Effect of external stress on γ nucleation and evolution in TiAl alloys

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Abstract

We have shown in a previous paper that twin boundary area fraction in lamellar γ -TiAl based alloys depends on collective or correlated nucleation induced by long-range elastic interactions among nucleating precipitates or between pre-existing and nucleating precipitates. In this study we investigate the effects of applied stresses on γ nucleation and evolution by elastic interaction energy calculations and phase field simulations. It is found that the elastic interaction energy reaches minimum when two variants are twin related because of their self-accommodation of the coherency strain. When external stresses are present, such autocatalysis during nucleation could be either enhanced or suppressed. Compression perpendicular or tension parallel to the basal plane of the α phase is found to enhance the internal elastic interactions between nucleating precipitates, promote twin variant selection and reduce the overall nucleation rate, while isostatic pressing, compression parallel, tension perpendicular to the basal plane does the opposite. Shear along the interface

always reduce the twin boundary area fraction, regardless of the shear direction. These findings could shed light on optimizing processing routes to increase twin boundary area fraction in lamellar microstructures of γ -TiAl based alloys.

Keywords: A. Intermetallics; B. Phase transformation; D. Microstructure; E. Phase field modeling;

1. Introduction

The mechanical properties of a fully lamellar structure depend not only on the grain size [1, 2] and texture [3, 4], which related strongly to the size and orientation of $\gamma+\alpha_2$ lamellae, but also on the fractions of different types of interfaces in the lamellae such as twin, pseudo-twin and ordered domain boundaries between γ lamellae and also γ/α_2 interfaces [5], therefore the lamellar microstructure control is a key to obtaining balanced mechanical properties of γ -TiAl based alloys. The twin boundaries have been found beneficial to the room temperature ductility [6] and high temperature microstructure stability [5] of TiAl alloys. Our previous studies [7] have shown that in the absence of external stresses, the frequency of twin boundary depends on whether the nucleation is independent, collective or correlated as a result of the interplay among several factors including long-range elastic interactions between nucleating precipitates or between pre-existing and nucleating precipitates. However, certain external stress exists in many circumstances during phase transformation. First, for many applications, thermal mechanical processing such as extrusion [8, 9] or rolling [10, 11] is needed as steps to form the parts and optimize the properties of the alloy.

Under such condition, the α to γ phase transformation may take place under external stresses of various type. Second, in the cases of components made from powder metallurgy (PM) or casting, the hot isostatic pressing (HIPping) is often a necessary step for shape formation and defect elimination, the microstructure may also be altered by adjusting temperature and pressure during or after HIPping [12]. Third, the applied stress could influence the microstructure stability during service such as under high temperature creep [13]. However, it is unclear how external stresses affect the frequency of various types of lamellar boundaries under these conditions.

In the present paper, we study the effect of external stresses on the nucleation and growth of γ precipitates and analyze the change of lamellar boundary area fraction as a function of the magnitude and type of applied stresses using the phase field method [14-18]. The nucleation and growth of precipitates under different stress conditions including tension, compression, isostatic pressing and shear are simulated using the phase field method. And then the elastic interaction energies between a pre-existing and nucleating precipitates are calculated by using phase field microelasticity theory. The fractions of various types of interfaces are obtained from the simulation and analyzed with the help of the interaction energy calculation.

2. Orientation relationship and phase field formulation of $\alpha \rightarrow \gamma+\alpha_2$ phase transformation in TiAl

For Ti-Al alloys, within the Al concentration range of 39–46.5 at.%, the phase transformation path is assumed to be $\alpha \rightarrow \gamma+\alpha_2$ [19]. The α phase has HCP structure,

and will order into an HCP based α_2 phase at lower temperature, and the γ (TiAl) and α_2 (Ti₃Al) phases have L1₀ and D0₁₉ crystal structures, respectively. The two phases follow the Blackburn orientation relationship (OR) [20]: $(0001)_{\alpha_2} // \{111\}_{\gamma}$, $\langle 11\bar{2}0 \rangle_{\alpha_2} // \langle 1\bar{1}0 \rangle_{\gamma}$. In this study, we define the coordinate system using the α_2 phase as a reference, i.e., X $[11\bar{2}0]$, Y $[\bar{1}100]$ and Z $[0001]$. Thus the six γ variants can be formed by the transformation shear with respect to the basal plane of the α_2 phase, e.g., variant one (V_1) corresponds to a shear along the positive direction of Y and its twin related variant V_2 corresponds to a shear along the negative direction of Y. The twin pair will be described as $V_1-0^\circ/V_2-180^\circ$ hereafter, where the angles represent the shear direction for γ variants forming measured as the angles of rotations around the Z axis, starting from the Y axis. Similarly, the other twin pairs are $V_3-120^\circ/V_4-300^\circ$ and $V_5-240^\circ/V_6-60^\circ$. Three orientation relationships will be formed between different neighboring γ variants from such transformation, and three types of lamellar boundaries are formed accordingly, i.e., twin (T), pseudo-twin (PT) and ordered-domain (OD) boundaries. The interfaces formed from different variants are listed in Table 1. The stacking sequence ABABAB of HCP becomes either ABCABC or ACBACB through shear during phase transformation. Group γ_I and γ_{II} represent ABC and ACB stacking sequence, respectively. The V_1 , V_3 and V_5 form group γ_I , while the other variants belong to group γ_{II} . The misorientation angles of the pseudo-twin, ordered-domain and twin related variants are 60° , 120° and 180° , respectively.

In the phase field model of $\alpha \rightarrow \gamma + \alpha_2$ transformation in TiAl alloys [7], an arbitrary

microstructure is characterized by introducing six structural order parameter fields and one concentration field. Within the composition and temperature range of our simulation, the α and α_2 phases have similar Gibbs free energy, and they also have similar crystal structure and lattice parameter. Then it is not necessary to distinguish them by the order parameters, for simplicity, we use one set of structural order parameters to describe α/α_2 . The order parameters 0 or 1 represent the α/α_2 or γ phase, respectively. The evolution of the microstructure is described by the time-evolution of the structural order parameter fields and concentration field following the time-dependent Ginzburg-Landau equation and the non-linear Cahn-Hilliard diffusion equation, respectively, i.e.,

$$\frac{\partial \eta_p(\mathbf{r}, \tau)}{\partial \tau} = -\left[\frac{\partial f_a}{\partial \eta_p(\mathbf{r}, \tau)} - \sum_{ij=1}^3 \beta_{ij}^p \frac{\partial^2 \eta_p(\mathbf{r}, \tau)}{\partial u_i \partial u_j} + \phi \frac{\delta E_{\text{el}}}{\delta \eta_p(\mathbf{r}, \tau)}\right] + \xi_p(\mathbf{r}, \tau) \quad (1)$$

$$\frac{\partial c(\mathbf{r}, \tau)}{\partial \tau} = \nu \nabla^2 \left[\frac{\partial f_a}{\partial c(\mathbf{r}, \tau)} - \alpha \nabla^2 c(\mathbf{r}, \tau)\right] + \zeta(\mathbf{r}, \tau) \quad (2)$$

where $\xi_p(\mathbf{r}, \tau)$ and $\zeta(\mathbf{r}, \tau)$ are the random noise terms related to fluctuations in the structural order parameter and concentration, respectively.

$$f_a(c, \eta_1, \eta_2, \dots, \eta_6) = \frac{a_1}{2}(c - c_1)^2 + \frac{a_2}{2}(c_2 - c) \sum_{p=1}^6 \eta_p^2 - \frac{a_3}{4} \sum_{p=1}^6 \eta_p^4 + \frac{a_4}{6} \left(\sum_{p=1}^6 \eta_p^2\right)^3 \quad (3)$$

is the local chemical free energy [21, 22], a_1 to a_4 are constants determined by the difference Δf of the free energy from Pandat shown in Fig. 2(b).

$$\begin{aligned} E_{\text{el}} = & \frac{V}{2} C_{ijkl} \bar{\epsilon}_{ij} \bar{\epsilon}_{kl} - V C_{ijkl} \bar{\epsilon}_{ij} \sum_{p=1}^6 \epsilon_{kl}^{p0} \overline{\eta_p^2(\mathbf{r})} \\ & + \frac{V}{2} C_{ijkl} \sum_{p,q=1}^6 \epsilon_{ij}^{p0} \epsilon_{kl}^{q0} \overline{\eta_p^2(\mathbf{r}) \eta_q^2(\mathbf{r})} \\ & - \frac{1}{2} \sum_{p,q=1}^6 \oint \frac{d^3 g}{(2\pi)^3} B_{pq}(\mathbf{n}) \{\eta_p^2(\mathbf{r})\}_g^* \{\eta_q^2(\mathbf{r})\}_g \\ & - V \sigma_{ij}^{ex} \bar{\epsilon}_{ij} \end{aligned} \quad (4)$$

is the elastic energy under an applied stress, σ_{ij}^{ex} , following the microelasticity theory for multi-phase coherent solids originated from Khachaturyan [23-25]. In the above equations $\overline{(\dots)}$ represents the volume average of (...), V is the total volume of the system, C_{ijkl} is the elastic moduli tensor, ε_{ij} is the elastic strain tensor, ε_{ij}^{p0} is the stress-free transformation strain tensor of variant p of the γ -phase when the corresponding order parameter assumes its equilibrium value, the same as that in reference [21]. $B_{pq}(\mathbf{n})$ is a two-body elastic interaction potential given by

$$B_{pq}(\mathbf{n}) = n_i \sigma_{ij}^{p0} \Omega_{jk}(\mathbf{n}) \sigma_{kl}^{p0} n_l \quad (5)$$

where $\mathbf{n} = \mathbf{g}/g$ is a unit vector in reciprocal space and n_i is its i th component, $\sigma_{ij}^{p0} = C_{ijkl} \varepsilon_{kl}^{p0}$ and $\Omega_{ij}(\mathbf{n})$ is a Green function tensor which is inverse to the tensor

$$\Omega_{ij}^{-1}(\mathbf{n}) = C_{iklj} n_k n_l \quad (6)$$

The elastic interaction between pre-existing and nucleating variants can be expressed as [16-18, 23]:

$$\Delta E_{el} = \sum_p \sum_q \int \frac{d\mathbf{g}}{(2\pi)^3} B_{pq}(\mathbf{n}) \{\eta_p(\mathbf{r})\}_g \{\Delta\eta_q(\mathbf{r})\}_g^* \quad (7)$$

where $\Delta\eta_q(\mathbf{r})$ is the change in the profile of the q th structural order parameter field associated with the nucleation events, and $\{\Delta\eta_q(\mathbf{r})\}_g^*$ is its Fourier transformation.

The parameters ϕ , β_{ij}^p and α characterize the magnitudes of specific strain energy and gradient energies with respect to the chemical driving force. ϕ is defined as $4G\varepsilon_0^2/|\Delta f|$ [22, 26], where G is the shear modulus, ε_0 is the largest component in the misfit strain tensor between α_2 and γ , and Δf is the chemical driving force. According to [27], the shear modulus G decreases with increasing temperature from 103.5 GPa at 298 K to 94.8 GPa at 723 K. By linear extrapolation, G is assumed to be 62.4 GPa at 1373K.

According to [28], ε_0 is 0.03. The chemical driving forces considered in this paper are 360, 450 and 540 J/mol, which correspond to 34, 43 and 51×10^6 J/m³, respectively. Therefore the ϕ value should be in the range of 4.4 to 11 under these conditions. Since the effect of elastic energy contribution has been investigated in our previous study [7], the ϕ value is set to a fixed value of 10 throughout the current study. The gradient energy coefficients β_{ij}^p are assumed to be variant independent and only diagonal terms are non-zero. According to Veeraraghavan et al [29], the energy of the disordered unfaceted α/γ interfaces typically ranges from 300 to 600 mJ/m², while the energy of the coherent or partially coherent faceted interfaces ranges from 25 to 200 mJ/m². These values yield an anisotropy parameter, $\beta_{11}(\beta_{22})/\beta_{33}$, ranging from 3 to 12. From our simulations it is found that the width to thickness ratio of the lamella obtained with $\beta_{11}(\beta_{22})/\beta_{33} = 5$ agrees with what was observed experimentally [30], thus, $\beta_{11}(\beta_{22})/\beta_{33} = 5$ (i.e., $\alpha = \beta_{11} = \beta_{22} = 5\beta_{33}$) is chosen in all simulations presented in this study. The value of ν determines whether the growth of nucleus is diffusion or interface controlled and it is assumed to be 1, which means it is mainly interface controlled. c_1 and c_2 are chosen to be 0.38 and 0.48 respectively, which are the equilibrium Al concentrations in the α_2 and γ phases, respectively, at 1100°C, this temperature is 25°C below the eutectoid temperature, which yields a chemical driving force of 450 J/mol according to the thermodynamic database used in the current study and the mean composition of the alloy is assumed to be 0.46. All the Voigt elastic constants are zero except that: $C_{11} = C_{22} = 175$ GPa; $C_{33} = 220$ GPa; $C_{12} = 88.7$ GPa; $C_{13} = C_{23} = 62.3$ GPa; $C_{44} = C_{55} = 62.2$ GPa; $C_{66} = (C_{11} - C_{12})/2$ [31]. Periodic

boundary condition is imposed along all three dimensions.

3. Simulation results

3.1. Nucleation and evolution of γ precipitates under normal stresses

Experimentally, most HIPping operations are carried out under stress levels of 300 MPa or below, e.g. 130 [32], 175 [33], 207 [12], 275 [34] and 300 MPa [35]. Stress levels of 125, 175, 275, 325, 375 and 425 MPa are considered in our simulations for different types of external stresses for this parametric study. The chemical driving force considered is 450 J/mol. In TiAl alloys, all lamellae formed during the $\alpha \rightarrow \gamma + \alpha_2$ transformation are parallel to the basal plane of the matrix phase, and the stress applied along the X $[11\bar{2}0]$ and Y $[\bar{1}100]$ will be referred to as “along” or “parallel” to the basal plane of the matrix or lamellar interface, while along the Z $[0001]$ will be referred to as “perpendicular” to the basal plane or lamellar interface. The applied stresses include compression or tension along Z (ComZ or TenZ), compression or tension along X (ComX or TenX), and isostatic pressing (ISP). The effect of shears along both 0° (the negative shear (shearN) indicates the right part of the matrix shear upward $[\bar{1}100]$ relative to the left part) and 180° (the positive shear (shearP $[1\bar{1}00]$) is the reversal of shearN) are discussed in section 3.2.

The typical spatial distribution and particle morphology of γ precipitates during the nucleation stage at reduced time $\tau = 1$ (real time $t = \tau \times 10^3$ s) under 125 MPa compression along the X $[11\bar{2}0]$ are shown in Fig. 1(a). Most precipitates formed consist of twin related variants, as marked by the arrows. When the random noise is

turned off after $\tau = 1$, some small precipitates disappear and the rest grow. Finally only some prevailing precipitates survive and form multi-twin lamellae. The typical microstructures at different stages are shown in Figs. 1(b) to (f).

Fig. 2(a) shows the evolution of composition distribution along the direction perpendicular to the laths during the transformation. The aluminum concentration of initial γ precipitates (c_i) is about 0.55. It decreases as the γ phase grows. When most matrix α transforms to γ , the aluminum concentration in the residual α decreases, and this will promote the transformation of α to α_2 . Finally, the concentrations of γ and α_2 phases approach their equilibrium values of 0.47 and 0.38, respectively. The volume fraction of α_2 decreases as γ grow with time, and finally go steady to about 0.1 after $\tau = 4.2$, in agreement with thermodynamic estimation. As shown in Fig. 2(b), the equilibrium concentration of α_2 and γ phases are determined by the common tangent of the two free energy curves, point c_1 and c_2 on line 1. However the c_i can only be determined by the equilibrium with a starting composition of α phase as the average composition of the alloy (c_a), utilizing the equal partial free energy or chemical potential of Al (defined as the derivative of the free energy to the Al concentration) in the two phase. The c_i determined this way is shown in figure 2(b), as the cutting point of line 2' with the free energy curve of γ phase. The obtained c_i agree with our phase field simulation.

Two parameters are introduced to describe the microstructure character, i.e., the twin area fraction (TAF) and volume fraction (VF). The TAF is defined as total twin boundary area over total γ/γ boundary area. VF is the ratio of total volume of

precipitates to the system volume. The TAFs are calculated at different moments from the simulation results obtained under different types and levels of external normal stresses. The results are shown in Fig. 3. The TAFs are nearly 100% in the early nucleation stage at $\tau < 0.6$, which indicates that all multi-layer precipitates consist of twin-related variants from either correlated or collective nucleation [7] under the influence of elastic interactions. When $0.6 < \tau < 1$, the TAFs decrease quickly because of the impingement of the growing precipitates that leads mostly to the formation of non-twin related boundaries. From the moment when the random noises are turned off at $\tau = 1$ to the moment when $\tau \sim 2.2$, the TAFs may increase or decrease, depending on the interplay between the dissolution of small precipitates and the growth and impingement of large precipitates. During $2.2 < \tau < 4.2$, the TAFs decrease sharply because the survived precipitates grow and impinge upon each other quickly, forming large amount of non-twin related interfaces. After $\tau \sim 4.2$, the TAFs change slowly and reach more or less constant after $\tau \sim 7$ when lamellar microstructures are fully developed, and the volume fraction of the product phase (γ) is larger than 0.9.

The TAF at the final stage of the simulation is about 70% for stress-free condition, as shown in Fig. 3(a). It is within the range of some experimental observations of 67–77% [36] and 72.4% [37]. The influence of different types of external stresses on the TAF can be seen clearly from Fig. 3(a). The TAF increases significantly under ComZ and TenX while it decreases slightly under isostatic compression, ComX and TenZ. In the end, the TAF under ComZ is 17% larger than that in the absence of external stress, while the TAF under TenZ is 5% smaller. The effect of stress amplitude on TAF

variation is shown in Figs. 3(b) to (d). Stress along the Y axis is not discussed in this work because its effects are similar to that caused by external stress along the X due to the similar strain components, Fig. 3(a).

The volume fractions (VFs) of different variant (V_1 to V_6) at $\tau = 1$ and their summation under different levels of compression (negative) and tension (positive) along X and Z, and isostatic pressing are shown in Figs. 4(a), (b) and (c), respectively. When the external stress is applied along X, the VF of every variant decreases monotonically when the stress changes continuously from compression to tension (Fig. 4(a)), while the opposite is true when the external stress is applied along Z (Fig. 4(b)). The effect of isostatic pressing on the VF is relatively small (Fig. 4(c)).

The above simulations are carried out with the chemical driving force of 450 J/mol (Δf shown in Fig. 2(b) corresponds to $c = 0.46$ and at 1100°C). It should be noted that the twin area fraction also varies with the chemical driving force [7]. The larger the chemical driving force, the smaller the TAF. The TAFs as functions of both applied stress level and chemical driving force level, 360, 450 and 540 J/mol (corresponds to different temperature for the alloy with a concentration $c = 0.46$), are shown in Figs. 5(a), (b) and (c), respectively for tension and compression along X and Z, and isostatic pressing at $\tau = 9$. It can be seen that the TAF increases significantly under tension along X (i.e., parallel to the lamellar interface) and compression along Z (i.e., perpendicular to the lamellar interface), decreases slightly under compression along X and tension along Z, and remains more or less constant under isostatic pressing. For example, under the medium chemical driving force (450 J/mol) the TAF increases

from 70% (without any external load) to 84% under 375 MPa tension along X, decreases from 70% (under no load) to 68% under 375 MPa compression along X. With the increase in chemical driving force, the TAF decreases and the effects from all the applied stresses are reduced. For example, the range of TAF under different levels of stress along X is roughly 70–88% when the chemical driving force is relatively small (360 J/mol), and decreases to 60–69% when the chemical driving force increases to 540 J/mol.

3.2. Nucleation and evolution of γ precipitates under shear stresses

The above simulations are carried out under normal stresses. We now consider shear stresses. The volume fractions of different variants and their total volume fraction as functions of levels of negative (N, $[\bar{1}100]$) and positive (P, $[1\bar{1}00]$) shear stresses at reduced time $\tau = 1$ are shown in Fig. 6. The volumes of twin related variants are very close to each other if there is no external load (Fig. 6 at 0 MPa). A negative shear is found to promote the formation of V_1 , V_4 and V_6 , and suppress V_2 , V_3 and V_5 , while a positive shear is found to have just the opposite effects. This is because the positive shear applied is along the direction of the transformation shear of V_2 and in the reverse direction of that of V_1 . At the same time, the transformation shear direction of V_3 and V_5 is closer to (60°) the positive shear applied, compared with V_4 and V_6 (120°) . Thus under positive shear, V_2 is promoted the most, V_3 and V_5 second, and V_1 is suppressed the most, V_4 and V_6 second.

The total volume fraction of γ precipitates increases while the TAFs decrease

monotonously during the precipitate growth under shear (Fig. 7), regardless of the direction of shear. The TAFs approach zero at $\tau = 3.4$ when the shear stress exceeds 375 MPa. Under large shear, the unfavored variants in any twinning pair will be consumed finally by shear transformation into its twin-related neighbor variants, and the TB between them will disappear. Therefore the thickness of the laths will be increased by shear, and the total number of the laths decreased compared to that without a stress.

Such shear deformation may occur in extrusion or rolling process in the two phase field, especially at the outer part of the extruded rod and near the surface of the rolled sheet. For grains with orientations favoring shear along lamellar plane, the γ transformation and detwinning of some variants may be promoted, forming certain textures containing fewer variants. Depending on the amplitude of applied stress, such shear induced transformation may be coupled with lamellar fragmentation and recrystallization, as observed experimentally during extrusion of a rectangular billet [38].

4. Discussions

The above simulations show that isostatic pressing promotes γ nucleation during the $\alpha \rightarrow \gamma + \alpha_2$ phase transformation, as shown in Fig. 4(c). This agrees with the experimental observation where the HIPping suppresses the dissolution kinetics of γ into α [12]. A similar pressure-assisted phase transformation has also been found in $\beta/B2 \rightarrow \gamma + \alpha_2$ transformation, where the $\beta/B2$ phase becomes unstable during HIPping

and shows a strong tendency to transform to a more closed packed $\gamma+\alpha_2$ structure [39]. These results are not surprising because the compression promotes the volume decreasing phase transformation and suppresses the volume increasing ones. The average atomic volume of γ phase ($16.3 \times 10^{-30} \text{ m}^3$) is smaller than that of the α/α_2 phase ($16.7 \times 10^{-30} \text{ m}^3$) or the $\beta/\text{B2}$ phase ($17.3 \times 10^{-30} \text{ m}^3$) in Ti-46Al-2Cr-2Mo-0.25Si-0.3B (at.%) at the HIPping temperature (about 1000 °C) [39].

The high twin area fraction shown in Figs. 3 and 5 is attributed partly to the elastic strain energy relaxation. The distributions of interaction energies between a pre-existing and nucleating precipitates in 3-D space are shown in Fig. 8. The position and size of the pre-existing plate (V_1) is shown by the dashed circle. The calculations indicate that the most negative elastic interaction energy exists between twin related variants, Fig. 8(b). Though the negative elastic interaction energy exists between ordered-domain related variants (Figs. 8(c) and (e)), the negative value regions are small compared to that of twin related variants. The minimum is $-1.0 \times 10^5 \text{ J/m}^3$, significantly smaller than that between the two twin related variants ($\text{Min}I_T = -1.2 \times 10^5 \text{ J/m}^3$).

Furthermore, the twin area fraction changes under different types of applied stresses are shown in Fig. 3, 5 and 7. The changes of interaction energy extrema under 275 MPa of different types of stresses are summarized in Table 2. All extrema show slight increases under ComZ and TenX as compared to those in the absence of external stresses, and slight decreases under isostatic pressing, ComX and TenZ. $\text{Min}I_T$ and $\text{Min}I_{OD}$ decrease while $\text{Max}I_{\text{SELF}}$ and $\text{Max}I_{PT}$ increase under shearP. The effect of

shearN on the interaction energy is just opposite to that of shearP. The relative magnitude of interaction energy extrema changes under applied stress. For example, the ratio of interaction energy minimum between twin related variants (MinI_T) and ordered-domain related variants (MinI_{OD}) under ComZ is slightly larger than that without applied stress, i.e., $(\text{MinI}_T/\text{MinI}_{OD})_{\text{ComZ}} > (\text{MinI}_T/\text{MinI}_{OD})_{\text{No}}$, as shown in Table 2. This means that the interaction between twin related variants becomes stronger relatively under ComZ, so twin related nucleation is more preferred in this condition, and therefore, higher twin area fraction will be obtained, see Figs. 3(a) and 5(b).

Based on the simulations and discussions above, the changes of twin frequency in TiAl alloys induced by the applied stress during thermo-mechanical processing or by the residual stress after that should be considered. In fully lamellar polycrystals, the twin frequency in different grains should be more or less alike theoretically if the $\alpha \rightarrow \gamma + \alpha_2$ transformation take places in the absence of external stress. While when the alloy is under thermo-mechanical processing such as flat rolling, the stress magnitude and orientation are generally different from the surface to interior in the plate during rolling [40] and so is the residual stress after the processing [41]. Therefore, the stress state is different from one grain to another, and the twin frequency of different grains after phase transformation should be diverse under these stresses.

Taking advantage of the effect that the twin frequency can be altered by external stress, the twin frequency refinement through stress control during phase transformation is possible. But the difficulty is that the grains in the matrix should

have similar orientations at least. Fortunately, the grain orientation can be modified through heat flux control during casting or some thermo-mechanical processes. For TiAl alloy bars after α field hot extrusion, most c axis of grains tend to be perpendicular to the extrusion direction [42]. TiAl alloy bars extruded through a rectangular die in the two phase field and then solution treated below α transus exhibit a dominant texture with the c axis parallel to the transverse direction [38]. After extrusion, compression perpendicular to the basal plane of the matrix applied during the $\alpha \rightarrow \gamma + \alpha_2$ phase transformation may be beneficial to improving the twin frequency.

In this simulation, the homogeneous modulus approximation is used for simplicity, i.e., both the parent and product phases have the same elastic constants. Therefore, the mismatch between them will not change under applied stress. Although with inhomogeneous modulus model [43], the soft phase will have slightly larger strain under applied stress and more accurate elastic strain energy will be obtained, acceptable results can be obtained fast and easily with the present model. The interaction energy calculation with pre-existing precipitates under applied stresses shows mainly the effect of applied stresses on the autocatalytic transformation rather than that on variant selection during nucleation. Quantitative characterization of the latter requires a combination of the phase field simulation with the free-end NEB method [44], with supercritical particles obtained in the phase field simulations being used as the end images.

The macro-strain from the transformation shear differs from system to system. In the

case of Co-Ni disordered alloy, only one variant can be formed by any combination of the three lattice shift on the same plane, different macro-strain may be induced by different combination of partial slips, depending on the composition and condition [45, 46]. While for $\alpha \rightarrow \gamma + \alpha_2$ (HCP $\rightarrow$ L1₀+D0₁₉) transformation in TiAl alloys, the stacking sequence ABABAB of HCP becomes either ABCABC or ACBACB through different shear. Atomic ordering takes place at the same time, so that one of the six variants can be formed by only one shear. Experimental study in TiAl alloys has shown that [47], surface relief can be formed during transformation. This provides direct experimental evidence in support of the displacive nature of $\alpha \rightarrow \gamma + \alpha_2$ transformation with macro-strain. In this investigation, the partial dislocations forming one variant have the same Burgers vector, therefore relatively large transformation strain is involved. Small variants may disappear due to elastic strain energy and interfacial energy minimization. With further evolution, one variant may occupy the whole lath due to the small system size along X and Y direction. While for the larger systems simulated, two or more different variants may exist within one lamella, with interfaces roughly perpendicular to the lamella formed between them, although the area fraction is small compared with the interface between different layers.

The heterogeneous nucleation from various defects may have significant influence on the final microstructure, as shown experimentally that the stacking faults formed from quenching or deformation can serve as nucleation sites for γ plates and induce fine lamellar structure [48], however, due to the complexity of deformed microstructure,

the detailed dislocation or stacking fault structure during high temperature deformation is practically unknown to us, therefore, only homogeneous nucleation is considered in the present simulations. The present simulation is valid only in the elastic deformation range, or the former deformation induced defect structure is extremely random that it can be described by the random noise used in the simulation here.

Plastic deformation can be effective in generating dislocations to promote precipitates nucleation and microstructure control [49]. Furthermore, the lamellar structure may also be tailored by controlling the dislocation distribution taking advantage of nanoscale plastic deformation produced by nano-imprinting, nano-indentation and nano-grooving [50]. However, the phase field models taking into account of the plastic deformation [51] and dislocation [15] are out of the scope of the present work and will be discussed separately.

5. Conclusions

The effects of applied stresses on γ nucleation and evolution in TiAl alloys during $\alpha \rightarrow \gamma + \alpha_2$ phase transformation have been investigated through elastic interaction energy calculations and phase field simulations. It is found that:

(1) Among the six types of interactions, the most negative elastic interaction energy exists between twin related variants in the absence of external stress. Compression perpendicular and tension parallel to the basal plane raise all interaction energies; while tension perpendicular and compression parallel to the basal plane or isostatic

pressing make them lower.

(2) The variant selection due to autocatalytic nucleation may be enhanced or suppressed by applied stresses. Compression perpendicular and tension parallel to the basal plane reduce the overall nucleation rate and promote twin formation, while isostatic pressing, compression parallel, tension perpendicular to the basal plane does the opposite. Shears parallel to the basal plane always increase the overall nucleation rate, but reduce twin boundary area fraction.

(3) The influence of external stress on twin area fraction is compared to that of chemical energy change for phase transformation. The twin area fraction increases 15% under 275 MPa compression perpendicular to the basal plane, while such increase can be obtained equally without stress by a chemical driving force reduction of 180 J/mol.

(4) For a highly textured sample where most grains have similar basal plane orientations, compression perpendicular to the basal plane applied during $\alpha \rightarrow \gamma + \alpha_2$ phase transformation is beneficial to increasing lamellar twin boundary area fraction.

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Table 1. Boundary types between different γ variants.

Group γ_I	Group γ_{II}	Relationships
V_1	V_2	twin $\longleftrightarrow 180^\circ$
V_3	V_4	twin $\longleftrightarrow 180^\circ$
V_5	V_6	twin $\longleftrightarrow 180^\circ$
ordered $\uparrow$ domain $\downarrow 120^\circ$	ordered $\uparrow$ domain $\downarrow 120^\circ$	pseudo $\swarrow \searrow$ -twin $\swarrow \searrow 60^\circ$

The horizontal double headed arrows indicate that the pairs of variants in the same row are twin related while the vertical double headed arrows indicate that ordered-domain boundary formed between any of the two variants in the same column when they are in contact. The cross of double headed arrows indicate that pseudo-twin boundary formed between any of the two variants in column I and II (from different rows) when they are in contact.

Table 2. Changes in the extrema (J/m^3) of interaction energy between a pre-existing (V_1) and nucleating precipitates (V_1 – V_6 , respectively), under 275 MPa of different types of external stresses relative to those in the absence of external stress whose values are listed in the column “NoStress”.

	ShearP	ComZ	TenX	NoStress	ISP	ComX	TenZ	ShearN
$MaxI_{SELF}$	267	59	24	142141	–12	–26	–60	–266
$MinI_T$	–250	55	24	–123692	–11	–23	–55	250
$MinI_{OD}$	–149	66	45	–105853	–13	–45	–66	150
$MaxI_{PT}$	152	67	46	106937	–13	–46	–68	–152

Fig. 1. Typical precipitate distribution and morphology at reduced time $\tau = 1$ (a) and their evolution at different stages (b - f). All multi-layer precipitates consist of twin related variants; typical configuration is marked by arrows. Different colors stand for different variants and the α/α_2 phase are made invisible.

Fig. 2. (a) The concentration profiles along the direction perpendicular to the lamellar plane at different reduced time of 1, 2.2 and 4.2, respectively. (b) The Gibbs free energy curves of α and γ phases at 1100°C. Line 1 is the common tangent of free energy curves of α and γ . The c_1 and c_2 stand for the equilibrium concentration of α_2 and γ phase. The c_a stands for the average aluminum concentration of the alloy. The line 2 is the tangent of the free energy curve of α through c_a . The line 2' which parallel to the line 2 is the tangent of free energy curve of γ . The cutting point c_1 indicates the concentration of initial precipitate of γ phase.

Fig. 3. (a) Comparison of the TAF evolution under tension, compression and zero stress. TAF evolutions under stress of different level (MPa) along Z (b) and X (c), and under isostatic pressing (d). The stress direction relative to the basal/lamellar plane is shown schematically.

Fig. 4. Volume fractions of V_1 to V_6 at $\tau = 1$ and their summation under different levels of compression (negative) and tension (positive) along X (a), Z (b) and isostatic pressing (c).

Fig. 5. TAFs under different levels of chemical driving force (J/mol) and external stress along X (a), Z (b) and isostatic pressing (c) at reduced time $\tau = 9$.

Fig. 6. The volume fractions of the six variants and their summation obtained at reduced time $\tau = 1$ as functions of external shear stresses.

Fig. 7. Comparison of TAF evolution (a) and TAF at reduced time $\tau = 5$ (b) under different shear (MPa).

Fig. 8. 3-D iso-surfaces and contour plot (central cross section) of interaction energies between a pre-existing plate V_1 (shown with arrow in (a) and outlined by the dashed lines in (a) - (f)) and nucleating precipitates (V_1 to V_6 , respectively). The interaction energy between the same variants (V_1 - V_1 in (a)) is denoted by I_{SELF} , and those between twin (V_1 - V_2 in (b)), pseudo-twin (V_1 - V_4 , V_1 - V_6 in (d) and (f)) and ordered-domain (V_1 - V_3 , V_1 - V_5 in (c) and (e)) related variants are denoted by I_T , I_{PT} and I_{OD} , respectively. The numbers of contour legend shall be multiplied by 10^5 (J/m³). The scale on the X, Y and Z axis in (a) is 10 nm, respectively.