostly confined in the (010) channels (channels that are perpendicular to the [010] direction). This can be noticed from both the face-on view in the (010) cross-section and the edge-on views in the (100) and (001) cross-sections. This simulation result compares favorably with previous individual dislocation tracking simulations [127], where for \(+0.3\%\) misfit under
Table 5.3: The distribution of plastic deformation strain related to the 8 active slip systems under 152MPa tension along [001] for a negative misfit (−0.3%) alloy. (Illustrated by cross-sections along {001}.)

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Table 5.3 Continued

<table>
<thead>
<tr>
<th>n = \frac{1}{\sqrt{3}}(111), b = \frac{1}{2}[\overline{1}01][b]</th>
<th>S</th>
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<tr>
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<tr>
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<tr>
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<tr>
<td><img src="image9.png" alt="Diagram" /></td>
<td><img src="image10.png" alt="Image 9" /></td>
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</table>
### Table 5.4: The distribution of plastic deformation strain related to the 8 active slip systems under 152MPa tension along [001] for a positive misfit (+0.3%) alloy. (Illustrated by cross-sections along {001}.)

<table>
<thead>
<tr>
<th>Negative misfit (+0.3%) under 152MPa tension</th>
<th>(100)plane</th>
<th>(010)plane</th>
<th>(001)plane</th>
</tr>
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<tbody>
<tr>
<td>Microstructure</td>
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<td><img src="image2" alt="Image" /></td>
<td><img src="image3" alt="Image" /></td>
</tr>
<tr>
<td>Slip system</td>
<td>( \mathbf{n} = \frac{1}{\sqrt{3}}(111), \quad \mathbf{b} = \frac{1}{2}[011]</td>
<td>)</td>
<td>( \mathbf{n} = \frac{1}{\sqrt{3}}(111), \quad \mathbf{b} = \frac{1}{2}[011]</td>
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<td><img src="image12" alt="Image" /></td>
<td><img src="image13" alt="Image" /></td>
<td><img src="image14" alt="Image" /></td>
<td><img src="image15" alt="Image" /></td>
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Continued
Table 5.4 Continued

| n = \frac{1}{\sqrt{3}}(111), b = \frac{1}{2}[011][b] | S | S | S |
| n = \frac{1}{\sqrt{3}}(111), b = \frac{1}{2}[101][b] | S | S | S |
| n = \frac{1}{\sqrt{3}}(111), b = \frac{1}{2}[011][b] | S | S | S |
| n = \frac{1}{\sqrt{3}}(111), b = \frac{1}{2}[101][b] | S | S | S |
| n = \frac{1}{\sqrt{3}}(111), b = \frac{1}{2}[011][b] | S | S | S |
| n = \frac{1}{\sqrt{3}}(111), b = \frac{1}{2}[101][b] | S | S | S |

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Figure 5.7: 3D view of the plastic strain distribution caused by slip system: $1/2[011](11ar{1})$ after initial dislocation $\gamma$-channel filling (negative misfit alloy ($-0.3\%$) under 152MPa tension along [001]).
152MPa tension, the dislocations only enter one of the vertical channels which contains 60 degree dislocations.

According to Eq. 5.14, the macroscopic plastic strain generated by the channel dislocations is given by

\[
\bar{\varepsilon}^p = \begin{bmatrix}
-0.07569\% & 0 & 0 \\
0 & -0.07586\% & 0 \\
0 & 0 & 0.1515\%
\end{bmatrix}
\]

for the negative misfit under tension case and

\[
\bar{\varepsilon}^p = \begin{bmatrix}
-0.06556\% & 0 & 0 \\
0 & -0.06630\% & 0 \\
0 & 0 & 0.1319\%
\end{bmatrix}
\]

for the positive misfit under tension case.

Apparently, the macroscopic plastic strain caused by initial dislocation channel filling is quite small which is on the same order of magnitude as the elastic strain.

### 5.3.3 Rafting under the influence of channel plasticity

When the local plastic strain field stops evolving, which indicates that the initial \(\gamma\)-channel plastic deformation (dislocation channel filling) process is complete, the evolution of the \(\gamma/\gamma'\) microstructure is described by the time-evolution of the concentration field and the order-parameter fields, which occurs at a much longer time scale as compared to the evolution of the plastic strain field associated with dislocation glide. During the simulation, the plastic strain fields, \(\eta_i(x)\), were updated constantly as the \(\gamma/\gamma'\) microstructural evolved. Figure 5.8 shows the time-evolution of the \(\gamma/\gamma'\) microstructure on one of the (100) cross-sections for a negative misfit alloy (−0.3%) under 152MPa tension along [001]. An N-type rafting (i.e., \(\gamma'\) particles evolve into rafts normal to the applied tension direction) was produced.
Figure 5.8: The time evolution of microstructure on one of the (100) plane cross-section for a negative misfit alloy (−0.3%) under 152MPa tension along [001] under these conditions. Figure 5.9 is the 3D view of the rafted $\gamma'$ microstructure at $t=5.67$ hours.

The spatial distribution of the plastic strains in the $\gamma$-channels with respect to each slip system at $t=5.67$ hours are listed in Table 5.5. The macroscopic plastic strain during the rafting process, $\bar{\varepsilon}^p_{33}$, calculated from the plastic strain fields (Eq. 5.14) is plotted in Fig 5.10 as a function of time after the external stress was applied.

The macroscopic plastic strain tensor at $t=5.67$ hours is

$$\bar{\varepsilon}^p = \begin{bmatrix} -0.1669\% & 0 & 0 \\ 0 & -0.1672\% & 0 \\ 0 & 0 & 0.3341\% \end{bmatrix}$$

which corresponds to an average dislocation spacing of 30nm on (001) $\gamma'/\gamma'$ interface [129]. (Considering all 8 active slip systems, on the (001) $\gamma'/\gamma'$ interface, dislocation’s line direction will be along 2 different $\langle 110 \rangle$ orientations (4 slip systems
Figure 5.9: 3D view of the microstructure for a negative misfit alloy (−0.3%) under 152MPa tension along [001] after t=5.67 hours.

Figure 5.10: Plastic strain-creep time curve (negative misfit: −0.3% under 152MPa tension)
Table 5.5: The plastic deformation strain distribution with respect to 8 activated slip systems after 5.67 hours evolution under 152MPa applied tension along [001] for a negative misfit (−0.3%) alloy.
Table 5.5 Continued

<table>
<thead>
<tr>
<th>( \mathbf{n} = \frac{1}{\sqrt{3}} (111), )</th>
<th>( \mathbf{b} = \frac{1}{2} [\bar{1}01] )</th>
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<td>( \frac{1}{2} [011] ) ( \mathbf{b} )</td>
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<td>( \frac{1}{2} [011] ) ( \mathbf{b} )</td>
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<td>( \frac{1}{2} [101] ) ( \mathbf{b} )</td>
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<td>( \frac{1}{2} [101] ) ( \mathbf{b} )</td>
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</tbody>
</table>

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for each \langle 110 \rangle direction). Thus, by assuming an even distribution of dislocations from all 8 slip systems in the \( \gamma \) channel, the distance between neighboring dislocations on the \{001\} \( \gamma/\gamma' \) interface is \( d_0 = \frac{|b|}{\varepsilon_{33}/V_f\gamma} = 30 \text{nm} \), where \( d_0 \) is the distance between neighboring dislocations which belong to the same slip system, Burgers vector \( |b| = 0.25 \text{nm} \) and the volume fraction of the \( \gamma \) phase \( V_f\gamma = 40\% \)

After the initial dislocation channel filling process, the increase of plastic strain comes from the evolution of the \( \gamma/\gamma' \) microstructure which changes the width and arrangement of the \( \gamma \)-channels. Thus the misfit stress distribution is altered accordingly which makes further plastic deformation of the \( \gamma \)-channels possible. When the rafts fully develops, the microstructural evolution slows down dramatically, which can be noticed from Fig 5.10 as the slope of the creep curve decreases. Increasing further the simulation time will not produce more useful information with regard to the creep deformation when it enters a steady-state stage (marked by the completion of rafting) where further creep deformation involve dislocation entering the \( \gamma' \) phase, which is not considered in this model.

Figure 5.11 shows the evolution of microstructure on one of the (100) plane cross-section for a positive misfit alloy (+0.3\%) under 152MPa tension along [001]. A P-type rafting was developed in which the \( \gamma' \) particles evolved into rafts parallel to the applied tension direction. Figure 5.12 is a 3D view of the microstructure at \( t = 5.67 \text{ hours} \). The plastic strains in the \( \gamma \) channels with respect to the eight activated slip systems at \( t = 5.67 \text{ hours} \) are listed in Table 5.6. The plastic strain component \( \varepsilon_{33}^p \) is plotted in Fig. 5.13 as a function of time after the external stress is applied. The
macroscopic plastic strain at 5.67 hours is:

$$\bar{\varepsilon}_p = \begin{bmatrix} -0.1085\% & 0 & 0 \\ 0 & -0.1028\% & 0 \\ 0 & 0 & 0.2114\% \end{bmatrix}$$

which corresponds to an average dislocation spacing of 47nm on (100) and (010) $\gamma/\gamma'$ interfaces [129].

5.4 Discussion

5.4.1 Comparison of rafting completion time and aspect ratio change with previous simulation results

The average chord length [132] of the $\gamma'$ phase was measured along the rafting direction and its transverse direction for both N-type and P-type rafting and plotted in Fig. 5.14 as a function of time. The average chord length for the starting microstructure is 192nm along both directions, which corresponds to the average edge length of the initial cuboidal shaped $\gamma'$ particles before the external stress was
Figure 5.12: 3D view of the microstructure for a positive misfit alloy (+0.3%) under 152MPa tension along [001] after $t=5.67$ hours.

Figure 5.13: Plastic strain-creep time curve (positive misfit: +0.3% under 152MPa tension)
Table 5.6: The plastic deformation strain distribution with respect to 8 activated slip systems after 5.67 hours evolution under 152MPa applied tension along [001] for a positive misfit (+0.3%) alloy.
Table 5.6 Continued

<table>
<thead>
<tr>
<th>( \mathbf{n} ) = ( \frac{1}{3\sqrt{3}} (111) ), ( \mathbf{b} = \frac{1}{2} [\mathbf{011}] \mathbf{b} )</th>
<th>( \mathbf{n} ) = ( \frac{1}{3\sqrt{3}} (111) ), ( \mathbf{b} = \frac{1}{2} [\mathbf{011}] \mathbf{b} )</th>
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<tr>
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<th>( \mathbf{n} ) = ( \frac{1}{3\sqrt{3}} (111) ), ( \mathbf{b} = \frac{1}{2} [\mathbf{011}] \mathbf{b} )</th>
<th>( \mathbf{n} ) = ( \frac{1}{3\sqrt{3}} (111) ), ( \mathbf{b} = \frac{1}{2} [\mathbf{011}] \mathbf{b} )</th>
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</table>
applied. From the volume fraction of the γ phase, the average γ channel width was estimated to be 36nm. One interesting finding is that the width of the γ rafts, regardless whether P-type or N-type, remains more or less constant during the rafting process, while the length of the rafts increases with time. Such an observation cannot be made simulations [127] where perfectly aligned periodical array of identical γ particles were considered. If one starts with a single particle in a computation cell with periodic boundary condition, then the volume of the particle is conserved during rafting and the corresponding aspect ratio will jump to infinite when the rafting process completes with the coalescence of neighboring particles. For the simulations in the current study where multiply particles with variations in their size and spatial distribution were considered, stress-induced coarsening of γ particles plays a significant role [3]. Experiment studies also indicated that by considering particle size and spatial distribution, a better description of the rafting process could be achieved as it was observed that the width of the rafts did not appear to become smaller comparing to the edge length of the original cuboidal-shaped γ particles, while in the mean time, the γ-channels grow significantly wider [3].

The average aspect ratio of the γ particles for the current study is plotted in Fig. 5.15. The aspect ratio increases almost linearly initially and gradually slows down. The rafting completion time could then be defined as the moment when the increase of the aspect ratio begins to slow down [66]. For the N-type rafting, it happened around 3.87 hours while for the P-type rafting, it happened around 4.72 hours. As compared to the previous rafting simulation results obtained for perfectly aligned periodical array of identical γ particles (11 hours for N-type and 7
hours for P-type), these rafting completion times are significantly shorter. There are several reasons that may have caused this discrepancy. First of all, the $\gamma'$ size and spatial arrangement in the starting microstructures are significantly different. As we already mentioned earlier, stress-induced coarsening plays an important role in the current study while it is absent in the previous study [127]. As the $\gamma$-channels widen during coarsening, more dislocations will enter the $\gamma$-channel leading to higher channel dislocation density that further enhance the rafting process. Moreover, for the simulations reported in [127], the starting microstructure contains $\gamma'$ particles with edge length of 400nm and $\gamma$-channel width of 93nm. As $\gamma'$ rafting is a diffusion-controlled process, the rafting completion time, $t$, can be related to the typical diffusional transport distance, which is of the order of the $\gamma'$ cube edge length $L$, i.e., $t \propto \frac{L^2}{D_{eff}}$ [28], where $D_{eff}$ is the effective diffusivity. So, starting with
Figure 5.15: $\gamma'$ particle aspect ratio as a function of time. (diamond: N-type rafting; square: P-type rafting)

Larger $\gamma'$ particles mean longer diffusion time to form rafts. In addition, the $\gamma'$ volume fraction in the current study (60%) is higher than that used in the previous study (53%). A benchmark test for a negative misfit alloy ($\delta = -0.3\%$) under 152MPa [001] tension was carried out using the current plastic strain field model but with identical conditions as the one used in the previous simulation [127]. Starting with the same uniform particle size and perfect alignment with 2 slip systems operating, the completion of rafting takes about 8 hours. Even though it is still faster than the previous prediction, this is because the dislocation density is higher (neighboring dislocations distance around 36nm) in this test than the previous case when dislocations are confined in pre-defined crystal planes with dislocations separation distance 65nm. This discrepancy is already explained in Section 5.2.2.
Another interesting observation in the current study is that the P-type rafting took even longer time to complete than the N-type rafting, which could be attributed to the spatial variation in $\gamma'$ particle alignment in the starting microstructure as it will be less probable for $\gamma'$ rafts (P-type) to meet head on with adjacent rafts as compared to the case of N-type rafting. That is the reason why the P-type rafted microstructure may not appear as well aligned as the N-type rafted microstructure just as observed in the experiments [3].

5.4.2 Comparison with results using effective plastic deformation strain

The effective plastic strain method [128, 129] is based on the assumption that the $\gamma$ phase is uniformly deformed. However, experimental observations [15], theoretical analyses [51] and simulations of $\gamma$-channel filling process [127] have shown that under a given stress different $\gamma$-channels will have different dislocation contents. Therefore, to evaluate the effect from non-uniform distribution of dislocations in different $\gamma$-channels, simulations with uniform average plastic deformation strain in the $\gamma$ phase were carried out for the case with negative and positive lattice misfit ($\delta = \pm 0.3\%$) under tension (152MPa). The values of the macroscopic plastic strain for the system were obtained from the results of the initial dislocation channel filling which was presented in Section 5.3.2. As the plastic deformation occurs only in the $\gamma$-channels, the effective plastic strain corresponding to the $\gamma$ phase depends on its volume fraction $V_{f\gamma}$. For $\delta = -0.3\%$:

$$\varepsilon_{\text{eff},\gamma} = \frac{1}{(V_{f\gamma})} \begin{bmatrix}
-0.07569\% & 0 & 0 \\
0 & -0.07586\% & 0 \\
0 & 0 & 0.1515\%
\end{bmatrix}$$

$$= \begin{bmatrix}
-0.1892\% & 0 & 0 \\
0 & -0.1897\% & 0 \\
0 & 0 & 0.3788\%
\end{bmatrix}$$
for $\delta = +0.3\%$

$$\epsilon^{eff} = \frac{1}{(Vf)} \begin{bmatrix} -0.06556\% & 0 & 0 \\ 0 & -0.06630\% & 0 \\ 0 & 0 & 0.1319\% \end{bmatrix}$$

$$= \begin{bmatrix} -0.1639\% & 0 & 0 \\ 0 & -0.1658\% & 0 \\ 0 & 0 & 0.3298\% \end{bmatrix}$$

which indicates an averaged channel dislocation spacing on the \{001\} $\gamma/'$ interface about 66nm for $\delta = -0.3\%$ and 76nm for $\delta = +0.3\%$ [128, 129] with $Vf = 40\%$.

By using $\epsilon^{eff} = \epsilon^0 - \epsilon^{eff,0}$ [128, 129], the effective stress-free transformation strain of $\gamma'$ phase can be calculated. For example, for $\delta = -0.3\%$,

$$\epsilon^{eff,0} = \begin{bmatrix} -0.1108\% & 0 & 0 \\ 0 & -0.1103\% & 0 \\ 0 & 0 & 0.6788\% \end{bmatrix}$$

and for $\delta = +0.3\%$,

$$\epsilon^{eff,0} = \begin{bmatrix} 0.4439\% & 0 & 0 \\ 0 & 0.4658\% & 0 \\ 0 & 0 & -0.02980\% \end{bmatrix}$$

The microstructure evolutions obtained under such effective stress-free transformation strains are shown in Fig. 5.16 for $\delta = -0.3\%$ and Fig. 5.18 for $\delta = +0.3\%$. The corresponding 3D views of the microstructures at t=5.67 hours are shown in Fig. 5.17 and Fig. 5.19, respectively.

By comparing the microstructural evolutions shown in Figs. 5.16 and 5.18 with those shown in Figs. 5.8 and 5.11, one can readily see that the rafting process predicted by using the uniform plastic deformation strain is much faster in terms of the change of the aspect ratio of individual $\gamma'$ particles. However, after the $\gamma'$ rafts form, further coarsening leads to similar microstructure as compared to the result obtained using the non-uniform plastic deformation strain fields. This is because
Figure 5.16: The time evolution of microstructure on one of the (100) plane cross-section for a negative misfit alloy (−0.3%) using effective plastic deformation strain obtained from plastic deformation strain fields calculation for 152MPa [001] tension.

Figure 5.17: 3D view of the microstructure (at t=5.67 hours) for a negative misfit alloy (−0.3%) using effective plastic deformation strain.
Figure 5.18: The time evolution of microstructure on one of the (100) plane cross-section for a positive misfit alloy (+0.3%) using effective plastic deformation strain obtained from plastic deformation strain fields calculation for 152MPa [001] tension.

Figure 5.19: 3D view of the microstructure (at t=5.67 hours) for a positive misfit alloy (+0.3%) using effective plastic deformation strain.
only one type of γ-channel is left after rafting and the plastic strain in the γ-channels become uniform. Therefore the effective transformation strain approach can be used to simulate microstructural evolution after the rafting has completed. The advantage of the effective transformation strain approach is that it allows for large scale simulations. One such example is shown in Fig. 5.20 to Fig. 5.22, where the simulations were carried out in a 3D system of 256 × 256 × 256 grid points, which represents a system size of (5.12μm)³. The starting microstructure (Fig. 5.20) is obtained from 4.7 hours aging at 1300K after solution treatment (misfit |δ| = 0.3%) with the phase field method. Using the same effective transformation strains as those used above for the smaller systems, the N- and P-type rafted microstructures obtained after 5.67 hours aging at 1300K under 152MPa tension along [001] for a negative-misfit (−0.3%) and a positive-misfit (0.3%) alloys are shown in Fig. 5.21 and 5.22, respectively.

The effective plastic deformation strain approach is established upon the assumption of uniform deformation of the γ matrix which exaggerates the influence from channel dislocations as they only prefer certain γ-channels due to the misfit stress. When using plastic deformation strain fields approach, the gradual elongation of γ′ by consuming small γ′ particles dominates the rafting process, which means the rafts maintain constant thickness while in the same time coarsen along a certain direction at the expense of the dissolution of smaller precipitates. While for effective plastic strain case, all γ′ particles undergo dramatic aspect ratio change immediately after the diffusion starts, which makes the early formed rafts appear even finer than the starting microstructure, and then begin to coarsen along lateral direction after rafts completely form by running into their neighboring particles.
Figure 5.20: Microstructure after 4.7 hours aging at 1300K.

Figure 5.21: Negative misfit (−0.3%) alloy rafting under 152MPa tension for 5.67 hours using effective plastic deformation strain.
After a certain period time, the two different approaches will demonstrate similar features for the rafted microstructure. The effective deformation strain can still be proven worthwhile when computational efficiency is concerned because it does not involve calculation of the plastic deformation from individual slip systems.

5.4.3 Precipitate-matrix inversion

For typical single crystal Ni-base superalloys for blade applications, the volume fraction of the $\gamma'$ phase usually far exceeds 50%. If there is no coherency elastic strain caused by lattice misfit between the $\gamma/\gamma'$ phases and no anti-phase boundary (APB) effect (i.e., a thin layer of $\gamma$ phase will separate neighboring $\gamma'$ particles of different anti-phase domains as long as the APB energy is higher than twice of the $\gamma/\gamma'$
interfacial energy [133]), the γ phase will become the matrix phase after extended period of thermal exposure. In reality, while the misfit stress and APB maintain the cuboidal shape of the γ particles, an applied stress introduces dislocation contents into certain types of γ-channels (horizontal or vertical channel, depending on the applied stress direction and the sign of lattice misfit) which partially relax the misfit stresses. As the microstructure evolves under the influence of the applied stress, rafts begin to form. The elastic contribution from the back stresses generated by the γ-channel dislocations is significant enough to drive neighboring γ particles coming into contact even when they belong to different anti-phase domains, which leaves APBs in between. During this rafting process, the majority phase (γ) percolates through the system and becomes the matrix phase. Thus, the precipitate and matrix are topologically inverted.

Such a microstructural inversion of the γ matrix and γ precipitates during the rafting could be characterized by different methods, for instance, by the ratio of area densities of transverse terminations of γ and γ lamellae [134]. One example is demonstrated in Fig. 5.23 which was performed on a 2D cross-section of the N-type rafting microstructure at t=5.67 hours with red circles at the transverse terminations of γ phase and blue triangles at the transverse terminations of γ phase. As the topological inversion has already occurred in this microstructure, the ratio of the transverse termination is \( \frac{\text{Terminates}(\gamma)}{\text{Terminates}(\gamma')} = 1.9 \), i.e., γ' is the matrix. In order to achieve better efficiency using digitized data with fewer human interventions, the quantification of the inversion process in the current study was performed through the statistics of the isolated particles number ratio between the two phases. That is, within the simulation unit cell, numbers of isolated γ and γ' particles are counted.
during the microstructure evolution. The analysis was performed using 2D cross-section plots obtained at different time moments (Fig. 5.8 and 5.11). As an example, Fig. 5.24 shows one $\gamma$ and one $\gamma'$ particle on the cross-section at $t=2.83$ hours for the $-0.3\%$ misfit under tension case. The ratio of $\gamma'$ particle number to $\gamma$ particle number as a function of time for $-0.3\%$ and $+0.3\%$ lattice misfit under 152MPa tension are shown in Fig. 5.25 and 5.26 respectively, from this, the inversion process can be quantified as a function of rafting time. Using the moment when this ratio changes from greater than one to smaller than one as the time when the inversion completes, the N-type rafting microstructure inverts before 4.72 hours and P-type microstructure inverts before 2.83 hours. Compare to the rafting completion time, P-type rafts percolate faster than N-type because most P-type rafts are rod-shaped $\gamma'$ aligned along the external stress direction (this has been verified by previous perfectly aligned $\gamma'$ particles test [127]). While due to the spatial variation of the initial microstructure, it takes longer for the already percolated P-type $\gamma'$ rods to complete the coarsening (rafting) process.

The ratio of $\gamma'$ particle number to $\gamma$ particle number for $-0.3\%$ and $+0.3\%$ lattice misfit under 152MPa tension are plotted in Fig. 5.25 and 5.26 respectively. From which, the inversion process can be quantified as a function of rafting time.

5.4.4 Limitations of the model and future development

Besides the $\gamma'$ precipitates, there is no other hardening mechanism in the current model. An interesting observation is that if the volume fraction of the $\gamma'$ phase is small, some of the dislocations will not encounter any $\gamma'$ particle on their glide
Figure 5.23: Illustration of the transverse termination count of the $\gamma$ and $\gamma'$ phases for a system with $-0.3\%$ lattice misfit under 152MPa tension after 5.67 hours.

Figure 5.24: Illustration of isolated $\gamma$ and $\gamma'$ particles for a system with $-0.3\%$ lattice misfit under 152MPa tension after 2.83 hours relaxation.
Figure 5.25: The ratio of $\gamma'$ particle number to $\gamma$ particle number for a system with $-0.3\%$ lattice misfit under 152MPa tension as a function of time.

Figure 5.26: The ratio of $\gamma'$ particle number to $\gamma$ particle number for a system with $+0.3\%$ lattice misfit under 152MPa tension as a function of time.
plane. In this case, the plastic deformation strain will reach infinite under the periodic boundary conditions employed. Further improvement of the model should include specifying certain hardening laws which take into account dislocation interactions.

5.5 Summary

A new phase field method of plastic deformation based on the evolution of plastic strain fields is developed and validated against discrete dislocation method. This new approach is implemented for γ‘ rafting study in single crystal Ni-Al, which is able to capture the main driving force (the channel plasticity) that dictates the rafting type and rafting kinetics, as well as to incorporate realistic experimental parameters such as the γ‘ paricle size and volume fraction, dislocation density and inter diffusivity at the creep temperature. From the input parameters, the plastic deformation and microstructural evolution can be simulated in 3D demonstrating the correlations between rafting type and kinetics with respect to different materials and processing parameters. Moreover, with the help of the KS model, large length scale is achieved for simulations containing hundreds of γ‘ precipitates with statistical variations in particle sizes and spatial distributions.

It is found that when the γ‘ particle size and spatial distributions are considered, while the cause of the symmetry breaking of the microstructure still comes from the solute-stress interaction, coarsening becomes more important as the average raft width maintains constant during the rafting process which agrees with the experimental observations. Also, P-type rafted microstructure appears less aligned comparing with the N-type rafted microstructure. Though the matrix-precipitate
inversion reflects the progress of the rafting process, it does not exactly coincide with the completion of rafting, which means certain material properties relating to the percolated phase may change before or after the completion of rafting.

Using this new method, given the composition of the alloy, the $\gamma/\gamma'$ interfacial energy, the lattice misfit, the elastic constants and its working condition (temperature and external stress), microstructure evolution (rafting and matrix-precipitate inversion) and plastic deformation process can be predicted quantitatively with the support of commercially available thermodynamic and kinetic databases, making possible more cost effective new alloy designs.
CHAPTER 6

SUMMARY AND FUTURE WORK

6.1 Summary

Three-dimensional phase field simulations of coupled \(\gamma/\gamma'\) microstructural evolution and plastic deformation were carried at different length scales. For the first time the relative contributions from modulus mismatch, channel plasticity and the combination of the two to \(\gamma'\) rafting have been discriminated by the dislocation-level simulations. Results from quantitative comparisons of the rafting completion time, driving force variations and microstructural evolution among different cases can be briefly summarized as follow:

I) When both channel plasticity and modulus mismatch are considered, the type of rafting is determined solely by the direction of the applied stress and the sign of the lattice misfit (i.e., the sign of \(\frac{\sigma^{c}}{\delta}\)). The sign of the modulus mismatch does not change the rafting type, which is true even when there is a large difference in the shear modulus (e.g., \(\frac{C_{11} - C_{12}}{C_{11} - C_{12}} = 85\%\)).

II) Normal type rafting occurs for a positive misfit alloy under compression or a negative misfit alloy under tension, while Parallel type rafting occurs for a positive misfit alloy under tension or a negative misfit alloy under compression.
III) Modulus mismatch between precipitate and matrix does alter the rafting kinetics. If the precipitate phase has higher elastic stiffness than that of the matrix, the rafting process will take less time to complete as compared to a system having softer precipitates (assuming the same applied stress, misfit and dislocation density). This is true irrespective of the signs of the applied stress and lattice mismatch.

IV) Dislocations prefer normal channels (normal to the direction of applied load) for a positive misfit alloy under compression, and a negative misfit alloy under tension, while parallel channels were preferred by dislocations in a positive misfit alloy under tension and a negative misfit alloy under compression.

V) Dislocation density will also affect the kinetics of rafting. The higher the channel dislocation density, the faster the rafting process. The density of dislocations is determined by the magnitude of the applied stress, lattice misfit and the starting microstructure (γ particle size and γ channel width).

Based on these findings, micrometer-scale simulations were carried out that takes into account plastic deformation in γ-channels described by local channel dislocation densities from individual active slip systems. The rafting kinetics, precipitate-matrix inversion process and the corresponding creep deformation can be characterized at different values of applied stress, lattice misfit and precipitate volume fraction through phase field simulation. With the assistance of the phase field model, the interplay between elastic inhomogeneity and channel plasticity can be characterized and utilized to offer opportunities for possible new design strategy to better control the rafting process.
6.2 Future work

The phase field dislocation model currently used in this study only deals with perfect dislocations, while in reality, as Ni-base superalloys usually have medium/low stacking fault energy, dislocation dissociation is not uncommon. Thus, the treatment of partial dislocations needs to be incorporated into the model for more accurate description of the channel filling process.

In order to answer the question how those complicate dislocation networks form, dislocation climbing mechanism must be addressed properly in the model. Combining with the cross-slip mechanism which has already been developed for phase field dislocation model, it will be interesting to explore how dislocations weave into the network and rearrange themselves through cross-slip and climb and even dislocation reaction to better accommodate the lattice misfit and applied stress. By incorporating these mechanisms, the creep curve for the steady state stage can also be obtained from phase field simulations.

As a consequence of the rafted microstructure, further plastic deformation of γ channel is blocked by the continuous γ' phase, thus, as local stress begins to build up, some dislocations may start to shear γ'. And by incorporating γ'-surface into the description of dislocation crystal energy, different kinds of fault energies such as anti-phase boundary energy (APB), stacking fault energy(CF), complex stacking fault energy(CSF), superlattice intrinsic stacking fault energy(SISF) and superlattice extrinsic stacking fault energy(SESF) can be introduced into the model to study the γ' shearing process which becomes important at the late stage of creep.

The ultimate goal of the phase field simulation of Ni-base single crystal alloys is to develop a model from which the creep property of the alloy can be predicted.
as a function of the alloy chemistry, heat treatment history and working condition (temperature and loading condition). For new superalloy designers, this model can serve as a cost-effective tool to optimize alloy composition and heat treatment procedures.
BIBLIOGRAPHY


