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MODELING OF PRECIPITATION EFFECTS ON THE THERMOMECHANICAL BEHAVIOR OF NITI SMA

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ABSTRACT

The effect of precipitation on Ni-rich NiTi Shape Memory Alloys on the thermomechanical properties is investigated. Particular, the relation between the chemical evolution of the alloy and phase transformation characteristics are studied in order to evaluate the impact on the global mechanical response. The evolution of chemical composition of the alloy is computed using a diffusion law, according to the precipitates population characteristics. The mechanical response is simulated using a Representative Unit Cell which takes into account the structural effect of the precipitates as well as the effect of the composition change. The results show an important effect on the global mechanical behavior even for a small volume fraction of precipitates.

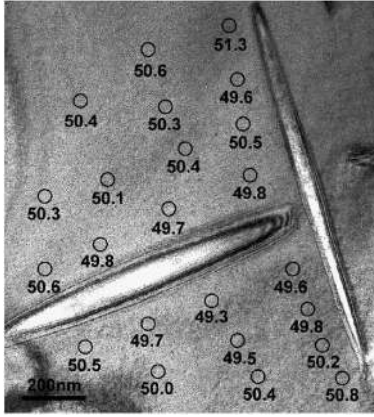
INTRODUCTION

It is well known that shape memory alloy behavior is very sensitive to microstructural parameters. At given chemical compositions, application of suitable thermomechanical treatments can strongly modify the SMA behavior characteristics, like hysteresis size, plastic yield limit, transformation temperatures, transformation and reorientation slopes or occurrence of residual strain [1]. Precipitation is among the major microstructural features responsible to this sensitivity. According to the nature of the alloy system considered, second phase precipitation or de-

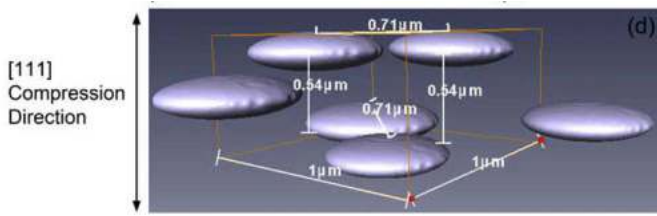
composition into equilibrium phases may occur, leading to strong modifications in the alloy overall behavior. Many metallurgical studies are devoted to these precipitation phenomena and their effects on martensitic transformation [1–3]. Many models are also derived to describe the SMA behavior. But, up to now, only a few attempts were made to model the influence of precipitates on this behavior despite its importance in reaching a better understanding of strain mechanisms in SMAs and for practical purposes in application design.

In Ni-rich NiTi SMAs, when precipitation occurs (especially the appearance of Ni_4Ti_3 precipitates), the Ni concentration in the material is affected. In this work Ni-rich SMA denotes a NiTi with more than 50.2 % Ni. Because Ni_4Ti_3 is a high Ni-content phase (57.1%), the growing of precipitates induces a Ni depletion in the matrix around precipitates through a diffusion process. A concentration gradient appears, until the thermodynamical chemical balance is reached (lowest bound for Ni concentration). This effect has been experimentally observed by Schryvers *et al.* [4]. Figure 1b shows the spot measurements of the Ni concentration around Ni_4Ti_3 precipitates, where it is clear that the Ni content decreases in the vicinity of the precipitates. Several authors have observed the size and shape of these Ni_4Ti_3 precipitates [2, 5–7]. Lenticular-shape precipitates appears with an aspect ratio and size which are strongly dependent on the annealing duration time and temperature. In a general manner, the thickness - diameter ratio decreases with the precipitates growth. Figure 1b shows

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(a) Spot measurement of Ni concentration around Ni_4Ti_3 precipitates [4]



(b) 3D reconstruction of Ni_4Ti_3 precipitates [5]

FIGURE 1. Morphology of Ni_4Ti_3 precipitates and Ni depletion in the matrix

a 3D reconstruction of these precipitates from microstructural observations [5], where the lenticular shape of the precipitates clearly appears.

Several authors have related the transformation temperatures to the Ni concentration in quasi equiatomic NiTi SMAs. The composition change in an alloy, due to an annealing process, can strongly modify the transformation temperatures. The thermo-mechanical behavior is then impacted, and the objective of this work is to better understand how the mechanical response is affected. This work focuses only on the Ni concentration gradient impact. The effect of cold-working [8], which can also shift the transformation temperatures is not taken into account.

To this end, Section 1 will discuss the model being utilized, from the determination of the concentration of Ni, the impact of the concentration change on the transformation characteristics and the development of the numerical model. Also, the model utilized for the simulation of the SMA phase is briefly presented. Section 2 will present the mechanical behavior of the different cases simulated, and discuss the effect of the different features implemented in the numerical model. Concluding remarks will be given in the last section.

1 MODELING

To describe the behavior of SMAs containing precipitates, an appropriate modeling scheme must be developed. Several approaches can be used:

One can consider a micromechanical model, considering a two-phase composite: the Ni_4Ti_3 precipitates and the NiTi matrix. Using the mean field approach, the concentration in the matrix is supposed to be homogeneous. This assumption is valid only for long annealing times, where the material reaches the thermodynamic equilibrium. However these models can be implemented in FEA softwares to determine the response of annealed structures in real conditions, for example the influence of such Ni depletion after shape setting.

One other approach is to consider a Representative Unit Cell, defined by a precipitate embedded in a surrounding matrix composed of several layers of NiTi SMA, each one with a different composition. A diffusion law is necessary to determine the mean concentration of each layer in this case. The concept of Representative Unit Cell has been developed for periodic microstructures [9, 10]. Even if it is not the case here, this method can still give a good estimation since the inclusions are well distributed. The advantage of this method, in comparison with the previous one, is the introduction of the distribution of the Ni concentration in the model. The drawback is the high computational time cost, as well as the difficulty in finding the behavior in the case of complex loadings.

As the main objective of this work is to study how the composition change of the alloy will impact the transformation behavior, it is important to include most of the features related to this phenomenon in the model. Moreover, several studies had showed that the incremental Mori-Tanaka scheme is not appropriate to describe the behavior of elastic-plastic or SMA matrix with stiff elastic inhomogeneities [11, 12]. For these reasons, the Unit Cell approach is preferred. Particularly, the effect of the gradient of the Ni concentration can be included. The developed model will help to study the effect of the volume fraction of precipitates, as well as their morphology and the effect of the composition change.

Ni concentration determination

In order to determine the Ni concentration of the matrix, several characteristics about the precipitates (volume fraction, shape, mean interdistance) have to be known. An exhaustive study of the effect of annealing on the precipitates characteristics has been conducted by Khalil Allafi *et al.* [2]. In this study, the influence of the Ni concentration gradient on multistage transformation was determined using an uniaxial diffusion law. The same law is used in this work, however applied to the three dimensional case of ellipsoidal precipitates. Moreover, the diffusion coefficient is identified by the balance between the average concentration given by the diffusion law and the one determined

from the experimental determination of the volume fraction of the precipitates. Indeed, knowing the volume fraction of precipitates and the Ni concentration of those precipitates, the mean Ni concentration of the matrix \bar{C}_{Ni} takes the form:

$$\bar{C}_{Ni} = \frac{c_0 - f_p \cdot c_{Ni_4Ti_3}}{1 - f_p} \quad (1)$$

where c_0 corresponds to the initial concentration of the initial material, and $c_{Ni_4Ti_3}$ is the Ni concentration of the Ni_4Ti_3 precipitated phase of volume fraction f_p .

Also, the diffusion law gives the isoconcentration value of an ellipsoidal surface around a precipitate, at a given distance of the surface of the precipitate. By integrating all concentrations on the total matrix volume, the mean concentration is computed. By iterating this procedure, varying the diffusion coefficient until the mean concentration value is equivalent with the one derived from the volume fractions determination, the diffusion coefficient is obtained [13].

Effect of Ni depletion

Several experimental studies show a strong impact of Ni concentration on martensitic transformation temperature M_s for NiTi materials. A review of these tests has been presented by Tang [14]. These experiments show that under 50% at. Ni, there is no relation between Ni concentration and transformation temperatures and a constant value (350K) of M_s is observed. Above 50% at. Ni, M_s values fall down drastically, reaching a theoretical transformation limit temperature (0K) for 51.5% at. Ni concentration. These tests also highlight the same trend for the end of the reverse transformation temperature A_f .

From these results an evolution law is proposed for the evolution of M_s as a function of the Ni concentration. By considering M_s^p the plateau temperature for Ni concentration under 50% at., and $c_{T=0}$ the theoretical concentration limit which correspond to $M_s = 0K$, the following power law for M_s is proposed:

$$M_s = M_s^p \left(1 - \left(\frac{c - c_T}{c_{T=0} - c_T} \right)^\gamma \right) \quad (2)$$

where $c_T = 48.5\%$, which corresponds to the minimal value of Ni concentration. A value of $\gamma = 5$ is calibrated using experimental data (Figure 2).

By combining the diffusion law with the M_s temperature evolution law, one can determine the evolution of transformation temperatures around Ni_4Ti_3 precipitates, due to Ni depletion.

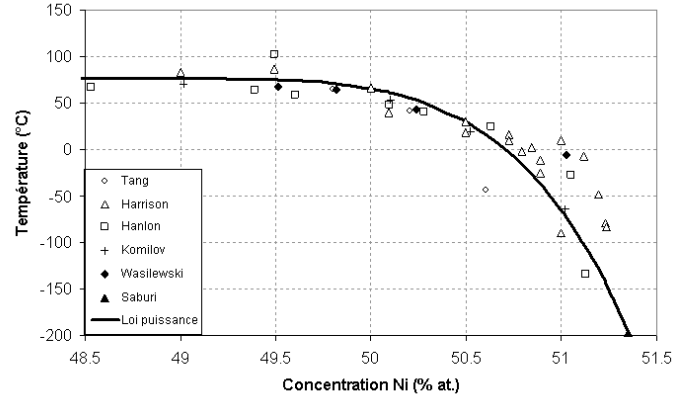


FIGURE 2. Comparison between the power law and experimental data [13]

Representative Unit Cell definition

In order to determine the macroscopic behavior of NiTi which contains precipitates, a model has to be defined with relevant information such as concentration gradient, volume fraction of precipitates, material properties and phases morphology. A Representative Unit Cell is defined to account for these characteristics. Representative Unit Cells have been used for homogenization models of periodic microstructures [10] and as a reference solution for elastic-plastic matrix containing elastic precipitates [11]. The Representative Unit Cell is defined according to the morphology of the precipitates, illustrated in Figure 1b. Since the precipitates have approximately the same shape and size, a unique Unit Cell is defined to represent the annealed material. In this work we choose a parallelepipedic Representative Unit Cell which contains one lenticular precipitate. To define the optimal geometry of this Unit Cell, several criteria are used:

1. The volume fraction of the precipitate inside the Representative Unit Cell is set to be equal to the volume fraction of the precipitates in the material.
2. According to the diffusion law, Ni isoconcentration ellipsoids are defined. Knowing the volume fraction of precipitates, a thickness for the surrounding matrix of each precipitate is defined. This thickness, as well as the shape of the precipitate, are used to define the dimensions of the parallelepipedic Unit Cell.

According to geometrical and uniaxial symmetries, the behavior of 1/8 of the Cell is simulated. For uniaxial loadings, boundary conditions are applied in such a way that they are identical to the periodicity configurations [11].

Phase constitutive laws

To account for the behavior of the precipitate and the SMA matrix layers respectively, two different laws are used. An isotropic elastic law is used to account for the precipitate behavior. While the precipitates can not be isolated from the NiTi matrix, elastic constant measurements are difficult to perform, so in these simulations the isotropic moduli being used ($E = 104\text{GPa}$, $\nu = 0.39$) have been defined using first principles calculations performed by Wagner and Windl [15]. A thermodynamical macroscopic constitutive law defined by Chemisky *et al.* is used to represent the behavior of the SMA matrix [16]. This law has already been used to model the behavior of SMA-based composites, such as SMA-fiber reinforced elastomer composites [17]. This specific law is based on an additive decomposition of the macroscopic strain, according to the small-strain assumption:

$$\mathbf{E} = \mathbf{E}^e + \mathbf{E}^T + \mathbf{E}^{twin} \quad (3)$$

where \mathbf{E}^e is the elastic strain, \mathbf{E}^T is the martensitic transformation strain and \mathbf{E}^{twin} is the twin accommodation strain, which accounts for the twin accommodation mechanism at the interface between two variants, as observed by Liu *et al.* [18]. The macroscopic behavior is then represented considering three internal variables to account the different strain mechanisms: the martensite volume fraction f , the mean transformation strain $\bar{\boldsymbol{\epsilon}}^T$ inside the martensite phase and the mean twin accommodation strain $\bar{\boldsymbol{\epsilon}}^{twin}$ inside the self-accommodated martensite. The relation 3 can be rewritten in the following form:

$$\mathbf{E} = \mathbf{E}^e + f\bar{\boldsymbol{\epsilon}}^T + \gamma_m\bar{\boldsymbol{\epsilon}}^{twin} \quad (4)$$

The definition of these internal variables and the thermodynamical forces related to each internal variable are given in [16]. This law is implemented into the FEA package Abaqus, using Simula+ Mathematical Object Library tool [19].

Material parameters used for the NiTi matrix correspond to common parameters used for NiTi materials [13] (see Table 1), except for the transformation temperatures, related to the Ni concentration gradient (see Table 2).

2 Simulations

Two Unit Cells are studied and each one is defined according to experimental data for a specified annealing time (1h and 10h, see Table 3). These data are obtained from TEM experiments performed on 50.7% at. Ni annealed NiTi by Khalil Al-lafi *et al.* [3]. To represent the transformation temperature gradient inside the Unit Cell corresponding to the 1h heat treatment,

four layers are considered, with different transformation temperatures (according to the Ni concentration average for each layer). As the Ni concentration throughout the matrix for the 10h treatment is quite constant, while the thermodynamical equilibrium is reached almost everywhere in the SMA matrix, a unique set of transformation temperatures is considered for the whole SMA matrix. Two different loading paths are performed: the first one is a uniaxial tensile loading in the direction of the diameter of the precipitate (Figure 4), while the second one is in the direction of the thickness of the precipitate (Figure 5). For each loading case, four temperatures are considered (20, 40, 60 and 80°C) in order to cover several different behaviors for the NiTi matrix phase (superelasticity, reorientation of self-accommodated martensite and combined transformation/reorientation mechanisms).

Results obtained for the various Unit Cell simulations are compared with the initial material, for isothermal tensile loading paths. From these results, the general trend observed is that the critical stress for inducing transformation decreases when the annealing time increases. This effect is explained by the depletion of the Ni concentration in the matrix, which raises the transformation temperatures. Therefore, the average stress limit to induce the martensitic phase transformation decreases.

For the higher testing temperature (80°C), the behavior of the initial material and the 1h annealed material are superelastic. In that case, where the behavior is driven by the transforma-

TABLE 1. Material properties for a common NiTi alloy (The temperatures are indicated in a separated table for each concentration considered)

Parameter	Value
E	70 GPa
ν	0.3
$\boldsymbol{\epsilon}_{tensilemax}^T$	0.05
$\boldsymbol{\epsilon}_{compmax}^T$	0.04
B	0.25 MPa/°C
M_s	NC°C
A_f	NC°C
r_f	0.6
F_ϵ	200 MPa
$H_f(\text{MPa})$	2 MPa
$H_{\epsilon T}(\text{MPa})$	1500 MPa
$H_{twin}(\text{MPa})$	30000 MPa

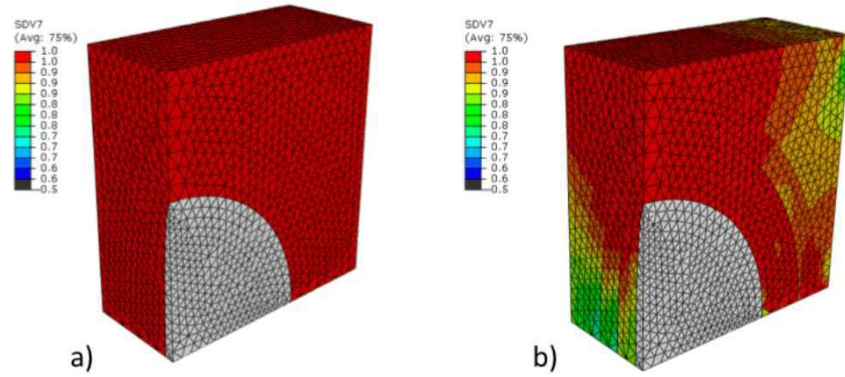


FIGURE 3. Volume fraction of martensite for the Unit Cell (treatment 1h) at 5% tensile strain at a) 20°C b) 80°C

TABLE 2. Transformation temperatures

Treatment 1					
	Layer 1	Layer 2	Layer 3	Layer 4	Average
M_s	51°C	39°C	25°C	9°C	19°C
A_f	81°C	69°C	55°C	39°C	49°C
Treatment 2					
Average					
M_s	55°C				
A_f	85°C				

TABLE 3. Characteristics of the Ni_4Ti_3 precipitates, from [3]

Annealing time at 500°C	1h	10h
Diameter $D(nm)$	115 ± 30	335 ± 120
Thickness $e(nm)$	7.7 ± 3.0	32 ± 7
Mean interparticules distance $l(nm)$	145 ± 20	300 ± 20
Volume fraction of precipitates $f_p(\%)$	1.7 ± 1.0	6.7 ± 3.5

tion of austenite to stress-induced martensite, the transformation hardening is not affected by the effect of annealing at the onset of the transformation. On the other hand, the end of transformation starts at a lower deformation state. A smoother transition between the transformation and the elastic behavior of the martensite is observed. The same effect is observed at the beginning of the reverse transformation. The hysteresis is also slightly

increased. This hardening effect is more pronounced for the 10h annealed material. Moreover, in that case, the hysteresis is significantly increased. Since this effect can be ascribed to the effect of combined reorientation and transformation, it is difficult to evaluate the impact of the precipitate on the transformation part of the hysteresis. For both annealed materials, the hardening effect is reduced in the case of loading in the direction of the thickness of the precipitate compared to the case of the loading in the direction of the diameter of the precipitate. The same trends are observed for the simulations at 60°C, even if in this case both annealed materials are no longer superelastic. For the lower testing temperatures (20 and 40°C), the reduction of the stress limit is less pronounced. This is mainly due to the fact that in these configurations, the mechanical behavior is driven by the reorientation process (which is independent of the transformation temperatures) instead of the transformation process. The variation of the transformation temperatures has then a small impact on the mechanical response.

To illustrate these explanations, Figure 3 shows the martensite volume fraction evolution for the 20°C and 80°C, at the end of the transformation (5% total strain). One can observe a full transformation for loading at 20°C. For the simulations at 80°C, the transformation is localized on the top of the precipitates, and propagates to the upper surface of the Unit Cell. When the transformation strain is fully completed in this area, the elastic behavior of the martensite increases the stiffness of the Unit Cell. This explains the slight increase of the stress at the end of the transformation. This effect is also observed for the second treatment, although there is no transformation temperature gradient in the matrix, and is then mainly due to the structural effect of the precipitate. The trends observed in this study are also experimentally observed by Frick *et. al.* [20], for a different NiTi composition and different set of heat treatments. For high volume fraction and increased size of the precipitates, the stress level to

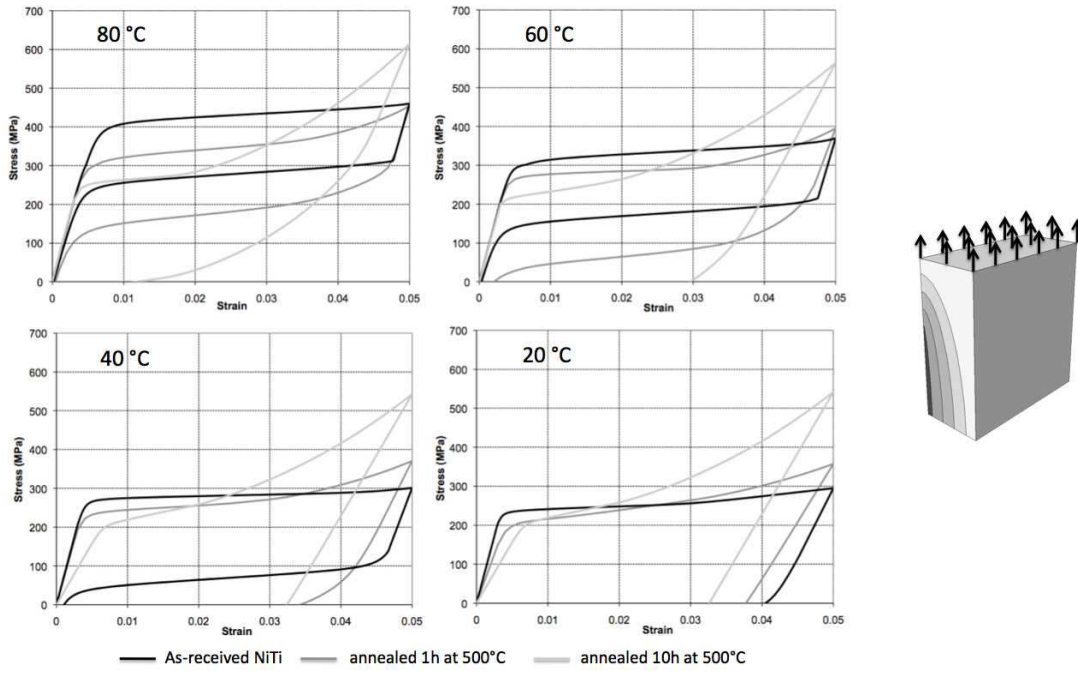


FIGURE 4. Stress strain response for the initial and annealed configurations at various temperatures. The Representative Unit Cell is loaded in the direction of the diameter of the precipitate.

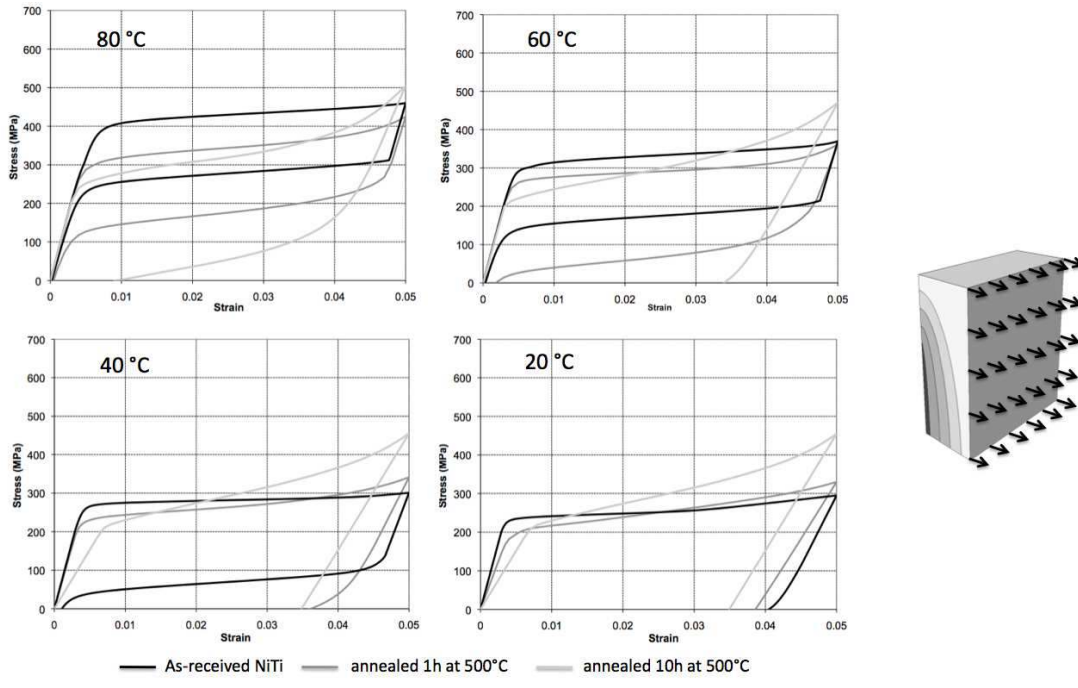


FIGURE 5. Stress strain response for the initial and annealed configurations at various temperatures. The Representative Unit Cell is loaded in the direction of the thickness of the precipitate.

initiate the transformation decreases significantly, as an important increase of the transformation/orientation hardening at the end of the loading path is observed.

3 Conclusions

A methodology has been defined in order to estimate the impact of heat treatment on the macroscopic behavior of Ni-rich NiTi SMAs. Definition of the Ni concentration around precipitates led to the definition of a Representative Unit Cell. Results show a strong influence of the precipitates, as a function of the annealing time, which determine the volume fraction, the size and the aspect ratio of the precipitates. Two main features are involved, both related to the precipitates growth: a) the Ni concentration decreases around precipitates, inducing a change of the transformation temperatures and b) precipitates, through their structural effect, cause a hardening due to their important stiffness. Further investigation will be conducted to study the effect of the internal stress effect due to precipitates coherency as well as the effect of the precipitates distribution.

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