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NUMERICAL CHARACTERIZATION OF THE TRANSFORMATION DYNAMICS IN SHAPE MEMORY ALLOY

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ABSTRACT

Numerical results on the transformation states of shape memory alloy (SMA) wire type samples under periodic thermal loading are reported. The mathematical model used for computation is based on a Landau-Ginzburg free energy density and nonequilibrium thermodynamics at the microscopic scale. A three-dimensional nonlinear finite element model has been developed to solve the initial boundary value problem. The model consistently captures the essential properties of the multivariant phase transformation dynamics. We analytically and computationally show how some of the important parameters, such as the lattice transformation strain, thermal loading rates as well as their amplitudes influence the austenite to martensite transformation.

Keywords: Phase transformation, Lattice, Kinetics, Gibbs free energy, Finite element.

INTRODUCTION

Shape Memory Alloys (SMAs) have various applications in transducer devices and in control of dynamical systems. However, despite the availability of numerous phenomenological models, design and analysis involving new SMA structures remain to be an extremely difficult task. Current phenomenological models vary drastically depending on material constants, conditions at which experiments are done, effective dimensions and size of the sample and the nature of thermo-mechanical loading. A better mathematical understanding of the origin of SMA microstructures under deformation has been developed over the past few decades (see e.g. [1-7]), and computational challenges in the application of related theories to modelling of SMA structures have been reviewed recently in [8]. It has been pointed out in [8] that the sequence of minimization processes over a set of austenite-martensite energy wells gives rise to the observed microstructure that has precise analytical description. Furthermore, it is known that at the macroscopic scale, these local

microstructural deformations, which are equivalent to transformations of Austenite (A) to variants of Martensite (M) and the reverse transformations, give rise to the shape-memory effect. Nevertheless, up to date little is known on how the three-dimensional and time-dependent dynamics of the real-scale SMA devices could be studied by linking the above theoretical framework to practical engineering applications. The main difficulties along this avenue are largely numerical in nature. They are aggravated by the underlying interdependence of following two aspects: (1) the sequence of energy minimizations may not lead to the global minimum under the prescribed dynamic loading and macroscopic constraints (the discrete version of the process is known as *quasi-convexification* of the original non-convex minimization problem) and (2) the conditions required to match the spatial discretization with the evolved interfaces dynamically lead to the necessity of the efficient implementation of a mesh adaptation scheme within the variational framework (e.g. adaptive finite elements).

A major challenge in resolving the phase transformation (PT) dynamics in 3D is connected with the fact that most of the available phenomenological models are applicable at the macroscopic and mesoscopic scales. Indeed, a macroscopic model requires the description of the 3D geometry and variational minimization of an appropriate potential involving the thermoelastic energy and the energy due to the dynamic self-accommodation of the martensitic variants. Whereas a mesoscopic model, such as the Falk-Kanopka free energy model [9], requires the description of the free energy as a function of the strain invariants of a single crystal and its point group symmetry. The strain field in such models is beyond the resolution of bain strain. A more detailed description of the PT requires a microscopic model, such as the model recently proposed by Levitas and Preston [10], where the invariance of the free energy with respect to the crystallographic point group symmetry is preserved but the discontinuous nature of the deformation across the individual habit planes is introduced. Such information is essential to describe the microstructural evolution. A model similar to those reported in [10-11] but with additional consistency condition on partitioning the chemical energy and the transformation energy in stress-space has been developed as part of the present work.

In the present paper we apply the Landau-Ginzburg theory to study first-order phase-field evolution applicable to SMAs (see e.g. [9,12-14]) and discuss its link with the microstructural compatibility [15] under the framework of macroscopic continuum deformation theory. A major advantage of the proposed approach is that the requirement of mesh adaptation scheme mentioned above in (2) is relaxed when we interpolate the phase field itself (represented by the order parameter η). However, a consistent evolution criterion needs to be derived from the mathematical description of the microstructure. In this paper, we outline possible steps to achieve this. Among other major issues we are concerned with is the problem of how to describe the energy wells $G(\eta)$ in case of multivariant PT without destroying the essential properties of frame-indifference and material symmetry. Further conditions that need to be imposed are related to the uniqueness of the evolution asserting that only a single variant is formed at a given point in space and time. A systematic approach to impose such conditions can be found in details in [10-11]. The approach allows to study, at least in principle, the evolution of the ordered states, or equivalently the A-M compatible microstructure, that is governed by a first-order kinetics based on the Ginzburg-Landau model [14].

However, in order to complete the description of this micro-meso-macro model, the value of the spontaneous strains and certain material parameters are to be estimated from a molecular dynamic simulation. The underlying step here is to obtain, on one hand the atomistic reordering without diffusion and the resulting free energy $G(\eta)$ (or the related constants and spontaneous strain at equilibrium temperature) based on the Born-Oppenheimer approximation, and on the other hand express this free energy (equivalently $G(F)$) according to the Cauchy-Born hypothesis. Here F is the deformation gradient. The order parameters (η), therefore, describe the phenomenological link between the above two dynamics. This approach deviates from the notion of phase fractions developed in [16,17] and provides a consistent framework for microstructure evolution in multivariant situations. Given the problem complexity, it should not come as a surprise that up to date there is no robust mathematical model and well-defined computational steps that can deal with general time-dependent dynamics of SMA over a range of deformation, stress and temperature. However, rigorous analytical statements on the stability and bounds of the numerical solutions have been drawn (e.g. [18-19]).

LANDAU-GINZBURG FREE ENERGY MODEL FOR MULTIVARIANT PHASE TRANSFORMATIONS

We approximate the free energy density in the Landau theoretic framework by satisfying essential properties of phase transformations. In this framework, we view a polynomial structure in such a way that the stability of the austenitic phase (A) and martensitic variants (M_j), non-minimum diffusion barrier and nucleation can be described in the stress-temperature space. The details are given in [20]. While extending this approach to multivariant cases, several nontrivial difficulties arise due to the inherent nonlinear interplay between different variant contributions.

Let us first consider a single variant of martensite and single order parameter $\eta \in [0,1]$. First we define the Gibbs free energy density in stress-temperature space (σ, θ) as

$$G(\eta) = -\sigma : \lambda : \sigma / 2 - \sigma : \varepsilon_t \varphi(\eta) + f(\theta, \eta), \quad (1)$$

where λ is the constant fourth-rank elastic compliance tensor, ε_t is the transformation strain tensor at the thermodynamic equilibrium of the martensite (obtained from crystallography), $\varphi(\eta)$ is a monotonic function with $\varphi(0) = 0$ indicating stable A phase and $\varphi(1) = 1$ indicating stable M phase. $f(\theta, \eta)$ is the chemical part of the free energy with property: $f(\theta, 1) - f(\theta, 0) = \Delta G^0(\theta)$, where ΔG^0 is the difference between the thermal parts of the Gibbs free energy density of the M and A phases, which can be obtained indirectly from experiments through the relation [21]

$$\Delta G^0 = -\Delta s_e(\theta - \theta_e) - \Delta c\theta[\ln(\theta / \theta_e) - 1] - \Delta c\theta_e, \quad (2)$$

where Δc is the difference between the specific heat of the phases, Δs_e is the jump in the specific entropy at the equilibrium temperature (θ_e). The objective is to obtain the unknown functions φ and f by satisfying their properties mentioned above and the conditions of extremum of the energy for existence of equilibrium of A and M phases: $\partial G / \partial \eta = 0$ at $\eta = 0, 1$. Thus, the main step in the subsequent formulation that follows [20] is to assume the extremum in the form

$$\frac{\partial G}{\partial \eta} = G_0 \eta(\eta - 1)(\eta - \eta_b), \quad (3)$$

so that the roots $\eta = 0, 1$ correspond to the extrema and the root $\eta = \eta_b$ represents the $A \leftrightarrow M$ PT barrier. Here, $G_0 = a \Delta s_e (\theta_e - \theta_c)$, where $a < 0$ is constant, independent of stress and temperature, but possibly dependent on experimentally measured values that include the effect of dislocation and creep in polycrystals. Integrating Eq. (3) and imposing the combined properties of φ and f stated earlier as

$$G(\sigma, \theta, 0) - G(\sigma, \theta, 1) = \sigma : \varepsilon_t - \Delta G^\theta, \quad (4)$$

we get

$$\eta_b = -6\sigma : \varepsilon_t + 6\Delta G^\theta + G_0 / 2. \quad (5a)$$

After some algebraic steps, we explicitly obtain the polynomial structure in $\varphi(\eta)$ as

$$\varphi(\eta) = 3\eta^2 - 2\eta^3 \quad (5b)$$

and then $f(\theta, \eta)$. For $A \rightarrow M$ PT, the criteria for the loss of stability of A phase is $\partial^2 G / \partial \eta^2 \leq 0$ at $\eta = 0$. Similarly, for $M \rightarrow A$ PT, the criteria for the loss of stability is $\partial^2 G / \partial \eta^2 \leq 0$, at $\eta = 1$. This procedure is extended to the multivariant case in the next section.

CRYSTALLOGRAPHIC VARIANTS OF MARTENSITE

In order to model realistic situations and macroscopic samples of SMA, it is essential to incorporate the effects of (1) martensitic variants (M_k), (2) thermal strain, (3) unequal compliances across the interfaces and the resulting inhomogeneity, (4) polycrystals with several grain boundaries. In this paper, we focus on cubic-tetragonal transformations. In this case, there are three variants of martensite according to the point group of crystallographic symmetry. The Gibbs free energy density, thus, should possess the invariance properties. In the mathematical model, this can be cross-checked by interchanging the variant indices (k). We assume the same order of variation in the compliance tensor and the thermal expansion

tensor as in $\varphi(\eta)$. The Gibbs free energy density for cubic-tetragonal transformation having three variants $k=1,2,3$ is expressed as

$$G(\sigma, \theta, \eta) = -\sigma : \left[\lambda_0 + \sum_{k=1}^N (\lambda_k - \lambda_0) \varphi(\eta_k) \right] : \sigma / 2 - \sigma : \sum_{k=1}^N \epsilon_{tk} \varphi(\eta_k) \\ - \sigma : \left[\epsilon_{00} + \sum_{k=1}^N (\epsilon_{0k} - \epsilon_{00}) \varphi(\eta_k) \right] + \sum_{k=1}^N f(\theta, \eta_k) + \sum_{i=1}^{N-1} \sum_{j=i+1}^N \bar{F}_{ij}(\eta_i, \eta_j) \quad (6)$$

where λ is the second-order forth-rank compliance tensor (λ_0 is for A phase), $\epsilon_{0k} = \alpha_k(\theta - \theta_e)$, α_0 and α_k are thermal expansion tensor in the A and M_k phase respectively. N is the total number of variants. $\bar{F}_{ij}(\eta_i, \eta_j)$ is an interaction potential required to preserve the frame-invariance of G with respect to the point group of symmetry and uniqueness of the multivariant PT at a given material point. The description of PT can now be generalized with three sets of ordered states: $\bar{0} = \{0, \dots, \eta_k = 0, \dots, 0\}$, $\bar{1} = \{0, \dots, \eta_k, \dots, 0\}$ and $\bar{\eta}_k = \{0, \dots, \eta_k, \dots, 0\}$. The extremum property of the free energy density requires

$$\frac{\partial G}{\partial \eta_k} = G_0 \eta_k (\eta_k - 1)(\eta_k - \eta_{bk}) = 0, \quad \eta_k = \bar{0}, \bar{1} \quad (7)$$

$$\frac{\partial^2 G}{\partial \eta_k^2} \leq 0, \quad \eta_k = \bar{0} \quad (A \rightarrow M_k) \quad (8)$$

$$\frac{\partial^2 G}{\partial \eta_k^2} \leq 0, \quad \eta_k = \bar{1} \quad (M_k \rightarrow A). \quad (9)$$

The transformation energy associated with $A \rightarrow M_k$ is

$$G(\sigma, \theta, \bar{0}) - G(\sigma, \theta, \bar{1}) = \sigma : \epsilon_{tk} - \Delta G^0. \quad (10)$$

Combining Eqs.(7) and (10) with similar steps described in Sec.2 and discussed in detail in [20], we get the energy barrier across the 0-k transformation surface as

$$\eta_{bk} = -6\sigma : \epsilon_{tk} - \sigma : (\lambda_k - \lambda_0) : \sigma / 2 - 6\sigma : (\epsilon_{0k} - \epsilon_{00}) + 6\Delta G^0 + G_0 / 2. \quad (11)$$

Following the steps discussed in [11], we arrive at the frame-invariance

$$G(\eta_i, \eta_j) = G(\eta_j, \eta_i) \quad (12)$$

as well as symmetry preserving polynomial structure of the interaction potential of the form

$$\bar{F}_{ij} = \eta_i \eta_j (1 - \eta_i - \eta_j) [B\{(\eta_i - \eta_j)^2 - \eta_i - \eta_j\} + D\eta_i \eta_j] + \eta_i^2 \eta_j^2 (\eta_i Z_{ij} + \eta_j Z_{ji}) \quad (13)$$

where B, D are material constants to be estimated from experiments, $Z_{ij} = Z_{ji}$ (B, D, σ) are known. Conditions for the transformation energy associated with $M_i \leftrightarrow M_j$ are already satisfied in this approach. Finally, the uniqueness of multi-variant PT at a material point is now imposed, which leads to

$$\frac{\partial}{\partial \sigma} \left[\sum_{k=1}^{N-1} \sum_{j=k+1}^N \frac{\partial \bar{F}_{kj}}{\partial \eta_k} \right] = \sum_{k=1}^3 \left[\boldsymbol{\varepsilon}_{tk} \frac{\partial \varphi(\eta_k)}{\partial \eta_k} + (\boldsymbol{\varepsilon}_{0k} - \boldsymbol{\varepsilon}_{00}) \frac{\partial \varphi(\eta_k)}{\partial \eta_k} + (\lambda_k - \lambda_0) \frac{\partial \varphi(\eta_k)}{\partial \eta_k} : \boldsymbol{\sigma} \right] \quad (14)$$

The constants Z_{ij} are estimated with the help of Eq.(14). It can be noted that the free energy density is represented in stress-temperature space. In this way the direct use of the deformation gradient associated with the phase transformation is avoided. As a consequence, the analysis in stress space without explicitly addressing the stress-strain hysteresis becomes convenient. On the other hand, if the atomic reordering during transformation is expressed in terms of continuum deformation gradient $\mathbf{F} = \nabla \mathbf{u}(\mathbf{x})$, as is usually done [15], then the Green strain tensor for the two different frameworks can be related as

$$\frac{1}{2} (\mathbf{F}^T \mathbf{F} - I) = -\frac{\partial G}{\partial \sigma} = \boldsymbol{\varepsilon}_{elt} + \sum_{k=1}^N \boldsymbol{\varepsilon}_{tk} \varphi(\eta_k) \quad (15)$$

where $\boldsymbol{\varepsilon}_{elt}$ is the linear elastic part of the Green strain in the present model, i.e. from the right hand side of Eq.(15). The general argument for microstructural evolution [1] can be formulated: for a compatible microstructure that minimizes $G(\mathbf{F}) \subset K$ at the austenitic well, where K is the space of sequence of energy wells, it follows from the Cauchy-Born hypothesis that

$$\hat{\mathbf{e}}^m = \mathbf{F}^m \mathbf{U}_m \mathbf{F}^a \hat{\mathbf{e}}^o = \mathbf{F} \hat{\mathbf{e}}^o \quad (16)$$

where $\hat{\mathbf{e}}^m$ and $\hat{\mathbf{e}}^o$ are the lattice vectors for the m th variant of M and the undeformed A lattice vector, respectively. At zero stress, $\mathbf{F}^m = \mathbf{F}^a = \mathbf{R}$ becomes a rigid rotation. \mathbf{U}_m is the Bain matrix or the *transformation matrix* [15]. At the onset of spontaneous transformations, we can drop the elastic strain part in Eq.(15) and relate to Eq.(16) to provide an analytical framework to the problem of microstructure formation. Further details on the compatibility conditions in terms of the deformation gradient across the $A-M_k$ or twin interfaces (domain walls) can be resolved numerically at macroscopic length scales using this approach. We skip the details and focus our discussion on the computational aspects of the problem.

We have so far described the phase transformation by the order parameters $(\eta_k, k = 1, \dots, N)$. The physical implication is that the statistical ensemble of the order variables (the state of the constituent atoms) is expressed in a spatially homogenized sense where their correlation in the atomistic length scale is approximated by ensemble average.

The transformation kinetics can be written in the form of Ginzburg-Landau equation, where the coefficients in the first-order system are related to the ensemble aggregate of the atomistic reordering. Most interestingly, this kinetics plays the main role in controlling the coupling between the thermodynamic conservation law and the momentum balance. In our study, we have analyzed the governing system of partial differential equations (PDEs) obtained from the present model, and these PDEs dynamically switch their characteristics at the onset of transformation spanned over a significantly smaller time-scales compared to the time scales of the SMA device dynamics. It poses one of the most challenging problems in computational mathematics. It has been suggested in [8] that one way to avoid the inherent ill-posedness of the computational version of the energy minimization problem is to relax the error-estimates. In our study, the additional difficulty is the fast time scale at which the transformation occurs. One possibility is to develop a singular perturbation scheme within the finite element framework. In the following sections we focus our discussion on developing a discrete version of the problem. Resolving the temporal events at the atomistic scale remains outside the scope of the present paper.

We first write the governing system of partial differential equations as follows. The transformation kinetics is represented by the Ginzburg-Landau equation

$$\frac{\partial \eta_k}{\partial t} = -\sum_{k=1}^N \mathbf{L}_{kp} \left[\frac{\partial G}{\partial \eta_p} + \boldsymbol{\beta}_p : \nabla \nabla \eta_p \right] + \theta_k, \quad (17)$$

where \mathbf{L}_{kp} are positive definite kinetic coefficients, $\boldsymbol{\beta}_p$ are positive definite second rank tensor. θ_k is the thermal fluctuation satisfying the dissipation-fluctuation theorem arising in context of non-equilibrium thermodynamics. While Eq.(17) governs the evolution process, the macroscopic energy conservation law is governed by the heat conduction

$$\frac{\partial H}{\partial t} - \nabla \cdot (\boldsymbol{\sigma} \cdot \frac{\partial \mathbf{u}}{\partial t} + \mathbf{q}) = h_0 \quad (18)$$

where $H = G + W$, W is the potential energy per unit volume due to stress work density. Note that the energy associated to the jump in entropy during phase transformation is included within our Gibbs free energy representation. Therefore, H is equivalent to the Helmholtz free energy per unit reference volume plus the stress work. h_0 is the point-wise heat source, $\mathbf{q} = -\kappa \nabla \theta - \alpha \kappa' \nabla \dot{\theta}$ is the heat flux vector with first order relaxation. The momentum balance equation is given by

$$\rho \frac{\partial^2 \mathbf{u}}{\partial t^2} = \nabla \cdot \boldsymbol{\sigma} + \mathbf{p} \quad (19)$$

where ρ is the mass density, \mathbf{u} is the displacement vector and \mathbf{p} is the body force. In a quasi-static sense, the framework so far requires the minimization of the total energy as discussed in [1,15,22], that is to seek

$$\text{Min} \left[\int_{\Omega} G(\nabla \mathbf{u}(\mathbf{x}), \theta) d\mathbf{x} + \text{Loading device energy} \right]$$

in the deformed configuration of the lattice. But in our approach, we are dealing with the physics-based model of the multivariant phase transformations via the additional set of variables $\eta_k, k = 1, \dots, N$. Thus, in our model, the order parameters, which appear in the Green strain tensor and related to the macroscopic deformation as in Eq.(15), provide the most important connection between the transformation of the lattice vector and the continuum deformation gradient. Moreover, the entire description in the present approach is with respect to the reference coordinate system, where η_k represents the atomic reordering (which is indeed the case) rather than quasi-steady nature of finite deformation of the lattice as generally assumed. We note also that in our model we have to deal with jump conditions (Eqs.(8) and (9)) which require special care during numerical implementation. Numerical discretization has been carried out using a 8-node/element 7 d.o.f./node hexahedral element.

NUMERICAL SIMULATION

Here we report simulated thermally-induced cubic to tetragonal transformations in NiAl under harmonic flux q applied normally to the cross-sectional surface of a bar. $u = 0, \theta = \theta_0$ are assumed at the opposite end ($x=0$). The surfaces are assumed to be in ambient environment ($\theta = 298K$), $\theta_e = 215K$.

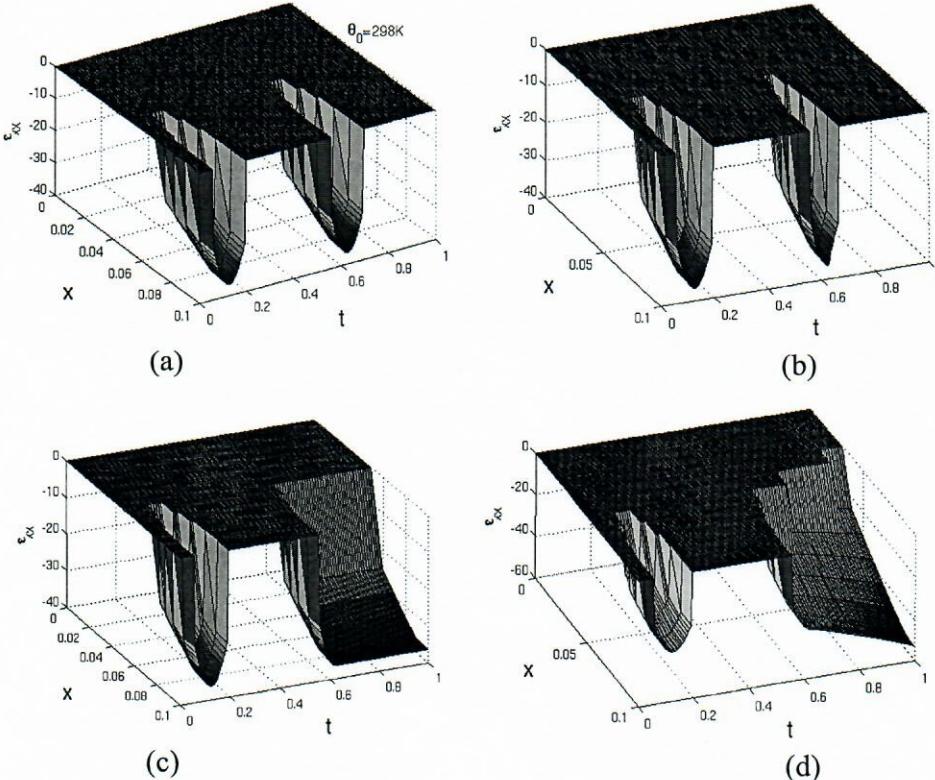


Fig. 1. Thermally induced % strain histories due to (a) harmonic flux (b) harmonic flux switched to exponential decay (c) harmonic flux switched to constant and (d) harmonic flux switched to exponential increase.

Fig. 1(a) shows the time history of longitudinal strain ε_{xx} over the length of the bar showing the evolution of the martensite along x . Figs.(b)-(d) respectively show the strain histories due to the evolution of martensite along x , when the harmonic flux is switched to exponentially decaying, constant, and exponentially increasing fluxes.

CONCLUSIONS

A Landau-Ginzburg free energy model and a computational finite-element-based framework to analyze the multivariant phase transformations have been developed and applied to study thermally induced phase transformations. Representative computational results on the evolution of martensitic variants under varying thermal fluxes have been presented. The developed model and its computational implementation is a valuable tool in a better understanding of the thermomechanical characteristics of SMA devices and their optimization.

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