

## **MICROMECHANICS OF STRESS-INDUCED MARTENSITIC TRANSFORMATION IN MONO- AND POLYCRYSTALLINE SHAPE MEMORY ALLOYS; Ni-Ti**

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### **Abstract**

Stress-induced martensitic transformation in single crystals and polycrystals are examined on the basis of micromechanics. A simple method to find a stress- and elastic energy-free martensite plate (combined variant), which consists of two variants, is presented. External and internal stresses preferentially produce a combined variant, to which the stresses supply the largest work upon its formation. Using the chemical energy change with temperature, the phase boundary between the parent and martensitic phases is determined in stress-temperature diagrams. The method is extended to a polycrystal, modeled as an aggregate of spherical grains. The grains constitute axisymmetric multiple fiber textures and a uniaxial load is applied to the fiber axis. The occurrence and progress of transformation are followed by examining a stress state in the grains. The stress is the sum of the external stress and internal stress. The difference in the fraction of transformation and, thus, in transformation strains between the grains causes the internal stress, which is calculated with the average field method. After a short transition stage, all the grains start to transform, and the external uniaxial stress to continue the transformation increases linearly thereafter. The external stress at the end of the transition is defined as the macroscopic yield stress due to the transformation in polycrystals. The yield stress tends to saturate, as the number of the textures increases.

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## Introduction

A traditional way to examine the mechanics of martensitic transformation is to follow the phenomenological crystallographic theory of martensitic transformation [1, 2]. We think that if only the shape deformation and crystallography of a martensite plate are concerned, the theory is useful. This is also the case when one examines the effect of an external stress on the transformation (stress-induced transformation) in a single crystal.

When martensitic transformation occurs in polycrystals, particularly in fine-grained samples, a martensite plate is blocked by a grain boundary and cannot extend to sufficient length and internal stresses accumulate and influence further transformation. The phenomenological theory cannot provide a method to evaluate these stresses. Thus, in order to examine the stress-induced transformation in a polycrystal, we have to adopt another method with which we can evaluate internal stresses.

The final objective of the present study concerns stress-induced martensitic transformation in a polycrystal and is also to obtain external stress-macroscopic strain relationships during transformation-induced deformation; transformation plasticity. In this subject, the internal stresses caused by differences in transformation strain between differently oriented grains play an essential role. Therefore, we adopt a micromechanics approach, with which the internal stresses are evaluated. While the phenomenological theory follows a finite deformation theory, the present study, thus, uses an infinitesimal theory, as usually adopted in micromechanics [3].

To be consistent, we first examine the mechanics of martensite plate formation with the infinitesimal theory (micromechanics). Next, we analyze the stress-induced martensitic transformation in a polycrystal by evaluating the internal stresses caused by the transformation. The amount, transformation strains and orientation of martensites and internal stress development depend on the orientation of constituting grains. To our best knowledge, the latter subject has not been fully studied in detail yet, except for the work by Lue, Tomota, Taya, Inoue and Mori [4]. The present study is an extension of this work, but also amends an unsatisfactory approximation adopted by it. An equi-atomic Ni-Ti alloy has been used as a model alloy to demonstrate our method, but the method is applicable to any material which undergoes martensitic transformation.

## Combined Variant

When a precipitate or martensite plate has a transformation strain,  $\varepsilon_{ij}^T$ , it, in general, produces an internal stress, thus resulting in non-vanishing elastic energy. However, a special case of causing no internal stresses and having no elastic energy is known; an infinitely extended flat plate. The plate must have the components of the transformation strains given as

$$\varepsilon_{11}^T = \varepsilon_{22}^T = \varepsilon_{12}^T = 0. \quad (1)$$

(Here, the  $X_3$ -axis is perpendicular to the flat plane.) The other components can take any values [5]. This case holds when the elastic constants of the plate differ from those of the matrix and is valid for an anisotropic material. Condition (1) is also written as

$$\det(\varepsilon_{ij}^T) = 0, \quad \varepsilon_{ii}^T \varepsilon_{jj}^T - \varepsilon_{ij}^T \varepsilon_{ij}^T \leq 0, \quad (2)$$

when the transformation strain is referred to a general coordinate system.

The transformation strain in Ni-Ti calculated from  $a_0=0.3015\text{nm}$  (parent phase, B2) and  $a=0.2889\text{nm}$ ,  $b=0.4120\text{nm}$ ,  $c=0.4622\text{nm}$  and  $\beta=96.8^\circ$  (martensite phase), as usually adopted by studies using the phenomenological theory [6,7], does not satisfy condition (2). Thus, we seek combinations of two variants ( $\varepsilon_{ij}^T(M)$  and  $\varepsilon_{ij}^T(N)$ ) among 12 variants, so that the average transformation strain

$$\overline{\varepsilon_{ij}^T} = f_M \varepsilon_{ij}^T(M) + f_N \varepsilon_{ij}^T(N), \quad f_M + f_N = 1, \quad (3)$$

satisfies condition (2). Here,  $\varepsilon_{ij}^T(M)$  refers to the transformation strain of the M-th variant (M=1-12). The possible combinations are classified into group A and group B. Group A has  $f_M=0.277$ , while group B has  $f_M=0.311$ . Each group has 24 crystallographically equivalent combinations. Thus, in total, there are 48 combinations, called combined variants. A combined variant in group A has the average transformation strains of

$$\overline{\varepsilon_{ij}^T} = \begin{pmatrix} 0.0212 & -0.0246 & -0.0422 \\ -0.0246 & 0.0212 & 0.0188 \\ -0.0422 & 0.0188 & -0.0418 \end{pmatrix}. \quad (4a)$$

The corresponding strains of a variant in group B are

$$\overline{\varepsilon_{ij}^T} = \begin{pmatrix} -0.0222 & -0.0120 & -0.0422 \\ -0.0120 & 0.0212 & 0.0248 \\ -0.0422 & 0.0248 & 0.0017 \end{pmatrix}. \quad (4b)$$

The average transformation strains of the other variants are given by the symmetric operation of (4a) and (4b).

Also, the good (stress free) matching on the interface of the M-th and N-th variants requires

$$\det(\delta\varepsilon_{ij}^T) = 0. \quad (5)$$

Here,  $\delta\varepsilon_{ij}^T = \varepsilon_{ij}^T(M) - \varepsilon_{ij}^T(N)$ . This requirement reduces the number of trial combinations among 12 variants; practical advantage. Since  $\delta\varepsilon_{ii}^T=0$ , such an inequality as that in (2) is not required in (5).

Equations (4a) and (4b) refer to the crystallographic axes of the parent phase, [100], [010] and [001]. Because of the uncertainty in the last digit of the lattice parameters in the parent and martensite phases, we should be aware that the last digit in the transformation strains has some ambiguity.

### Transformation in a Single Crystal under External Stress

If a martensite plate of a combined variant extends all across the section of a single crystal, no internal stress is induced. Accordingly, no increase in elastic energy occurs. Thus, the occurrence and continuation of stress-induced martensite follows the criterion

$$V + F^M + W^D = F^A. \quad (6)$$

Here,  $V$  is the change in the potential energy of an external loading device by transformation and is given by

$$V = -\sigma_{ij}^A \overline{\varepsilon_{ij}^T}, \quad (7)$$

where  $\sigma_{ij}^A$  is the external stress.  $F^M$  is the chemical free energy of the martensite phase and  $F^A$  that of the parent phase.  $W^D$  is the energy dissipation accompanied by the progress of the transformation. (Its presence causes temperature hysteresis in the transformation.) All the quantities in (6) refer to a unit volume of the parent phase.

If the left hand side in (6) is larger than the right hand side, the transformation does not proceed. Thus, the combined variant, most favored by the external stress, is the one which makes  $V$  lowest. For uniaxial loading along a unit vector  $\ell_i$ , (7) is equivalent to

$$V = -\sigma^A \ell_i \ell_j \overline{\varepsilon_{ij}^T}. \quad (8)$$

Here  $\sigma^A$  is a uniaxial stress;  $\sigma^A > 0$  for tension and  $\sigma^A < 0$  for compression. Thus, the most favored combined variant is the one which makes  $\ell_i \ell_j \overline{\varepsilon_{ij}^T}$  maximum in tension and minimum in compression.

$$\Delta F^C = F^A - F^M, \quad (9)$$

is a decreasing function of temperature.  $W^D$  is not so sensitive to temperature. For lack of reliable data, we will use the values used by [4] ---Ni-Ti-Cu---for these quantities. However, the possible error caused by this tentative assignment of  $\Delta F^C$  and  $W^D$  can be easily fixed and is a secondary concern in the present study. For example, the lines in Fig. 1 would be shifted to a higher or lower temperature side, when different functional forms of  $\Delta F^C$  and  $W^D$  are assigned.

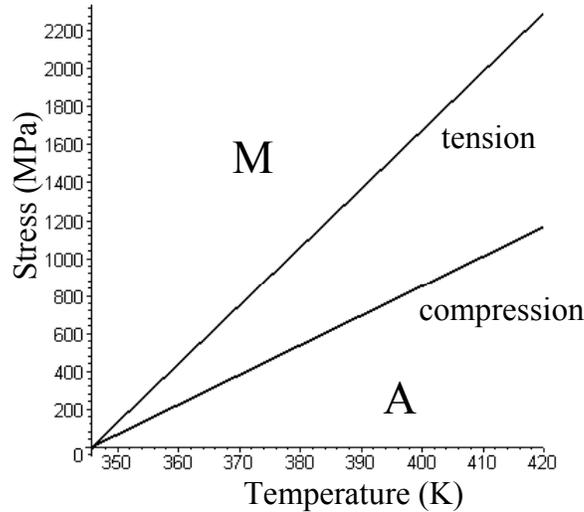


Figure 1: Stress-temperature relationship to induce martensitic transformation in a single crystal loaded along [001]: A is the parent phase domain, and B the martensite domain.

Figure 1 shows the magnitude of stress to induce the transformation as a function of temperature under uniaxial loading along [001], using the criterion given by (6). The magnitude of the tensile stress is larger than that of the compressive stress. This result is understood by considering the smallest and largest principal strains ( $\lambda_1, \lambda_3$ ) and their directions of the average transformation strains of the combined variants. Group A has the principal direction of  $\langle 0.427, 0.076, 0.901 \rangle$  for  $\lambda_1 = -0.0634$  and that of  $\langle 0.581, 0.388, 0.715 \rangle$  for  $\lambda_3 = 0.0640$ . In group B, these directions are  $\langle 0.621, 0.080, 0.780 \rangle$  for  $\lambda_1 = -0.0545$  and  $\langle 0.625, 0.445, 0.641 \rangle$  for  $\lambda_3 = 0.0552$ . Thus, the magnitude of a compressive stress becomes smaller than that of a tensile stress, when the same amount of martensite is formed under [001] loading. There are, of course, other directions where this observation is reversed. For example, when loading is along [111], tension induces the transformation at a lower stress than compression.

It is clear that the magnitude of a stress to induce the transformation becomes smallest when uniaxial loading is directed to the principal direction of the average transformation strains. In this conjunction, a variant belonging to group A plays a more important role. This is because the magnitude of the non-zero principal strains in the group A combined variants is larger than that of those in the group B combined variants. Also, the difference in the smallest stress between simple tension and simple compression is small, since the transformation accompanies a small volume change, less than 0.0007.

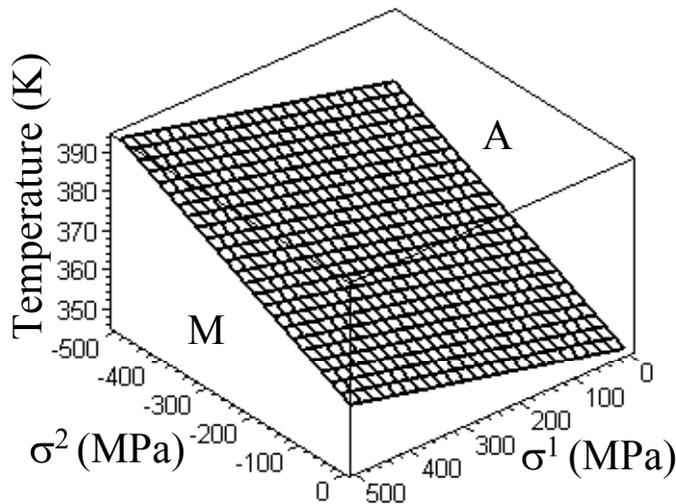


Figure 2: Phase boundary separating the parent and martensite phases in a single crystal biaxially loaded along [100] (tension,  $\sigma^1$ ) and [010] (compression,  $\sigma^2$ ).

The two lines in Fig.1 are the phase boundaries separating the parent and martensite phases during forward transformation. In the most general loading, the phases are separated by a boundary in a multi-dimensional space. As another simple example, the boundary is shown in Fig. 2 under biaxial loading; tension along [100] and compression along [010]. In this case, an identical combined variant is formed by those stresses. However, the combined variant formed by loading changes, in general, from a stress state to another on the boundary, on which the transformation occurs.

### Transformation in a Polycrystal under Loading

A martensite plate cannot extend to sufficient length in a polycrystal. A plate is blocked by the boundary of a grain, where the plate is formed. Thus, the internal stress develops. The

internal stress not only retards its growth but also influences the occurrence and progress of other martensite plates in the same and other grains. Since the elastic energy is induced by transformation in a polycrystal by transformation, the criterion for the occurrence and progress of the transformation is now written as

$$\delta(V + E + F^M + W^D) = \delta F^A, \quad (10)$$

instead of (6). Here,  $E$  is the elastic energy increase due to the transformation caused by the occurrence of the internal stresses. Equation (10) is written in a variational form for the variation,  $\delta f$ , (increase) in the fraction of the martensite formed in a grain. To facilitate the involved computation, we adopt two approximations. First, a grain is assumed spherical. Second, the uniform eigenstrain,

$$\varepsilon_{ij}^* = f \overline{\varepsilon_{ij}^T}, \quad (11)$$

is assigned to a grain where the martensite with the transformation strain,  $\overline{\varepsilon_{ij}^T}$ , occurs to the fraction of  $f$  ( $0 \leq f \leq 1$ ). With these approximations, the increase in the elastic energy in a particular grain is written as

$$E = -(\sigma_{ij}^S + \overline{\sigma_{ij}}) \overline{\varepsilon_{ij}^T} \delta f, \quad (12)$$

when the amount of the martensite increases by the fraction  $\delta f$ .  $\sigma_{ij}^S$  is the stress in the grain (self-stress), when only it transforms to  $f$  and other grains are hypothetically untransformed.  $\overline{\sigma_{ij}}$  is the stress caused by the transformation in the other grains.  $\sigma_{ij}^S$  is calculated by

$$\sigma_{ij}^S = C_{ijkl} (S_{klmn} \varepsilon_{mn}^* - \varepsilon_{kl}^*), \quad (13)$$

where  $C_{ijkl}$  is the elastic constant and  $S_{klmn}$  the Eshelby tensor.  $\overline{\sigma_{ij}}$  is calculated by

$$\overline{\sigma_{ij}} = -\sum_I g(I) \sigma_{ij}^S(I), \quad (14)$$

using the average field method [8,9]. Here,  $g(I)$  is the volume fraction of the I-th grain, which have an identical orientation and  $\sigma_{ij}^S(I)$  is the self-stress of these grains.  $\delta V$  is given as

$$\delta V = -\sigma_{ij}^A \delta \varepsilon_{ij}^* = -\sigma_{ij}^A \overline{\varepsilon_{ij}^T} \delta f. \quad (15)$$

Thus, (10) is rewritten as

$$-\left( \sigma_{ij}^A + \sigma_{ij}^S(J) - \sum_I g(I) \sigma_{ij}^S(I) \right) \overline{\varepsilon_{ij}^T}(J) + W^D = \delta F^C, \quad (16)$$

for the J-th grain which has the combined variant with the transformation strain,  $\overline{\varepsilon_{ij}^T(J)}$ . The macroscopic strain,  $\langle \gamma_{ij} \rangle$ , due to the transformation is simply given as

$$\langle \gamma_{ij} \rangle = \sum_I g(I) f(I) \overline{\varepsilon_{ij}^T(I)}, \quad (17)$$

where  $f(I)$  is the fraction of the transformation in the I-th grains. For a given set of differently oriented grains with respective  $g(I)$ , we can obtain  $f(I)$  as a function of  $\sigma_{ij}^A$  from (16). In turn, we can calculate the macroscopic strain as a function of  $\sigma_{ij}^A$  with (17). For example, we can construct a stress-strain curve in a simple tension. However, we have to find a combined variant which is formed in a group of grains with the same orientation with respect to the tensile axis. This procedure is rather involved. Thus, we have adopted a practical method, as will be demonstrated in the following sub-section.

### Double fiber textured specimen.

We will show how to use the above method in a simple problem as uniaxial loading. A tensile stress is applied to a specimen which has double fiber textures, axisymmetric with respect to the tensile axis. The grains in texture 1 have the tensile direction of  $\langle 0.5836, 0.3370, 0.7388 \rangle$ , while the grains in texture 2 have that of  $\langle 0.3948, 0.1058, 0.9127 \rangle$ .  $g(1)=0.492$  and  $g(2)=0.508$ . For simplicity, the specimen is assumed elastically isotropic; the shear modulus = 23GPa and the Poisson ratio = 0.43. Deformation temperature is 361 K.

#### (1) Start of transformation

The grains in texture 1 start to transform at a lower stress than those in texture 2. This initial stress,  $\sigma^0$ , is found, by using criterion (6) and (7) with  $\sigma_{ij} = 0$  and giving  $\sigma_{ij}^A = \sigma^0 \ell_i \ell_j$ . After these grains transform to the fraction of  $f$ , a grain in texture 1 has the self-stress of

$$\sigma_{ij}^S = \begin{pmatrix} \sigma_{11} & \sigma_{12} & \sigma_{13} \\ \sigma_{21} & \sigma_{22} & \sigma_{23} \\ \sigma_{31} & \sigma_{32} & \sigma_{33} \end{pmatrix}. \quad (18)$$

Here, each component is proportional to  $f$  and referred to the coordinate in which the X<sub>3</sub>-axis is along the tensile direction. Because of the rotational symmetry of the grains, belonging to one type of texture, the average of (18) over all the grains is calculated as

$$\overline{\sigma_{ij}^S(1)} = \begin{pmatrix} (\sigma_{11} + \sigma_{22})/2 & 0 & 0 \\ 0 & (\sigma_{11} + \sigma_{22})/2 & 0 \\ 0 & 0 & \sigma_{33} \end{pmatrix}, \quad (19)$$

when referred to the above coordinate system. Thus, when only texture 1 transforms, the average stress is written as

$$\overline{\sigma_{ij}} = -g(1) \overline{\sigma_{ij}^S(1)}. \quad (20)$$

The transformation in the grains in texture 1 proceeds, as long as

$$-\left(\sigma_{ij}^A + \sigma_{ij}^S(1) - g(1)\overline{\sigma_{ij}^s(1)}\right)\overline{\epsilon_{ij}^T(1)} + W^D = \Delta F^C \quad (21)$$

is satisfied.

#### (2) Start of transformation in texture 2

As the fraction of transformation in texture 1 increases, the stress in the grains in texture 2 increases, partly due to an increase in the external stress and partly due to the internal stress from the transformed grains in texture 1. Eventually, the transformation in texture 1 is initiated.

The condition for the grains in texture 2 to start transformation is given by solving

$$-\left(\sigma_{ij}^A - g(1)\overline{\sigma_{ij}^s(1)}\right)\overline{\epsilon_{ij}^T(2)} + W^D = \Delta F^C. \quad (22)$$

Equation (22) must hold together with (21). The combined variant for the grains in texture 2 is the one which minimizes the first term of the left-hand side of (22).

#### (3) Progress of transformation in textures 1 and 2

The two conditions must be satisfied. These are

$$-\left(\sigma_{ij}^A + \sigma_{ij}^S(1) - g(1)\overline{\sigma_{ij}^s(1)} - g(2)\overline{\sigma_{ij}^s(2)}\right)\overline{\epsilon_{ij}^T(1)} + W^D = \Delta F^C \quad (23)$$

for texture 1 and

$$-\left(\sigma_{ij}^A + \sigma_{ij}^S(2) - g(1)\overline{\sigma_{ij}^s(1)} - g(2)\overline{\sigma_{ij}^s(2)}\right)\overline{\epsilon_{ij}^T(2)} + W^D = \Delta F^C \quad (24)$$

for texture 2.

$\sigma_{ij}^S(1)$  and  $\overline{\sigma_{ij}^s(1)}$  are linearly related to  $f(1)$ , and  $\sigma_{ij}^S(2)$  and  $\overline{\sigma_{ij}^s(2)}$  to  $f(2)$ . By solving these linear equations,  $f(1)$  and  $f(2)$  are obtained as a linear function of  $\sigma^A$ . The macroscopic strain along the tensile direction is easily obtained as a function of  $\sigma^A$ .

#### (4) Completion of transformation in texture1 and progress in transformation in texture 2

The tensile stress for texture 1 to completely transform is obtained as the stress at which  $f(1)$  reaches 1. After this stage, only texture 2 proceeds to transform. The stress reaches a maximum when  $f(2)$  becomes 1.

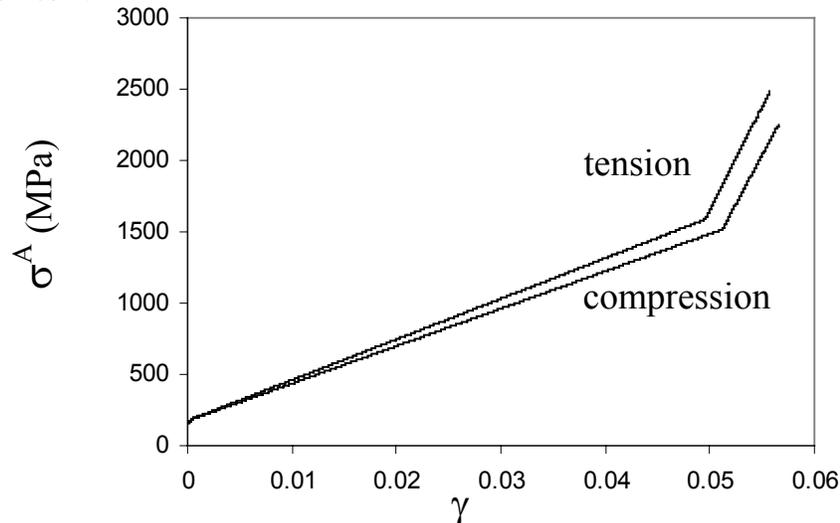


Figure 3: Stress  $\sigma^A$ -strain  $\gamma$  curves of a double textured specimen at 361K.

In Fig. 3, the tensile stress  $\sigma^A$ -tensile strain  $\gamma$  diagram, obtained by the above method, is drawn. The similar curve for compression is also given. As will be discussed below, the very beginning of the curves is not linear in a strict sense. However, we can clearly pin down the stress which is needed for transformation in all grains to undergo, as will be discussed below. This stress can be defined as the macroscopic yield stress,  $\sigma_M$ , due to transformation.

#### Initial part of transformation

In Fig. 4, the initial parts of flow curves in Fig.3 are enlarged. For example, let us examine the tensile loading. Until  $f(l)$  reaches 0.015 when  $\sigma^A$  becomes 189MPa and  $\gamma = 0.0005$ , only the grains in texture 1 transform. Below this stress, the flow curve is linear. After this stage, both textures transform, resulting in another linear hardening. Since the strain at the start of the latter linear hardening is so small that this point can be taken as the macroscopic yield stress,  $\sigma_M$ , due to transformation (transformation plasticity).

Even when a multiple textured specimen is examined,  $\sigma_M$  is clearly defined in a similar manner. For example, when the grains in a specimen with 16 textures all start to transform, the macroscopic strain is only 0.0016. (The fiber axis directions are approximately evenly distributed in the standard triangle; [001]-[101]-[111].) The fraction of transformation in the grains which have transformed most is only 0.042. The tensile stress in this point is  $\sigma_M$ . Before this stage is reached, the flow curve consists of piecewise linear curves, which covers strain negligibly smaller than the strain when all the grains complete transformation.

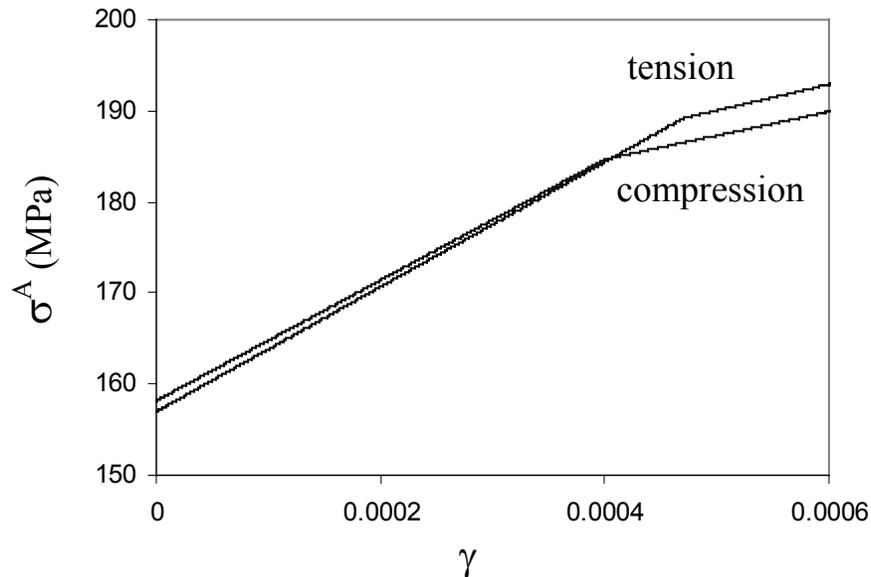


Figure 4: Enlargement of the initial part of the curves in Fig. 3.

#### Macroscopic yield stress, $\sigma_M$

We have examined similar flow curves in multiple textured specimens. We expect that as the number of textures increases, grains in a specimen becomes more randomly distributed. In Fig. 5, the macroscopic yield stress is plotted against the number of textures, thus examined. As the number increases, the yield stress increases, but certainly tends to saturate. It is necessary to determine the saturation stress, by increasing the number of textures.

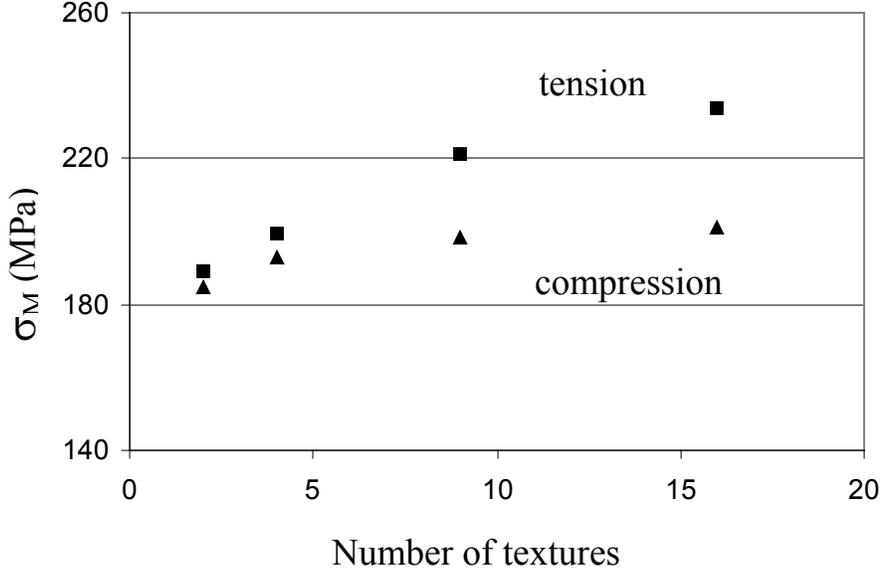


Figure 5: Macroscopic yield stress  $\sigma_M$  due to transformation against the number of textures.

## Discussion

First, we will compare the present method with that using the phenomenological theory. In the analysis of single crystal deformation, the two treatments are similar, except for difference caused by that between finite and infinitesimal deformation theories. If only the shape deformation and crystallography are concerned, the phenomenological theory gives more accurate prediction. However, the calculation involved in infinitesimal theory is much easier and less time-consuming. This is one reason why the present method adopted the latter. Another reason is that internal stress which develops in polycrystals and influences concurrent and subsequent transformation is neatly assessed in the present method. It must be noticed that the present method does not require the detailed information on the habit plane and shear strain direction. These are needed to determine the stress to induce transformation and a particular combined variant under loading in the phenomenological theory, as seen in a recent paper by Gall, Sehitoglu, Chumlyakov and Kireeva [10]. On the contrary, the present method only requires the magnitude of  $\ell_i \ell_j \overline{\varepsilon_{ij}^T}$  under uniaxial loading. This is invariant, so that the most favored combined variant and the stress to induce this variant are very easily determined under uniaxial loading. Of course, the habit plane normal and shear deformation direction are easily found in the present method, using the geometrical property of the representative quadratic of  $\overline{\varepsilon_{ij}^T}$  [11]. Further, finding two variants to form a stress-free combined variant is straightforward, using (2) - (4).

Next, we would like to mention that the deficiency in the previous work [4] has been remedied by the present study. The deficiency is demonstrated in a single textured specimen under uniaxial loading. Because of a crude approximation, the previous work could not take into account the internal stress in this case, while the present study has duly calculated the internal stress. Roughly speaking, the internal stress in this case is caused by difference in the

transformation strain along lateral directions between grains belonging to the same texture, even though the tensile component is all identical. Because of the development of the internal stress, the flow curve of a single textured specimen should develop linear hardening, while the previous study predicts the constant flow stress during the progress in transformation.

Finally, our examination of all 48 combined variants (groups A and B) will be discussed in conjunction with past studies. The finding of these variants is not new. In a phenomenological theory analysis, Matsumoto, Miyazaki, Otsuka and Tamura found two values of  $x$  [6], which plays the same role as  $f_M$  in (3). One is 0.271, which apparently corresponds to  $f_M=0.277$ . The other value was discarded as a result of experiments. Other papers investigating mechanics of transformation in Ni-Ti just employed the shape deformation for the case of  $x=0.271$  [10, 12,13]. However, we have examined all the combined variants in the present study,  $f_M=0.277$  and  $f_M=0.311$ . This is because we cannot see any reason to rule out the role played by the combined variants with  $f_M=0.311$ .

## Summary

The structure of a twinned martensite plate in Ni-Ti is examined on the basis of micromechanics. Combined with thermodynamics and energy dissipation accompanied by the movement of martensite/parent phase interfaces, the stress to induce martensitic transformation is determined as a function of temperature in single crystals. The progress of the transformation is also studied in polycrystals. A grain is modeled as a spherical inclusion and a specimen is assumed to have fiber textures. Further, a grain is assumed to have a uniform eigenstrain, which is the fraction of the transformation times the transformation strain of the plate like martensite, most favored by stresses. The stresses are the sum of external uniaxial stress, parallel to the fiber axis, and internal stress, which is evaluated with the Eshelby theory and the average field method. The external uniaxial stress, which is required to initiate the transformation in all the grains, is identified as the macroscopic yield stress due to the transformation. The macroscopic yield stress increases, as the number of the texture increases, but tends to saturate. The compressive yield stress is smaller than the tensile yield stress.

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