NEW METHOD FOR EVALUATING GIBBS FREE ENERGY IN SHAPE MEMORY ALLOYS

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Abstract

Smart materials or shape memory alloys have a wide range of important applications in our time, They are used in the medical field, the main parts that are used in surgical operations by computers made of these materials, because of unique properties of this kind of alloys where it can automatically retrieve forms and accurately with the change of ambient temperature or with the change of pressure imposed on it or a magnetic field (or electric) Surrounding. Thus, it becomes possible for a specialist to give the desired shape during the development of the alloy in the vicinity of a temperature equal to the temperature of the human body and then pull this alloy from the oven then in room temperature it will have the ingot form would facilitate the introduction into the patient's body through the events of wound smaller than if the process was conducted through regular, As in modern operations to widen the arteries and directed by gravel from the kidneys eliminate malignant tumors and other surgical procedures other. As also used in manufacturing aircraft wings where the wing changes shape with changing temperatures of the upper atmosphere. Also used in the very small motor industry because of the ability of its molecules to interact with the surrounding magnetic field or by changing the pressure imposed on it.

A new way for the evaluation of the Gibbs free energy for the shape memory alloys at any point within the specimen at the end of the transformation is developed.

In contrast with our previous model , which is limited in practice to the whole specimen , this model is valid for length scales specified by the a, b, c for the parent and the emberio lattice and controlled by the transformation matrices.

حساب الطاقة الحرة لشبائك المواد الصلبة الذكية نظريا

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الخلاصة

تستحوذ المواد الصلبة الذكية على مجال واسع من التطبيقات المهمة في عصرنا هذا، فهي تستخدم في المجال الطبي حيث تصنع من هذه المواد الأجزاء الرئيسية التي تستخدم في مجـال العمليـات الجراحيـة

بالكمبيوتر وذلك بسبب خواصها الفريدة من نوعها حيث تقوم تلقائيا باسترجاع إشكالها وبدقه متناهية مـع تغير درجة حرارة المحيط أو مع تغير الضغط المسلط عليها أو المجال المغناطيسي (أو الكهربائي) المحيط بها، وبهذا يصبح بإمكان الأخصائي إعطاء الشكل المطلوب أثناء وضع السبيكة في محيط درجة حرارتـه مساوية لدرجة حرارة جسم الإنسان ومن ثم إخراج هذه السبيكة من الفرن وفي درجة حرارة الغرفة تعطى السبيكة شكل يسهل معه إدخالها إلى جسم المريض من خلال إحداث جرح اصغر مما لو أجريت العمليـة بالطرق الاعتيادية، كما في العمليات الحديثة لتوسيع الشرايين وإخراج الحصى من الكلى استئصال الأورام الخبيثة وغيرها من العمليات الجراحية الأخرى. كما وتستخدم أيضا في مجال صناعة أجنحـة الطـائرات حيث يغير الجناح شكله مع تغير درجات حرارة طبقات الجو العليا. وأيضا تستخدم فـي مجـال صـناعة المحركات الصغيرة جدا بسبب قدرة جزيئاتها على التفاعل مع المجال المغناطيسي المحيط أو مـن خـلال تغيير الضغط المسلط عليها.

تم في هذا البحث استخدام طريقة رياضيه جديدة لحساب الطاقة اللازمة أو المتولدة ضمن أي نقطـة علـى الشبيكة في نهاية التحول لهذه المواد من طور إلى آخر و بالمقارنة مع النموذج السابق الذي يحسب الطاقة اللازمة لتحول العينة بشكل كامل هذا النموذج يصح لحساب الطاقة المتولدة أثناء التحول في أي نقطة على سبيكة تلك البلورة باستخدام أطوال الأضلاع المكونة لشبيكة تلك البلورة a, b, c بالنسبة لشبيكة الطـور الأول والطور المتحول إليه من خلال حساب مصفوفات التحول.

1- Introduction

Shape memory alloys (SMA) (also referred to as ***smart materials ,intelligent materials ,adaptive materials*** or ***structronic materials*****)**[1] are materials which have increasing range of engineering , aerospace, and biomedical applications. ***Smart materials can act as transducers and physical parameters converters***. Some of them can convert energy from one form into another such as mechanical energy (stress) into a change in length (strain) or electrical energy (supplied electric field) to electric displacement. Others convert the thermal energy (change in temperature) into a change in entropy and the magnetic energy (supplied magnetic field) to a magnetic flux. The chemical energy (change in concentration) may also be converted into a volumetric flux by SMA's.

Smart materials undergo solid to solid phase transformations from Austenite to Martensite .Mostly Austenite has cubic lattice structure while Martensite phases have lower symmetries which may be (trigonal, tetragonal, orthorhombic, or monoclinic) lattice structure. A number of theoretical and experimental studies has been concerned with this kind of transformation [2-19].

A continuum theory which explains the transformation strain in the microscopic scale, was described by Bhattacharya[3]. The change in symmetry (described by the changes in lattice vectors) are its only input. Also his theory discusses the energy of a specimen subjected to a deformation gradient ∇y at a temperature θ, according to the following equation:)

$$\psi = \int \Phi(\nabla y, \theta) dV \quad \text{--------(1)}$$

where Φ is called free energy density, and it means the stored energy density which depends on the lattice local distortion and measured by the deformation gradient and the temperature. Liang et. al. [8] constructed the transformation strain matrices for the NiTi SMA with a_o=0.3015 nm for the Cubic (Austenite) lattice while for Martensite monoclinic unit cell a=0.289 nm, b=0.412 nm, c=0.4622nm, and e=96.8^o, where a, b, and c are the lattice parameters and e is the angle between (b) and (c) vectors of Martensite. Hane and Shield [9] also studied the microstructure of the transformation from Cubic to Monoclinic (in NiTi) describing the transition by a uniform expansion, shuffling of atoms and shear. In this sense, they expressed the gradient of deformation for the Cubic to Monoclinic transformation in NiTi as:

$$\nabla \gamma = \begin{pmatrix} \tau & \chi \cos e & 0 \\ 0 & \chi \sin e & 0 \\ 0 & 0 & w \end{pmatrix} \quad \text{------(2)}$$

where τ, w, χ are the transformation stretches given by

$$\tau = a / a_o, w = b / \sqrt{2} a_\circ, \chi = c / \sqrt{2} a_\circ \quad \text{---(3)}$$

Lue et.al. [6] suggested a micromechanical model based on minimization of Gibbs free energy for single crystal TiNiCu. They calculated the free energy difference between Austenite and Martensite phase and minimized it with respect to spherical orientation angles (e, ϕ). They also put the strain matrices for all the possible variants for the Ti 4at % Ni -10 at % Cu ,SMA's. Reynolds [15] and Kloucek and Reynolds 2003[16] provided a mathematical model to describe the thermodynamic behavior Of SMA using the principles of mass conservation which is :

$$\rho_o^{\bullet} = 0$$

And the conservation of linear momentum

$$\partial_t(\rho_o u^{\bullet}) = Div(\sigma) + \rho_o B ------- (4)$$

where u is the deformation, B represent external body force ,σ is the stress tensor. They solved the following partial differential equations:

$$\rho_o \sigma = Div(\rho_o \partial_\gamma \psi + A\gamma^{\bullet}) + \rho_o B --(5)$$

$$\rho_o C_p \theta^{\bullet} = \rho_o \theta \partial_{\gamma\theta}{}^2 \psi . \gamma^o + Tr(\gamma^o A \gamma^o)$$

$$+ Div(K \det(\gamma) \nabla \theta) + \rho_o r ------ (6)$$

Where A is the symmetric viscous anisotropy matrix.

After solving these two equations they reached to the elastic strain energy formula:

$$\psi(\gamma,\theta) = W_M(\gamma) C_M(\theta) +$$

$$W_C(\gamma) C_C(\theta) + W_A(\gamma) C_A(\theta) --------- (7)$$

$$\psi(\gamma,\theta) = C_A(\theta) W_A(\gamma) + C_M(\theta) W_M(\gamma)$$

$$+ C_C(\theta) W_C(\gamma) + C_p(\theta - \theta \ln\theta) ------ (8)$$

Where $\psi(\gamma,\theta)$ is Helemoholtze free energy, W_M is the energy of Martensite, W_A is the energy of Austenite.

The prediction of the thermomechanical behavior of (SMA)[2,3,4,5,6,20], and magnetically actuated (SMA's) [18,,21,22,23] have attracted special attention ,in this respect modeling of interatomic scale is of interest to understand the behaviour of these alloys and its phase stability .

The mathematical framework for modeling phase transformations in the SMA's is based on Gibbs free energy $\Delta \boldsymbol{G}$ absorbed by or emitted from a spacemen subjected to a deformation gradient $\Delta \boldsymbol{\gamma}$ at a temperature $\boldsymbol{\theta}$. According to equation (1) we can say that

$$\nabla G = \int \Phi(\nabla y, \theta) dV --------- (9)$$

As it is cleared from equation (1) we have: $\Phi = \Phi(\gamma, \theta)$

A new model for the evaluation of the Gibbs free energy at any point at the specimen at the end of the transformation is developed, the new model based on our previous model in which we calculated the rate of change of the free energy per unit time ΔG during transformation between Austenite and Martensite. Where [24]:

$$\Delta G = \Delta\gamma(\frac{\sigma}{\rho_o}) - \Delta\theta \cdot C_P(\ln\theta + 1) \text{ --(10)}$$

Where γ is the deformation gradient

$\sigma = \sigma(\gamma, \theta)$ the stress tensor /area (Pa/m^2)

ρ_o the mass density (Kg/m^3)

$\Delta\theta$ the temperature difference (K^o)

C_P the heat capacity

θ the temperature (K^o)

In contrast with our previous model ,in which we calculate Gibbs free energy for the whole transformation from the beginning to the end ,this new model is valid in calculating Φ the stored energy density for a length scales specified by the a, b, c for the lattice and controlled by the transformation matrices. By minimizing ΔG from equation (9), we minimize the bulk energy of the considered structure, as a function of the deformation tensor, and temperature $\Delta G(\gamma, \theta)$, also one can apply this procedure at the measoscale specially when we know that equation (10), applied on both the macroscale and the measoscale.

Precise definition of the free energy is highly important task to account for the movement of the atoms rows with respect to the other without breaking any chemical bonds (dislocation movement).

In this paper we construct a mathematical model for the free energy of the phase transformation in SMA materials ,the free energy that depends on the local distortion in the lattice measured by the deformation gradient (γ) and the temperature (θ).

2- Mathematical Problem

Transitions between solid phases involve relatively large amounts of energy specially in the case of SMAs. These materials produce thermal energy when they are bent or subjected to a suitable stress. They also change their crystal shape under certain applied conditions. Through evaluating this shape change, one can

calculate the free energy density ,because the mathematical framework of modeling phase combination in shape memory materials is based on the solution of the variational problem with respect to a frame indifferential nonconvex free energy function $\Phi(\nabla\gamma,\theta)$, see equation (1).

By differentiating equation (9), an expression for the stored energy function can be obtained,.

3-Mathematical Tools

The elastic energy required to deform a lattice from its reference type identified by a bounded domain $\Omega(R^3)$ by $U:\Omega\to R^3$ is given as :

$$\Delta G=\int\Phi(DU)dX \text{ ----------(11)}$$

where U represents the deformation gradients:

$$DU=(\frac{\partial u^i}{\partial x^j})\text{----------(12)}$$

The energy change in an isothermal quasistatic deformation is characterized as follows:

Firstly, we suppose that $\Delta E\to\min$ [2]

where

$$\Delta E=\Delta W+\Delta\Phi\text{ ----------(13)}$$

ΔW is the total work increament supplied to the deformed body ,and $\Delta\Phi$ is the increament in potential energy of the loading device (assumed conserved).

The Cauchy-Born rule implies that the stored energy density is the energy per unit reference volume required to perform an affine deformation $x\to F_x$ [21, 24].

Also, suppose that this stored energy is invariant under rigid transformation (frame indifference) ,and under changes that correspond lattice invariant rotations [3].

$$\Phi(QF)=\Phi(F)\quad\forall\quad Q\in SO(3)$$

$$\Phi(FR)=\Phi(F)\quad\forall\ R\in P\in SO(3)$$

where P is the point group for the lattice which reflects the symmetry properties, So: It is convenient to normalize Φ such that ($\min\Phi=0$), then the set

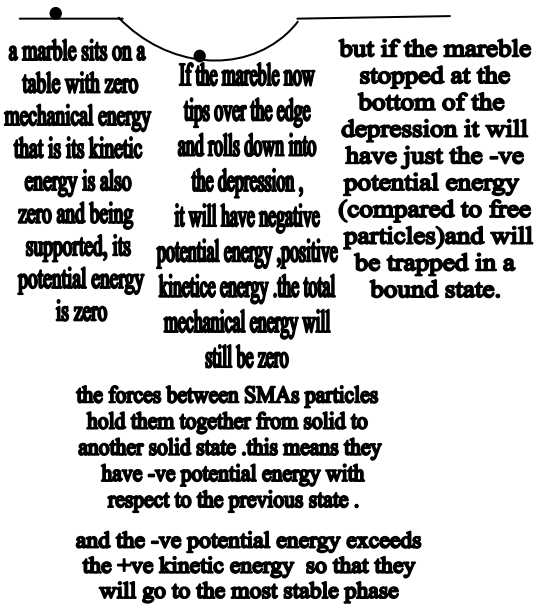
$$K=\{F:\Phi(F)=0\}$$

contains exactly the zero –energy affine deformation of the lattice.

Physically, it is essential to know the reason behind having $\Phi=0$.

This kind of transformation is diffusionless the lattice-distortive phase transformation, i.e, during the transformation the atoms of highly ordered crystal are rearranged in a coordinated manner leading to the formation of a new crystalline phase [24]. This might be attributed to the expectation that this kind of transformation is totally an energy-related process where the potential energy is treated as a negative quantity. An analogy is put forward when a mechanical system with gravitational potential energy and kinetic energy might be adequate for understanding the implications of the negative energy concept.

It seems logical to choose the zero potential energy such that the free particles at rest have zero energy and a bounded particle at rest has negative potential energy.



Then if $\Phi_{\min}=0$, Then the set k consists of one or several disjoint copies of $SO(3)$

$$K=SO(3)U1-------Um\text{ ----(14)}$$

Using equation 3

For cubic to monoclinic transformation ,the strain matrices [2]

$$U_1=\begin{pmatrix}\alpha&\delta&\in\\ \delta&\alpha&\in\\ \in&\in&\beta\end{pmatrix}---------(15a),$$

$$U_2=\begin{pmatrix}\alpha&\delta&-\in\\ \delta&\alpha&-\in\\ -\in&-\in&\beta\end{pmatrix}------(15b)$$

$$U_3=\begin{pmatrix}\alpha&-\delta&-\in\\ -\delta&\alpha&\in\\ -\in&\in&\beta\end{pmatrix}--------(15c),$$

$$U_4=\begin{pmatrix}\alpha & -\delta & \in\\ -\delta & \alpha & -\in\\ \in & -\in & \beta\end{pmatrix}---(15d)$$

$$U_5=\begin{pmatrix}\alpha & \in & \delta\\ \in & \beta & \in\\ \delta & \in & \alpha\end{pmatrix}----(15e),$$

$$U_6=\begin{pmatrix}\alpha & -\in & \delta\\ -\in & \beta & -\in\\ \delta & -\in & \alpha\end{pmatrix}----(15f)$$

$$U_7=\begin{pmatrix}\alpha & -\in & -\delta\\ -\in & \beta & \in\\ -\delta & \in & \alpha\end{pmatrix}----(15g),$$

$$U_8=\begin{pmatrix}\alpha & \in & -\delta\\ \in & \beta & -\in\\ -\delta & -\in & \alpha\end{pmatrix}---(15h)$$

$$U_9=\begin{pmatrix}\beta & \in & \in\\ \in & \alpha & \delta\\ \in & \delta & \alpha\end{pmatrix}----(15i),$$

$$U_{10}=\begin{pmatrix}\beta & -\in & -\in\\ -\in & \alpha & \delta\\ -\in & \delta & \alpha\end{pmatrix}----(15j)$$

$$U_{11}=\begin{pmatrix}\beta & -\in & \in\\ -\in & \alpha & -\delta\\ \in & -\delta & \alpha\end{pmatrix}---(15k),$$

$$U_{12}=\begin{pmatrix}\beta & \in & -\in\\ \in & \alpha & -\delta\\ -\in & -\delta & \alpha\end{pmatrix}---(15l)$$

Where α, β, ε, and δ can be written as follows [9]:

$$\alpha=\frac{1}{2}\frac{\chi(\chi+\tau\sin(e))}{\sqrt{\tau^2+\chi^2+2\tau\chi\sin(e)}}+w\text{-------(16)}$$

$$\beta=\frac{1}{2}\frac{\tau(\tau+\chi\sin(e))}{\sqrt{\tau^2+\chi^2+2\tau\chi\sin(e)}}\text{.----------(17)}$$

$$\varepsilon=\frac{1}{\sqrt{2}}\frac{\tau\chi\cos(e)}{\sqrt{\tau^2+\chi^2+2\tau\chi\sin(e)}}\text{--------(18)}$$

$$\delta=\frac{1}{2}\frac{\chi(\chi+\tau\sin(e))}{\sqrt{\tau^2+\chi^2+2\tau\chi\sin(e)}}-w\text{-----(19)}$$

By substituting equation(14) in (16), (17), (18), and (19) we get:

$$\alpha=\frac{1}{2}\frac{\frac{c^2}{2a_o}+\frac{ac}{\sqrt{2}a_o^{\,2}}\sin(e)}{\sqrt{\frac{a}{a_o^{\,2}}^{2}+\frac{c}{2a_o^{\,2}}^{2}+\frac{\sqrt{2}ac}{a_0^{\,2}}\sin(e)}}+\frac{b}{2\sqrt{2}a_o}\text{ -- (20)}$$

$$\beta=\frac{\frac{a^2}{2a_o}+\frac{ac}{\sqrt{2}a_o^{\,2}}\sin(e)}{\sqrt{\frac{a}{a_o^{\,2}}^{2}+\frac{c}{2a_o^{\,2}}^{2}+\frac{\sqrt{2}ac}{a_0^{\,2}}\sin(e)}}+\frac{b}{2\sqrt{2}a_o}\text{ ---.(21)}$$

$$\varepsilon=\frac{\frac{ac}{\sqrt{2}a_o^{\,2}}\cos(e)}{\sqrt{2\frac{a}{a_o^{\,2}}^{2}+\frac{c}{2a_o^{\,2}}^{2}+2\frac{\sqrt{2}ac}{a_0^{\,2}}\sin(e)}}\text{---------(22)}$$

$$\delta=\frac{\frac{c^2}{2a_o}+\frac{ac}{\sqrt{2}a_o^{\,2}}\sin(e)}{2\sqrt{\frac{a}{a_o^{\,2}}^{2}+\frac{c}{2a_o^{\,2}}^{2}+\frac{\sqrt{2}ac}{a_0^{\,2}}\sin(e)}}-\frac{b}{2\sqrt{2}a_o}\text{ ----.(23)}$$

If we take one row from any transformation matrix, for example, the first row which represents the forces acting along the x-axis

$$\sigma_x=(\frac{1}{2}\frac{\frac{c^2}{2a_o}+\frac{ac}{\sqrt{2}a_o^{\,2}}\sin(e)}{\sqrt{\frac{a}{a_o^{\,2}}^{2}+\frac{c}{2a_o^{\,2}}^{2}+\frac{\sqrt{2}ac}{a_0^{\,2}}\sin(e)}}+\frac{b}{2\sqrt{2}a_o})\hat{i}$$

$$+(\frac{\frac{c^2}{2a_o}+\frac{ac}{\sqrt{2}a_o^{\,2}}\sin(e)}{2\sqrt{\frac{a}{a_o^{\,2}}^{2}+\frac{c}{2a_o^{\,2}}^{2}+\frac{\sqrt{2}ac}{a_0^{\,2}}\sin(e)}}-\frac{b}{2\sqrt{2}a_o})\hat{j}\text{---(24)}$$

$$+(\frac{\frac{ac}{\sqrt{2}a_o^{\,2}}\cos(e)}{\sqrt{2\frac{a}{a_o^{\,2}}^{2}+\frac{c}{2a_o^{\,2}}^{2}+2\frac{\sqrt{2}ac}{a_0^{\,2}}\sin(e)}})\hat{k}$$

the first term represents the strain along the x-direction for compression or stretching (a_o) and to be transformed to (a), while the second and the third terms from the above equation are the strain forces applied to change the angle between a and b, and the angle between a and c respectively.

And to find the energy required to transform a_o to a we substitute σ_x in equation 8, and differentiate it with respect to x , to get ϕ_x .and differentiate it with respect to y to get ϕ_y and ϕ_z .

Results

1- We can estimate the contributions of the energy by understanding the length variation and the fine geometry of the microstructure.
If we have the *a, b, c* for any lattice crystal undergoing martensitic transformation, and the kind of this transformation, we can get Φ.
Where $\Phi = \phi_x + \phi_y + \phi_z + \phi_{angle}$ ----(25)
For example, in cubic to monoclinic transformation (for example NiTi alloy):

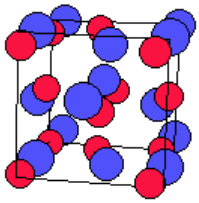


Figure a: cubic NiTi lattice (Austenite phase)

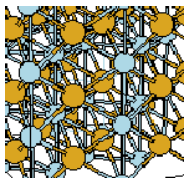


Figure b: monoclinic NiTi lattice (Martensite phase).

$$\phi_x = 4.12\times10^{-21}\cdot A\cdot Z + \frac{3.616\times10^{-21}}{A}\cdot Z^2 + \frac{1.952\times10^{-21}}{A}\cdot X\cdot Z \text{ ----------(26)}$$

Where A is a constant and equal to :

$$A=\sqrt{1.1\times10^{7}\cdot a^2+4.54\times10^{-18}\cdot c^2+5.9\times10^{6}\cdot a\cdot c} \text{-(27)}$$

$$\phi_y = \text{A----------(28)}$$

$$\phi_z = 5.85\times10^{-20} + A - \frac{1.862*10^{-18}}{A^{3/2}}\cdot X^2 - \frac{4.123*10^{-21}}{A^{3/2}}\cdot X\cdot Z\cdot(9.07\times10^{-18}\cdot Z + 5.9\times10^{6}\cdot X) \text{ -(29)}$$

Now, assume that the effective force in compressing or stretching the parameter a along the x-axis is the force resulting from the x-direction.(we ignore the other)
And after simplifying the above equation for ϕ_x,and made the same calculation in the case of ϕ_y and ϕ_z we find that

$$\phi_x = (0.3141e-21)\,Joul$$

2- in addition to that there is ϕ_{angle} which is the energy required to change the angle of the lattice.
3- by calculating the energy of the transformation from the simple consepts in physics (throw calculating the number of lattices in one gram . estimating that the total energy for NiTi transformation is 6 Joul/gram (experimentally) [24]. we find that

$$\phi = 4.24e-21\,Joul$$

Conclusions

1- The stored energy density function can be described as follows:

$$\Phi = \phi_x + \phi_y + \phi_z + \phi_{angle}$$

2- ϕ_x is the energy required to compress a_0 to be a along the x-axis and ϕ_y is the energy required stretches a_0 to reach b along the y-axis. ϕ_z is the energy required also stretches a_0 to reach c along the z-axis.

Here, if we apply the values of a, b, and c at any time during transformation of x, y, and z positions, the total energy required for or emitted from NiTi lattice at the end of the transformation may be deduced.

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